

Review reply to 2nd review:

Sensitivity estimations for cloud droplet formation in the vicinity of the high alpine research station Jungfrauoch (3580 m asl) by E. Hammer et al.

We would like to thank to Reviewer #1 and the editor for their time and the comprehensive comments. Please find below the replies by the authors in green text.

In the previous review, I have called for a major revision of the paper, pointing out:

1. Limited reproducibility due to lack of access to software and data the study is based on, as well as due to the lack of sufficient detail in the description of simulation input parameters.
2. Lack of proper context even though the paper subject has been widely studied in the literature, the initial version of the manuscript lacked references to previous studies.
3. Paper composition flaws in the initial version of the manuscript.
4. Ambiguities in the description of the methodology that I tried to highlight by asking some specific questions.

In my opinion, the authors did address points 2 and 3 (as well as several points raised by the other reviewers) to an extent that makes the manuscript considerably better. Some of my specific questions from point 4 remain unanswered, and being convinced that clarifying these issues will improve the manuscript, I am repeating these questions in my comments to the authors' reply below. There is no improvement with regard to the first point - reproducibility. The authors have added in the revised manuscript an e-mail address of the model author. I do not consider it any better than having an e-mail address of the corresponding author. There is no information on the software version. The provided references do not give information on how the considered equations are numerically solved. Let me quote the ACP guidelines again: "Copernicus Publications encourages authors to also deposit software, algorithms, model code, and other underlying material on suitable repositories/archives whenever possible. These materials should be referenced in the article and preferably cited via a persistent identifier as a DOI."

We are still unable, for reasons previously stated, to publish the model. The ACP guidelines "encourage" authors to make models available "whenever possible". In this case it is not possible. We have checked back with the editor to confirm that this is acceptable. However, in the case of the MeteoSwiss data, we have found that after a registration process, while not publically available, it is available to the research community, free of charge (for anyone with an email address from a research institute or educational facility), via IDAWEB <https://gate.meteoswiss.ch/idaweb> . We have included this information in the paper.

Replies to the authors' comments

In the comments below, I address selected points from the authors' reply. The comments below contain:

quotes from my original review doubly indented and typeset in blue;
indigo-coloured quotes from the authors' reply with single indentation;
my present comments in black and with no indentation.

. . . subsaturated growth of the aerosol is governed by the kappa-koehler 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 . . .

Then, in my understanding, the κ -Köhler parameterisation has to be used as well above saturation. If that is correct, please do not state that it concerns only subsaturated growth.

If that is not correct, please clarify how the aerosol solubility/composition is taken into account above saturation.

This part of the paper was indeed still poorly worded and has now been modified.

At and above a saturation of .99, the κ -Köhler parameterisation is still used – from this the saturation over the droplets or aerosol is determined, allowing the calculation of the condensational flux of water. The text around equation 2 has been updated to clarify this (see also response to similar question about time step below).

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.

Please at least provide a persistent identifier and version information that can help to acquire the data from the mentioned entities. Please also note that according to the previously cited ACP guidelines, Authors are required to provide a statement on how their underlying research data can be accessed. This must be placed as the section "Data availability" at the end of the manuscript before the acknowledgements. If the data are not publicly accessible, a detailed explanation of why this is the case is required."

This section has been added as requested:

"The meteorological data used in this manuscript were provided by MeteoSwiss, the Swiss Federal Office of Meteorology and Climatology. They may be obtained via <https://gate.meteoswiss.ch/idaweb>, after a registration process. The model used here, ZOMM, may be obtained by contacting Dr. Beiping Luo (beiping.luo@env.ethz.ch). Data measured during the CLACE2011 campaign are available from the authors."

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."

First, I cannot find the added sentence in the revised manuscript.

This sentence was missing from the revised manuscript. As part of the changes made in response to a question above, it now reads "At $S < 0.99$ with respect to liquid water, the aerosol is assumed to be in equilibrium with the gas phase, and the model time step is one second. At $S > 0.99$, equation 2 is used to calculate the equilibrium vapour pressure over the aqueous phase droplets, and the flux of water between the droplets and the gas phase is calculated accounting for gas phase diffusion as well as the Kelvin effect. Based on the calculated flux, the model time step is determined, such that the water content of the droplet can change by no more than 2% per time step, enabling the model to resolve even high frequency fluctuations. "

Second, let me ask the authors again: does this condition result in a timestep value needed to resolve the response of condensation kinetics to the imposed 20 Hz frequency temperature fluctuations? Please mention it in the paper.

Yes, if flux is such that at any stage during a 20Hz fluctuation, the composition of the droplet would change by more than 2%, then the model will resolve it. See answer to previous point above.

The very last summarising paragraph of the paper starts with a statement saying that "*small-scale temperature fluctuations are revealed to be the strongest effect on cloud formation process beside the updraft velocity, which is influenced by the temperature fluctuations.*"

- I suggest rephrasing the above-quoted sentence so it is made clear that the employed model actually does not differentiate the temperature and velocity fluctuations. Done
- Such sentence also calls for a comment and reference[s] clarifying if that is in agreement or not with previous studies (see e.g. the summary and references in the review article on "Growth of Cloud Droplets in a Turbulent Environment" by Grabowski and Wang, 2013). We have added some references, stating that our findings broadly match those of previous studies, although fluctuations were not always found to be the first most important factor in aerosol activation. The Grabowski and Wang paper has been included in the changes related to the next point below.
- I also suggest supplementing the conclusions with a brief reiteration of the limitations of the employed modelling method: (i) assumed equilibrium state at RH=99%; (ii) no direct coupling between the latent heat release and the parcel heat budget; (iii) subjecting all particles to the same fluctuations.

The first paragraph of the Conclusions has been adapted to include these caveats.

Please also consider revising the abstract so that the list and priority of conclusions there matches the one from the final section of the paper. I suggest removing from the abstract the mention of the division by 4 of the velocity.

We would like to stick with the statement of the division by 4 of the updraft velocity since we think that this is an important finding for the JFJ location. As quite a few groups use data from the JFJ, this statement is interesting to a moderate number of people.

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, Kulmala et al. 1997, Feingold 2003, Lance et al., 2004, Chuang 2006, Ditas et al. 2012, Partridge et al. 2012.

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.

Let me point out that the list of references I provided was composed having in mind the subject of the sensitivity of CCN activation models, and in particular the sensitivity to the small-scale fluctuations of vertical velocity and temperature. Yet, in the revised manuscript, it is presented as a list of studies dealing with modelling of CCN activation in general. Please reformulate the first sentence on page 4 so it includes the mentions of sensitivity and small-scale fluctuations.

We reformulated the sentence according to your suggestions.

Also, I do encourage the authors to fill in the gap in the referenced literature between 1979 and 1997.

We have added some new citations from the late 80's and early 90's, including some parameterisations which were based only on aerosol number (for stratus clouds).

Citing Kulmala 1997 in my previous comment, I provided a quote from the paper in J. Aerosol Sci 28, and not the one in Nature (that's just to ensure if the different choice was intentional).

We used the wrong citation tag, we had intended to cite the JAS paper. This has now been corrected. 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)".

My point was to encourage shortening and/or rephrasing it.

Although the section is similar, we included it to save a reader from looking up the previous paper. As suggested by the editor, we removed the new sentence (quoted above) and added the reference at the end of the paragraph. We made some minor changes to the wording too.

Further comments We added all the comments listed below. Where no extra comment is added in green, the point has been addressed as requested.

