Atmos. Chem. Phys. Discuss., 15, C4428–C4434, 2015
www.atmos-chem-phys-discuss.net/15/C4428/2015/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemical and physical influences on aerosol activation in liquid clouds: an empirical study based on observations from the Jungfraujoch, Switzerland" *by* C. R. Hoyle et al.

Anonymous Referee #2

Received and published: 7 July 2015

The authors of the manuscript have considered experimental data, obtained during four measurement campaigns, on droplet activation in warm clouds. The measurement campaigns were conducted at a Global Aerosol Watch (GAW) station located at a high altitude site Jungfraujoch, Switzerland. Utilizing measurement data on particle size distribution, trace gas concentrations and meteorological conditions, the authors present a simple statistical model that reproduces relatively well the experimentally inferred cloud droplet number concentrations (CDNC). The performance of the model is evaluated not only against the whole data set but separately for specific wind directions and individual measurement campaigns. Also, the predictions of the derived model are

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

compared with those of the two previously developed, empirical CDNC parameterizations. The results of the study can be viewed as at least partial synthesis of long-term cloud measurements conducted at the Jungfraujoch GAW station. Manuscript also provides insight into the factors determining CDNCs in warm clouds formed on free tropospheric aerosol, and the derived statistical model can be, in principle, used in e.g. computationally expensive numerical models to calculate CDNC. In my opinion, however, the manuscript has major shortcomings which should be addressed before I can recommend it for publication in ACP, as detailed below.

Major comments

1. The authors state in abstract (line numbers 12-13) that "...we expect that this model is applicable to warm, free tropospheric clouds over the European continent.". Similarly, at the end of the Introduction (line numbers 80-81) it is stated that "...allows the construction of relationships which are applicable to a wide range of conditions." and at the end of Conclusions section (line numbers 477-478): "... we expect Equation 2 and Equation 3 [derived parameterizations for CDNC] to be broadly applicable to the European free troposphere."

However, the authors present no evidence to justify these claims. To address this, I would recommend that the authors compare, if possible, their results to previous cloud droplet observations on free troposphere in order to see how the derived parameterization compares against other data sets. Second, a comparison should be made with numerical or semi-empirical, established CDNC parameterizations such as those developed by Abdul-Razzak and Ghan (2000), Nenes et al. (2003) and Kivekäs et al.(2008) by initializing these parameterizations with "typical" free tropospheric aerosol. This would give insight into the applicability and limitations of the derived parameterization. Also, such comparison is needed as one of the real advantages of the parameterization over some of these numerical schemes derived here is computational efficiency. In order to further strengthen the manuscript, the authors should also discuss what new their results bring anything else new on the table compared to previous

ACPD
15, C4428–C4434, 2015

cloud parameterizations.

2. The authors should discuss more extensively about some of the limitations of their approach. First, does the topography of the site limit the applicability of the derived parameterization? For example, is there any information on the formation mechanisms of the sampled clouds – are they formed orographically or via convective activity? If they are formed through former mechanism, how it would impact the applicability of the derived parameterization (in large scale models, for example, clouds are often assumed to be formed via convection)? Related to this, the derived parameterization contains a parameter that is specific to the measurement site: vertical distance of the site from the cloud base, H (see equation 2). To me, it seems that this contradicts with the authors claim on the general applicability of their statistical model, and therefore this issue should be discussed.

Second, eq. 2 implies linear dependence of CDNC on the CCN concentration. Is it expected that such relationship holds on warm clouds formed on free tropospheric aerosols in general?

Third, as stated in Section 3: "The present study utilises data collected during four summer campaigns, in 2002, 2004, 2010, and 2011". The considered data set is thus limited to the summer season. Do the characteristics of the cloud droplet activation observed at the site depend strongly on the season? And does this limit applicability of the derived parameterization?

3. Usage of English in the manuscript should be improved considerably. Some of the issues with language are pointed out below, but I'd suggest that somebody with an excellent command of the language would review the usage of English in the manuscript.

Minor and technical comments

1. Introduction, line numbers 33-34. The studiesof Abdul-Razzak and Ghan (2000) and Kivekäs et al. (2008) should be cited here. Also, Petters and Kreidenweis (2007) did not

ACPD
15, C4428–C4434, 2015

present a parameterization for CDNC but for the impact of the particle hygroscopicity to CCN activation. Finally, I'd move the content of lines 26-34, i.e. description various CDNC parameterizations, to the end of the Introduction following the discussion on the experimental CCN and cloud formation studies.

2. Introduction, lines 36-37. The studies of the Henning et al. (2002) and Verheggen et al. (2007) cited here did not consider CCN but only cloud droplet formation.

