Atmos. Chem. Phys. Discuss., 14, C10572–C10575, 2014
www.atmos-chem-phys-discuss.net/14/C10572/2014/
© Author(s) 2014. This work is distributed under
the Creative Commons Attribute 3.0 License.

Atmospheric Chemistry and Physics Discussions

ACPD
14, C10572–C10575, 2014

Interactive Comment

Interactive comment on "Time-dependent freezing rate parcel model" *by* G. Vali and J. R. Snider

Anonymous Referee #2

Received and published: 29 December 2014

I'll begin by stating that I am supportive of publication. The ideas and data are appropriately presented and are a contribution to the literature. That said, I do have some philosophical differences with the approach presented here. These are offered in the spirit of a *Discussions* section. I am not asking for changes in the paper based on these. I do have a few questions and minors points which the authors should consider. Those are in the sections following the next one.

Philosophical Points

There's a fundamental disconnect between the two things being presented here. Contrast our understanding of the physics underlying the parcel model with what we understand of heterogeneous nucleation, and in particular a cooling rate dependent freezing rate. We know why and how the temperature within an adiabatic parcel decreases

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

as it rises through the atmosphere. The first law of thermodynamics tells us that the work of expansion comes from internal energy in an adiabatic parcel. The decrease in temperature can then be calculated from a knowledge of the gas's heat capacity. (We even understand the heat capacity of the gas on a fundamental level.) In contrast, the physics and chemistry of nucleation are much, much less certain. Classical Nucleation Theory is the most commonly used approach, though there are problems with it. The general idea of a nucleus or seed which spontaneously forms in the metastable phase, triggering the phase transition if the fluctuation is large enough, seems to be sound. It should be noted that the details of how this happens, what properties the seed has, and how those properties differ from the bulk are not always captured by the classical theory (Oxtoby 1998).

There's no debate as to why temperature changes in the way that it does in an adiabatic ascent because we all agree on the underlying physical mechanisms. There is still considerable debate on mechanisms in heterogeneous (or even homogeneous) nucleation. Stochastic? Singular? Some hybrid? The pages of this journal have hosted that debate over the past few years and we still haven't drawn a definitive conclusion.

Personally, I think the final statement of the Abstract, "The results here presented can help guide decisions on whether to include a time-dependent ice nucleation scheme or simpler singular description in models.", is premature. I know that both the singular and time dependent model have their roots in observations, but I don't think that we, as a community, have reached the point of making these sorts of judgments yet. There's very little physical basis on which to base them. I've not seen any reference to a fundamental mechanism for a cooling rate dependent heterogeneous nucleation rate. (If the authors know of one, please include it.) I'm not against cloud models that include ice formation processes. We need that of course. But until we nail down the relevant physics and chemistry, we shouldn't be adopting one framework over another.

Points for the authors to consider

ACPD
14, C10572–C10575, 2014

CC BY

The parcel is labeled as adiabatic. Is that adiabatic, or pseudo-adiabatic? Changes in the liquid water content are explicitly calculated. Is the latent heat of condensation accounted for? Is the latent heat of freezing?

The ice nucleation parameterization is based on measurements that are presented in terms of ice nucleating particles per unit mass of water. That is used to initialize the parcel. Is it adjusted as more water condenses? I don't think it is, but then why calculate changes in liquid water content (see previous question)? For a given ice nucleating particle, more water surrounding it (i.e. a bigger droplet) doesn't make a difference (once you have bulk water).

There are a few places in the manuscript which state that the stochastic description overestimates the ice concentration in the parcel. Overestimated in comparison to what? There's a difference between the stochastic description and the one you are using here, but you can't say one is correct and the other isn't because you don't have an observation of a parcel rising through the atmosphere with ice concentrations recorded as a function of time and/or height.

A comparison of the results presented here to those presented in Knopf and Alpert (2013) would be warranted. They present results from a stochastic based model that seem quite reasonable.

Minor points, typos, etc...

Pg. 29308, line 9: "is" should be "are".

Herbert et al. (2014) is not in the list of references.

Pg. 29319, line 10: "effected" should 'affected"

References

D. Oxtoby, Nucleation of First-Order Phase Transitions, *Acc. Chem. Res.*, **31**, 91-97,

ACPD

14, C10572–C10575, 2014

Interactive Comment

1998.

D. Knopf and P. Alpert, A water activity based model of heterogeneous ice nucleation kinetics for freezing of water and aqueous solution droplets. *Faraday Discuss.*, **165**, 513–534, 2013.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 29305, 2014.

CC BY