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Interactive comment on "Supersaturation, dehydration, and denitrification in Arctic cirrus" *by* B. Kärcher

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This is a comment about the author's use of a fixed deposition coefficient. Actually, the use of a fixed deposition coefficient is very common in cloud modeling, so this is a general comment about all such papers. My point is that the deposition coefficient (which I often call the condensation coefficient in my papers), is fixed in only special cases, and in such cases, it is probably very nearly unity.

First of all, I define the term 'deposition coefficient' for the purposes here. If a crystal surface has an adjoining vapor density of N (and providing certain conditions that should be satisfied for a crystal in the atmosphere), then the maximum possible growth rate of the surface, normal to the plane of the surface, equals (volume per molecule in the solid)*(mean molecular vapor molecule speed/4)*(N-Neq), where Neq is the value of N under equilibrium conditions. Call this growth rate Rm. Typically, the actual rate R is less than Rm, with the deposition coefficient, hereafter alpha, defined as R/Rm.

With the above definition of alpha, it is a common misconception in cloud modeling that crystal surfaces have a single value of alpha for a given temperature. First and foremost, alpha depends on the surface growth mechanism. Moreover, for a given mechanism, alpha depends on (N-Neq)/Neq, hereafter surface supersaturation. In some cases, this dependence on surface supersaturation is supersensitive. For example, it is theoretically plausible that only a 10% change in surface supersaturation near the critical value can change the growth rate by a factor of 100 when growth is by step nucleation. Thus, with step nucleation, the key parameter is the critical supersaturation. I wrote above that this dependence is ’theoretically plausible’ only because it has not been directly measured. But this supersensitivity is crucial for explaining many features of the snow crystal growth habit at temperatures above -20 C; moreover, the supersensitive dependence on supersaturation is well established experimentally in other systems. Also, although we did not measure the superaturation dependence of alpha in our experiments, we established that the growth rate was supersensitive to supersaturation (Nelson and Knight, J. Atmos. Sci. 55, p. 1452 (1998)) because the growth effectively shut off when the supersaturation was below the critical value.

Other experiments have shown directly that alpha depends on surface supersaturation for ice growth from the vapor (e.g., Sei and Gonda, J. Cryst. Gr. 94 p. 697 (1989)). Many experimental studies report a limiting R/Rm that is less than 1. However, most such experiments are plagued by surface heating, and reanalysis of many such experiments show that R/Rm approaches unity at surface supersaturations above some (temperature- and face-dependent) characteristic value, a result that is consistent with theoretical arguments (Nelson, J. Cryst. Gr. 132 p. 538 (1993)). However, in the atmosphere, the surface supersaturation is probably significantly less than the characteristic value.

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There is much indirect evidence that step nucleation is a dominant growth mechanism at temperatures above -20 C for humidities near liquid water saturation. Also, the existence of Shimizu's long column diamond dust crystals (e.g., Grenfell and Warren, J. Geophys. Res. 104 D24 p. 31,697 (1999)), indicate a barrier to step nucleation on the prism faces for some crystals in the much colder regime in the atmosphere over the South Pole. However, for imperfect crystals that are either small (20 microns or less) or are at low supersaturations, the spiral-step mechanism may be the dominant growth mechanism. With the spiral step mechanism, alpha increases from zero to unity as the surface supersaturation increases, as does step nucleation, but the sensitivity to supersaturation is much less than that of step nucleation. This was first described in the classic paper by Burton, Cabrera, and Frank (Phil. Trans. R. Soc. A 243 p. 299 (1951)), probably the most cited paper in all of crystal growth, and the spiral-step mechanism has since been supported by numerous experimental studies. One of the first persons to use the spiral-step mechanism in an atmospheric ice-crystal-growth model was Kuroda (J. Meteor. Soc. Japan, 62 p. 552 (1984)). As far as I know, only one study has used realistic supersaturation-dependent alpha functions in a cloud model (Wood, Baker, and Calhoun, J. Geophys. Res. 100 p. 4845 (2001)). Although the evidence is strong that alpha for the vapor-growth of ice depends on surface supersaturation, we do not have good estimates of the characteristic supersaturations for ice at the temperatures relevant for the present contribution. In fact, the growth mechanism is not even known for low temperatures.

When the crystal size of of order the surface ad-molecule migration distance (probably about one micron), even more uncertainties enter the problem. In this size range, there may not even be an established crystal growth model. I studied this case in my dissertation (Nelson, University of Washington, Seattle, 1994) and found that the effective alpha could be much smaller than 0.1 under sufficiently low supersaturations; however, the growth model in my dissertation had major simplifications. Thus, this regime is in dire need of a reliable crystal-growth model.

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But if the crystals do not have facets or vicinal faces, then alpha should be 1 or practically so (with the exception of the case when there is surface impurity).

The experiments that were cited in the present contribution found that alpha was about 0.5, and thus the present study used alpha = 0.5. As far as I recall, those experiments were done far from equilibrium. Thus, they measured a much different thing than that which applies to the near-equilibrium conditions of ice in the atmosphere. Given the sensistivity of alpha to small departures from equilibrium for some growth mechanisms, this means that such measured results are not even a good first approximation.

So, what do I suggest for the present contribution? If the results significantly change when alpha changes from 0.5 to, say 0.05, then the author should be worried about the accuracy of the model. (However, one cannot fix the problem by reassigning the value of alpha, because it usually responds to supersaturation as described above.) Regardless, the author should mention that a fixed alpha is a simplification. Hopefully, we will soon learn more about the small-crystal, low-temperature growth regime so that future modeling studies can use a more realistice supersaturation-dependent deposition coefficient.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 1829, 2005.