- page 2, line 21: please add unit to 0.46 (Hz?) - last sentence of the abstract
- page 2, lines 13,23: repetition of "can significantly alter CCN activation"
- page 4, line 14: wrong tense for "find"
- page 5, line 18: "To develop effective models . . ." - please specify what kind of models
- page 6, line 17: I suggest moving the paragraph with reference to Table 1 from section 2 (Methods) to the end of section 1 (Introduction) and extending it to a brief description of the paper structure.
- page 7, line 14: parameter already mentioned while the work of Petters & Kreidenweis is cited only on page 10
- page 8, line 10: there is a "when when" repetition
- page 8, line 11: "updraught" spelling, while elsewhere "updraft"
- page 10: section 2.2 contains just a single subsection 2.2.1 { please skip subsectioning here
- page 10, line 17: isn't there a cause/effect mismatch in "Water removal due to precipitation is negligible since it is assumed that the total water content is preserved"
- page 11, line 11: please specify that the assumption refers to the dry size spectrum

- page 11, line 26: "i.e. equation" seems unneeded?
- page 12, line 13: what does "but" refer to in the last sentence of section 2.2.1? (also probably replacing "fast" with "fastest" will match authors' intent)
- page 14, 20, sect 2.3.4: sensitivity of on SS should probably be changed to sensitivity of SS
- on page 15: title and content of section 2.4: please replace "reference model" with "reference simulations" { the model is the same page 16: is "solid" (i.e. non-dashed) meant when referring to "black" lines?
- page 16, line 15: isn't it the velocity that has an effect on SS, and not the other way round?
- page 18, line 15: please rephrase "Thus, for the sensitivity..." to something like "To assess the sensitivity..."
- page 19, line 11: I suggest removing the "describing the Raoult term of the Kohler equation"
- page 20: last paragraph: please rephrase so that the reader is aware what "ratios" the text refers to
 We added: "The evaluation of the applied real-time fluctuation to the linear cooling rate compared to the reference model simulation revealed a dependence of these ratios on SS_{peak}^{ref} ."
- page 22 (vs. 10): ZOMM acronym is differently deciphered
- page 23, line 27: unit missing for 0.46
- references:
 - please differentiate between the two Hammer et al. 2014 papers when citing (e.g.,: 2014a, 2014b)
 - Clark and Hall 1979 entry is out of alphabetical order in the reference list
 - the Gerber 1991/JAS reference should likely be Gerber 1991/Apl. Opt., 30

Hope that helps!

Yes, it certainly did, and has led to further improvement of the manuscript.

Editor comment: While I think you have addressed well all comments by Reviewer #2 and #3 in the response to the reviews, there seems to be a mismatch of the response and the revised manuscript. I cannot find the following text fragments (that you had marked in blue) in the revised manuscript.

Reviewer #2:

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.

... effective peak supersaturation (SS_{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_{peak}

This sentence can be found at the end of the 2nd paragraph of the Introduction. However, the first sentence has been altered for better clarity.

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?

“(…) 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.”

We added this sentence now to the manuscript.

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.”

This sentence can be found in the first paragraph of Section “Modelled updraft velocity” in an altered way: “..., which was considered to be sufficient for the propagation of SS_{peak} values.

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.

This sentence can be found in the 3rd paragraph of Section “Influence of the updraft velocity, particle size distribution and hygroscopicity on the effective peak supersaturation” in an altered way: “In Sect. 2 it is described that the topography at the JFJ defines two main wind directions, NW and SE wind. The variability of the particle number concentration as well as the particle size is expected to be on the same order of magnitude as the difference between 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_{peak}^{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)”

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, 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_{peakref}, where the updrafts are generally higher, the critical saturation of the largest particles plays less of a role in determining the SS peak.”

We have now improved the explanation in this section to read as follows:

An increase in the size of the particles or an increase in the number concentration leads to a decrease in the SS_{peak}, relative to the SS_{peakref}, due to earlier activation of more particles. Similarly, smaller particles or lower number concentrations lead to an increased SS_{peak}. It is interesting to note that by changing the number concentration, the ratio between SS_{peak} and SS_{peakref} is modified by a fairly constant amount (approximately a 5% increase for lower number concentrations and a 5% decrease for higher number concentrations). Changing the diameter of the aerosol however, results in a greater change in SS_{peak} for points where SS_{peakref} is low (left hand side of Fig. 6). As shown by the colour coding, points with low SS_{peakref} were generally also those where the updraft velocity was smaller. Thus at low updraft velocity, a change in the aerosol size has a greater effect on the SS_{peak} reached, than at higher updraft velocity. This is because at low updraft velocities, the activated particles have more time to deplete the gas phase, and the SS_{peak} that is reached will be that required to activate only the largest particles. Any change in the size of the largest particles will translate directly into a change in the SS_{peak}. At higher updraft velocities however, a higher SS_{peak} is achieved, and more of the particle size distribution is activated. Thus the SS_{peak} reached is less sensitive to the size of the largest particles. 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 et al, 1989). Therefore the effect is relatively constant across the range of SS_{peakref}.

Editor comment: The only difference between this text and the text in the original manuscript is the beginning where 'This is because' was replaced by 'This can be explained by the fact', but even this change is not reflected in the revised version. In addition, I would like to see a more thorough response to the reviewer's comment.

Editor comment: Also, here I cannot find the following changes in the revised manuscript.

Reviewer #3:

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 $w_{actestim}$. 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 ...”

The sentence can be found in Sect. in an altered way: “This difference can most likely be explained by the fact that w_{act}^{estim} is an upper limit for the updraft velocity and w_{act}^{mod} is based on the simulated number of cloud droplets and the number of cloud residuals at the JFJ (as described in Sect. 2.3.2). Thus, w_{act}^{mod} is not an upper limit but the “true” updraft velocity at the point of aerosol activation.”

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 to the decrease ...”

The sentence can be found in Sect. 2.3.1 in an altered way for better clarity: “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 to the JFJ at time (t), the 20 Hz time resolved updraft

velocities measured by the sonic anemometer (w_{act}^{meas}) were applied to the linear temperature decrease derived from the lapse rate (T_{lin}).”

Editor comments (all addressed as requested, extra information added in green if necessary)

In addition, I have a few more comments you should consider in a revised manuscript.

(page and line numbers refer to the manuscript without marked changes)

p. 2, l. 18: do you mean 'if SS_{peak} is between 0.2-0.4%' ? (Otherwise, it might be read as 0.2-0.4% being the 'maximum influence')

p. 2, l. 20: independently of

p. 4, l. 14: find → found

p. 4, l. 18-p. 5, l.27: I appreciate that you added more references and context to the introduction.

However, a more thorough discussion and summary of the previous findings might be better. For example, you mention several times that various studies found highest sensitivity for polluted conditions. I suggest a more structured discussion instead of listing each finding from individual papers separately. We have made a few changes to the structure of this part of the introduction to group papers by topic better and link the similar findings. We have added a few more references too.

p. 6, l. 17: 'To make the readability of the manuscript easier' is an odd expression. Better 'For clarity' or simply remove this fragment. The description of the table has been shifted to the end of the introduction, following the request of the reviewer, and the odd fragment has been deleted.

p. 7, l. 24: If rosemount is a manufacturer name, it should be capitalized

p. 8, l. 9: Do you mean 'In the current study' by 'In here'? We have addressed all “In the current study” in the manuscript.

p. 8, l. 9-13: Something seems wrong with this sentence. Maybe the confusion arises because one of the 'when' (l. 10) should be a verb? This paragraph has been improved and now makes sense.

p. 8, l. 15: This is an odd expression in a paper. Is this paragraph taken from Hammer et al. (2014) or rephrased? If the former, it should be shortened; if the latter, simply add the reference at one or two places in the text.

p. 9, l. 17: lead → leads

p. 9, l. 19: hygroscopically grown

p. 9, l. 22: is there an 'of' missing between 'comparison' and 'SS(peak,mod)'?

p. 10, l. 3, 4: Since there is no Section 2.2.2, the headers should be combined and only a Section 2.2. should exist.

p. 10, l. 17/18: Shouldn't be this sentence the other way around? It is assumed that the total water content is preserved, since water removal due to precipitation is negligible.

