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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Conceptual study on nucleation burst evolution in the convective boundary layer – Part III: Physico-chemical characterization" *by* O. Hellmuth

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1. Response to specific comments of RC2

1. RC3: Page 11521, line 22; "Hence, even if ...", sentence hard to understand
   AC: Excuse me, I try it again:
   "While the nucleation rate determines the *absolute amount* of newly formed particles, the parameter dependency [1], mainly affects the UCN *variation* in space and time. As the different nucleation rates *behave* similar, the spatiotemporal

[1]Here, the shape of the function is meant, i. e, its steepness, smoothness etc..

variation of the UCN is expected to be, at least, more reliable than the absolute amount of predicted new particles."

2. RC3: Page 11522, line 5; Low background concentration
   AC: Revised. See response to part I.
   "As described in section 5 of part I, here two scenarios are considered with an emission source at the ground but very low background concentrations of aerosol, henceforth called 'clean-air mass' scenarios. Such situations are suspected to occur in anthropogenically influenced CBLs in connection with frontal air mass change and postfrontal advection of fresh polar and subpolar air."

3. RC3: Page 11522, line 13; Emission rate
   AC: The sentence
   "Therefrom the average emission mass flux can be calculated."
   is replaced with
   "The corresponding average emission rates are $3.24 \times 10^{11} \mathrm{molecules\,cm^{-2}\,s^{-1}}$ for SO_2 and $1.83 \times 10^{11} \mathrm{molecules\,cm^{-2}\,s^{-1}}$ for total NH_3, respectively. The SO_2 emission rate is approximately 50 % of that used by Pirjola et al. (1998a, Table 2) in their urban case scenario U3."

4. RC3: Page 11522, line 18; Empirical relation for the hydroxyl radical
   AC: Quotation added.
   In part I on page 11424, line 7-8:
   "To reduce the chemical mechanism, the OH evolution is diagnostically prescribed (Liu et al., 2001) (see appendix D 1.1, Eq. (D2))"
   In part III on page 11522, line 18:
   "The time-height cross section of the photochemically driven hydroxyl radical concentration is prescribed using an empirical relation (Liu et al., 2001) (see also part I, section 3.2.2, and appendix D1.1, Eq. (D2)).

5. RC3: Page 11523, line 13; Threshold of sulfuric acid concentration

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AC: Here, the reference to a "certain threshold" is only a qualitative evaluation, based on visual inspection. To give a numerical value, the data must be reevaluated. It has not been done and deserves a detailed analyses.
The word "exceeds a certain threshold" has been replaced with:
"apparently exceeds some threshold value [2]"

6. RC3: Page 11524, line 11; Sentence could not be understood.
AC: Excuse me. The fact is discussed in Napari et al. (2002b). The reference is added as follows:
"It should be noted, that the parameterisation formula of the ternary nucleation rate cannot be used to obtain the binary water vapour-sulfuric acid limit (vanishing ammonia concentration) or the water vapour - ammonia limit (vanishing sulfuric acid concentration). This results from the fact, that the natural logarithm, being part of that parameterisation, is infinite for argument zero (i.e., for vanishing concentrations) (Napari et al., 2002b)".

7. RC3: Page 11524, line 17; ... sulfuric insert acid.
AC: To be inserted in the final version. Thanks.

8. RC3: Page 11528, line 19-26; Confusing paragraph, should be rewritten
AC: Excuse me. Revised. The old paragraph
"The differences in UCN number concentration ... largest one according to the law of error propagation."
is replaced with the revised one:
"The differences in UCN number concentration for the binary and ternary case, respectively, leads to corresponding differences in the mass concentrations of pre-existing Aitken (compare Fig. 5c with Fig. 5d) and accumulation mode particles (compare Fig. 5e with Fig. 5f). Aitken mode aerosols serve as a coagulation sink for UCN and as a deposition sink for condensable vapours. Accumula-

[2]Based on visual inspection.

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tion mode aerosols serve as a coagulation sink for both UCN and Aitken mode aerosols, and as a deposition sink for condensable vapours. Hence, differences in UCN number concentration propagate upward across the mode cascade to affect the larger size modes, i. e., changes in UCN number concentration influence the Aitken and accumulation mode, even small [3]."

9. RC3: Page 11541, line 23; Conclusion should be reworded with respect to the special character of the simulated scenarios
AC: Conclusions revised as follows:
Page 11540, line 17: The word "typical ... scenario" is replaced with "special cases of synoptically possible ... scenario"
Page 11541, line 22-23: The old sentence
"... and give insight in the genesis of NPF in the boundary layer."
is replaced with:
"... and give insight from a modelling point of view in the in the genesis and special conditions, under which NPF is likely to occur in the boundary layer."
Page 11541, line 23-29: The old paragraph
"Although the simulated scenarios claim to consider typical conditions leading to NPF burst ... self-consistent modelling approach"
is replaced with:
"The simulated scenarios consider special conditions, under which NPF was hypothesised to occur in the boundary layer. These scenarios represent possible, but not typical CBL conditions, especially with respect to the low aerosol background concentration assumed. Moreover, the scenarios are restricted to clas-

[3]Example: Assuming for simplicity some nonlinear dependency $y_j = f_{1,j}(x_i)$ between a set of dependent variables $y_j, j = 1, \ldots, N$ and a set of independent variables x_i, $i = 1, \ldots, n$. Then, the change of y_j due to variations in x_i can be approximated according to the *law of error propagation*, i. e., $\Delta y_j \approx \sum_i^n (\partial f_{1,j}/\partial x_i)\Delta x_i$. Here, Δy_j are the differences in y_j resulting from differences ("errors") in x_i. The sensitivity of y_j against x_i depends on the first partial derivatives $\partial f_{1,j}/\partial x_i$.

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sical homogeneous nucleation involving only up to three species (water vapour, sulfuric acid vapour). Even though not typical, the assumed scenarios of an anthropogenic emission source in a postfrontal "clean air mass" are instructive to elucidate the interactions between CBL turbulence, sulfur and ammonia chemistry and aerosol dynamics by means of a self-consisting modelling approach. Deviations from the scenarios considered here will be discussed in a technical note prepared for Atmos. Chem. Phys. Discuss."

10. RC3: Page 11525, line 23; exceedance of maximum
    AC: To be corrected into "reaching the maximum". Thanks.

11. RC3: Rescaling of Figs. 3b-d, 10c
    AC: Will be rescaled for the final version.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 11517, 2005.