

# Response to Anonymous Referee #1

We thank the reviewer for the very helpful and detailed comments. We greatly appreciate the time she/he took in making such a detailed review of our paper, and we believe that the manuscript has been substantially improved as a result of these suggestions. Comments are summarized and responses are in italics below.

## 1. Limited reproducibility

Following the ACP guidelines, “A paper should contain sufficient detail and references to public sources of information to permit the author's peers to repeat the work” and “the data should be held in persistent public repositories”.

The paper does not fulfill either of those requirements. There is no information as to where the observational data can be accessed. There are no pointers to the source code of the programs used to obtain the results. The model input parameters are not described in sufficient detail to reproduce the simulations independently.

The numerical model used is said to be described in:

- Luo et al. 2003, GRL 30: “Dehydration potential of ultrathin clouds at the tropical tropopause”,
- Hoyle et al. 2005, JAS 62: “The origin of high ice crystal number densities in cirrus clouds”,
- Hoyle et al. 2013, ACP 13: “Heterogeneous formation of polar stratospheric clouds...”.

The first reference should probably read “Luo et al. 2003, JGR 108” but anyhow none of the two papers contain formulation of the model presented in a way allowing result reproducibility (i.e. governing equations, numerical schemes). The second reference also lacks full model formulation as it mentions that “The details of the model are given in (Luo et al. 2003)”. The third paper does feature a full-length section on the model formulation, but as the titles of all mentioned papers suggest, it is focused on ice microphysics, and contributes little to the reproducibility of the presented study.

*As mentioned in the author's response on 30 September 2014, we are unable to place a copy of the model online (the author of the original model version is not happy to do so).*

*Nevertheless, an interested reader may contact Beiping Luo for a copy of the model if they wish to reproduce the results. A statement to this effect has now been added to the manuscript text.*

*The current model operates in the same way as described in the references above, with the exception that solid phases (e.g. ice) cannot form, and that the subsaturated growth of the aerosol is governed by the kappa-coehler equation (this difference is now mentioned in the paper). The kinetic uptake of water from the gas phase is done in a very standard way, and we now direct the interested reader to chapter 12 of Seinfeld and Pandis 2006, which describes the approach used in the model, and provides all the equations necessary to re-create this part of the model.*

*The meteorological data is provided by MeteoSwiss such that we are not permitted to make it publicly accessible. Nevertheless, all other data (such as aerosol parameters measured by the PSI) are available to the interested public by writing an e-mail to the contact author.*

*We corrected the reference from Luo et al. 2003, GRL 30 to Luo, B. P., C. Voigt, S. Fueglistaler, and T. Peter, 2003: Extreme NAT supersaturations in mountain wave ice PSCs: A clue to NAT formation. J. Geophys. Res., 108, 4441, doi:10.1029/2002JD003104.*

Calculation of supersaturation in the model is central to the presented study. Yet, the given sources do not provide information on:

- how the supersaturation budget is calculated in the parcel (with prescribed temperature evolution and the latent heat release not accounted for);
- the model timestep choice and integration method (how it copes with the stiffness of the drop growth equations? how it copes with the timestep requirement for simulating the small-scale fluctuations with frequencies up to 20Hz?);  
*we have added the following text to the model description section “At  $S < 0.99$ , the model time step is one second, and at  $S \geq 0.99$ , it is calculated such that the water content of the droplet can change by no more than 2% per time step.”*
- the form of drop growth equation used (are the latent heat effects accounted for here? how the molecular/ continuum regime transition is accounted for?).  
*This has been answered in the response to questions above, and the appropriate text has been added to the model description section.*

*We have added the following text to the model description section: "After the droplet growth is calculated, the total water content of the droplets is deducted from the total water content of the air parcel, and the saturation of the gas phase is re-calculated. The total water content (i.e. gas plus condensed phase water) is conserved throughout the simulation." And "The latent heat release due to condensation of water is accounted for implicitly in the use of a moist adiabatic lapse rate to calculate the temperature trajectory along which the model is run."*

None of those papers refer to the  $\kappa$ -Köhler parameterisation used in the present study. Few model constants are explicitly mentioned in all four papers combined, and some of those assumptions should be discussed in the context of a sensitivity study (e.g. the mass accommodation coefficient of unity as mentioned in [Hoyle et al. 2013](#)). I do not mean that the model formulation/documentation should be part of this very paper. It can be published, for instance, in an e-print repository like arXiv, but giving the readers access to it is essential to *permit the author's peers to repeat the work*. Let me underline that, reproducibility principles aside, access to the model documentation and code would bring answers to several question listed below.

*As mentioned above the model ZOMM so far was only used for ice microphysics, thus, the  $\kappa$ -Köhler parameterisation was implemented in the model for this study. We described this implementation in Sect. 2.2.1 with reference to [Petters and Kreidenweis \(2007\)](#).*

## 2. Lack of proper context

The paper reports on the sensitivity of cloud droplet activation process, in particular the sensitivity to the small-scale fluctuations of vertical velocity and temperature. This is a widely studied topic and the paper clearly lacks references to other studies discussing analogous tools, methodologies and results, e.g.:

- [Clark and Hall 1979](#) with remarks on the deficiencies of the approach to "simulate the effects of turbulent mixing by applying a highly time-dependent  $w$  to a Lagrangian parcel calculation of condensation growth";
- [Kulmala et al. 1997](#) with investigation on "the effects of fluctuations of saturation ratio on droplet (cloud condensation nuclei) growth by stochastic approach employing an advanced growth model for cloud droplets";
- [Feingold 2003](#) where "an adiabatic parcel model has been used as a tool to investigate the relative sensitivity of the radiatively important cloud drop effective radius to [...] parameters such as updraft velocity ...";
- [Lance et al., 2004](#) where "a detailed numerical cloud parcel model [...] is used to] determine a most probable size distribution and updraft velocity for polluted and clean conditions of cloud formation";
- [Chuang 2006](#) - a study on "Sensitivity of cloud condensation nuclei activation processes to kinetic parameters" that also uses an adiabatic parcel model;
- [Ditas et al. 2012](#) where "sensitivity of the supersaturation on observed vertical wind velocity fluctuations is investigated with the help of a detailed cloud microphysical model";
- [Partridge et al. 2012](#) with discussion on local vs. global sensitivity analyses and the applicability of inverse modelling approach to droplet formation sensitivity studies.

As of now, the discussion of the methodology and results is left without proper context. This also makes it hard for the reader to understand where the novelty of the presented results lies.

*Thank you for providing these references. The reviewer is correct in that the introduction did not provide a detailed enough discussion of previous work. We have included a discussion of all of these papers in the introduction (sizable section of text not reproduced here), as well as in the discussion of figure 5, which shows similar features to what was found in the studies of [Chuang](#), [Feingold](#) and [Partridge](#).*

## 3. Paper composition

There are numerous flaws in paper composition, for instance:

- the overlap with the [Hammer et al. 2014](#) paper published earlier this year in ACP is excessive:
  - most of section 2.1.1 "Measurement set-up" is composed of material from section 2.2 "Instrumentation" therein,  
*We rephrased the overlapping material accordingly.*

- the whole section 2.1.2 bears well too much similarity to section 3.4 (with the same title) from Hammer et al. 2014, ACP,  
*We added a sentence at the beginning of 2.1.2 to make it clear that this section was put from Hammer et al. (2014): "(This section is composed by a summary of section 3.4 from Hammer et al. (2014).)" and removed "see detailed explanation in Hammer et al., 2014)".*
- there are cases where the section contents clearly do not match the section titles:
  - first (and only) two paragraphs of section "2. Methods" are only related to "2.1 Observational data",  
*We added the first two paragraphs in "2. Methods" to "2.1 Observational data".*
  - some of the methodology of model initialisation is presented in the last two paragraphs of "2.1.1 Measurement setup",  
*We moved the last two paragraphs of "2.1.1 Measurement setup" to a 2.2.1. and rephrased it accordingly.*
  - section "2.2.1 Box model description (ZOMM)" contains a paragraph on filtering the observational data for entertainment;  
*We moved the paragraph "Cloud periods that exhibited evidence of substantial entrainment or mixing were not included in the analysis. Such clouds were detected by analysing the activated fraction of the aerosol particles as a function of aerosol size. Periods where the largest size bins were not at least 90% activated were excluded. This is the same procedure to that used by Hammer et al. (2014)." to a new Section "Defined cloud periods" after 2.1.1.*
- there are repetitions in the text, e.g.:
  - definition of  $SS_{peak}$  is given thrice:
    - 1: *The highest supersaturation that a particle experiences for a sufficiently long time to grow to a stable cloud droplet is defined as the effective peak supersaturation*
    - 2.3.1: *The effective peak supersaturation ( $SS_{peak}$ ) is the highest saturation encountered within an air parcel, which leads to activation of aerosol*
    - 2.3.1: *the  $SS_{peak}$  is defined as the highest supersaturation that a particle experiences for a sufficiently long time to grow to a stable cloud droplet**We removed the subsequent definitions after Section 1 and rephrased the sentences accordingly.*
  - the very same sentence "Previous studies have found that a high  $SS_{peak}$  can be caused by ..." is used to begin subsections 2.3.3 and 3.2;  
*We rephrased the sentence beginning the subsection 3.2 as follow: "According to previous studies, low  $SS_{peak}$  can be caused by small updraft velocity or a large number of potential CCN. Conversely, a high  $SS_{peak}$  can be caused by a high updraft velocity or a low number of potential CCN (see Sect. 2.3.3)."*
  - almost the same wording is used in section 3.3.2 and in the conclusions: "combinations of amplitudes and frequencies ... small-scale fluctuations in the vicinity of the JFJ"  
*We rephrased the wording in the conclusions as follow: "Furthermore, small-scale fluctuations in the vicinity of the Jungfraujoch were simulated based on several sinus functions with combinations of amplitudes and frequencies."*
- some statements seem incoherent (i.e. need rephrasing):
  - **abstract:** "It was found that the updraft velocity, defining the cooling rate of an air parcel, is the parameter with the largest influence on  $SS_{peak}$ "  
*We rephrased it: "The updraft velocity, which defines the cooling rate of an air parcel, was found to have the greatest influence on  $SS_{peak}$ ."*
  - **conclusions:** "On average small-scale variations are raising the  $SS_{peak}$  values to a larger extent than the other investigated parameters in this study"  
*Here the connection to the next sentence was missing. Therefore we rephrased it as: "On average small-scale variations of temperature raise the  $SS_{peak}$  values to a larger extent than the other investigated parameters in this study:"*
  - "effect of  $SS_{peak}$  on updraft velocity" & "influence that the vertical wind potentially has on the  $SS_{peak}$ "  
*We replaced "potentially" with "most likely".*
- there are numerous vague/ambiguous/unclear statements:
  - "... turbulence applied to a small linear cooling rate" (what does it mean to apply turbulence?)  
*we replaced turbulence with "small-scale temperature fluctuations"*
  - "... (median dry activation of CLACE2011)"  
*There was a word missing, we added: "median dry activation diameter of CLACE2011"*

- "... corresponding dew point temperature of the LWC" (suggests that LWC has a temperature)  
*We rephrased it as follow: "Assuming all the water is in vapour phase, the dew point temperature, was calculated via the ideal gas law and the Clausius-Clapeyron equation".*
- "... temperature and the corresponding pressure trajectory" (what is a pressure trajectory?)  
*We replaced all wordings "pressure trajectory" to "pressure along the air parcel trajectory".*
- "It is not feasible to measure the updraft velocity at the point of aerosol activation." (sometimes it is! please add "at JFJ")  
*We added "at the JFJ".*
- "To investigate the importance of the fluctuations to the decrease of temperature ..." (only decrease?)  
*It was meant to the mean decrease of temperature. However, we rephrased it to: "To investigate the influence of the small-scale fluctuations of  $SS_{peak}$  on the temperature ( $T_{turb}$ ) and pressure along the air parcel trajectory from the initialization point ...".*

### Further comments and questions

- In Hammer et al. 2014, the model-derived  $SS_{peak}$  is defined as simply "highest SS reached along the trajectory". Here, it is defined using the 2  $\mu\text{m}$  diameter threshold. I guess that the change was needed due to employment of the fast-varying input data resulting in supersaturation fluctuations - this should be explicitly mentioned and discussed.

*We added to Sect. 2.3.1.: "It is important to note that in Hammer et al. (2014) the definition of the  $SS_{peak}^{mod}$  simply was the "highest SS reached along the trajectory". The new definition described above is needed for investigating the small-scale fluctuation described in Sect. 3.3. The comparison  $SS_{peak}^{mod}$  obtained by the two definitions respectively was within 10%."*

- If I understand correctly, the model is stopped at different heights above cloud base (but always at the altitude of JFJ). Thus, the time the droplets are given to grow differs from simulation to simulation. Yet, the abovementioned  $SS_{peak}$  definition features a threshold on final droplet size (?) Isn't it incompatible?

*This seems to be a misunderstanding. It is true that the model is always stopped at the altitude of the JFJ which is always at different altitudes above the cloud base. However, the threshold is not defined on final droplet size but on the threshold of the droplet size at the point droplets grow fast (i.e. at point of activation). We therefore added: "The threshold is, therefore, defined on final droplet size but on the threshold of the droplet size at the point droplets grow fast (i.e. at point of activation)."*

- What does the model calculate between the starting point at  $RH=90\%$  and the point of  $RH=99\%$  at which the equilibrium assumption is lifted?

*Between  $RH=90\%$  and  $99\%$ , the model calculates the water content of the aerosols, assuming that they are in equilibrium with the gas phase. It does this using the k-Koehler equation, as described in the text immediately above equation 2. We have slightly modified the text to be clearer that we mean that the water content is calculated when we say the growth is calculated.*

- Even though very small-scale fluctuations of air thermodynamic properties are considered, all droplets in the model are exposed to the same conditions. Worth mentioning/discussing.

*We added in Sect. 3.3.1: "It was assumed that each particle experienced the same real-time fluctuations."*

- The Hammer et al. 2014 paper features an error estimate of the measurement-derived  $SS_{peak}$  of  $> \pm 30\%$ . Why not mention it in sections 3.2 and 3.3 when discussing sensitivity of model-predicted  $SS_{peak}$  values.

*We agree that it is important to note the error estimate of measurement-derived  $SS_{peak}$  and therefore added in Sect. 2.3.1: "A relative uncertainty of about  $\pm 30\%$  was estimated for  $SS_{peak}$ "*

- Why not give the reader a hint on the uncertainty of the model predictions with respect to such parameters as the timestep, bin layout, bin number and the debated values of constants (e.g. mass accommodation coefficient).

*A description of the limitations on the model time step has been added, as described above. The following text has been added to the model description section: "The aerosol size observed in a single SMPS measurement has an uncertainty of 10% (Wiedensohler et al., 2012); however the input distributions used in the basic model simulations consist of median size distributions taken over the CLACE 2011 campaign. The bin resolution used in the model is the same as that measured by the SMPS. Any uncertainties in the model calculation resulting from the resolution of the bin sizes or the aerosol size distribution would be much smaller than the differences in simulated peak supersaturation caused by varying the number and size of the aerosols, as is done in Fig 6.*

- In the paper, the parcel model is fed with a prescribed temperature profile instead of an adiabatic one that results from the simulated droplet growth. In my understanding, the only reasons to do so would be that an actual temperature profile is accurately known or that the intended profile differs significantly from an adiabatic one. Here, the profile was not measured, and instead an approximate one is used. Why?

*The background trajectory (i.e. without the fluctuations) is essentially an adiabatic trajectory, with the cooling rate corresponding to an average rate which is present when latent heat is released by the droplets. The simulations were done in this way as the model does not contain a calculation of latent heat release from growing droplets.*

- As a side note to the above point: in Hammer et al. 2014, the adiabatic lapse rate is assumed to be  $0.6 \text{ K (100 m)}^{-1}$ , while here the value of  $0.65$  is used. Why?

*This was simply an arbitrary difference in the number of significant figures used for the value in the different studies. We agree that using  $0.6\text{K}$  here too would have been more consistent, however the difference of  $0.05\text{K}$  will not influence the results in a significant way.*

- The model used features ice microphysics. I assume (although it is not said explicitly in the paper) that ice microphysics was turned off for the presented simulations as the whole discussion relates liquid clouds. Yet, the simulation parameters cover negative cloud-base temperatures. Would the model predict ice nucleation if it was turned on in the model?