P. 11, l. 16: ...size distributions at/below the cloud base? (Word missing)

p. 11, l. 19: Before, the campaign was only called 'CLACE', not 'CLACE2011' – please use consistently one of them

p. 12, l. 14/15: I don't understand this sentence.

p. 12, l. 25: Add space between 'Fig2' and 'with'

p. 13, l. 10: This sounds odd, as if a model run had been detected.

p. 14, l. 23/24: The two papers should be cited as (Hammer et al., 2014a, b). 'a)' should be the paper that is first cited in the manuscript, 'b)' the other one. Make sure, that throughout the whole manuscript you refer correctly then to either paper.

p. 16, l. 5-8: This sentence seems very convoluted. Either restructure it or split into two.

p. 18, l. 26: 0.4% We have substantially edited this section to clarify the explanation, as requested above.

p. 19, l. 26: 'Köhler theory' should be 'Köhler equation' as a theory does not have terms.

p. 22, l. 19: Specify 'it' – SS_{peak} or the updraft velocity?

p. 23, l. 27: 0.46%

p. 24, l. 7: Should 'influenced' be 'influential'?

p. 24, l. 5-10: The last sentence seems repetitive with the first one.

Thank you for these comments which have led to a further improvement of the manuscript.

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 (GLACECLACE2011) 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_{peak}), an important parameter for cloud activation, to key aerosol and dynamical parameters was investigated. The updraft velocity, which defines the cooling rate of an air parcel, was found to have the greatest influence on SS_{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_{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_{peak} . ~~Furthermore, there is a maximum of influence from turbulence on SS_{peak} between~~ The aerosol number concentration and hygroscopic properties were found to be less important than the aerosol size in determining SS_{peak} . Furthermore turbulence is found to have a maximum influence when SS_{peak} is between approximately 0.2 ~~–0.4~~ and 0.4 %.

Simulating the small-scale fluctuations with several amplitudes, frequencies and phases, revealed that independently ~~on~~ of the amplitude, the effect of the frequency on SS_{peak} shows a maximum at 0.46 Hz (median over all phases) and at higher frequencies, the maximum SS_{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_{peak} ; Hammer et al., 2014a) 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_{peak} , which is dependent on the cooling rate of the air parcel.

5 The influence of physical and chemical quantities on the [sensitivity of the number and size of cloud droplets to small scale fluctuations in temperature and saturation](#) 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~~ [supersaturation](#) 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~~ [found](#) 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.

20 [At higher particle number densities, the number of activated aerosol particles has been found to become more sensitive to the updraft velocity. For example,](#) Feingold et al. (2003) applied an adiabatic parcel model and ~~find~~ [found](#) that in non-precipitating stratocumulus cloud, at higher aerosol number densities (above a number corresponding to an extinction of approximately 0.008 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](#) [Similarly, 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.

With regard to the chemical composition of the aerosol, Lance et al. (2004) used a cloud parcel model to show 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. Likewise, 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. Under less polluted conditions, 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.

Several studies also exist which present good predictions of cloud droplet numbers by empirical equations based on the number of aerosol larger than a certain size. These studies imply that at least for stratus clouds, the aerosol number is the most important factor in determining the activated fraction and the number of cloud droplets formed (e.g. Raga and Jonsson

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%. Ranges of supersaturations were also measured by Politovich and Cooper (1988),

finding standard deviations in the 10 m average supersaturation of around 0.1–0.4 % depending on the amount of entrainment. They suggest that the range of the observed droplet size distributions may be accounted for by the variation in supersaturation.

The present study builds on the work of Hammer et al. (2014a), which showed that there is a strong link between SS_{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_{peak} . However, the study was not able to shed light into which extent each parameter contributed to SS_{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_{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., 2014a).

To develop cloud microphysical 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 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 of the turbulent strength and the updraft velocity on the cloud activation is investigated using a cloud parcel model.

In the next section, the methods used to collect observation data and calculate the derived quantities are described. A description of the model is also provided. The results of the study are discussed in section 3 and conclusions are presented in section 4. An

[overview of all the notation used in this manuscript, grouped into calculated, measured and modelled parameters is given in Table 1.](#)

2 Methods

2.1 Observational data

5 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 ([GLACECLACE2011](#)) 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](#)
10 [modelling study in here.](#)

Due to the topography around the Jungfraujoch (JFJ) mainly northwest (NW) and south-east (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).
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[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.](#)
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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 con-
25

tent 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°C). The total inlet sampled 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°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. 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 (κ ; [Petters and Kreidenweis \(2007\)](#)) was inferred (Hammer et al., 2014a).

The 3-dimensional wind speed vector at the JFJ with a time resolution of 20 Hz 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 wind 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.

The horizontal wind direction was obtained with the [rosemount Rosemount](#) pitot tube anemometer. This instrument is mounted at the top of a 10 m mast located at around

75 m away from the ultrasonic anemometer. 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 thermohygrometer Thygan VTP-37 (Meteolabor AG).

5 Cloud presence and LWC were measured with a particle volume monitor (PVM-100; Gerber, 1991).

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 % activated were excluded. This is the same procedure to that used by Hammer et al. (2014a).

15 ~~In here~~Here, only clouds ~~reaching the JFJ with~~observed at the JFJ under NW wind directions are considered. ~~Relatively few measurement points when when SE wind was present, and also because the clouds coming from the NW~~The clouds formed under NW wind conditions are mostly found to be formed locally by rapid ~~updraughts~~updrafts, in contrast to the clouds ~~from the south~~formed under southerly wind conditions, which are often stratus, ~~which has that have~~ been advected from further away. ~~This makes the clouds~~There are also rather few measurement points when SE wind was present, as the prevailing wind direction at the JFJ is from the NW~~more suitable for our study~~.

20

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. (2014a).)~~

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

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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 ($v_{\text{JFJ}}^{\text{h}}$):

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

where α 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 CLAGE2011~~ therefore this angle was used in the updraft estimation (Hammer et al., 2014a).

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_{eq}) over a solution droplet is described considering the Raoult (solute) and Kelvin laws. The critical supersaturation (SS_{crit}) of a particle with a certain size and composition (κ ; (Petters and Kreidenweis, 2007)) defines the point of activation from particle to cloud droplet. Therefore, all particles in an air parcel having a SS_{crit} smaller than SS_{peak} are able to activate and grow to cloud droplets. In the box model the S_{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 $\text{SS}_{\text{max}}^{\text{mod}}$. The simulated effective peak supersaturation, $\text{SS}_{\text{peak}}^{\text{mod}}$, however is below $\text{SS}_{\text{max}}^{\text{mod}}$. $\text{SS}_{\text{peak}}^{\text{mod}}$ was obtained by finding the highest water vapour saturation which ~~lead~~ leads 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~~ hygroscopically grown particles from cloud droplets (Jurányi et al., 2011; Henning et al., 2002).

It is important to note that in Hammer et al. (2014a) 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~~ fluctuations described in Sect. 3.3.1. The comparison of $SS_{\text{peak}}^{\text{mod}}$ obtained by the two definitions respectively, was within 10%.

