Betreff: Re: corrections for acp-2022-434
Von: Daniel Knopf <daniel.knopf@stonybrook.edu>
Datum: 06.01.23, 17:46
An: Peter Spichtinger <spichtin@uni-mainz.de>, ACP Editorial <editorial@copernicus.org>

Dear Prof. Peter Spichtinger,

Indeed, there is significant repetition of context in this paragraph. After careful reviewing and looking at Fig. 2, I am fine with your suggestion to delete the highlighted text sections in the accepted manuscript.

This note and email could be added to your page proofing document to inform the Editorial.

With best regards,

Daniel Knopf

On Fri, Jan 6, 2023 at 7:13 AM Peter Spichtinger <spichtin@uni-mainz.de> wrote:

> Dear Dr Knopf,
>
> I apologize for bothering you again.
>
> During the preparation of the files for the production office I realized that we wrote more or less the same content three times within one paragraph. Therefore I would like to delete some parts of the text. Since this is a change in the already accepted manuscript, I wanted to ask you if this is OK with you. I have attached the relevant page, the marked text would be deleted.
>
> Please let me know if this is OK with you and if (and how) I have to communicate this to the editorial office.
>
> Thank you very much in advance.
>
> Best regards,
> Peter
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(ii) a Taylor expansion at a prescribed value y_0. While the first approach is just a fitting procedure in the relevant range $0.26 \leq \Delta a_w \leq 0.34$, the second approach relies on an a priori choice for the evaluation point $y_0 \in [0.26, 0.34]$ and it is not evident from the outset which value should be used to provide an accurate approximation. For this, we investigate the sensitivity of p_3 to a small perturbation $\varepsilon = y - y_0$, i.e. we consider

$$p_3(y) = p_3(y_0+\varepsilon) = p_3(y_0) + \left.\frac{\mathrm{d}p_3}{\mathrm{d}x}\right|_{y_0} \varepsilon + \mathcal{O}\left(\varepsilon^2\right) \quad (35)$$

$$\approx b_{t0} + b_{t1}\cdot y = p_{t,y_0}(y) \quad (36)$$

with the coefficients

$$b_{t0} = p_3(y_0) - \left.\frac{\mathrm{d}p_3}{\mathrm{d}x}\right|_{y_0}\cdot y_0 \quad \text{and} \quad b_{t1} = \left.\frac{\mathrm{d}p_3}{\mathrm{d}x}\right|_{y_0}. \quad (37)$$

The Taylor approximation provides a range for the slope of the linear approximation; these values motivate the sensitivity analysis in section 4.5.2. In the relevant range $0.26 \leq y \leq 0.34$ for $y = \Delta a_w$ we obtain slopes in the range $221 \leq b_{t1} \leq 453$. This investigation gives us a hint about possible variations in the slope which will be used later for the sensitivity analysis in section 4.5.2.

In contrast, using a least square fitting routine for $0.26 \leq \Delta a_w \leq 0.34$ we obtain a linear function

$$p_{\mathrm{ls}}(x) = b_{\mathrm{ls},0} + b_{\mathrm{ls},1}\cdot x \quad (38)$$

with $b_{\mathrm{ls},0} = -62.19267$ and $b_{\mathrm{ls},1} = 254.7749$. For the further investigations, we only use the linear fit from eq. (38). We observe that the linear fit $p_{\mathrm{ls}}(x)$ best approximates p_3 close to the inflection point $x_{\mathrm{infl}} \approx 0.30756$ (see figure 2, left panel).

For each linear approximation $p(x) = b_0 + b_1\cdot x$ of $p_3(x)$, the exponent of the nucleation rate and the saturation ratio threshold become, as demonstrated in section 4.2,

$$\begin{aligned} j(S_i,T) &= j_0 + \underbrace{b_1 a_w^i(T)}_{:=A(T)}(S_i - S_c(T)), \\ S_c(T) &= \frac{1}{a_w^i(T)}\frac{j_0 - b_0}{b_1} + 1. \end{aligned} \quad (39)$$

Since a_w^i is a rather complicated function of temperature, it is particularly useful in the context of analytical investigations to have simpler approximations of this quantity. This motivates to approximate a_w^i and its inverse $\frac{1}{a_w^i}$ in the relevant temperature range $190 \leq T \leq 230\,\mathrm{K}$ by polynomials $q(T)$ of degree $\deg q \leq 2$. Similarly, we can approximate the nucleation threshold $S_c(T)$ by polynomials $s(T)$ of degree $\deg s \leq 2$. For the approximations we use a least square procedure within the temperature range $190 \leq T \leq 230\,\mathrm{K}$.The results are presented in figure 2 (middle and right panels). Note that the thresholds, either exact or approximate, are quite similar to the former approximations by Ren and Mackenzie (2005), while there is a larger difference to the approximation by Kärcher and Lohmann (2002).

Combining the approximations $q(T)$ and $s(T)$ yields the formulation

$$\begin{aligned} j(S_i,T) &= j_0 + b_1 q(T)(S_i - s(T)) \\ &\approx j_0 + A(T)(S_i - S_c(T)) \end{aligned} \quad (40)$$

of $\log_{10}(J)$. As can be seen in Figure 2, the nucleation threshold is accurately approximated by a linear relation (deviation is smaller than 0.3%). In former studies (e.g. Kärcher and Lohmann, 2002; Ren and Mackenzie, 2005) linear fits were derived for the nucleation thresholds; however, these fits deviate significantly more from the reference in comparison to ours (see figure 2). The deviation depends on the respective formulation (or approximation) of a_w^i.

Also former studies use (empirical) linear approximation for the saturation threshold. Comparing these linear approximations with ours in Fig 2 it is evident that they deviate significantly.

4.4 Thresholds for prescribed nucleation rate values

The threshold description in section 4.3 was based on the choice $j_0 = 16$, corresponding to a nucleation rate $J = 10^{16}\,\mathrm{m^{-3}s^{-1}}$. As already mentioned, the choice of j_0 is quite arbitrary, and these high values of J are very often not reached in the numerical simulations (see section 4.5). For a better diagnostics of the nucleation events and the relative strength of nucleation events, we introduce a similar concept for nucleation thresholds, based on a prescribed nucleation rate value $J \sim 10^{x_0}$. For this purpose we use eq. (40) of the nucleation threshold based on the linear approximation of the nucleation rate with a fixed but arbitrary value $x_0 > 0$ for the nucleation rate value; hence, we can write

$$\begin{aligned} x_0 = j(S_0,T) &= j_0 + A(T)\,(S_0 - S_c(T)) \\ \Leftrightarrow\quad S_{cx0}(T) = S_0 &= \frac{x_0 - j_0}{A(T)} + S_c(T) \end{aligned} \quad (41)$$

where the function $A(T) = b_1 a_w^i(T)$ depends only on the linear approximation of J as stated in section 4.2. Note that obviously $S_{cx0}(T) = S_c(T)$ for $x_0 = j_0$. This leads to the formulation of the nucleation rate

$$j(S_i,T) = x_0 + A(T)\,(S_i - S_{cx0}(T))\,. \quad (42)$$

with a general nucleation value x_0 and its associated threshold function $S_{cx0}(T)$. The threshold function is just shifted by the value $\frac{x_0 - j_0}{A(T)}$, i.e. the type of the threshold function remains the same. This formulation will be used for the theoretical investigations using small perturbations (see section 4.6)

4.5 Numerical simulations of nucleation events for different approximations

In the following we investigate the impact of our approximations of $\log_{10}(J)$ on nucleation events. The setup is as follows: We use the simple bulk ice physics model as described