*It's correct that the ice microphysics was turned off for the presented simulations. This was because 1) The current ice parameterisation is not tested for warm, mixed phase clouds, and 2) We specifically excluded cases where partial glaciation had occurred from the simulations by only using input data from cloud periods where the activated fraction was greater than  $0.9$  above the activation threshold diameter. We now state explicitly in the paper that we did not include ice formation in the simulations.*

- In section 2, the two prevailing wind directions are mentioned, while it seems that beginning from section 2.1.2 only the NW advection is considered - why?

*That's correct. Apparently, we did not explicitly state that. The reason that we only considered clouds from NW wind direction is that there are relatively few measurement points when southeast wind was present, and also because the clouds coming from the northwest are mostly found to be formed locally by rapid updraughts, in contrast to the clouds from the south, which are often stratus, which has been advected from further away. This makes the clouds from the northwest more suitable for our study. We added: "In here, only clouds reaching the JFJ with NW wind directions are considered. Relatively few measurement points when when SE wind was present, and also because the clouds coming from the NW are mostly found to be formed locally by rapid updraughts, in contrast to the clouds from the south, which are often stratus, which has been advected from further away. This makes the clouds from the NW more suitable for our study."*

- Figure 3 (Now Fig. 4) includes cloud-period labelling which is not used elsewhere in the paper.

*That's true; however, we thought that this is better than showing a broken time series. We hope that this is acceptable. We have now added text to the caption of Fig 4 to state that the lines and the labels serve to identify different cloud periods between which there are gaps of non-cloudy time.*

## **Response to Anonymous Referee #2**

**We thank the reviewer for the helpful and detailed comments. We appreciate the time she/he took in reviewing our paper. Comments are summarized and responses are in italics below.**

General comments:

The objective defined on Page 25969, Line 17–24 is relatively incremental and does not bring in the big picture. In particular, the manuscript starts with pointing out that it is important to understand aerosol-cloud interactions and to reduce uncertainty in aerosol radiative forcing, but how this work can be applied to address this overall goal is not clearly put. Why is it important to understand how effective peak supersaturation varies with those factors? Based on the finding, do the authors conclude that climate modellers should consider including these effects? Why and why not? I believe that the authors have very specific science questions in mind, but it would help readers to put this work into the context if those science questions could be better defined and described. The majority of the figures are about the ratio of effective peak supersaturation in “modified conditions” to that in control run. What is missing yet important question to ask is: do we need to care about a ratio of 1.1, 1.2, or we only need to worry if the ratio goes up to 5 or 10, for example? What are their consequences? It would be extremely valuable for both readers and the authors if a clear implication and path forward can be provided in the manuscript. Additionally, since this manuscript is about sensitivity, I feel that the authors need to provide stronger/more rigorous justifications about certain choice/threshold used in the paper. Generally, the manuscript can be written more concisely and can be structured a bit better. Quite a few bits are disorganised; some bits of text are duplicated and interrupted the flow. I would suggest that the authors take another careful look at the manuscript and reorganise some awkward paragraphs that seem to be misplaced somehow.

*We reorganized the paragraphs as suggested in the specific comments.*

*We agree, that some information was not clearly put into the big picture: 1) why it is important to understand the variation of the effective peak supersaturation, 2) why the modellers should consider these effects, 3) what the importance of the range of the ratios says and 4) more rigorous justifications about certain choice/threshold used in the manuscript. Thus, we implemented this into the manuscript as follow:*

*To answer the other reviewer's question, we deleted the last paragraph in Sect. 1 but added to make 1) and 2) clearer: “To develop effective models it is important to know the influence of the variation of several key aerosol parameters influencing the cloud droplet formation. It has been pointed out by Boucher et al. (2013) and Spichtinger et al. (2008) that the main uncertainties in the aerosol radiative forcing are due to aerosol-cloud interaction dynamical factors such as turbulent strength and entrainment controlling the cloud condensation rate, and the key aerosol parameters such as aerosol number concentration and size distribution, and to a much lesser extent, the composition. I.e. the interplay of dynamics versus effects purely attributed to aerosols remains highly uncertain. Thus, in this study the influence of the variation the turbulent strength and the updraft velocity on the cloud activation is investigated using a cloud parcel model.”*

*To answer 3) we added in Sect. 4, Conclusions at the very end: “Summarizing, small-scale temperature fluctuations are revealed have the strongest potential effect on cloud formation processes . The variation of aerosol number concentration and hygroscopic properties had a lesser influence than the aerosol size.*

To 4):. The ratios chosen for the updraft velocity (2 and 5) were not properly discussed in the manuscript. The mean of the modelled updraft velocities is  $\sim 1 \text{ ms}^{-1}$  with a maximum value of  $5.6 \text{ ms}^{-1}$  and a minimum value of  $0.03 \text{ ms}^{-1}$ . The maximum value of updraft velocities deviating from the mean therefore is  $\sim 5$  (5 times 1). We added in Sect. 3.2: "The ratio 5 describes the maximum deviation from the mean value of  $w_{\text{act}}^{\text{mod}}$  and the ratio 2 is given from the 75th and 25th percentile of  $w_{\text{act}}^{\text{mod}}$ , which are about a factor of 2 from the mean value."

Specific comments:

1) Page 25970, Line 1–9: I would recommend reorganising this paragraph, because duplicated information is given in the 1st and this paragraph, and the flow and the connection with the previous paragraph are just not very good.

*We deleted the according paragraph and added some sentences of it to the 1<sup>st</sup> paragraph as follow: "(...Cherian et al.,2014;Dufresne et al.,2013;Levy et al.,2013). It has been pointed out by Boucher et al. (2013) that the main uncertainties in the aerosol radiative forcing are due to aerosol–cloud interaction dynamical factors such as turbulent strength and entrainment controlling the cloud condensation rate, and the key aerosol parameters such as aerosol number concentration and size distribution, and to a much lesser extent, the composition.*

*... effective peak supersaturation ( $SS_{\text{peak}}$ ; Hammer et al. 2014). Small-scale fluctuations in vertical velocity can alter the cooling rate of an air parcel and thereby also the corresponding  $SS_{\text{peak}}$ .*"

2) Page 25971–25972: It is OK to list/explain a number of measurements in 2.1.1 one by one, but it would be much better if certain connections and reasons behind these measurements can be given in this section, so readers can start linking these measurements with model input. For example, which measurements are exactly used as model input? Additionally, it wasn't clear why suddenly temperature, pressure trajectory, and the regime of 90% needed to be calculated. Readers could figure out eventually, but this kind of connection is sort of the authors' responsibility to make it clear.

*We added the following sentence at the beginning of section 2.1.1*

*A number of quantities measured at the JFJ were either used as model input directly, or were used to calculate model input parameters. These included the aerosol size distribution, the temperature and pressure, wind speed and direction, and the total water content of the air.*

*We also agree that the description why the regime of  $RH=90\%$  was calculated is a bit out of context. We deleted this paragraph as the wish from another reviewer:*

3) Page 25974, Line 2: Could the authors please explain why a 6-min time period is chosen? How sensitive is the overall result to the averaging time period?

*The 6-min period was chosen according to the duration of measuring one size distribution with the SMPS..The main influence this time resolution has on the results is on the number of points in the plots. A longer averaging period would have resulted in fewer model runs being performed, and thus probably also a smaller range of results simply due to the lower probability of capturing extreme values. There would be no systematic shift in the results though. We added an explanation about that to the according sentence:*

*"(...) given in six minute averages. The six minute periods were chosen according to the instrument with the lowest time resolution which is the SMPS instrument measuring the dry particle size distribution."*

4) Page 25975, Line 13–15: While this manuscript focuses on effective peak supersaturation, how to measure it is not mentioned until Page 25977 with a very short statement that refers to Hammer et al. (2014). I would recommend providing a brief review in Sect. 2.1.1, because this variable is the key of the manuscript!

*According to your suggestion we added a brief review of the effective peak supersaturation in Sect. "Effective Peak Supersaturation.: The  $SS_{\text{peak}}$  was retrieved as follow: 1) the activation threshold diameter was determined from the measurements of the total and interstitial number size distributions*

2) the aerosol hygroscopicity was obtained from the simultaneous CCNC measurements 3) the activation threshold diameter was combined with the aerosol hygroscopicity to infer the effective peak supersaturation. A relative uncertainty of about 30% was estimated for  $SS_{peak}$ . A detailed description how the  $SS_{peak}$  was estimated from the measurements performed at the JFJ can be found in Hammer et al. (2014).”

5) Page 25976, Sect. 2.2.2: I found this section is disorganised and it is quite hard to understand what the authors try to convey. Could the authors please consider using a schematic illustration or Figure 2 to explain/link Eq. (3) and a lot of variables in the text? Additionally, a very minor suggestion – it doesn’t mean anything for readers if the model run is #516 or #1. If this number has a specific meaning or important implication, perhaps the authors could clearly describe it. Otherwise, I would suggest removing the number to make the manuscript read better and more concise.

*We tried to link Figure 2 with Eq. (3): “This was done by superimposing a time series of temperature fluctuations measured at the JFJ upon the linear temperature trajectory along which the model was run (see black line in Fig. 2). The time series of fluctuations was chosen to be simply that, which was measured at the JFJ during the time taken for the air parcel to ascend from the point where the model was initialized (indicated in Fig. 2 with RH=90%), to the JFJ (indicated in Fig.2 with JFJ).”*

*There is no specific meaning for the model run Nr. Thus, we removed the number from Fig. 2 and have rewritten the last sentence in Sect. 2.1.1. as follow: “... for the model run detected at the JFJ on 8 August 2011 18:20UTC.”*

6) Page 25977, Page 3: Could the authors please explain why choosing to find the highest water vapour saturation which lead to droplets larger than 2 microns in diameter? Any physical basis for the choice of 2 microns?

*According to several cloud studies, e.g. Juranyi et al. (2011) and Henning et al. (2002), a diameter of 2 microns is a good threshold to distinguish hygroscopic grown particles from cloud droplets. We therefore added: “(...) larger than 2  $\mu\text{m}$  in diameter. In earlier studies it was found that a diameter of 2  $\mu\text{m}$  is a good threshold distinguishing the hygroscopic grown particles from activated cloud droplets (Juranyi et al., 2011; Henning et al., 2002)”*

7) Page 25977, Line 11: Similarly, Reasons for choosing 2%?

*This was a relatively arbitrary choice, balancing calculation time with accuracy. Reproducing the exact value would have been theoretically possible, but would have required impractical amounts of computer time. “..., which was considered to be sufficient for the determination of  $SS_{peak}$  values, without consuming impractical amounts of computer time.”*

8) Page 25977, Line 24: Could the authors please clarify “what” exactly is independent of  $w$  here?

*We rephrased the sentence to make it clearer as follow: “The aerosol-limited regime is characterized by a relatively high ratio of  $w/N_{CN}$ , by a high activated fraction of aerosol particles (larger than 90%) and the aerosol-limited regime is basically independent of  $w$ .”*

9) Page 25981, Line 1: Could the authors please describe what kind of  $w/N_{cn}$  range is here?

*We added: “(...) is lower. Thus, the ratio of  $w/N_{CN}$  at these low  $SS_{peak}$  values is relatively low (at about 0.003) and is increasing with an increase in  $SS_{peak}^{ref}$ . (up to about 0.03)”*

10) Page 25981, Line 5–7: I am afraid that I don’t understand what the authors mean.

*We rephrased to make it clearer: “In Sect. 2 it is described that the topography at the JFJ defines two main wind directions, NW and SE wind. As shown by Hammer 2014, the particle number concentration and size measured at the JFJ differs between these two wind directions, with more and*



larger particles being measured during SE wind conditions. The variability of number and size is smaller within data collected from a single wind direction than the difference between the two wind directions. Therefore, we test the influence of particle number and size by varying these parameters over a similar range as the difference between values measured during SE and NW wind conditions.

11) Page 25981, Line 23–38: I am not sure how “updrafts are generally smaller: and only the largest particles activate” explain “more pronounced effect at low effective peak supersaturation. Could the authors please elaborate on this a bit more?”

*We rephrased to make it clearer: “This can be explained by the fact, that changing the size of the particles, changes the minimum supersaturation at which the particles can activate. At low  $SS_{peak}^{ref}$ , updrafts are generally smaller (colour coding in Fig. 6), and only the largest particles activate. At these large size ranges, usually a low particle number concentration is present and therefore if the particles are smaller (larger)  $SS_{peak}$  will be higher (lower). At higher  $SS_{peak}^{ref}$ , where the updrafts are generally higher, the critical saturation of the largest particles plays less of a role in determining the  $SS_{peak}$ .”*

12) Page 25984, Line 4–6: Could the authors please provide proper metrics to support the evidence of “Improves the relationship”?

*We added: “... slightly improves the  $SS_{peak}^{fluc} - W_{mod}^{act}$  relationship at lower updraft velocities as can be seen in Fig. 9. At updrafts of  $0.1$  to  $5 \text{ ms}^{-1}$ , the  $SS_{fluc}^{peak}$  to  $W_{mod}^{act}$  relationship is improved slightly, with 44% of the points lying within the range of the 25-75th percentile of the measured values, compared with 40% when fluctuations are not included”*

13) Page 25996, Figure 6 and related text on Page 25981 and 25982: The way of writing could be misleading – one may thought that the changes in diameter AND in number concentration are made simultaneously, which I don't think is true. The authors may wish to consider rewriting it more precisely. Additionally, since the observations support higher number concentration AND larger particles, it would be interesting to demonstrate when these two factors are combined, how will effective peak supersaturation change?

*To make it clearer we added: “... (see Fig. 6). The effects of changing the particle number size distribution and the particle number concentration were investigated separately.”*

14) Figure 8: Could the authors please explain what causes the spread of the ratio at a given effective peak supersaturation? Why a factor of 5 and 10 is a reasonable choice for sensitivity test?

*We added the following to describe the spread of the ratio: “Nevertheless, there is also a spread of the ratio at a given  $SS_{peak}^{ref}$ . This is explained by the variable nature of the temperature fluctuations – at the point where aerosol activation occurs, the cooling rate will sometimes be greatly modified by the temperature fluctuation, in some cases it will be rather close to the average cooling rate. In the latter case, the  $SS_{peak}$  from the simulation including fluctuations will be close to the  $SS_{peak}$  calculated from the reference simulation ...”*

*We believe that the reason for the factor of 5 and 10 is already well described with the following sentence: “The factors 5 and 10 are resulting in a similar range of temperature amplitudes used for the sinus curve simulations described in Sect. 2.2.2.”*

15) The authors may wish to be consistent to use either “peak effective supersaturation” or “effective peak supersaturation” throughout the paper.

*We checked that and use “effective peak supersaturation” throughout the paper.*

### **Response to Anonymous Referee #3**

**We thank the reviewer for the helpful and detailed comments. We appreciate the time she/he took in reviewing our paper. Comments are summarized and responses are in italics below.**

**General comments:**

One of the biggest issues with this study is that it uses the box model to fine tune a calculated updraft velocity based on the observations, and then, as far as I understand, compares it to the effective supersaturation determined by the same box model using the same trajectory. It is therefore not surprising that the comparison with the model improves when the same model is also used to predict all the parameters. Of course we can still learn a lot from the model output, but the text should clearly state which parameters were derived from the model versus derived from basic atmospheric assumptions. This would allow the reader to understand the significance of the findings.

This lack of clarity is mostly because of two technical issues. The first is that this study relies heavily on the results from the authors' previous study, but it does not explain how all the parameters were calculated. This makes it difficult for the average reader to follow how an estimated updraft velocity is different from a model updraft velocity, for example. (Note - to me, both of these values are modelled, only one uses a much more sophisticated model than the other.) This is not appropriate for a stand-alone paper and can be easily remedied by additional text briefly, but thoroughly, describing the key parameters derived in the previous study and used in this one. The second technical issue is that the notation is difficult to follow and I found myself needing to make a table just to keep track of all the variations of the parameters. If the authors could provide a table that logically grouped the parameters based on their origins, this would greatly increase the readability of the manuscript. Based on this, I would recommend that this paper be accepted subject to the important, but minor, revisions stated above, and the specific comments below.