5 The SS_{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 $\pm 30\%$ was estimated for SS_{peak} . A detailed
10 description how the SS_{peak} was estimated from the measurements performed at the JFJ can be found in Hammer et al. (2014a).

2.2 Box model ~~simulations~~ description (ZOMM)

2.2.1 ~~Box model description~~ (ZOMM)

The Zurich optical and microphysical model (ZOMM) was used in this study to simulate the
15 effect of aerosol properties and atmospheric dynamics on liquid cloud formation (~~please note, that no~~ ice formation was not simulated). ZOMM is a box model which calculates the evolution of an initial aerosol distribution along a temperature and pressure trajectory. A further basic 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.

20 For the initialisation of the model, the cloud periods detected at the JFJ were divided into six minute periods. 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. 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°C .

25 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 It is assumed that the total water content is preserved, since water removal due to precipitation was negligible (Hammer et al., 2014a). Assuming all the water is in the vapour phase, the dew point temperature T_d 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 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. (2014a).

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. To initialize the model under clearly subsaturated conditions, the temperature, and the corresponding pressure on the air parcel trajectory was calculated at $\text{RH} \approx 90\%$ to the cloud base of $\text{RH} = 100\%$ assuming a dry adiabatic lapse rate of $\Gamma_{\text{dry}} = 0.98 \text{ K (100 m)}^{-1}$. 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 $\Gamma_{\text{wet}} = 0.65 \text{ K (100 m)}^{-1}$. Implicit in this initialisation is the assumption that the dry aerosol size distribution observed at the JFJ is the same as that which was present below at the cloud base. As it is not feasible to measure the aerosol size distributions below the cloud base at the JFJ, this assumption 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 below 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 ~~GLAGE-2011~~ CLACE2011 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, The equilibrium water content of the aerosol~~ (i.e. ~~water content of the aerosol~~ the hygroscopic growth) is calculated according to the κ -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_{dry} the dry diameter, $\sigma_{\text{s/a}}$ the surface tension of the solution/air interface, ~~(0.072 Jm⁻², following Petters and Kreidenweis, 2007)~~, M_{w} the molar mass of water, R the ideal gas constant, T the prevailing air temperature and ρ_{w} the density of water.

~~At $S > 0.99$ At $S < 0.99$~~ with respect to liquid water, ~~or at high cooling rates, the aerosol is assumed to be in equilibrium with the gas phase and the model time step is one second. At $S > 0.99$, equation 2 is used to calculate the equilibrium vapour pressure over the aqueous phase droplets, and the flux of water between the droplets and 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. Based on the calculated flux, the model time step is determined, such that the water content of the droplet can change by no more than 2% per time step, enabling the model to resolve even high frequency fluctuations.

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 effective peak supersaturation pre-

dicted 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 modelled peak supersaturation is defined based on the droplet size at the point droplets grow fast~~ point of activation of the smallest aerosol (i.e. at point of activation), the highest supersaturation reached which resulted in droplet activation), not on the final droplet size.

2.3 Investigated parameters

2.3.1 Simulated small-scale temperature fluctuations

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 to the JFJ at time (t), the 20 Hz time resolved updraft velocities measured by the sonic anemometer ($w_{\text{act}}^{\text{meas}}$) were applied to the linear temperature decrease derived from the lapse rate (T_{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 solid 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:

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

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_{start} (the time at which the model was initialized) to t_{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 calculated by multiplying $w'(t)$ with the wet adiabatic lapse rate ($\Gamma_{\text{wet}} = 0.65 \text{ K (100 m)}^{-1}$). Adding $T'_{\text{turb}}(t)$ to $T_{\text{lin}}(t)$ leads then to $T_{\text{turb}}(t)$. $T'_{\text{turb}}(t)$ ranged from -0.8 to 1.1 K with a 25th percentile of 0.01 K , a 75th per-

centile of 0.05 K and a median of 0.03 K. Figure 2 shows an example of T_{turb} for the model run [detected at the JFJ](#) [simulated for conditions](#) on 8 August 2011 18:20 UTC [at the JFJ](#).

2.3.2 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_{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 SS_{peak} (see results in Sect. 3.2).

2.3.3 Aerosol- and updraft-limited regimes

Previous studies have found that a high SS_{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 SS_{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 different regimes depending on the ratio between the updraft velocity and the particle number concentration (w/N_{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_{CN} , by a high activated fraction of aerosol particles (larger than 90%) and the aerosol-limited regime is basically independent of w . The high updraft velocities lead to high SS_{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_{CN} (smaller than 20%), saying that only a few particles are activated

to cloud droplets due to low SS_{peak} values. In this regime the cloud droplet number concentration exhibits a linear dependence on w and a weak dependence on the N_{CN} . The aerosol- and updraft-sensitive regime is characterized by w/N_{CN} values lying between the two other regimes. Depending on SS_{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 SS_{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 SS_{peak} (see results in Sect. 3.2).

2.3.4 Hygroscopicity parameter

The hygroscopicity parameter, κ , stays rather constant over time at the Jungfraujoch at around 0.2 (Jurányi et al., 2011). To investigate the sensitivity of κ on SS_{peak} , a typical κ 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. (2014a) and Hammer et al. (2014b) revealed only a small influence of the κ value on the calculated SS_{peak} .

2.4 Reference model simulations for sensitivity analysis

For the sensitivity studies shown in Sects. 3.2 and 3.3.1 a set of reference model simulations was used. These reference simulations were performed using the dataset measured at the JFJ during CLACE2011 as input variables. For this purpose an average, constant κ 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. 2.3.2. All output parameters of the reference model simulations are depicted with a superscript $^{\text{ref}}$, 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_{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. (2014a) simulated SS_{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_{peak} was generally overestimated for a particular $w_{\text{act}}^{\text{estim}}$ (see Fig. 3; red circles and solid 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. 2.3.2. 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. (2014a), 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_{peak} (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_{peak} . ~~The fact that when~~ When the model is constrained to reproduce the observed number of droplets, a lower updraft velocity is found, ~~causing~~. This then causes a better agreement between modelled and estimated SS_{peak} , ~~suggests~~ suggesting that the updraft velocity estimated from wind speed measurements at the JFJ is indeed overestimated.

The black curves in Fig. 3 represent the box model simulations of $SS_{\text{peak}}^{\text{mod}}$ obtained by running the simulations for a range of constant updraft velocities. In the upper, dashed line, the aerosol size distribution was chosen so that the number and sizes of the aerosol and κ value were representative of the 75th percentile of those observed during CLACE 2011. The bottom CLACE2011. The bottom, dashed 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 updraft velocity on SS_{peak} on-updraft-velocity can be seen. The solid 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 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 most likely has on the SS_{peak} .

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

According to previous studies, a 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).

In Sect. 3.1 it was shown that $w_{\text{act}}^{\text{mod}}$ is on average a factor of 4 lower than the estimated $w_{\text{act}}^{\text{estim}}$. 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. 2.3.2). 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_{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_{peak} is on average a factor of 2 larger compared to using w^{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_{peak} was observed. Low SS_{peak} values are less affected by the updraft velocity because for low SS_{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_{act} is slightly larger when the potential CCN number concentration 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 $SS_{\text{peak}}^{\text{ref}}$ (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.

In Sect. 2 it is described that the topography at the JFJ defines two main wind directions, NW and SE wind. The variability of the particle number concentration as well as the particle

size is expected to be on the same order of magnitude as the difference between 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 To assess 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 An increase in the size of the particles or an increase in the number concentration leads to a decrease in the SS_{peak} , relative to 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.