3. Introduction, fourth, fifth and sixth paragraphs. Here previous studies on CCN activation and cloud droplets formation are discussed in mixed order. I'd reorganize the discussion so that the CCN studies are treated first, followed by the cloud formation studies.

4. Line number 61. I'd write "...the mechanisms through which..." instead of "...the way which...", for example.

5. Line number 64. "...particles and thus their activation into cloud droplets..." instead of "... aerosol droplets and thus activation to form droplets...", for example.

6. Line number 101. "...is influenced...", instead of "...will be influenced...".

7. Line number 179. ". Without entrainment, all particles above a particular size will be activated.". This is strictly true only for internally mixed aerosols. Otherwise, particle population may contain a mixture of large, non-hygroscopic and hygroscopic particles so that the former particles do not activate, leading to a non-monotonic activation curve.

8. Line numbers 179-181. The definition of the activation diameter given here is extremely confusing. To me, it does not even sound right – critical diameter cannot be defined solely based on "critical diameter of the least hygroscopic particle" as the particle dry size plays a role too. Please clarify.

9. Line numbers 216-218. Another definition is given here for activation diameter. Please define the concept in consistent and precise manner.

ACPD
15, C4428–C4434, 2015

10. Line 235. What is “potential CCN”? Please clarify.

11. Lines 243-247. This sentence should be split into two or more sentences.

12. Section 4.3. Please give a short description of the applied statistical method as it would benefit understanding the results of this study. Also, in line no. 274, there is a question mark in parenthesis following “. . .developed by. . .”.

13. Line numbers 278-279. The end of the sentence (“. . .to investigate its general applicability”) can be omitted.

14. Line number 293. The expression “. . .is not complete,” is not fitting in this context.

15. Line numbers 293-294. “. . .activation appears to be lower. . .”. See the previous comment.

16. Line number 307. Parenthesis should be removed from the end of the line.

17. Line number 323, equation 2. According to this equation increasing ozone levels lead to increases in CDNC, while increasing [CO] decreases CDNC. Is there any physical explanation for these trends? Also, the derived parametrization does not contain any variable that would directly reflect the particle chemical composition. Does this limit applicability of eq. 2 as cloud droplet concentrations depend, at least to some extent, on the chemical composition of CCN?

18. Section 5.2. Is there any explanation as for the worse performance of the parameterization against data from SE wind sector?

19. Line number 340. “. . .in order to see. . .”, instead of “. . .to see. . .”.

20. Line number 348. Remove comma from “. . .cloud droplets, to a. . .”

21. Line numbers 351-352, sentence “Therefore there appears to be no systematic bias introduced by considering both wind directions in the model together.”. However, according to Fig. 5C the parameterization tends to underpredict observed CDNC when

ACPD
15, C4428–C4434, 2015

CDNC >500 /cm3. Please explain.

22. Line number 356. What does “compact correlation” mean?

23. Line number 360. “. . . to which. . .” instead of “. . .by which. . .”.

24. Line numbers 369. “(Fig. 8).”

25. Line numbers 370. See comment 22 above.

26. Line numbers 370-373, sentence starting with “In this case. . .”. I do not understand how good performance of the tuned parameterization against data from 2002 campaign implies that the underestimation of CDNC is not due to saturation effect. Please clarify.

27. Line number 378. What does “without replacement” mean in this context?

28. Line numbers 400-404. The sentence starting with “Further. . .” should be split into two or more sentences.

29. Line numbers 405-408. Here it is concluded that ”. . .temperature dependent inïňĆuences of surface active compounds do not play a signiïňĄcant role in cloud droplet activation.”. This is based on the lack of correlation between CDNC and cloud base temperature. I would argue that such effects (or their absence) cannot be inferred from ambient data series as the key parameters, such as exact chemical composition of CCN, that determine the cloud droplet activation process cannot be controlled in atmospheric measurements. Also, such effects could also be masked by changes in CDNC due to other temperature-related factors.

30. Line number 444. See comment no. 22 above.

31. Table 2, page 21. The description of the table was hard to understand, please clarify.

References

Abdul-Razzak, H., and S. J. Ghan (2000), A parameterization of aerosol activation: 2.

ACPD

15, C4428–C4434, 2015

Multiple aerosol types, J. Geophys. Res., 105, 6837–6844.

Kivekäs, N., V.-M. Kerminen, T. Anttila, H. Korhonen, H. Lihavainen, M. Komppula, and M. Kulmala (2008), Parameterization of cloud droplet activation using a simplified treatment of the aerosol number size distribution, J. Geophys. Res., 113, D15207, doi:10.1029/2007JD009485.

Nenes, A., and J. H. Seinfeld (2003), Parameterization of cloud droplet formation in global climate models, J. Geophys. Res., 108(D14), 4415, doi:10.1029/2002JD002911.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 15469, 2015.