*We agree that it might be more useful to have the whole explanation of the parameters effective peak supersaturation and estimated updraft velocity in this manuscript. Therefore, we added the following descriptions:*

*to Section Effective peak supersaturation: "The  $SS_{peak}$  was retrieved from the measurements at the JFJ as follows: 1) the activation threshold diameter was determined from the measurements of the total and interstitial number size distributions 2) the aerosol hygroscopicity was obtained from the simultaneous CCNC measurements 3) the activation threshold diameter was combined with the aerosol hygroscopicity, and the estimated cloud base temperature to infer the effective peak supersaturation. A relative uncertainty of about 30% was estimated for  $SS_{peak}$ . A detailed description how the  $SS_{peak}$  was estimated from the measurements performed at the JFJ can be found in Hammer et al. (2014)."*

*Following your suggestion, we added the following table to Sect. 2:*

**Table 1.** List of important symbols

Parameter	Notation
General parameters	
SS	supersaturation
SS <sub>peak</sub>	effective peak supersaturation (Hammer et al., 2014)
SS <sub>crit</sub>	critical supersaturation (Köhler, 1936)
$w$	updraft velocity
Measured parameters	
$w_{act}^{meas}$	measured updraft velocity
Estimated parameters	
SS <sub>peak</sub> <sup>estim</sup>	estimated effective peak supersaturation derived from measurements (see Sect. 2.3.1)
$w_{act}^{estim}$	estimated updraft velocity derived from measurements and topography (see Sect. 2.1.2)
Modelled parameters	
SS <sub>peak</sub> <sup>mod</sup>	modelled effective peak supersaturation
SS <sub>peak</sub> <sup>ref</sup>	effective peak supersaturation obtained from the reference model simulation
SS <sub>mod</sub> <sup>max</sup>	maximum relative water vapour pressure between the model initialization point and the JFJ
$w_{act}^{mod}$	modelled updraft velocity
$w_{mod}^{divX}$	modelled updraft velocity divided by $X$
$w_{mod}^{mulX}$	modelled updraft velocity multiplied by $X$
SS <sub>peak</sub> <sup>fluc</sup>	modelled effective peak supersaturation applying the real-time fluctuations
SS <sub>peak</sub> <sup>fluc,sin</sup>	modelled effective peak supersaturation with a sinus function

**Specific comments:**

Page 25972, line 12

*You should state that you are assuming that none of the water is lost to precipitation and reasons to back up this assumption.*

*We added: "... before cloud formation. As in (Hammer et al., 2014), it was assumed that water removal due to precipitation was negligible.*

Page 25974, line 11

*What is the estimated time between droplet activation and the observation site? Would you expect coalescence to occur?*

*The median cloud base height was calculated at about 270 meters below the station. Taking the median estimated updraft velocity of 0.8 m/s, a transit time of 216 seconds results. Therefore, coalescence can't be ruled out but the transit times are pretty short, so we don't think that it has a big effect.*

Page 25972, line 26

*What is the basis of this wet adiabatic lapse rate? Are there no observations that you can use?*

*No, unfortunately there are no measurements available for retrieving the lapse rate. Due to the steep slope towards the North side (its basically a crumbly rock wall) it is not possible to mount any instruments there. There are also few meteorological stations in the area, and those that exist tend to be within a valley, or located in other places where the complex terrain dictates that they would not be influenced by the same air masses as those approaching the JFJ. For example, during the CLACE campaign in 2010 there was a ceilometer and a windprofiler installed at a site just below the Jungfraujoch. The study of Ketterer et al. (2014) showed that despite the proximity, the air passing over this site was not reaching the JFJ*

Page 25976, line 7

*The last statement on this line seems unintuitive. Normally one would think that if your model does not account for latent heat, then the wet adiabatic lapse rate would be the one that would be unnecessary.*

*The model runs from a pre-calculated temperature trajectory. Since latent heat release during condensation is not accounted for in the model (i.e. the temperature trajectory is not modified during the calculation), it needs to be accounted for in the trajectory. Therefore we use the wet adiabatic lapse rate.*

Page 25977, line 5

A brief, but thorough, description of the calculations used to determine  $SS_{peak}$  from measurements should be included here. It is unfair to the reader to expect that they have the manuscript from the previous study readily available.

We answered this comment above in the general comments.

Page 25978, line 13

How does using an average  $\kappa$  affect your results? It is unclear from the previous description that you are even using a varying  $\kappa$ . For this reason, it is important to include a description of your calculations, as mentioned above.

To make the section investigated parameters clearer, we rearranged some sections as follow:

- Section "The effective peak supersaturation" was put as a new section after observational data
- In Section "Investigated parameters" we put "Simulated small-scale temperature fluctuations" as a subsection.
- Added a new subsection for "Hygroscopicity parameter"

We added in Section "Modelled updraft velocity": "The modelled updraft velocity,  $w_{act}^{mod}$ , was used for the reference model simulation (see Sect. 2.4). This parameter,  $w_{act}^{mod}$ , was then varied to investigate the sensitivity of the updraft velocity on  $SS_{peak}$  (see results in Sect. 3.2)."

We added in Section "Aerosol- and updraft-limited regimes": "Thereby, the aerosol number concentration and size was varied by  $\pm 15\%$  to investigate the sensitivity of the aerosol- and updraft-limited regimes on  $SS_{peak}$  (see results in Sect. 3.2)."

For the reference simulation we use a constant kappa, for the sensitivity tests we vary kappa to answer exactly this question – what the influence of kappa on the results would be (Fig 7). We added into the new section "Hygroscopicity parameter": "The hygroscopicity parameter,  $\kappa$ , stays rather constant over time at the Jungfraujoch at around 0.2 (Juranyi et al., 2011). To investigate the sensitivity of  $\kappa$  on  $SS_{peak}$ , a typical  $\kappa$  value for an aerosol size distribution with a larger fraction of organics ( $\kappa=0.1$ ; Dusek et al., 2010) and for a continental aerosol ( $\kappa=0.3$ ; Andreae et al, 2008; Pringle et al., 2010) was used (see results in Sect. 3.2). It is important to note, that the studies (Hammer et al., 2014a) and (Hammer et al., 2014b) revealed only a small influence of the  $\kappa$ -value on the calculated  $SS_{peak}$ ."

Page 25979, line 7

Please expand on why these two updraft velocities are so different. This is a very important point since the remainder of the paper only relies on the modelled updraft velocity.

We added: "... than the estimated  $w_{act}^{estim}$ . As discussed in Hammer et al. 2014, The reason that the modelled  $w_{act}$  is much lower than the estimated value is possibly due to the air mass being accelerated as it passes through the narrow pass where the JFJ is located. This may lead to higher windspeeds being measured at the JFJ than those which were actually present when the cloud was formed at a lower altitude. To investigate the sensitivity ..."

Page 25979, line 12

It is not surprising that the modelled data points are closer to the model simulations for the median case. It is true that the signal is clearer between  $SS_{peak}$  and  $w$  than in the previous study. However, the results presented here reflect the model, and in fact, our previous understanding of updraft velocity and supersaturation. This Figure does not really reveal any new understanding that is not already represented in the model. Figure 4 Since you are studying the effects of updraft velocity on peak supersaturation, the axes on this Figure should be reversed.

The point of Figure 4, was to show that the improved agreement between modelled and estimated peak SS suggests that the estimated  $w_{act}$  is indeed too high. We agree that this was not clearly stated in the text, and have now added "...which is related to  $SS_{peak}$ . The fact that when the model is constrained to reproduce the observed number of droplets, a lower updraft velocity is found, causing a better agreement between modelled and estimated  $SS_{peak}$ , suggests that the updraft velocity

*estimated from wind speed measurements at the JFJ is indeed overestimated.” We have left the axes in the same configuration so that the figure may easily be compared with that of Hammer et al 2014.*

*Page 25979, line 11*

*How was  $SS_{estim}^{peak}$  derived from measurements?*

*We added: “... $SS_{peak}^{estim}$  (derived from measurements; see Sect. 2.3.1).” as we added a description about  $SS_{peak}^{estim}$  according to the general comments.*

*Page 25979, line 19*

*Shouldn't the black lines by definition run through the green points since they are the median? While the black lines do seem to fit the green points better, the residuals are by no means centred around zero. The bias in the fit, and possible sources, should be discussed in this section.*

*It is possible that there has been a misunderstanding here. The black lines were calculated by running the model with number and size and kappa values representative of the 25 and 75<sup>th</sup> percentile of those observed, and the median. The model, using these values as input, was run for a range of updraft velocities, yielding the three lines in Fig 4. The observed points on the left hand side of the plot, showing low peak supersaturations at relatively high updraft velocities in comparison with the three modelled lines, result from times when the aerosol number was high (i.e. above the 75<sup>th</sup> percentile), preventing a high supersaturation from being reached. Likewise, on the right hand side of the plot, lower aerosol numbers (below the 25<sup>th</sup> percentile) would lead to the systematic overestimation displayed by the three lines. The variation of Kappa provides further scatter.*

*We have changed the description slightly to make this clearer:*

*“The black curve in Fig.4 represents the box model simulations of  $SS_{peak}^{mod}$  obtained by running the simulations for a range of constant updraft velocities. In the upper line, the aerosol size distribution was chosen so that the number and sizes of the aerosol and  $k$  value were representative of the 75<sup>th</sup> percentile of those observed during CLACE 2011. The bottom line was calculated similarly using aerosol properties representative of the 25<sup>th</sup> percentile, with the middle line calculated using aerosol properties representative of the median.*

*Page 25979, line 27*

*The points shift down in your current Figure.*

*We changed it to: “...the points shift substantially closer towards the black line, ...”*

*Page 25981, line 7*

*Was  $\kappa$  also kept constant over size?*

*Yes, however this text fragment is deleted due to the answer to another reviewer.*

*Section 3.3*

*These results are really quite interesting. It would be worthwhile to consider moving this section earlier so that it is not passed over by an inattentive reader.*

*Actually, we would like to end the paper with one of the main result, which is Section 3.3. Therefore, we would like to keep it here but added the following to the abstract to give the results more weight: “Furthermore, there is a maximum of influence from turbulence on  $SS_{peak}$  between 0.2 - 0.4%. Simulating the small-scale fluctuations with several amplitudes, frequencies and phases, revealed that independently on the amplitude, the effect of the frequency on  $SS_{peak}$  shows a maximum at 0.46 (median over all phases). It was found that an increase in amplitude of the small-scale variations in the cooling rate, can significantly alter the CCN activation.”*

*Page 25984, line 4*

*To which of the modelled values are you referring?*

*We changed it to: “... showed that for small  $w_{act}^{mod}$  the model was slightly underestimating the  $SS_{peak}$ .”*

*Page 25984, line 5*

*This is really quite remarkable. What percentage of the points now fall within the 25th and 75th percentile lines?*

*We changed this as follows: "... slightly improves the  $SS_{peak}^{fluc} - W_{act}^{mod}$  relationship at lower updraft velocities as can be seen in Fig. 9. 44% of all the data points are now lying within the 25<sup>th</sup> and 75<sup>th</sup> percentile range (compared to 41% before) and the other points are also much closer to the band."*

#### **Technical Comments**

*Page 25972, line 15*

*Change to "the ideal gas law and the Clausius-Clapeyron equation".*

*Page 25974, line 21*

*Change "can not" to "cannot".*

*Page 25975, line 23*

*Remove the comma between "that" and "which".*

*Page 25976, line 3*

*Change "while" to "where".*

*Page 25976, line 9*

*Change "was ranging" to "ranged".*

*Page 25976, line 16*

*Sentence should be "According to Köhler theory".*

*Page 25978, line 9*

*Consider changing the title of this section to "Reference model for sensitivity analysis" so that the reader can easily refer back to this section later.*

*Page 25978, line 12*

*This sentence should read "For this purpose" if you are referring to the reference simulation.*

*Page 25979, line 1*

*Figure 4 is mentioned before Fig. 3.*

*Page 25984, line 17*

*Change wording to "is faster than the time".*

*Page 25984, line 18*

*Remove "also" from the sentence to make it less awkward.*

*Page 25984, line 25*

*Consider removing "being able" from the sentence.*

*Page 25985, line 18*

*Change "indicates" to "results in" or "causes".*

*Page 25985, line 22*

*Consider changing the sentence to "particle size had a stronger influence on".*

*Page 25986, line 7*

*Remove "presumably" from the sentence.*

*Page 25986, lines 10-11*

*Consider changing the text to "are more strongly influenced by small-scale variations. The decreasing influence..."*

*Page 25986, lines 16-17*

*The present tense should be used to emphasize the results.*

*Page 25986, line 22*

Change the text to “independent of the amplitude”.

*Thank you for all these technical comments. We applied all of these in the text accordingly.*

*Page 25975, line 17*

*To what fluctuations are you referring? This sentence is vague.*

*We changed it to: “To investigate the importance of the small-scale fluctuations of  $SS_{peak}$  to the decrease ...”*

*Page 25976, line 25*

*Your wording of “corresponds to the  $SS_{mod}^{max}$ ” suggests that this variable has been used before, whereas you are actually introducing it here.*

*We changed it to: “is expressed as  $SS_{mod}^{max}$  ...”*

# **Sensitivity estimations for cloud droplet formation in the vicinity of the high alpine research station Jungfrauoch (3580 m a.s.l.)**

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## Abstract

Aerosol radiative forcing estimates suffer from large uncertainties as a result of insufficient understanding of aerosol–cloud interactions. The main source of these uncertainties are dynamical processes such as turbulence and entrainment but also key aerosol parameters such as aerosol number concentration and size distribution, and to a much lesser extent, the composition. From June to August 2011 a Cloud and Aerosol Characterization Experiment (CLACE) was performed at the high-alpine research station Jungfraujoch (Switzerland, 3580 m a.s.l.) focusing on the activation of aerosol to form liquid-phase clouds (in the cloud base temperature range of  $-8$  to  $5$  °C). With a box model the sensitivity of the effective peak supersaturation ( $SS_{\text{peak}}$ ), an important parameter for cloud activation, to key aerosol and dynamical parameters was investigated. ~~It was found that the~~ The updraft velocity, ~~defining which defines~~ the cooling rate of an air parcel, ~~is the parameter with the largest was found to have the greatest~~ influence on  $SS_{\text{peak}}$ . Small-scale variations in the cooling rate with large amplitudes can significantly alter CCN activation. Thus, an accurate knowledge of the air parcel history is required to estimate  $SS_{\text{peak}}$ . The results show that the cloud base updraft velocities estimated from the horizontal wind measurements made at the Jungfraujoch can be divided by a factor of approximately 4 to get the updraft velocity required for the model to reproduce the observed  $SS_{\text{peak}}$ . Furthermore, there is a maximum of influence from turbulence on  $SS_{\text{peak}}$  between 0.2–0.4 %. Simulating the small-scale fluctuations with several amplitudes, frequencies and phases, revealed that independently on the amplitude, the effect of the frequency on  $SS_{\text{peak}}$  shows a maximum at 0.46 (median over all phases) and at higher frequencies, the maximum  $SS_{\text{peak}}$  decreases again. It was found that an increase in amplitude of the small-scale variations in the cooling rate, can significantly alter the CCN activation.

# 1 Introduction

The interactions between aerosols and clouds are the largest contributors to uncertainty in the calculation of aerosol radiative forcing (Boucher et al., 2013). Aerosols with a certain size, shape and chemical composition are able to form a cloud droplet, if they are exposed to air which is supersaturated with respect to water vapour. Particles that are able to activate and become cloud droplets are called cloud condensation nuclei (CCN). The number concentration of CCN is determined by the aerosol number size distribution, the hygroscopic properties of the aerosol and the supersaturation in the surrounding air. Thus, to address the aerosol–cloud interaction processes in detail, all these properties need to be known. However, present climate models are not capable of representing these aerosol properties in the required detail. Thus, compromises and assumptions that accurately address the most important aerosol effects within the constraints of application are required (Cherian et al., 2014; Dufresne et al., 2013; Levy et al., 2013). It has been pointed out by Boucher et al. (2013) that the main uncertainties in the aerosol radiative forcing are due to aerosol–cloud interaction dynamical factors such as turbulent strength and entrainment controlling the cloud condensation rate, and the key aerosol parameters such as aerosol number concentration and size distribution, and to a much lesser extent, the composition.