, due to earlier activation of more particles. Similarly, smaller particles or lower number concentrations lead to an increased SS_{peak} . It is interesting to note that while by changing the number of the particles has a relatively constant effect on the modelled concentration, the ratio between SS_{peak} , changing the size of the particles has a much more pronounced effect at low and $SS_{\text{peak}}^{\text{ref}}$ is modified by a fairly constant amount (approximately a 5% increase for lower number concentrations and a 5% decrease for higher number concentrations). Changing the diameter of the aerosol however, results in a greater change in SS_{peak} for points where $SS_{\text{peak}}^{\text{ref}}$ is low (left hand side of Fig. 6). As shown by the colour coding, points with low $SS_{\text{peak}}^{\text{ref}}$ were generally also those where the updraft velocity was smaller. Thus at low updraft velocity, a change in the aerosol size has a greater effect on the SS_{peak} reached, than at higher updraft velocity. 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 at low updraft velocities, the activated particles have more time to deplete the gas phase, and the SS_{peak} that is reached will be that required to activate only the largest particles activate. If they are smaller (larger) SS_{peak} will be higher (lower). Any change in the size of the largest particles will translate directly into a change in the SS_{peak} . 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 updraft velocities however, a higher SS_{peak} is achieved, and more of the particle size distribution is activated. Thus the SS_{peak} reached is less sensitive to the size of the largest particles.

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_{peak} is the hygroscopicity parameter of the dry particles, κ , 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., 2014a) at $\kappa \approx 0.2$. To look into the sensitivity of SS_{peak} to κ , a typical κ 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_{peak} values compared to the reference size distribution ($SS_{\text{peak}}^{\text{ref}}$; see Fig. 7). On average the SS_{peak} is lowered by 6 %, however, for smaller $SS_{\text{peak}}^{\text{ref}}$ the effect of a larger κ value is stronger and lowers the SS_{peak} up to 15 %. The model simulations using a κ value of 0.1 show on average 11 % higher SS_{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 equation](#) (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 κ on small SS_{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_{peak} (see Fig. 7), the critical saturation at which the largest particles activate plays a more important role in determining the final SS_{peak} than it does at higher updraft velocities. The changes in κ result in changes in this critical saturation, therefore the changes in κ 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 added to the fluctuation) the SS_{peak} increases significantly: applying the real-time fluctuation to the cooling rate raises the SS_{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 added to the cooling rate by a factor of 5 increases the SS_{peak} by $\sim 87\%$ and multiplying the fluctuations by 10 increases the SS_{peak} by a factor of ~ 3.22 (see green and blue triangles in Fig. 8, respectively). The factors 5 and 10 [are resulting result](#) in a similar range of temperature amplitudes used for the sinus curve simulations described in Sect. 3.3.2.

[AThe evaluation of the applied real-time fluctuation to the linear cooling rate revealed a dependence of the ratios of modelled effective peak supersaturations applying small-scale](#)

fluctuations 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 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 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 $SS_{\text{peak}}^{\text{fluc. } x} : SS_{\text{peak}}^{\text{ref}}$ 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_{peak} from the simulation including fluctuations will be close to the SS_{peak} calculated from the reference simulation

Figure 3 showed that for small $w_{\text{act}}^{\text{mod}}$ the model was slightly underestimating the SS_{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 5 s^{-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

Figure 10 shows the dependency of SS_{peak} on simulated small-scale fluctuations added to the cooling rate using a certain frequency (f), amplitude (A) and phase (ϕ). 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 of 0

to 360° with 18° 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 $SS_{\text{peak}}^{\text{fluc,sin}}$ 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** decreases since the fluctuation is faster than the time required for significant droplet growth. Likely for the same reason the range of $SS_{\text{peak}}^{\text{fluc,sin}}$ (25th 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 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).

Several combinations of amplitudes and frequencies for sinus functions were found **being able to** that could represent the median small-scale fluctuations in the vicinity of the JFJ. Figure 12 shows the relationship of the modelled SS_{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 $f = 0.022 \text{ s}^{-1}$. The good 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 and 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. ZOMM is a air parcel model, and as such, does not simulate processes such as sedimentation of particles and mixing of air masses. As pointed out by Grabowski and Wang (2011) references therein, subjecting all particles or droplets to the same temperature fluctuations, as done in this study, does not fully represent the processes of turbulent or gravitation mixing of droplets which may lead to broadening of the droplet size distributions. ZOMM

calculates the flux of water to and from aerosol and droplets at RH above 99%, while assuming equilibrium with the gas phase at lower RH. There is no direct coupling between the latent heat release of the droplets and the parcel heat budget, although the latent heat release is taken into account when calculating the temperature trajectories used to run the model.

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

Another input parameter influencing the SS_{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_{peak} only to a small extent up to 21 %. It was observed that the 15 %-change in particle size had a stronger influence on the SS_{peak} values at lower updraft velocities than the 15 %-change in number concentration. The influence of the hygroscopicity on SS_{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 κ 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 κ showed a stronger influence on SS_{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_{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 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 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 of temperature raise the SS_{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_{peak} by $\sim 87\%$ and multiplying the fluctuations by 10 increases the SS_{peak} by a factor of ~ 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_{peak} shows a maximum at 0.46 Hz (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 Jungfraujoch were simulated based on several sinus functions with combinations of amplitudes and frequencies. 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 s^{-1} .

Summarizing, small-scale temperature fluctuations, produced by variations in vertical wind velocity, 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 activation.~~ Previous studies have also highlighted such fluctuations as having a strong effect on the number of aerosol activated, although it has not always been found to be the strongest factor influencing aerosol activation (e.g. Lance et al., 2004; Antilla et al., 2012). The aerosol number concentration and hygroscopic properties ~~occurred were found~~ to be less ~~influenced compared to~~ important than 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).~~ in determining activation and peak supersaturation.

Data Availability

The meteorological data used in this manuscript were provided by MeteoSwiss, the Swiss Federal Office of Meteorology and Climatology. They may be obtained via <https://gate.meteoswiss.ch> after a registration process. The model used here, ZOMM, may be obtained by contacting Dr. Beiping Luo (beiping.luo@env.ethz.ch). Data measured during the CLACE2011 campaign are available from the authors.

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Table 1. List of important symbols

| Parameter | Notation |
|--------------------------------------|----------------------------------------------------------------------------------------------------|
| General parameters | |
| SS | supersaturation |
| SS_{peak} | effective peak supersaturation (Hammer et al., 2014a) |
| SS_{crit} | critical supersaturation (Köhler, 1936) |
| w | updraft velocity |
| Measured parameters | |
| $w_{\text{act}}^{\text{meas}}$ | measured updraft velocity |
| Estimated parameters | |
| $SS_{\text{peak}}^{\text{estim}}$ | estimated effective peak supersaturation derived from measurements (see Sect. 2.3) |
| $w_{\text{act}}^{\text{estim}}$ | estimated updraft velocity derived from measurements and topography (see Sect. 2.3) |
| Modelled parameters | |
| $SS_{\text{peak}}^{\text{mod}}$ | modelled effective peak supersaturation |
| $SS_{\text{peak}}^{\text{ref}}$ | effective peak supersaturation obtained from the reference model simulation |
| $SS_{\text{mod}}^{\text{max}}$ | maximum relative water vapour pressure between the model initialization point and the current time |
| $w_{\text{act}}^{\text{mod}}$ | modelled updraft velocity |
| $w_{\text{mod}}^{\text{div}X}$ | modelled updraft velocity divided by X |
| $w_{\text{mod}}^{\text{mul}X}$ | modelled updraft velocity multiplied by X |
| $SS_{\text{peak}}^{\text{fluc}}$ | modelled effective peak supersaturation applying the real-time fluctuations |
| $SS_{\text{peak}}^{\text{fluc,sin}}$ | modelled effective peak supersaturation with a sinus function |

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 [mg m^{-3}] | | 4110 | 4750 | 5200 |

* Full width at half maximum.