One of the properties that can be used to characterize the CCN activity of an aerosol particle is the critical supersaturation, i.e. the lowest supersaturation at which the particle is activated to a cloud droplet. The critical supersaturation depends on the particle size and chemical composition and is described by Köhler theory (Köhler, 1936). Whether a particle is able to act as a CCN in the atmosphere depends, aside from the particle's chemical and physical properties, on the supersaturation of water vapour. As an air parcel rises, it cools and may become supersaturated. Those particles with a critical supersaturation at or below the supersaturation in the air parcel will activate to form cloud droplets. The highest supersaturation that a particle experiences for a sufficiently long time to grow to a stable cloud droplet is defined as the effective peak supersaturation ( $SS_{\text{peak}}$ ; Hammer et al., 2014) -and this value is important as it determines the minimum activation diameter

in a population of particles, and thus the number of particles which activate to form cloud droplets. Small-scale fluctuations in vertical velocity can alter the path of an air parcel and thereby also the corresponding  $SS_{\text{peak}}$ , which is dependent on the cooling rate of the air parcel.

~~This study~~ The influence of physical and chemical quantities on the number and size of cloud droplets has been the subject of model studies for some time. A three dimensional model was used by Clark and Hall (1979) to examine the effect of fluctuations in super-saturation on the droplet size distribution evolution. They suggest that the observed broadening in the droplet size distribution above cloud base in cumulus clouds cannot be fully reproduced by the addition of fluctuations to a Lagrangian model, as this ignores spatial correlations between population or thermodynamic characteristics. However they also performed Lagrangian simulations for comparison with their 3D modelling results, and find that these reproduce a lower bound to the population broadening caused by turbulence. Fluctuations in saturation ratio were also investigated with a stochastic model by Kulmala et al. (1997), who pointed out that under conditions that are, on average, sub-saturated, fluctuations may lead to the activation of aerosol, and that above saturation, variation in saturation ratio may lead to accelerated growth of droplets. Feingold et al. (2003) applied an adiabatic parcel model and find that in non-precipitating stratocumulus cloud, at higher aerosol number densities (above a number corresponding to an extinction of approximately  $0.008 \text{ km}^{-1}$  in their study), the updraft velocity begins to have an influence on the droplet effective radius, as more aerosol is activated and the available condensable water is shared among more growing droplets. Using a cloud parcel model, Lance et al. (2004) showed that the presence of organic surfactants enhances the sensitivity of the modelled droplet concentration to vertical wind velocity, increasing the number of droplets. Under polluted conditions, this effect was determined to be of the same scale as the influence of updraft velocity. Using data from a non-urban site, Dusek et al. (2010) have shown that the number of activated aerosol mainly depends on the details of the aerosol size distribution and not the chemical composition. An adiabatic parcel model was used by Chuang (2006) to show that aerosol activation is sensitive to

the mass accommodation coefficient below values of approximately 0.1 – 0.001, and that the sensitivity to updraft velocity is greater under polluted conditions than under conditions with low aerosol number concentrations. In the study of Partridge et al. (2012), an adiabatic cloud parcel model was used to show that under clean conditions, the number and size of aerosol in the accumulation mode was important in determining the number of cloud droplets, however under polluted conditions, aerosol activation was more sensitive to chemical composition. Also, the sensitivity of the cloud droplet formation to aerosol chemical composition was found to increase when the updraft was reduced.

Ditas et al. (2012) derived the fluctuations of supersaturation in marine stratocumulus, based on observational data, finding a peak to peak supersaturation fluctuation in the range of 1.5%. The present study builds on the work of Hammer et al. (2014), which showed that there is a strong link between  $SS_{\text{peak}}$  and the updraft velocity. Additionally, it was shown that the physical properties (number concentration and size) of the aerosol possibly also have a non-negligible influence on  $SS_{\text{peak}}$ . However, the study was not able to shed light into which extent each parameter contributed to  $SS_{\text{peak}}$ . In here a sensitivity study was performed to gain more knowledge of the contribution of different physical and chemical aerosol parameters as well as the dynamical history of the air parcel to  $SS_{\text{peak}}$ . This was done for a dedicated measurement campaign (CLACE2011; described in Sect. 2). Although only results from the campaign performed in 2011 are shown, all results shown in Sects. 3.1 and 3.2 are also applicable to the earlier campaign performed in 2010 as the chemical and physical properties of the aerosol, and the meteorological conditions encountered during the campaign were similar in 2010 and 2011 (Hammer et al., 2014).

~~Small-scale fluctuations in vertical velocity can alter the path of an air parcel and thereby also the corresponding  $SS_{\text{peak}}$ , which is dependent on the cooling rate of the air parcel.~~ To develop effective models, it is important to know the influence of the variation of several key aerosol parameters influencing the cloud droplet formation. It has been pointed out by Boucher et al. (2013) and Spichtinger and Cziczo (2008) that the main uncertainties in the aerosol radiative forcing are due to ~~aerosol-cloud~~ aerosol-cloud interaction dynamical factors such as turbulent strength and entrainment controlling the cloud condensation rate,

and the key aerosol parameters such as aerosol number concentration and size distribution, and to a much lesser extent, the composition. I.e. the interplay of dynamics versus effects purely attributed to aerosols remains highly uncertain. Thus, in this study the influence of ~~these key aerosol parameters, the~~ the variation of the turbulent strength and the updraft velocity on the cloud activation (~~i.e.  $SS_{peak}$~~ ) is investigated using a cloud parcel model.

## 2 Methods

### 2.1 Observational data

Measurements of aerosol and cloud properties were performed at the high-alpine site Jungfraujoch (3580 m a.s.l.) in Switzerland during summer 2011. This intensive measurement campaign was carried out within the framework of a CCloud and Aerosol Characterization Experiment (CLACE) campaign. The main focus of the campaign was to investigate the physical, chemical and optical properties of aerosols as well as the interaction of the aerosol particles with clouds, for a better quantification of the radiative forcing due to aerosol–radiation interactions (RFari) and the radiative forcing due to aerosol–cloud interactions (RFaci). These measurements provide the basis for the current modelling study.

Due to the topography around the Jungfraujoch (JFJ) mainly northwest (NW) and southeast (SE) wind directions are observed at the site. The topography approaching from the NW differs from that on the SE side as can be seen in Fig. 1. To the SE, the Great Aletsch glacier declines gradually from the JFJ (1500 m of altitude decrease over 18 km) while the NW side drops steeply, descending 1500 m over a horizontal distance of 4800 m (Ketterer et al., 2014).

### 2.2 **Observational data**

To make the readability of the manuscript easier, Table 1 gives an overview of all notations used in this manuscript grouped into calculated, measured and modeled parameters.

## 2.1.1 Measurement setup

A number of quantities measured at the JFJ were either used as model input directly, or were used to calculate model input parameters. These included the aerosol size distribution, the temperature and pressure, wind speed and direction, and the total water content of the air.

For sampling the aerosols and the hydrometeors on JFJ, an interstitial and a total inlet were installed on the roof of the laboratory. The interstitial inlet sampled only the non-activated particles by a size discriminator to remove droplets larger than  $2\ \mu\text{m}$  in aerodynamic diameter. In the laboratory the aerosol was dried to  $\text{RH} < 10\%$  as it was heated to room temperature (typically 20 to  $30\ ^\circ\text{C}$ ). The total inlet sampled ~~all particles (including hydrometeors)~~ the hydrometeors as well as the interstitial particles, i.e. all particles.

The condensed water of the hydrometeors and the aerosol particles was evaporated via heating the top part of the total inlet to approximately  $20\ ^\circ\text{C}$ . Thus, all dried aerosol particles (non-activated aerosols and the residuals of the cloud droplets) reached the laboratory. The difference between the number concentration measured behind the total inlet minus the number concentration measured behind the interstitial inlet corresponds to the number of cloud residuals, i.e. the number of particles that have been activated to cloud droplets.

~~Those can be compared to the number of cloud droplets directly observed in the ambient air.~~ Downstream of the inlets, two scanning mobility particle sizers (SMPS) measured the total and the interstitial dry particle number concentration, respectively. Additionally, a cloud condensation nuclei counter (CCNC; DMT CCNC-100, described by Roberts and Nenes, 2005) measured the polydisperse CCN number concentration at eight defined supersaturations (SS) behind the total inlet. Combining these measurements with the total dry particle number size distributions, measured with the SMPS behind the total inlet, the hygroscopicity parameter ( $\kappa$ ) was inferred (Hammer et al., 2014).

~~An ultrasonic anemometer (Metek USA-1) was used to measure the~~ The 3-dimensional wind speed vector at the JFJ with a time resolution of 20 Hz. ~~The ultrasonic anemometer was measured with an ultrasonic anemometer (Metek USA-1). This instrument~~ was installed on a 3 m pole pointing away from the JFJ building to reduce the

influence of the building on the measured wind fields, although this influence could not be totally eliminated. Therefore, the ~~horizontal wind speed and wind direction~~ direction and horizontal wind speed data of the ultrasonic anemometer were not further used in this study. Nevertheless, the high-time resolved vertical wind speed measured by the ultrasonic anemometer is still expected to provide information on the small-scale fluctuations of the air mass.

~~A largely undisturbed measurement of the~~ The horizontal wind direction was obtained with the rosemount pitot tube anemometer ~~that~~ This instrument is mounted at the top of a 10 m mast located at around 75 m away from the ultrasonic anemometer. ~~These~~ The measurements were performed as part of the SwissMetNet network of MeteoSwiss together with temperature and pressure measurements continuously obtained at the JFJ. The temperature is measured with a thermo-hygrometer Thygan VTP-37 (Meteolabor AG).

Cloud presence and LWC were measured with a particle volume monitor (PVM-100; Gerber, 1991). ~~For the initialization of the box model, it was important to know the altitude of cloud base. The cloud base altitude was inferred from the liquid water content (LWC) of the cloud observed at the JFJ assuming an adiabatic rise of the air parcel before cloud formation. Thereby the corresponding dew point temperature of the LWC, assuming all the water is in vapour phase, was calculated via the ideal gas law and the law of Clausius-Clapeyron (Goff and Gratch, 1946). Via the hypsometric equation, the cloud base can be determined by iteratively lowering the altitude. The cloud base was defined as the point where the water partial pressure (assuming all water is in the gas phase) is equal to the saturation vapour pressure over liquid water (corrected for the pressure difference between the cloud base and the JFJ). A detailed description can be found in Hammer et al. (2014).~~

~~The temperature and the corresponding pressure trajectory was then calculated from the subsaturated regime of  $RH \approx 90\%$  (the~~

### 2.1.2 Defined cloud periods

Cloud periods that exhibited evidence of substantial entrainment or mixing were not included in the analysis. Such clouds were detected by analysing the activated fraction of the aerosol particles as a function of aerosol size. Periods where the largest size bins were not at least 90 % ~~was chosen in order to initialize the model under clearly subsaturated conditions~~) to the cloud base of  $\text{RH} = 100\%$  assuming a dry adiabatic lapse rate of  $\Gamma_{\text{dry}} = 0.98$ . ~~The calculation of the temperature and the corresponding pressure trajectory from the cloud base to the JFJ was done assuming a wet adiabatic lapse rate of  $\Gamma_{\text{wet}} = 0.65$~~  activated were excluded. This is the same procedure to that used by Hammer et al. (2014).

In here, only clouds reaching the JFJ with NW wind directions are considered. Relatively few measurement points when when SE wind was present, and also because the clouds coming from the NW are mostly found to be formed locally by rapid updraughts, in contrast to the clouds from the south, which are often stratus, which has been advected from further away. This makes the clouds from the NW more suitable for our study.

### 2.1.3 Estimation of the updraft velocity at the cloud base

(This section is composed by a summary of section 3.4 in Hammer et al. (2014).)

It is not feasible to measure the updraft velocity at the point of aerosol activation at the JFJ. Thus, an estimate of the updraft velocity at the cloud base ( $w_{\text{act}}^{\text{estim}}$ ) was inferred from the horizontal wind speed at the JFJ, as measured by the Rosemount pitot tube anemometer by making the following assumptions: (1) the air approaching the JFJ research station strictly followed the terrain, i.e. the flow lines are parallel to the surface (at least in the lowest layers). (2) Neither horizontal convergence nor divergence of the flow lines occurred between cloud base and the JFJ. Thus, the horizontal wind speed component stays the same between cloud base and the JFJ. With these assumptions,  $w_{\text{act}}^{\text{estim}}$  is obtained from the horizontal wind speed measured at the JFJ ( $v_{\text{JFJ}}^h$ ):

$$w_{\text{act}}^{\text{estim}} = \tan(\alpha) v_{\text{JFJ}}^h, \quad (1)$$



where  $\alpha$  denotes the inclination angle of the flow lines at cloud base. According to the topography software “Atlas der Schweiz 3.0” from Swisstopo and ETH Zurich, the terrain has a mean inclination of  $\alpha \approx 46^\circ$  over the last 700 m altitude difference before reaching the JFJ for northwesterly advection, which is close to the estimated location of the median cloud base during CLACE2011 (see detailed explanation in Hammer et al., 2014).

#### 2.1.4 The effective peak supersaturation

The cooling of an air parcel below its dew point temperature results in the formation of a cloud. According to Köhler theory (Köhler, 1936) the equilibrium saturation vapour pressure ( $S_{\text{eq}}$ ) over a solution droplet is described considering the Raoult (solute) and Kelvin laws. The critical supersaturation ( $SS_{\text{crit}}$ ) of a particle with a certain size and composition ( $\kappa$ ) defines the point of activation from particle to cloud droplet. Therefore, all particles in an air parcel having a  $SS_{\text{crit}}$  smaller than  $SS_{\text{peak}}$  are able to activate and grow to cloud droplets. In the box model the  $S_{\text{eq}}$  is calculated for each time step along the temperature and pressure on the air parcel trajectory. The maximum relative water vapour pressure between the model initialization point and the JFJ is expressed as  $SS_{\text{max}}^{\text{mod}}$ . The simulated effective peak supersaturation,  $SS_{\text{peak}}^{\text{mod}}$ , however is below  $SS_{\text{max}}^{\text{mod}}$ .  $SS_{\text{peak}}^{\text{mod}}$  was obtained by finding the highest water vapour saturation which lead to droplets larger than  $2 \mu\text{m}$  in diameter. In earlier studies it was found that a diameter of  $2 \mu\text{m}$  is a good threshold distinguishing the hygroscopic grown particles from cloud droplets (Jurányi et al., 2011; Henning et al., 2002).

It is important to note that in Hammer et al. (2014) the definition of the  $SS_{\text{peak}}^{\text{mod}}$  simply was the “highest SS reached along the trajectory”. The new definition described above is needed for investigating the small-scale fluctuation described in Sect. 3.3.1. The comparison  $SS_{\text{peak}}^{\text{mod}}$  obtained by the two definitions respectively, was within 10%.

The  $SS_{\text{peak}}^{\text{mod}}$  was retrieved as follow: 1) the activation threshold diameter was determined from the measurements of the total and interstitial number size distributions 2) the aerosol hygroscopicity was obtained from the simultaneous CCNC measurements 3) the activation threshold diameter was combined with the aerosol hygroscopicity to infer the effective peak

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supersaturation. A relative uncertainty of about  $\pm 30\%$  was estimated for  $SS_{\text{peak}}$ . A detailed description how the  $SS_{\text{peak}}$  was estimated from the measurements performed at the JFJ can be found in Hammer et al. (2014).

## 2.2 Box model simulations

### 2.2.1 Box model description (ZOMM)

5 The Zurich optical and microphysical model (ZOMM) was used in this study to simulate the effect of aerosol properties and atmospheric dynamics on ~~cloud formation~~ liquid cloud formation (please note, that no ice formation was simulated). ZOMM is a box model which calculates the evolution of an initial aerosol distribution along a temperature and pressure trajectory. A further description of ZOMM can be found in Luo et al. (2003) and Hoyle et al. (2005, 2013). [The model is available on request via the following e-mail: beiping.luo@env.ethz.ch](#).

15 For the initialisation of the model, the cloud periods detected at the JFJ were divided into six minute periods. Therefore, all aerosol and cloud properties described in this study are given in six minute averages. The temperature range of the observed clouds was from  $-8$  to  $5^{\circ}\text{C}$ . ~~Cloud periods that exhibited evidence of substantial entrainment or mixing were not included in the analysis. Such clouds were detected by analysing the activated fraction of the aerosol particles as a function of aerosol size. Periods where the largest size bins were not at least 90% activated were excluded. This is the same procedure to that used by~~

20 [It was important to know the altitude of cloud base. The cloud base altitude was inferred from the liquid water content \(LWC\) of the cloud observed at the JFJ assuming an adiabatic rise of the air parcel before cloud formation. Water removal due to precipitation is negligible since it is assumed that the total water content is preserved \(Hammer et al., 2014\). Assuming all the water is in vapour phase, the dew point temperature, was calculated via the ideal gas law and the Clausius–Clapeyron equation \(Goff and Gratch, 1946\). Via the hypsometric equation, the cloud base can be determined by iteratively lowering the altitude. The cloud base was defined as the point where the water partial pressure \(assuming all](#)

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water is in the gas phase) is equal to the saturation vapour pressure over liquid water (corrected for the pressure difference between the cloud base and the JFJ). A detailed description can be found in Hammer et al. (2014).

The model was initialised with an aerosol size distribution, consisting of aerosol number concentrations in 100 size bins. The size distributions were taken from the SMPS measurements at the total inlet, and therefore include both activated and interstitial aerosol. As ZOMM is a box model, mixing and sedimentation processes are not accounted for, and the total water content of an air parcel is conserved during the simulation. The total water contents used in the simulations were determined from the sum of the gas and liquid phase water measured at the JFJ. ~~From the temperature and total water content observed at the JFJ, the location of the altitude where  $RH = 90\%$  was calculated, and the starting points (temperature and pressure) of the model trajectories were determined~~To initialize the model under clearly subsaturated conditions, the temperature, and the corresponding pressure on the air parcel trajectory was calculated at  $RH \approx 90\%$  to the cloud base of  $RH = 100\%$  assuming a dry adiabatic lapse rate of  $\Gamma_{dry} = 0.98 \text{ K (100 m)}^{-1}$ . The calculation of the temperature and the corresponding pressure on the air parcel trajectory from the cloud base to the JFJ was done assuming a wet adiabatic lapse rate of  $\Gamma_{wet} = 0.65 \text{ K (100 m)}^{-1}$ . Implicit in this initialisation is the assumption that the aerosol size distribution observed at the JFJ is the same as that which was present below the cloud base. As it is not feasible to measure the aerosol size distributions below the cloud base at the JFJ, this assumption ~~can not~~ cannot be tested. However in this study the analysis is not performed on single trajectories, rather the results of the simulations are examined together, therefore the variability of the size distributions observed at the JFJ should capture the variability of the size distributions the cloud base.

The aerosol size observed in a single SMPS measurement has an uncertainty of about 10% Wiedersohler et al. (2012); however the input distributions used in the basic model simulations consist of median size distributions taken over the CLACE 2011 campaign. The bin resolution used in the model is the same as that measured by the SMPS. Any uncertainties in the model calculation resulting from the resolution of the bin sizes or the

aerosol size distribution would be much smaller than the differences in simulated peak supersaturation caused by varying the number and size of the aerosols, as is done in Fig. 6.

Below saturation with respect to liquid water, the hygroscopic growth, i.e. water content of the aerosol is calculated according to the  $\kappa$ -Köhler parametrization of (Petters and Kreidenweis, 2007), i.e. equation, under the assumption of equilibrium between the gas and liquid phases:

$$S(D, \kappa) = \frac{D^3 - D_{\text{dry}}^3}{D^3 - D_{\text{dry}}^3(1 - \kappa)} \exp\left(\frac{4\sigma_{\text{s/a}}M_{\text{w}}}{RT\rho_{\text{w}}D}\right), \quad (2)$$

where  $D$  is the droplet diameter,  $D_{\text{dry}}$  the dry diameter,  $\sigma_{\text{s/a}}$  the surface tension of the solution/air interface,  $M_{\text{w}}$  the molar mass of water,  $R$  the ideal gas constant,  $T$  the prevailing air temperature and  $\rho_{\text{w}}$  the density of water.

At  $S > 0.99$  with respect to liquid water, or at high cooling rates, the kinetic uptake of water to the droplets from the gas phase is calculated, accounting for gas phase diffusion as well as the Kelvin effect. The new radius of each size bin is calculated, and the bins are allowed to evolve independently in radius space, i.e. they are not constrained to a particular distribution shape.

The simulation follows the time series of temperature and pressure values which is given as input, and the simulation ends once the conditions observed at the JFJ are reached. In this way, the number of activated droplets and the ~~peak-effective~~ effective peak supersaturation predicted by the model can be compared with the values determined from the JFJ observational data. The threshold is, therefore, defined on final droplet size but on the threshold of the droplet size at the point droplets grow fast (i.e. at point of activation).

## 2.3 Investigated parameters

### 2.3.1 Simulated small-scale temperature fluctuations

To investigate the ~~importance of the fluctuations to the decrease of~~ influence of the small-scale fluctuations of  $SS_{\text{peak}}$  on the temperature ( $T_{\text{turb}}$ ) and pressure along the air parcel trajectory from the initialization point to the JFJ at time ( $t$ ), the 20 Hz time resolved updraft ~~velocity-velocities~~ measured by the sonic anemometer ( $w_{\text{act}}^{\text{meas}}$ ) were applied to the linear temperature decrease derived from the lapse rate ( $T_{\text{lin}}$ ). This was done by superimposing a time series of temperature fluctuations measured at the JFJ upon the linear temperature trajectory along which the model was run ~~-(see black line in Fig. 2).~~ The time series of fluctuations was chosen to be simply that ~~;~~ which was measured at the JFJ during the time taken for the air parcel to ascend from the point where the model was initialized (indicated in Fig. 2 with RH=90%), to the JFJ ~~-(indicated in Fig. 2 with JFJ).~~ The relative vertical fluctuation calculated from the measured wind field at the JFJ ( $w'$ ) at time  $t$  was retrieved as follows:

$$w'(t) = w_{\text{act}}^{\text{meas}}(t) - (a + bt), \quad (3)$$

~~while where~~  $a$  and  $b$  are the  $y$  intercept and the slope, respectively, from the linear regression function of  $w_{\text{act}}^{\text{meas}}(t)$  for the time period from  $t_{\text{start}}$  (the time at which the model was initialized) to  $t_{\text{JFJ}}$  (the time at which the modelled trajectory reached the JFJ). The deviation from  $T_{\text{lin}}(t)$  due to the fluctuation ( $T'_{\text{turb}}(t)$ ) was then estimated-calculated by multiplying  $w'(t)$  with the wet adiabatic lapse rate ( $\Gamma_{\text{wet}} = 0.65 \text{ K (100 m)}^{-1}$ ; ~~the dry adiabatic lapse rate is not needed since the model does not account for latent heat~~). Adding  $T'_{\text{turb}}(t)$  to  $T_{\text{lin}}(t)$  leads then to  $T_{\text{turb}}(t)$ .  ~~$T_{\text{turb}}(t)$  was ranging~~  $T'_{\text{turb}}(t)$  ranged from  $-0.8$  to  $1.1 \text{ K}$  with a 25th percentile of  $0.01 \text{ K}$ , a 75th percentile of  $0.05 \text{ K}$  and a median of  $0.03 \text{ K}$ . Figure 2 shows an example of  $T_{\text{turb}}$  for the model run ~~#516, which was~~ detected at the JFJ on 8 August 2011 18:20 UTC.

## 2.4 Investigated parameters

### 2.3.1 The effective peak supersaturation

The cooling of an air parcel below its dew point temperature results in the formation of a cloud. According to the Köhler theory (Köhler, 1936) the equilibrium saturation vapour pressure ( $S_{\text{eq}}$ ) over a solution droplet is described considering the Raoult (solute) and Kelvin laws. The critical supersaturation ( $SS_{\text{crit}}$ ) of a particle with a certain size and composition ( $\kappa$ ) defines the point of activation from particle to cloud droplet. The effective peak supersaturation ( $SS_{\text{peak}}$ ) is the highest saturation encountered within an air parcel, which leads to activation of aerosol. Therefore, all particles in an air parcel having a  $SS_{\text{crit}}$  smaller than  $SS_{\text{peak}}$  are able to activate and grow to cloud droplets. In the box model the  $S_{\text{eq}}$  is calculated for each time step along the temperature and pressure trajectory. The maximum relative water vapour pressure between the model initialization point and the JFJ corresponds to the  $SS_{\text{max}}^{\text{mod}}$ . The simulated effective peak supersaturation,  $SS_{\text{peak}}^{\text{mod}}$ , however is below  $SS_{\text{max}}^{\text{mod}}$  since the  $SS_{\text{peak}}$  is defined as the highest supersaturation that a particle experiences for a sufficiently long time to grow to a stable cloud droplet.  $SS_{\text{peak}}^{\text{mod}}$  was obtained by finding the highest water vapour saturation which lead to droplets larger than 2 in diameter. A detailed description how the  $SS_{\text{peak}}^{\text{mod}}$  was estimated from the measurements performed at the JFJ can be found in Hammer et al. (2014).

### 2.3.1 Modelled updraft velocity

As well as being estimated from measurements ([see Sect. 2.1.3](#)), the updraft velocity can be modelled ( $w_{\text{act}}^{\text{mod}}$ ). With the ZOMM model, an initial model run was performed, and the number of simulated cloud droplets was compared with the observed number of cloud residuals at the JFJ. The cooling rate in the model was then iteratively adjusted until the simulated number of droplets was within 2% of the observed number of cloud residuals, which was considered to be sufficient for the propagation of  $SS_{\text{peak}}$  values.

The modelled updraft velocity,  $w_{\text{act}}^{\text{mod}}$ , was used for the reference model simulation (see Sect. 2.4). This parameter,  $w_{\text{act}}^{\text{mod}}$ , was then varied to investigate the sensitivity of the updraft velocity on  $\text{SS}_{\text{peak}}$  (see results in Sect. 3.2).

### 2.3.2 Aerosol- and updraft-limited regimes

5 Previous studies have found that a high  $\text{SS}_{\text{peak}}$  can be caused by a high updraft velocity or a low number of potential CCN (i.e. low number concentration of sufficiently large particles and/or low particle hygroscopicity). Conversely, a low  $\text{SS}_{\text{peak}}$  can be caused by small updraft velocity or a large number of potential CCN (i.e. high number concentration of large particles and/or high particle hygroscopicity). The study of Reutter et al. (2009) defined three  
10 different regimes depending on the ratio between the updraft velocity and the particle number concentration ( $w/N_{\text{CN}}$ ): (1) the aerosol-limited regime, (2) the updraft-limited regime and (3) the aerosol- and updraft-sensitive regime (transitional regime). The aerosol-limited regime is characterized by a relatively high ratio of  $w/N_{\text{CN}}$ , by a high activated fraction of aerosol particles (larger than 90 %) and the aerosol-limited regime is basically independent  
15 of  $w$ . The high updraft velocities lead to high  $\text{SS}_{\text{peak}}$  large enough to activate almost all of the particles except of the very small ones. The updraft-limited regime is characterized by a low ratio of  $w/N_{\text{CN}}$  (smaller than 20 %), saying that only a few particles are activated to cloud droplets due to low  $\text{SS}_{\text{peak}}$  values. In this regime the cloud droplet number concentration exhibits a linear dependence on  $w$  and a weak dependence on the  $N_{\text{CN}}$ . The  
20 aerosol- and updraft-sensitive regime is characterized by  $w/N_{\text{CN}}$  values lying between the two other regimes. Depending on  $\text{SS}_{\text{peak}}$ , the critical dry activation diameter for CCN activation ranges from very low up to the maximum of the dry particle size distribution. All these regimes will be discussed in Sect. 3.2 regarding the sensitivity study of  $\text{SS}_{\text{peak}}$  on updraft velocity, particle size distribution and hygroscopicity. Thereby, the aerosol number concentration and size was varied by  $\pm 15\%$  to investigate the sensitivity of the aerosol- and updraft-limited regimes on  $\text{SS}_{\text{peak}}$  (see results in Sect. 3.2).  
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## 2.4 Reference model simulation

### 2.3.1 Hygroscopicity parameter

The hygroscopicity parameter,  $\kappa$ , stays rather constant over time at the Jungfraujoch at around 0.2 (Jurányi et al., 2011). To investigate the sensitivity of  $\kappa$  on  $SS_{\text{peak}}$ , a typical  $\kappa$  value for an aerosol size distribution with a larger fraction of organics ( $\kappa = 0.1$ ; Dusek et al., 2010) and for a continental aerosol ( $\kappa = 0.3$ ; Andreae and Rosenfeld, 2008; Pringle et al., 2010) was used (see results in Sect. 3.2). It is important to note, that the studies Hammer et al. (2014) and Hammer et al. (2014) revealed only a small influence of the  $\kappa$  value on the calculated  $SS_{\text{peak}}$ .

### 2.4 Reference model for sensitivity analysis

For the sensitivity studies shown in Sects. 3.2 and 3.3.1 a ~~reference model simulation set of reference model simulations~~ was used. ~~This reference simulation was~~ These reference simulations were performed using the dataset measured at the JFJ during CLACE2011 as input variables. For ~~that this~~ purpose an average, constant  $\kappa$  value of 0.2 was used (Jurányi et al., 2011). For the updraft velocity, the simulated parameter  $w_{\text{act}}^{\text{mod}}$  was used as described in Sect. 2.3.1. All output parameters of the reference model simulations are depicted with a superscript <sup>ref</sup>, as e.g. for the effective peak supersaturation from the reference model simulation:  $SS_{\text{peak}}^{\text{ref}}$ .

## 3 Results and discussions

The sensitivity of the  $SS_{\text{peak}}$  to the particle's size distribution and hygroscopicity, cooling rate of the air parcel (i.e. updraft velocity), and the temperature fluctuations with time have been investigated.



### 3.1 Comparison of the estimated and the simulated updraft velocity

The study of Hammer et al. (2014) simulated  $SS_{\text{peak}}$  using  $w_{\text{act}}^{\text{estim}}$  as an upper limit for the updraft velocity at the point of aerosol activation (see Sect. 2.1.3) and the same model as in this study. It was observed that  $SS_{\text{peak}}$  was generally overestimated for a particular  $w_{\text{act}}^{\text{estim}}$  (see Fig. 3; red circles and black line). It was speculated that the estimated  $w_{\text{act}}^{\text{estim}}$  might overestimate the true updraft velocity at cloud base due to flow convergence in the approach to the narrow gap in which the JFJ is located (see Fig. 1), or due to flow lines that do not strictly follow the terrain. Thus, in the present work, the mean updraft velocity was simulated with ZOMM, as described in Sect. 2.3.1. In Fig. 4 the ratio of the modelled mean updraft velocity ( $w_{\text{act}}^{\text{mod}}$ ) to  $w_{\text{act}}^{\text{estim}}$  is shown for each model simulation. The ratios show that  $w_{\text{act}}^{\text{mod}}$  is a factor of 4 (median) lower than the estimated  $w_{\text{act}}^{\text{estim}}$ . In Fig. 3, the red dots show data from Hammer et al. (2014), where the effective peak supersaturation was plotted against the estimated updraft velocity,  $w_{\text{act}}^{\text{estim}}$ . There, it was found that there was a very weak correlation between updraft velocity and  $SS_{\text{peak}}^{\text{estim}}$  (derived from measurements; see Sect. 2.1.4). The modelled data points are substantially closer to the model simulations when  $w_{\text{act}}^{\text{mod}}$  (green circles) rather than  $w_{\text{act}}^{\text{estim}}$  (red circles), is plotted against  $SS_{\text{peak}}^{\text{estim}}$  as  $w_{\text{act}}^{\text{mod}}$  was calculated by constraining the model to the observed number of activated droplets, which is related to  $SS_{\text{peak}}$ . ~~Please note that none of these values account for possible small-scale fluctuations. The fact that when the model is constrained to reproduce the observed number of droplets, a lower updraft velocity is found, causing a better agreement between modelled and estimated  $SS_{\text{peak}}$ , suggests that the updraft velocity estimated from wind speed measurements at the JFJ is indeed overestimated.~~

The black ~~curve~~ curves in Fig. 3 ~~represents~~ represent the box model simulations of  $SS_{\text{peak}}^{\text{mod}}$  obtained by running the simulations for a ~~range~~ of constant updraft velocities ~~with 25th~~. In the upper line, the aerosol size distribution was chosen so that the number and sizes of the aerosol and  $\kappa$  value were representative of the 75th percentiles and median aerosol properties from CLAGE2011. percentile of those observed during CLAGE 2011. The bottom line was calculated similarly using aerosol properties representative of the 25th percentile,

with the middle line calculated using aerosol properties representative of the median. From this, the expected effect of  $SS_{\text{peak}}$  on updraft velocity can be seen. The black line lies near the middle of the cloud of green points, and the variability of the green circles about the line is a result of the different chemical and physical properties of the aerosol distributions in the different model simulations. The dashed curves represent the box model simulations of  $SS_{\text{peak}}^{\text{mod}}$  using the 25th and the 75th percentiles of the aerosol properties from CLACE2011 (see values in Table 2). While the data points showing the  $SS_{\text{peak}}^{\text{estim}}$  values derived from measurements vs. the  $w_{\text{act}}^{\text{estim}}$  values are spread across the upper left half of the plot, the relationship between  $SS_{\text{peak}}^{\text{mod}}$  and  $w_{\text{act}}^{\text{mod}}$  appears better defined, and the points shift substantially ~~to the right~~ closer towards the black line, with 39% being between the 25th and 75th percentile band of the values modelled with fixed aerosol size distributions. This substantial shift in the data illustrates the strong influence that the vertical wind ~~potentially-most likely~~ has on the  $SS_{\text{peak}}$ .