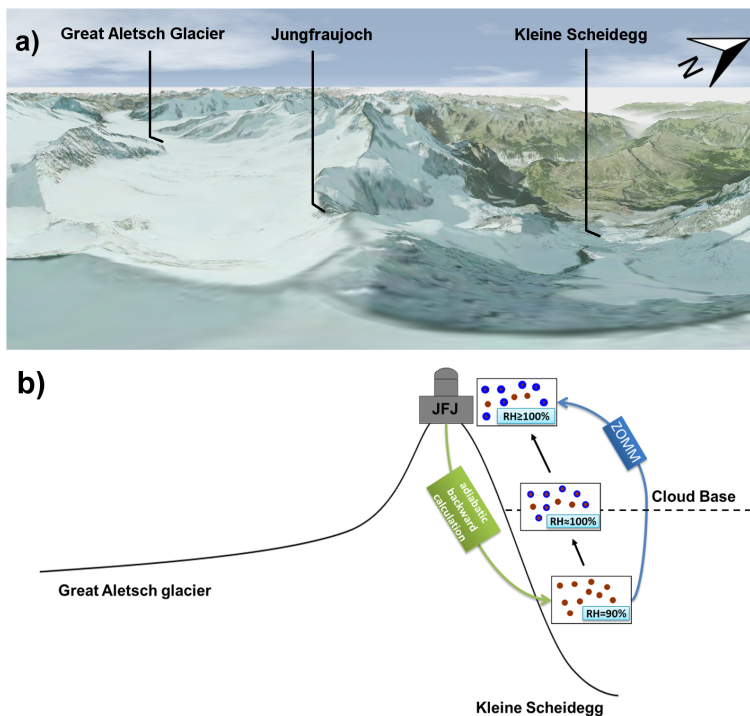


Figure 1. In (a) a panorama picture is shown to give an overview of the surrounding of the Jungfrauoch. The topography is shown in a sketch (b) along with the subsaturated conditions, conditions at the cloud base and at the Jungfrauoch. The green arrow shows the adiabatic backward calculations for the 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.

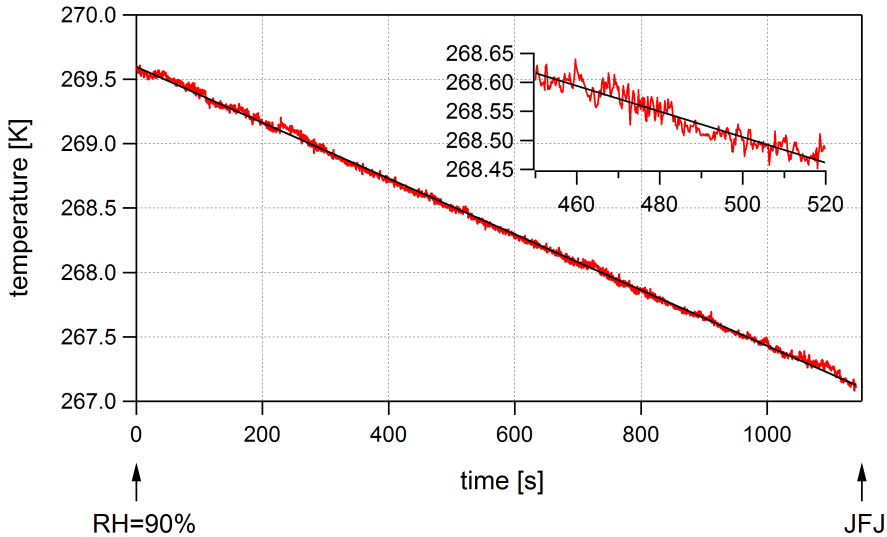


Figure 2. For the model run #516, which was detected on 8 August 2011 18:20 UTC, the temperature trajectory is shown with the added small-scale temperature fluctuation (T_{turb}) retrieved from the sonic anemometer measurements (see detailed description in Sect. 2.3.1). The inset shows the trajectory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

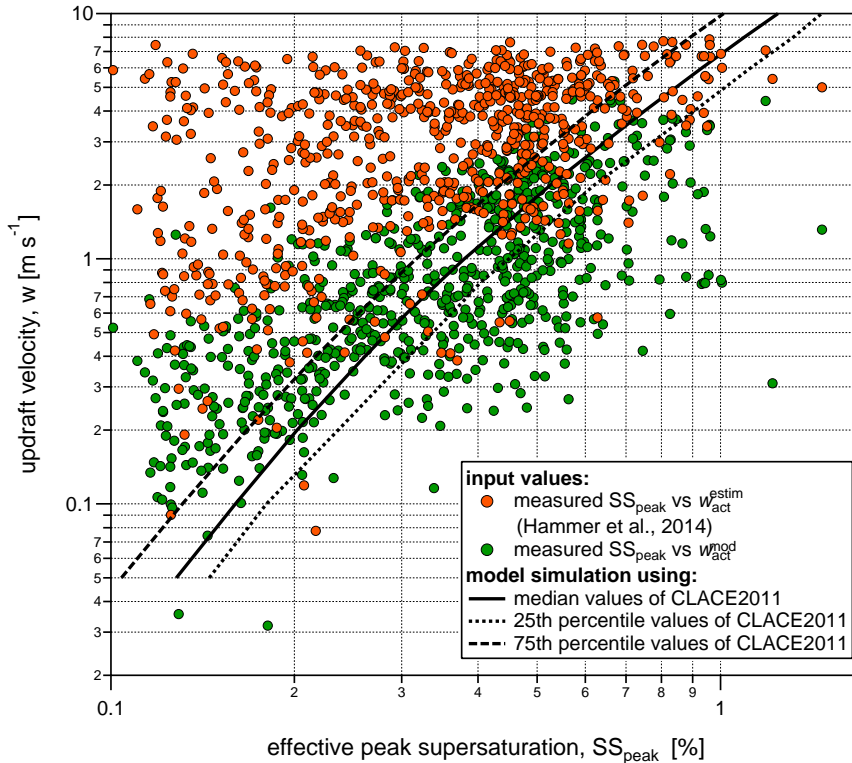


Figure 3. Each circle represents a trajectory calculation while the black lines show the trajectory calculations with the 25th, median and 75th values of the whole campaign given in Table 2. The relationship between the retrieved updraft velocity ($w_{\text{act}}^{\text{estim}}$) and effective peak supersaturation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

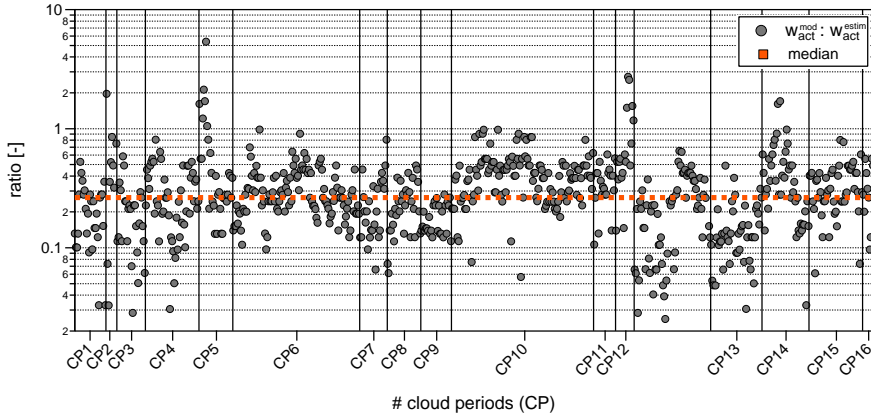


Figure 4. Ratio of the simulated updraft velocity (w_{act}^{mod}) and the estimated updraft velocity at the cloud base (w_{act}^{estim}) for each model simulation categorized for the different cloud periods (CP). The lines and the labels of CP serve to identify different cloud periods between which there are gaps of non-cloudy time. The orange line indicates the median ratio of $w_{act}^{mod} : w_{act}^{estim}$.