### 3.2 Influence of the updraft velocity, particle size distribution and hygroscopicity on the effective peak supersaturation

~~Previous studies have found that a high~~ According to previous studies, a low  $SS_{\text{peak}}$  can be caused by a high small updraft velocity or a low large number of potential CCN. Conversely, a low high  $SS_{\text{peak}}$  can be caused by small a high updraft velocity or a large low number of potential CCN (see Sect. 2.3.2).

In Sect. 3.1 it was shown that  $w_{\text{act}}^{\text{mod}}$  is on average a factor of 4 lower than the estimated  $w_{\text{act}}^{\text{estim}}$ . ~~Thus, to~~ This difference can most likely be explained by the fact that  $w_{\text{act}}^{\text{estim}}$  is an upper limit for the updraft velocity and  $w_{\text{act}}^{\text{mod}}$  is based on the simulated number of cloud droplets and the number of cloud residuals at the JFJ (as described in Sect. 2.3.1). Thus,  $w_{\text{act}}^{\text{mod}}$  is not an upper limit but the “true” updraft velocity at the point of aerosol activation. To investigate the sensitivity of  $SS_{\text{peak}}$  to the updraft velocity, the modelled value  $w_{\text{act}}^{\text{mod}}$  was divided by 2 ( $w_{\text{mod}}^{\text{div}2}$ ), divided by 5 ( $w_{\text{mod}}^{\text{div}5}$ ), multiplied by 2 ( $w_{\text{mod}}^{\text{mul}2}$ ) and 5 ( $w_{\text{mod}}^{\text{mul}5}$ ). The ratio 5 describes the maximum deviation from the mean value of  $w_{\text{act}}^{\text{mod}}$  and the ratio 2 is given from the 75th and 25th percentile of  $w_{\text{act}}^{\text{mod}}$ , which are about a factor of 2 from the

mean value. Figure 5 shows the ratio of  $SS_{\text{peak}}(w_{\text{mod}}^{\text{mul}x})$  using the modified updraft velocities as input parameters to the  $SS_{\text{peak}}^{\text{ref}}$  using the input parameter  $w_{\text{act}}^{\text{mod}}$ . All symbols are colour coded to show the number concentration in the size range of 96 (median dry activation diameter for CLACE2011) and 500 nm (upper limit of the SMPS). This value was used as an estimate for the potential CCN number concentration. It was found that using  $w_{\text{mod}}^{\text{div}2}$  as input parameter,  $SS_{\text{peak}}^{\text{ref}}$  is lowered on average by 25 % and using  $w_{\text{act}}^{\text{div}5}$  as input parameter lowers  $SS_{\text{peak}}^{\text{ref}}$  on average by 50 %. Using  $w_{\text{mod}}^{\text{mul}2}$  as input parameter the  $SS_{\text{peak}}^{\text{ref}}$  is raised by 38 % and with  $w_{\text{mod}}^{\text{mul}5}$  the  $SS_{\text{peak}}$  is on average a factor of 2 larger compared to using  $w^{\text{ref}}$  (i.e.  $w_{\text{act}}^{\text{mod}}$ ) as input parameter. Therefore, the relative influence of small and large changes in the updraft velocity is similar. Furthermore, an increase of the influence of  $w_{\text{act}}^{\text{mod}}$  from low to high  $SS_{\text{peak}}^{\text{ref}}$  on  $SS_{\text{peak}}$  was observed. Low  $SS_{\text{peak}}$  values are less affected by the updraft velocity because for low  $SS_{\text{peak}}$  values  $w_{\text{act}}^{\text{mod}}$  is already relatively low and therefore the absolute difference in  $w_{\text{act}}^{\text{mod}}$  due to a division by 2 or 5 is rather small and the rate of increase in saturation will not change substantially. Comparable to the aerosol-limited regime (Reutter et al., 2009) Fig. 5 shows that the effect of changes in  $w_{\text{act}}$  is slightly larger when the potential CCN number concentration is lower. Thus, the ratio of  $w/N_{\text{CN}}$  at these low  $SS_{\text{peak}}$  values is relatively low (at about 0.003) and is increasing with an increase in  $SS_{\text{peak}}^{\text{ref}}$  (up to about 0.03). This corresponds well with the results presented by Chuang (2006), and Feingold et al. (2003), who both found that under more polluted conditions, the characteristics of the droplet distribution are more sensitive to changes in the updraft velocity. In addition, the sensitivity of the peak supersaturation to a doubling (or increase by a factor of 5) in vertical wind velocity is slightly greater than the sensitivity to the corresponding decrease. This is similar to the findings of Partridge et al. (2012) for cloud droplet number concentration.

~~For a given supersaturation, the number concentration of CCN depends on the aerosol number size distribution and the particle hygroscopicity.~~

In Sect. 2 it is described that the topography at the JFJ defines two main wind directions, NW and SE wind. The variability of ~~either~~ the particle number concentration ~~or~~ as well as the particle size is expected to be on the same order of magnitude as the difference be-

tween NW and SE wind case. The dry number size distributions for the SE wind case during CLACE2011 showed on average 15% higher particle number concentration and 15% larger particles [than when the wind came from the NW](#). Thus, for the sensitivity of  $SS_{\text{peak}}^{\text{ref}}$  to the dry particle number size distribution the measured particle number size distribution was used as an input for the model simulations applying a 15% higher and lower particle number concentration and a 15% increase and decrease in diameter across all size bins, respectively (see Fig. 6). The [effects of changing the particle number size distribution and the particle number concentration were investigated separately](#). The higher/lower number concentration of larger particles decreases/raises the  $SS_{\text{peak}}^{\text{ref}}$ , respectively. The same was found for larger/smaller particle number concentration. 15% smaller and higher particle number concentration change the modelled peak supersaturation by approximately  $\pm 8\%$ , compared to the reference case. This ratio is rather constant over the whole diameter range. Using a 15% smaller and larger size distribution compared to the reference, a maximum difference of 21% was observed, however above a  $SS_{\text{peak}}^{\text{ref}}$  of about 0.4, the effect of changing the size or the number of the particles is similar.

It is interesting to note that while changing the number of the particles has a relatively constant effect on the modelled  $SS_{\text{peak}}$ , changing the size of the particles has a much more pronounced effect at low  $SS_{\text{peak}}^{\text{ref}}$ . This is because changing the size of the particles changes the minimum supersaturation at which the particles can activate. At low  $SS_{\text{peak}}^{\text{ref}}$ , updrafts are generally smaller (colour coding in Fig. 6), and only the largest particles activate. If they are smaller (larger)  $SS_{\text{peak}}$  will be higher (lower). At higher  $SS_{\text{peak}}^{\text{ref}}$ , where the updrafts are generally higher, the critical saturation of the largest particles plays less of a role in determining the  $SS_{\text{peak}}$ . Changing the number of the particles on the other hand does not affect the critical saturation needed to activate the largest particles, but rather influences just the condensation sink once the critical saturation has been exceeded (Rogers and Yau, 1989). Therefore the effect is relatively constant across the range of  $SS_{\text{peak}}^{\text{ref}}$ .

Another aerosol parameter influencing  $SS_{\text{peak}}$  is the hygroscopicity parameter of the dry particles,  $\kappa$ , describing the Raoult term of the Köhler equation (Petters and Kreidenweis, 2007). At the Jungfraujoch, it stays rather constant over time (Jurányi et al., 2011; Hammer

et al., 2014) at  $\kappa \approx 0.2$ . To look into the sensitivity of  $SS_{\text{peak}}$  to  $\kappa$ , a typical  $\kappa$  value for an aerosol size distribution with a larger fraction of organics ( $\kappa = 0.1$ ; Dusek et al., 2010) and for a continental aerosol ( $\kappa = 0.3$ ; Andreae and Rosenfeld, 2008; Pringle et al., 2010) was used as input for the model simulation. For the reference model simulation a  $\kappa = 0.2$  was used as input. Applying the aerosol size distribution with  $\kappa = 0.3$  as input for the model simulation results in lower  $SS_{\text{peak}}$  values compared to the reference size distribution ( $SS_{\text{peak}}^{\text{ref}}$ ; see Fig. 7). On average the  $SS_{\text{peak}}$  is lowered by 6 %, however, for smaller  $SS_{\text{peak}}^{\text{ref}}$  the effect of a larger  $\kappa$  value is stronger and lowers the  $SS_{\text{peak}}$  up to 15 %. The model simulations using a  $\kappa$  value of 0.1 show on average 11 % higher  $SS_{\text{peak}}$  values compared to the reference model simulation, whereas the maximum difference lies at 30 %. The larger increase of ratios of  $SS_{\text{peak}}(\kappa = 0.1) : SS_{\text{peak}}^{\text{ref}}$  compared to the decrease for  $SS_{\text{peak}}(\kappa = 0.3) : SS_{\text{peak}}^{\text{ref}}$  can be explained by the fact that a lower particle hygroscopicity results in a lower condensation of water vapour onto the particles and thus particles reach the size where the Kelvin term of the Köhler theory (Petters and Kreidenweis, 2007) becomes more important than the Raoult term and where particles activate to cloud droplets at larger sizes compared to higher particle hygroscopicity. The stronger influence of  $\kappa$  on small  $SS_{\text{peak}}$  values can be explained by the same reason as for the smaller/larger particle number concentration: at the small updraft velocities associated with small  $SS_{\text{peak}}$  (see Fig. 7), the critical saturation at which the largest particles activate plays a more important role in determining the final  $SS_{\text{peak}}$  than it does at higher updraft velocities. The changes in  $\kappa$  result in changes in this critical saturation, therefore the changes in  $\kappa$  have a larger effect at low  $SS_{\text{peak}}^{\text{ref}}$ .

### 3.3 Turbulence estimations and its influence on the effective peak supersaturation

#### 3.3.1 Measured turbulence

Turbulence is often present before cloud formation and within clouds. To address the influence of turbulence on the cloud activation, i.e. on the effective peak supersaturation, the linear cooling rate was modulated with the fluctuations obtained by a ultrasonic anemometer (Metek USA-1) that was located close to the other instruments at the site as described in

Sect. 2.3.1. Figure 8 shows the ratio of  $SS_{\text{peak}}^{\text{fluc} \cdot x}$  modelled applying the real-time fluctuations with a factor ( $x$ ) to the cooling rate vs.  $SS_{\text{peak}}^{\text{ref}}$  using a linear cooling rate (reference model simulation). It was assumed that each particle experienced the same real-time fluctuations.

Figure 8 shows that with stronger small-scale fluctuations (i.e. a larger  $x$  ~~applied~~ added to the fluctuation) the  $SS_{\text{peak}}$  increases significantly: applying the real-time fluctuation to the cooling rate raises the  $SS_{\text{peak}}$  by  $\sim 8\%$  (shown in Fig. 8 with the ratio of  $SS_{\text{peak}}^{\text{fluc}} : SS_{\text{peak}}^{\text{ref}}$ ). Multiplying the small-scale fluctuation ~~applied~~ added to the cooling rate by a factor of 5 increases the  $SS_{\text{peak}}$  by  $\sim 87\%$  and multiplying the fluctuations by 10 increases the  $SS_{\text{peak}}$  by a factor of  $\sim 3.22$  (see green and blue triangles in Fig. 8, respectively). The factors 5 and 10 are resulting in a similar range of temperature amplitudes used for the sinus curve simulations described in Sect. 3.3.2.

A dependence of the ratios on  $SS_{\text{peak}}^{\text{ref}}$  was observed. There is a maximum of the ratios at  $SS_{\text{peak}}^{\text{ref}}$  between 0.2–0.4%. The smaller ratios at higher  $SS_{\text{peak}}^{\text{ref}} \gtrsim 0.2\%$  are likely because fluctuations ~~applied~~ added to high cooling rates have a smaller relative influence on the cooling rate at the point of aerosol activation (updraft-limited regime; Reutter et al., 2009) than ~~turbulence applied~~ small-scale temperature fluctuations added to a small linear cooling rate. The reason for the smaller influence of the fluctuations in the air parcel for low  $SS_{\text{peak}}^{\text{ref}} \lesssim 0.2\%$  is likely due to the competition between the influence of cooling rates and aerosol properties (aerosol-limited regime; Reutter et al., 2009). Aerosol properties such as hygroscopicity, number and size are more important at lower cooling rates and thus lead to this maximum of the ratios  $SS_{\text{peak}}^{\text{fluc} \cdot x} : SS_{\text{peak}}^{\text{ref}}$  for  $0.2\% \lesssim SS_{\text{peak}}^{\text{ref}} \lesssim 0.4\%$ . Nevertheless, there is also a spread of the ratio at a given  $SS_{\text{peak}}^{\text{ref}}$ . This is explained by the variable nature of the temperature fluctuations – at the point where aerosol activation occurs, the cooling rate will sometimes be greatly modified by the temperature fluctuation, in some cases it will be rather close to the average cooling rate. In the latter case, the  $SS_{\text{peak}}$  from the simulation including fluctuations will be close to the  $SS_{\text{peak}}$  calculated from the reference simulation

Figure 3 showed that for small ~~updraft velocities~~  $w_{\text{act}}^{\text{mod}}$  the model was slightly underestimating the  $SS_{\text{peak}}$ . However, including small-scale fluctuations slightly improves the  $SS_{\text{peak}}^{\text{fluc}} - w_{\text{act}}^{\text{mod}}$ -relationship at lower updraft velocities as can be seen in Fig. 9. At updrafts of 0.1 to

$5s^{-1}$ , the  $SS_{\text{peak}}^{\text{fluc}}$  to  $w_{\text{act}}^{\text{mod}}$  relationship is improved slightly, with 44% of the points lying within the range of the 25-75th percentile of the measured values, compared with 40% when fluctuations are not included.

### 3.3.2 Sinus curve simulations of the effective peak supersaturation

5 Figure 10 shows the dependency of  $SS_{\text{peak}}$  on simulated small-scale fluctuations **applied**  
**added** to the cooling rate using a certain frequency ( $f$ ), amplitude ( $A$ ) and phase ( $\phi$ ). Three  
different amplitudes ( $A = 0.015, 0.022$  and  $0.04$  K) were used to simulate the small-scale  
fluctuations. The applied frequencies are in the range of 0.05 to 20 Hz. The variability on  
the  $y$  axis per  $f$  is given by the different phases of the sinus functions. They are in the range  
10 of 0 to  $360^\circ$  with  $18^\circ$  steps. Independently of the amplitude, the influence of the frequency  
on  $SS_{\text{peak}}^{\text{fluc, sin}}$  shows a maximum at  $f = 0.46$  Hz. Thus, the influence of  $f < 0.46$  on  $SS_{\text{peak}}$  is  
decreasing since  $f$  is too small to affect the cooling rate. For  $f > 0.46$ , the influence of  $f$  on  
 $SS_{\text{peak}}^{\text{fluc, sin}}$  is decreasing since the fluctuation is faster **compared with than** the time required  
for significant droplet growth. Likely for the same reason **also** the range of  $SS_{\text{peak}}^{\text{fluc, sin}}$  (25th  
15 and 75th percentiles) implied by the different phases is decreasing after the maximum of  
 $f = 0.46$  Hz. It was also found that larger amplitudes imply a larger range of  $f$  **being able**  
**to affect affecting** the  $SS_{\text{peak}}^{\text{fluc, sin}}$  as seen in Fig. 10. Furthermore, an increase in amplitude  
reveals an exponential increase in  $SS_{\text{peak}}^{\text{fluc, sin}}$  value (see Fig. 11). **At small amplitudes, high**  
**frequencies are affecting the  $SS_{\text{peak}}^{\text{fluc, sin}}$  values more significant than low frequencies.**

20 Several combinations of amplitudes and frequencies for sinus functions were found being  
able to represent the median small-scale fluctuations in the vicinity of the JFJ. Figure 12  
shows the relationship of the modelled  $SS_{\text{peak}}$  applying simulated small-scale fluctuations to  
the cooling rate ( $SS_{\text{peak}}^{\text{fluc, sin}}$ ) and  $SS_{\text{peak}}^{\text{fluc}}$ . The simulation of the small-scale fluctuations for the  
cooling rate was done using the example:  $A = 0.24$  K, frequency  $f = 0.022 s^{-1}$ . The good  
25 linear correlation (slope = 0.85, intercept = 0.06,  $r^2 = 0.88$ ) indicates that the combination  
of this amplitude and frequency is able to simulate the median small-scale fluctuations in  
the vicinity of the JFJ.

## 4 Conclusions

A sensitivity analysis was performed for the cloud activation at the high-alpine research station Jungfrauoch in Switzerland. The Zurich optical microphysical model (ZOMM) was used to simulate the effective peak supersaturation within the clouds using a set of input parameters, representative of the ambient air and aerosol properties at the JFJ during CLACE2011.

The analysis shows that  $SS_{\text{peak}}$  depends mainly on the updraft velocity, and not the physical properties of the aerosol. However, it is also the most difficult parameter to measure. It was observed that reducing the modelled updraft velocity,  $w_{\text{act}}^{\text{mod}}$ , by a factor of 2 lowers the  $SS_{\text{peak}}$  values on average by 25%, whereas a factor of 5 lowers the  $SS_{\text{peak}}$  on average by 50%. While multiplying  $w_{\text{act}}^{\text{mod}}$  by a factor of 2 and 5, increases the  $SS_{\text{peak}}$  by a factor of  $\sim 1.38$  and  $\sim 2$ , respectively. Thus, lowering or raising the updraft velocity to the same extent ~~indicates results in~~ a similar influence on  $SS_{\text{peak}}$ .

Another input parameter influencing the  $SS_{\text{peak}}$ , is the shape of the aerosol size distribution and its hygroscopicity. The sensitivity analysis showed that representative aerosol size distributions for the JFJ are influencing  $SS_{\text{peak}}$  only to a small extent up to 21%. It was observed that the 15%-change in particle size ~~was stronger influencing had a stronger influence on~~ the  $SS_{\text{peak}}$  values at lower updraft velocities than the 15%-change in number concentration. The influence of the hygroscopicity on  $SS_{\text{peak}}$  was investigated by taking  $\kappa = 0.1$ , as a typical value for a high organic fraction, and by taking  $\kappa = 0.3$  as a typical value for continental aerosols, as input parameter compared to the typical observed  $\kappa$  at the JFJ of 0.2. The average difference to the reference simulation was only  $\sim \pm 10\%$ , whereas the maximum difference goes up to  $\sim \pm 30\%$ . The lower  $\kappa$  showed a stronger influence on  $SS_{\text{peak}}$  compared to the higher one.

Small-scale temperature variations are always present at cloud formation processes. In this study the influence of small-scale variations on  $SS_{\text{peak}}$  was investigated by applying real-time fluctuations, measured with an ultrasonic anemometer, to the cooling rate. Although the fluctuations were measured at the JFJ, it is assumed that ~~presumably~~ conditions



that lead to greater fluctuations at the JFJ also lead to greater fluctuations at cloud base. Generally, it was found that small values of  $SS_{\text{peak}}^{\text{ref}}$  between approximately 0.2 and 0.4 % are ~~experiencing a stronger influence of small-scale variation. The decrease of the more strongly influenced by small-scale variations. The decreasing~~ influence of the small-scale fluctuations on  $SS_{\text{peak}}^{\text{ref}} \gtrsim 0.4\%$  could be explained due to the larger cooling rates which are less affected by small-scale variations. The decrease of the influence of the small-scale fluctuations on  $SS_{\text{peak}}^{\text{ref}} \lesssim 0.2\%$  is likely due to the higher competition of the small cooling rates with the aerosol properties, i.e. at these low  $SS_{\text{peak}}^{\text{ref}}$  values the aerosol properties such as hygroscopicity, number concentration and size become more important. On average small-scale variations ~~are raising of temperature raise~~ the  $SS_{\text{peak}}$  values to a larger extent than the other investigated parameters in this study. Multiplying the real-time fluctuation by a factor of 5 increases the  $SS_{\text{peak}}$  by  $\sim 87\%$  and multiplying the fluctuations by 10 increases the  $SS_{\text{peak}}$  by a factor of  $\sim 3.22$  compared to conditions without any small-scale fluctuations.

Simulating the small-scale fluctuations with several amplitudes, frequencies and phases, revealed that independently on the amplitude, the effect of the frequency on  $SS_{\text{peak}}$  shows a maximum at 0.46 (median over all phases). It was found that an increase in amplitude of the small-scale variations in the cooling rate, can significantly alter the CCN activation. Furthermore, ~~small-scale fluctuations in the vicinity of the Jungfrauoch were simulated based on~~ several sinus functions with combinations of amplitudes and frequencies ~~were found to represent the median small-scale fluctuations in the vicinity of the Jungfrauoch~~. The amplitudes are in the range of 0.01 and 0.09 K and the frequencies in the range of 0.05 and  $0.24 \text{ s}^{-1}$ .

Summarizing, small-scale temperature fluctuations are revealed to be the strongest effect on cloud formation processes beside the updraft velocity, which is influenced by the temperature fluctuations. The variation of aerosol number concentration and hygroscopic properties occurred to be less influenced compared to the aerosol size. Thus, this study revealed that of all investigated parameters the small-scale temperature fluctuation accompanied with the updraft velocity and the variation of aerosol size distribution has the strongest influence of cloud formation process (i.e. effective peak supersaturation).

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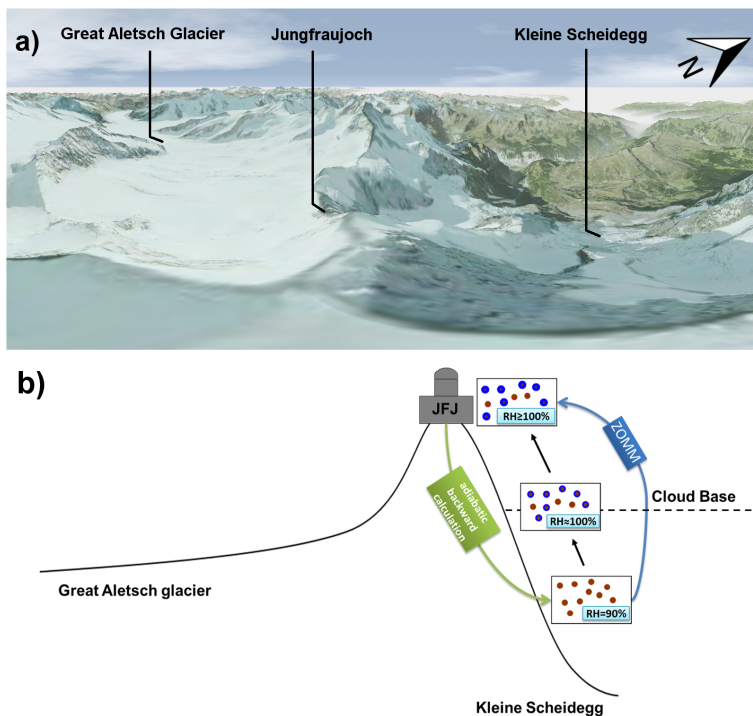
**Table 1.** List of important symbols

<u>Parameter</u>	<u>Notation</u>
<u>General parameters</u>	
<u>SS</u>	<u>supersaturation</u>
<u>SS<sub>peak</sub></u>	<u>effective peak supersaturation (Hammer et al., 2014)</u>
<u>SS<sub>crit</sub></u>	<u>critical supersaturation (Köhler, 1936)</u>
<u>w</u>	<u>updraft velocity</u>
<u>Measured parameters</u>	
<u>w<sup>meas</sup><sub>act</sub></u>	<u>measured updraft velocity</u>
<u>Estimated parameters</u>	
<u>SS<sup>estim</sup><sub>peak</sub></u>	<u>estimated effective peak supersaturation derived from measurements (see Sect. 4.1)</u>
<u>w<sup>estim</sup><sub>act</sub></u>	<u>estimated updraft velocity derived from measurements and topography (see Sect. 4.1)</u>
<u>Modelled parameters</u>	
<u>SS<sup>mod</sup><sub>peak</sub></u>	<u>modelled effective peak supersaturation</u>
<u>SS<sup>ref</sup><sub>peak</sub></u>	<u>effective peak supersaturation obtained from the reference model simulation</u>
<u>SS<sup>max</sup><sub>mod</sub></u>	<u>maximum relative water vapour pressure between the model initialization point and the top of the cloud</u>
<u>w<sup>mod</sup><sub>act</sub></u>	<u>modelled updraft velocity</u>
<u>w<sup>divX</sup><sub>mod</sub></u>	<u>modelled updraft velocity divided by X</u>
<u>w<sup>mulX</sup><sub>mod</sub></u>	<u>modelled updraft velocity multiplied by X</u>
<u>SS<sup>fluc</sup><sub>peak</sub></u>	<u>modelled effective peak supersaturation applying the real-time fluctuations</u>
<u>SS<sup>fluc,sin</sup><sub>peak</sub></u>	<u>modelled effective peak supersaturation with a sinus function</u>

**Table 2.** Box model input parameters used for Fig. 3. The 25th, median and 75th percentile of the dry aerosol number size distribution were calculated binwise. The median and 75th percentile of the dry aerosol number size distribution resulted in a bimodal distribution and thus two modes are given.

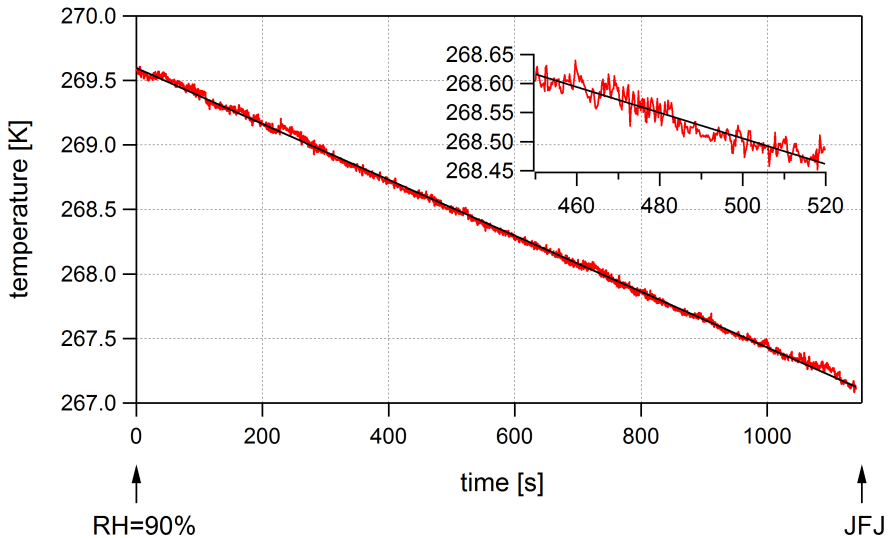
Measured parameter(s)		25th percentiles	median	75th percentiles
Hygroscopicity parameter [-]		0.19	0.26	0.37
Dry aerosol number size distribution	modes [nm] FWHM* [nm]	50 102	51, 131 179	46, 136 200
Temperature at the JFJ [K]		270	272	273
Pressure at the JFJ [hPa]		659.6	660.5	663.2
Total water content [ $\text{mg m}^{-3}$ ]		4110	4750	5200

\* Full width at half maximum.



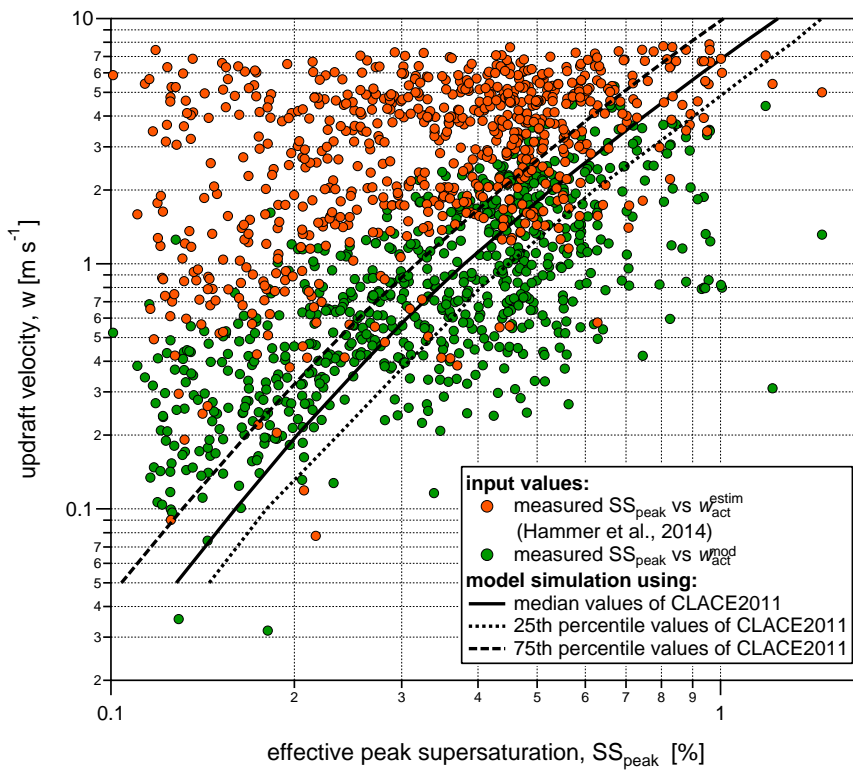
**Figure 1.** In (a) a panorama picture is shown to give an overview of the surrounding of the Jungfrauoch. The topography is shown in a sketch (b) along with the subsaturated conditions, conditions at the cloud base and at the Jungfrauoch. The green arrow shows the adiabatic backward calculations for the conditions at subsaturated conditions (initialization point of ZOMM;  $RH = 90\%$ ) with the measurements performed at the Jungfrauoch. The blue arrow shows the direction from the initialization point of the model until the end state of the simulation, which is at the Jungfrauoch. Brown dots indicate aerosol particles, blue dots cloud droplets.



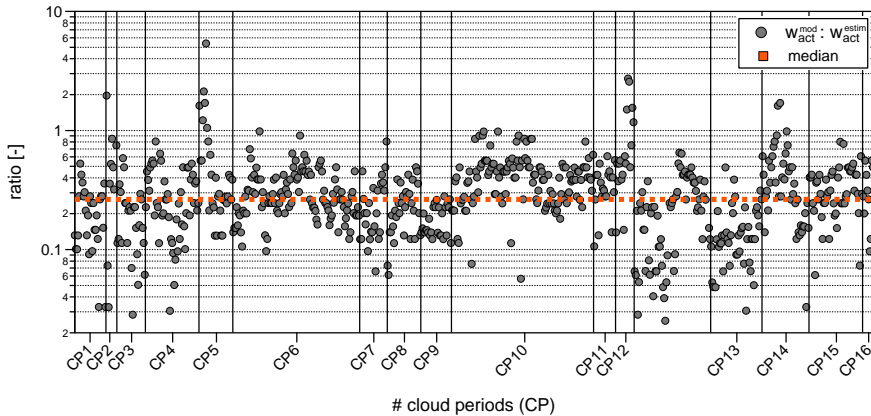


**Figure 2.** For the model run #516, which was detected on 8 August 2011 18:20 UTC, the temperature trajectory is shown with the **applied added** small-scale temperature fluctuation ( $T_{\text{turb}}$ ) retrieved from the sonic anemometer measurements (see detailed description in Sect. 2.3.1). The inset shows the trajectory on a smaller scale for a more quantitative view of the small-scale temperature fluctuations.