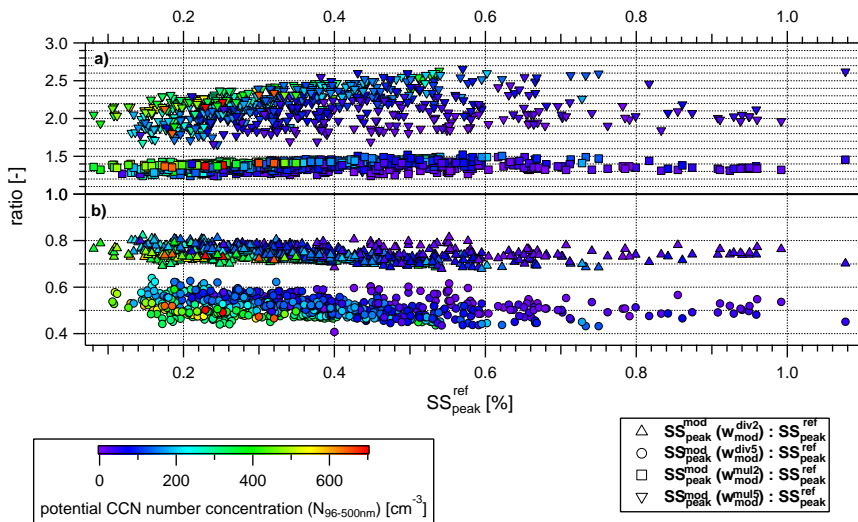


Figure 5. Ratio of modelled effective peak supersaturations using two different data sets of updraft velocities as input parameter: once divided and multiplied by 2 ($w_{\text{act}}^{\text{div}2}$, $w_{\text{act}}^{\text{mul}2}$) and once divided and multiplied by 5 ($w_{\text{act}}^{\text{div}5}$, $w_{\text{act}}^{\text{mul}5}$) to the reference updraft velocities (w^{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.

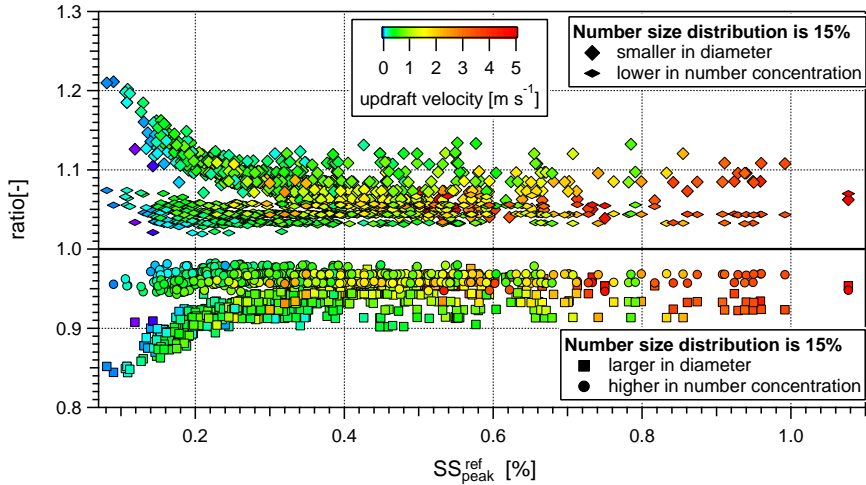


Figure 6. Ratio $SS_{\text{peak}} : SS_{\text{peak}}^{\text{ref}}$ using: 15 % higher (circles) and lower (flat diamonds) particle number concentration compared to the measured one, and 15 % larger (squares) and smaller (diamonds) particles compared to the measured size distribution. All symbols are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

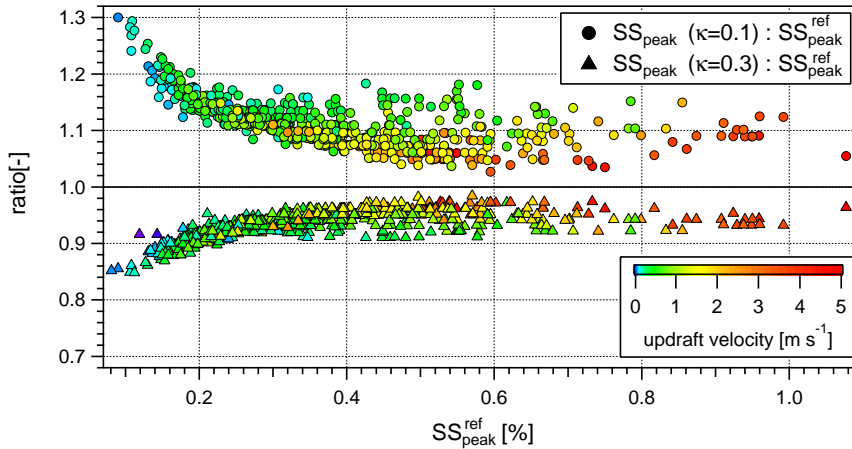


Figure 7. Ratio of effective peak supersaturation values using either a hygroscopicity value $\kappa = 0.1$ and 0.3 to the median hygroscopicity parameter measured at the Jungfraujoch of $\kappa = 0.2$. The points of the datasets are colour coded to show the modelled updraft velocities.

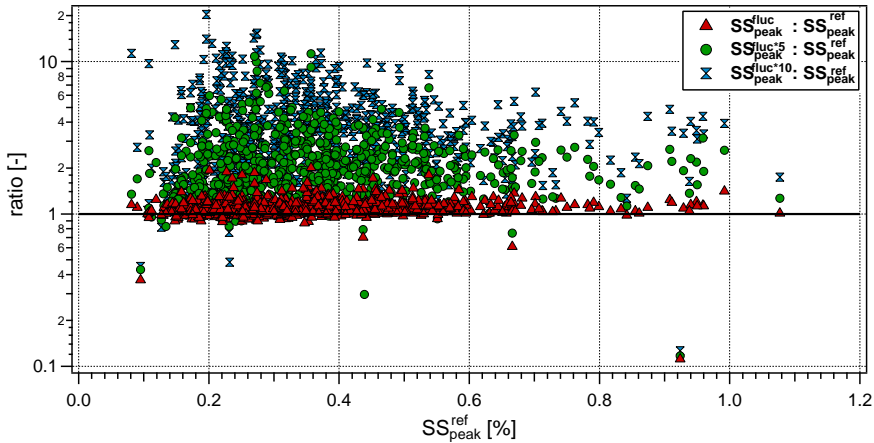


Figure 8. Ratio of modelled effective peak supersaturations applying small-scale fluctuations (obtained from the ultrasonic anemometer measurements) to the cooling rate, to the $SS_{\text{peak}}^{\text{ref}}$. The small scale-fluctuations are multiplied by 1 ($SS_{\text{peak}}^{\text{fluc}}$; red triangles),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