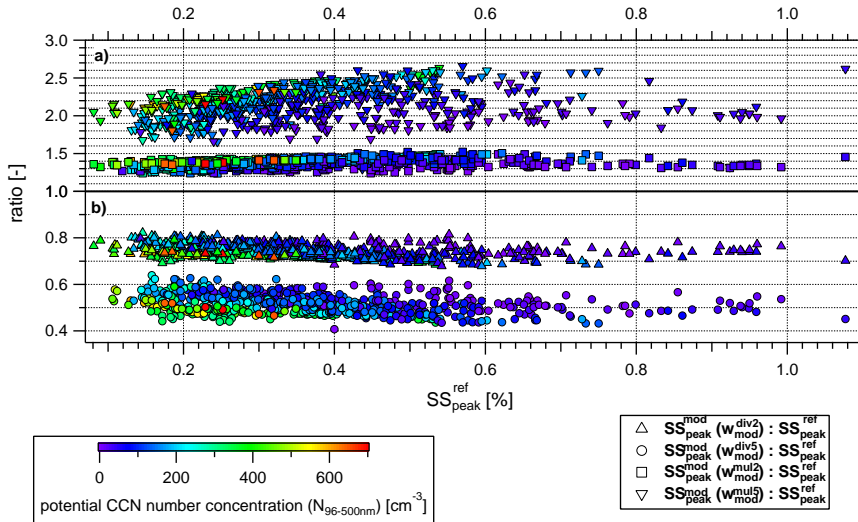
Ratio of the simulated updraft velocity ( $w_{\text{act}}^{\text{mod}}$ ) and the estimated updraft velocity at the cloud base ( $w_{\text{act}}^{\text{estim}}$ ) for each model simulation categorized for the different cloud periods (GP). The orange line indicates the median ratio of  $w_{\text{act}}^{\text{mod}} : w_{\text{act}}^{\text{estim}}$ .



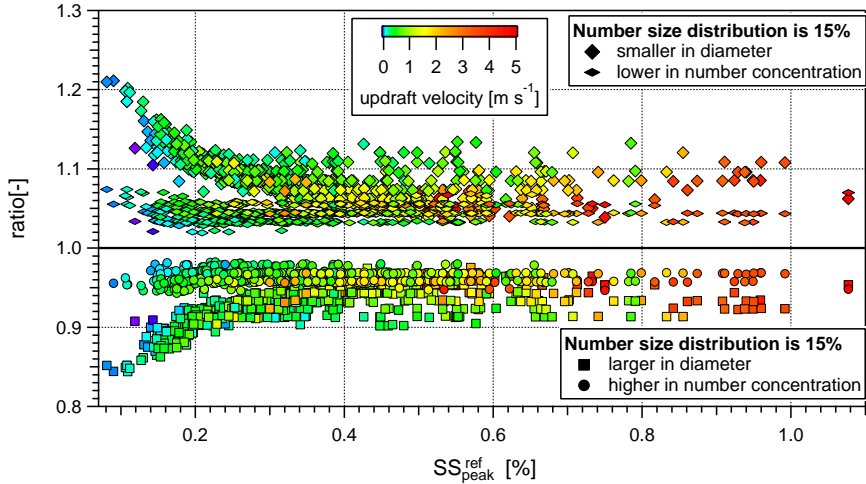
**Figure 3.** Each circle represents a trajectory calculation while the black lines show the trajectory calculations with the 25th, median and 75th values of the whole campaign given in Table 2. The relationship between the retrieved updraft velocity ( $w_{\text{act}}^{\text{estim}}$ ) and effective peak supersaturation ( $\text{SS}_{\text{peak}}^{\text{estim}}$ ) is given in red circles while the relationship of the simulated updraft velocity at cloud base ( $w_{\text{act}}^{\text{mod}}$ ) and  $\text{SS}_{\text{peak}}^{\text{estim}}$  is given in green circles.



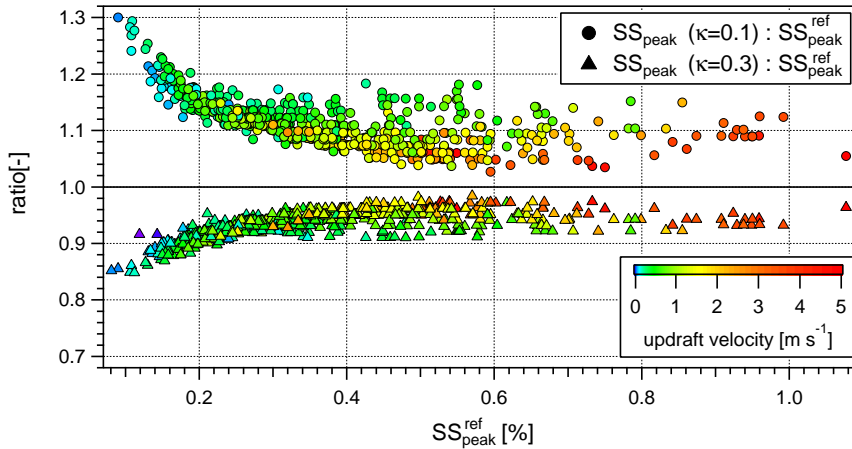
**Figure 4.** Ratio of modelled effective peak supersaturations using two different data sets of the simulated updraft velocities as input parameter: once divided and multiplied by 2-velocity ( $w_{act}^{div2}$ ,  $w_{act}^{mul2} w_{act}^{mod}$ ) and once divided and multiplied by 5 the estimated updraft velocity at the cloud base ( $w_{act}^{div5}$ ,  $w_{act}^{mul5} w_{act}^{estim}$ ) to for each model simulation categorized for the reference updraft velocities different cloud periods ( $w_{act}^{ref} CP$ ). The points are colour coded to show lines and the number concentration labels of particles in the size range of 96 (median dry activation of GLACE2011) CP serve to 500 (upper limit identify different cloud periods between which there are gaps of the SMPS) non-cloudy time. This is considered to be The orange line indicates the potential CGN number concentration median ratio of  $w_{act}^{mod} : w_{act}^{estim}$ .



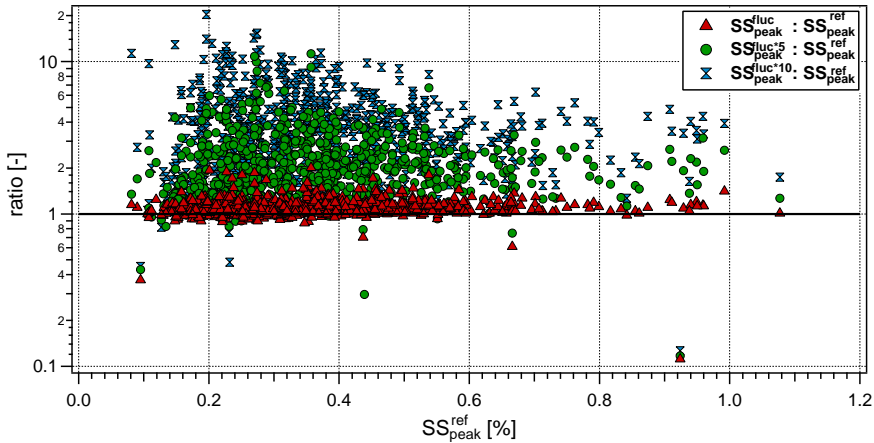
**Figure 5.** Ratio of modelled effective peak supersaturations using two different data sets of updraft velocities as input parameter: once divided and multiplied by 2 ( $w_{\text{act}}^{\text{div}2}$ ,  $w_{\text{act}}^{\text{mul}2}$ ) and once divided and multiplied by 5 ( $w_{\text{act}}^{\text{div}5}$ ,  $w_{\text{act}}^{\text{mul}5}$ ) to the reference updraft velocities ( $w^{\text{ref}}$ ). The points are colour coded to show the number concentration of particles in the size range of 96 nm (median dry activation diameter of CLACE2011) to 500 nm (upper limit of the SMPS). This is considered to be the potential CCN number concentration.



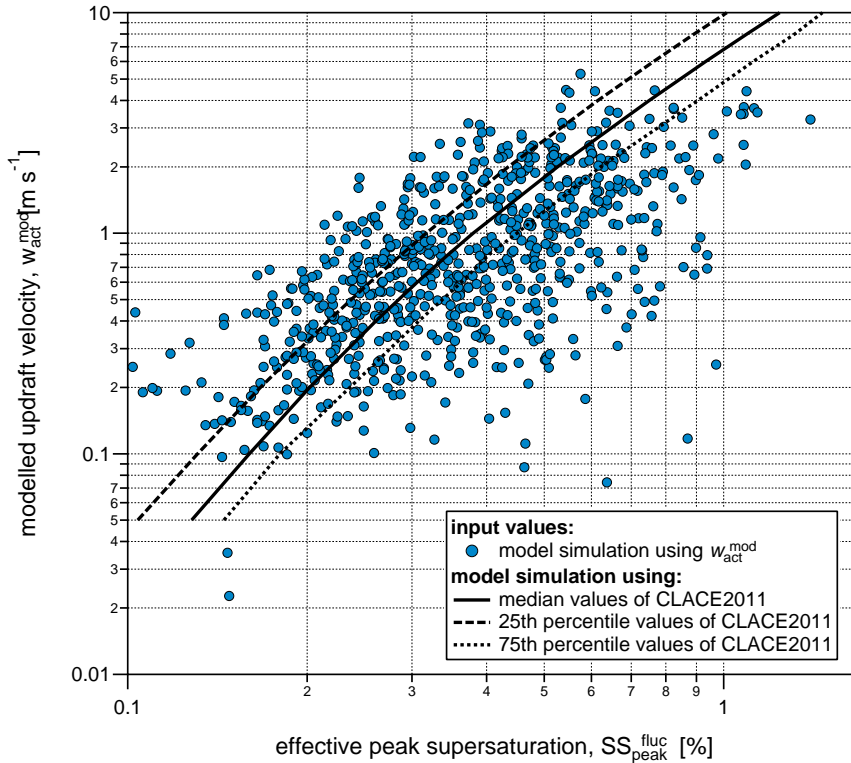
**Figure 6.** Ratio  $SS_{\text{peak}} : SS_{\text{peak}}^{\text{ref}}$  using: 15 % higher (circles) and lower (flat diamonds) particle number concentration compared to the measured one, and 15 % larger (squares) and smaller (diamonds) particles compared to the measured size distribution. All symbols are colour coded to show the modelled updraft velocities.



**Figure 7.** Ratio of effective peak supersaturation values using either a hygroscopicity value  $\kappa = 0.1$  and 0.3 to the median hygroscopicity parameter measured at the Jungfraujoch of  $\kappa = 0.2$ . The points of the datasets are colour coded to show the modelled updraft velocities.

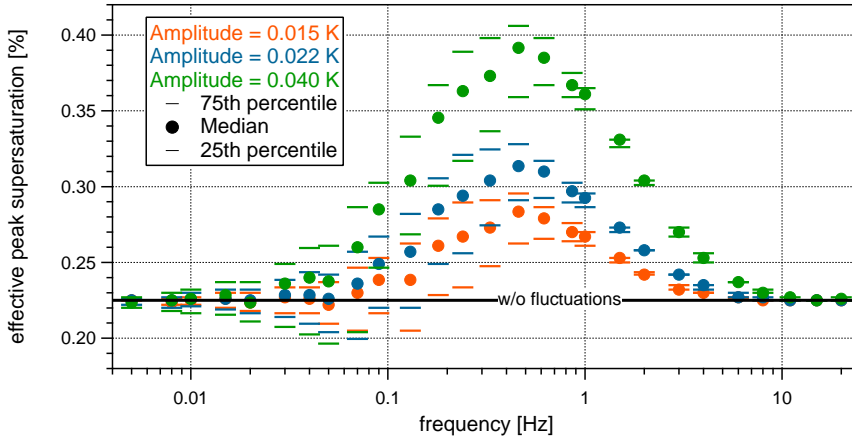


**Figure 8.** Ratio of modelled effective peak supersaturations applying small-scale fluctuations (obtained from the ultrasonic anemometer measurements) to the cooling rate, to the  $SS_{\text{peak}}^{\text{ref}}$ . The small scale-fluctuations are multiplied by 1 ( $SS_{\text{peak}}^{\text{fluc}}$ ; red triangles), 5 ( $SS_{\text{peak}}^{\text{fluc}\cdot 5}$ ; green triangles), and 10 ( $SS_{\text{peak}}^{\text{fluc}\cdot 10}$ ; blue triangles).

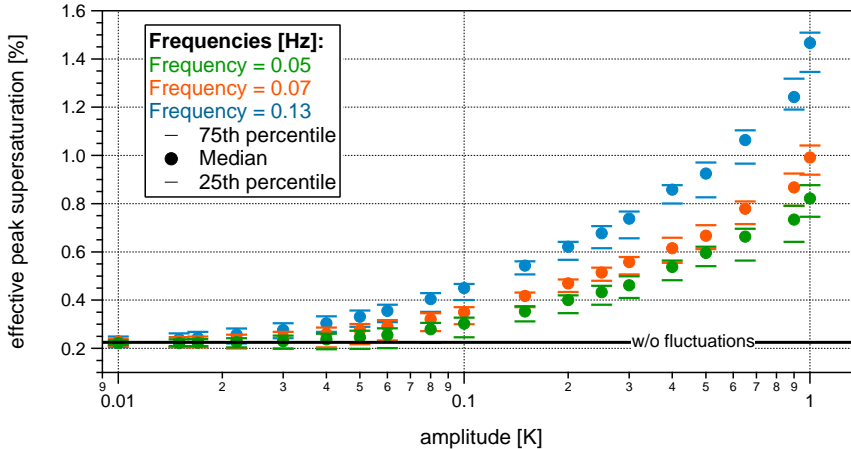


**Figure 9.** Same as Fig. 3 but the relationship between the modelled updraft velocity ( $w_{act}^{mod}$ ) and effective peak supersaturation taking into account the small-scale fluctuations ( $SS_{peak}^{fluc}$ ).

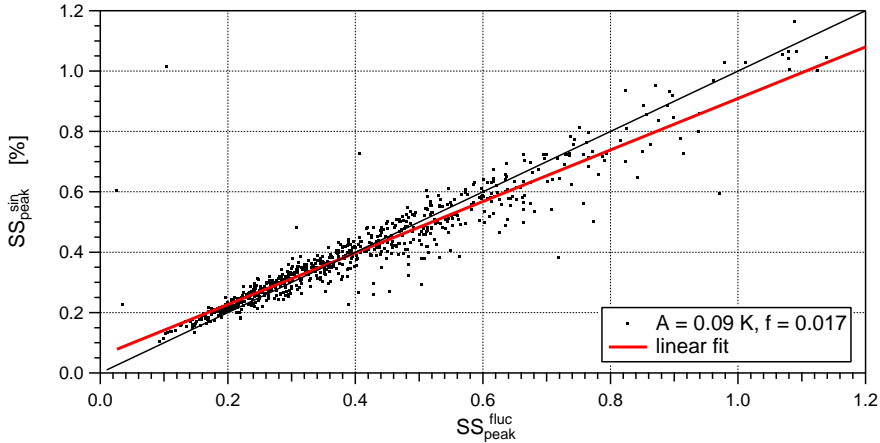




**Figure 10.** Dependency of the modelled effective peak supersaturations applying simulated small-scale fluctuations ( $SS_{\text{peak}}^{\text{fluc, sin}}$ ) to the cooling rate on the frequency. The applied small-scale fluctuation were obtained with a sinus function using three different amplitudes  $A = 0.015$  (red),  $0.022$  (blue) and  $0.04$  K (green), several frequencies in the range from  $0.05$  to  $20$  Hz and phases from  $0$  to  $360^\circ$  with  $18^\circ$  steps. The circles indicate the median values, while the bars show the 25th and 75th percentiles.



**Figure 11.** Dependency of the modelled effective peak supersaturations applying simulated small-scale fluctuations ( $SS_{\text{peak}}^{\text{fluc,sin}}$ ) to the cooling rate on the amplitude. The applied small-scale fluctuations were obtained with a sinus function using three different frequencies  $f = 0.05$  (green),  $0.07$  (red) and  $0.13$  Hz (blue), several amplitudes in the range from  $0.01$  to  $1.5$  K and phases from  $0$  to  $360^\circ$  with  $18^\circ$  steps. The circles indicate the median values, while the bars show the 25th and 75th percentiles.



**Figure 12.** Modelled effective peak supersaturations applying simulated small-scale fluctuations ( $SS_{\text{peak}}^{\text{fluc, sin}}$  obtained with a sinus function using an amplitude  $A = 0.022 \text{ K}$  and a frequency  $f = 0.24 \text{ s}^{-1}$ ) to the cooling rate vs. the one applying small-scale fluctuations obtained from the ultrasonic anemometer measurements ( $SS_{\text{peak}}^{\text{fluc}}$ ). The black line indicates the 1 : 1 line and the red line shows the linear fit.