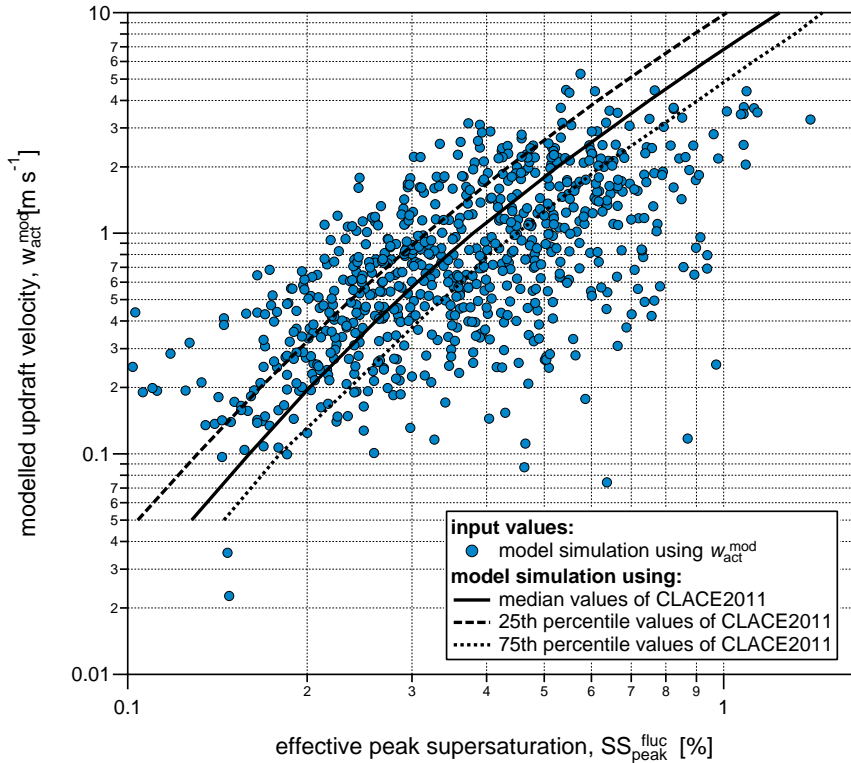


Figure 9. Same as Fig. 3 but the relationship between the modelled updraft velocity ($w_{\text{act}}^{\text{mod}}$) and effective peak supersaturation taking into account the small-scale fluctuations ($SS_{\text{peak}}^{\text{fluc}}$).

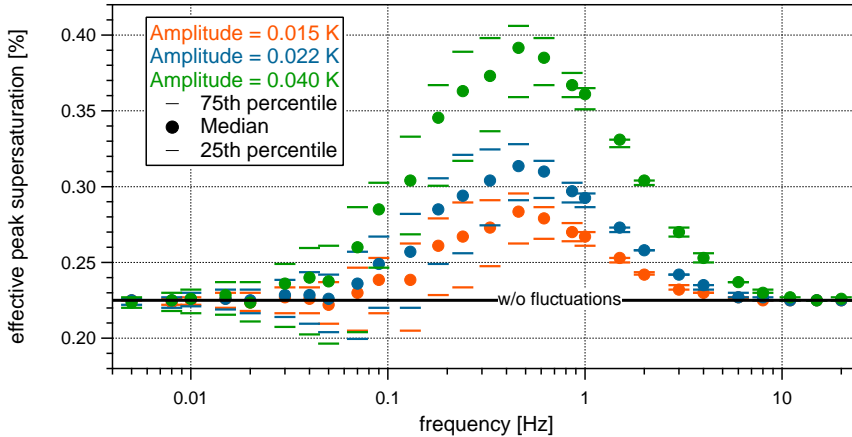


Figure 10. Dependency of the modelled effective peak supersaturations applying simulated small-scale fluctuations ($SS_{\text{peak}}^{\text{fluc, sin}}$) to the cooling rate on the frequency. The applied small-scale fluctuation were obtained with a sinus function using three different amplitudes $A = 0.015$ (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° with 18° steps. The circles indicate the median values, while the bars show the 25th and 75th percentiles.

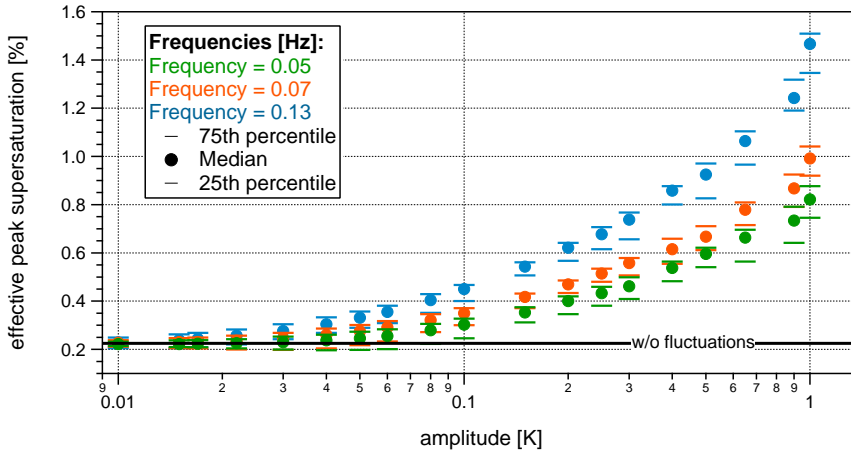


Figure 11. Dependency of the modelled effective peak supersaturations applying simulated small-scale fluctuations ($SS_{\text{peak}}^{\text{fluc,sin}}$) to the cooling rate on the amplitude. The applied small-scale fluctuation were obtained with a sinus function using three different frequencies $f = 0.05$ (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° with 18° steps. The circles indicate the median values, while the bars show the 25th and 75th percentiles.

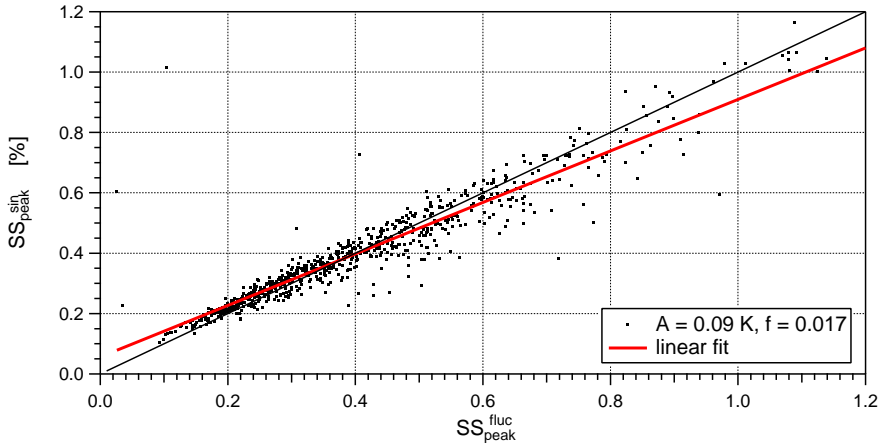


Figure 12. Modelled effective peak supersaturations applying simulated small-scale fluctuations ($SS_{\text{peak}}^{\text{fluc, sin}}$ obtained with a sinus function using an amplitude $A = 0.022 \text{ K}$ and a frequency $f = 0.24 \text{ s}^{-1}$) to the cooling rate vs. the one applying small-scale 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.