Atmos. Chem. Phys. Discuss., 14, C9457–C9460, 2014
www.atmos-chem-phys-discuss.net/14/C9457/2014/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "On the derivation of particle nucleation rates from experimental formation rates" *by* A. Kürten et al.

Anonymous Referee #1

Received and published: 24 November 2014

Review of Kuerten et al. (ACPD 14, 27233-27261, 2014)

This manuscript by the CERN and Frankfurt 'CLOUD-groups' presents methods to determine nucleation rates from measured particle formation rates at larger sizes. The methods presented seem mostly valid and definitely useful but I have some serious concerns about their novelty and how they are presented.

The paper consists basically of two parts. In the first, two simple analytical formulae are derived – one for the case of small particulate scavengers, the other for scavenging dominated by wall losses. The second part is a numerical technique that is stated as universal, i.e. should work for any nucleation+growth+coagulation+scavenging system.

My biggest concern is related to the motivation and writing of the first part of the paper,

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

which is basically 'an attack' against the (original) Kerminen&Kulmala-equation, published in 2002. The work on this topic by Kerminen and coworkers is actually a series of four papers, all listed as references in the manuscript: 1. Kerminen and Kulmala (J. Aerosol Sci. 33, p. 609, 2002) 2. Lehtinen et al. (J. Aerosol Sci. 38, p. 988, 2007) 3. Anttila et al. (J. Aerosol Sci. 41, p. 621, 2010) 4. Korhonen et al. (J. Aerosol Sci. 69, p. 13, 2014)

The original Kerminen and Kulmala equation is derived assuming large background particles as scavengers (assumption 2 on page 27236 of KK, that results in the power law exponent -2 for the sink), a size-independent growth rate and no self-coagulation. In the follow-ups this assumption was removed and a more general equation was derived that holds for any power law dependence (2), an approximate method to include self-coagulation was included (3), and analytical equations were derived for linear and power law type size dependent growth rates (4).

Why did I repeat these here? Simply because the authors in this manuscript by Kuerten et al. have included all of these four papers in their list of references (so they clearly know of all this work), but they choose to compare their method with the oldest, original one! Furthermore, they make their comparisons with cases for which the original KK paper was not even intended, scavenging by freshly nucleated particles and scavenging by wall losses! Comparing to a method with a case, which even the original authors clearly state that falls outside of their assumptions (and for which several improvements have been made since) makes very little or no sense!!!

The derived equations seem to be special cases of the more general ones already derived in Lehtinen et al. (2007). Equation (12) of Kuerten et al., for the case of size-independent sink, is directly obtained by setting m = 0 in equation (7) of Lehtinen et al., and, equation (19) for Kuerten et al., for the case of wall losses by turbulence enhanced diffusion, by setting m = -1. (Actually this is not perfectly true, since in eq. 7 of Lehtinen et al. by setting m = -1 one obtains 0/0. However, taking the limit m -> -1 gives the 'correct' equation.). Thus, if the authors would compare their two analytical equations

ACPD
14, C9457–C9460, 2014

with Lehtinen et al. instead of Kerminen and Kulmala, they should get identical results.

The second part presents a “universal method”, equation 25 on page 27244. If the method is indeed universal, it should work also for cases where self-coagulation is important. I am slightly worried that having equation (24) as a part of the derivation violates this. The justification of equation (24) is a reference to Lehtinen et al., presumably equation (3) in that paper, which states that J = GR * n. This, I believe, is true only for condensational growth, i.e. a propagating ‘wave’ in size space – and is incorrect when there is also self-coagulation? (In the derivation of Lehtinen et al., it is assumed that self-coagulation is negligible.) In addition, the size-discretization results in an equation (equation 25) where Nm does not depend on Nm-1 . This seems odd, as growth is considered? Is it possible that because of this, equation (25) works better for a steady-state case and not so well for time-dependent cases?

Minor comments:

a) Page 27234, line 19: The definition of critical size is slightly inaccurate. It is the smallest size at which GR is on average faster than ER. Individual particles can have GR > ER even at below the critical size – otherwise nucleation would not be possible?

b) Page 27234, line 25: The reference to Kuerten et al when talking about barrierless particle formation should be replaced or supported by more original work, e.g. McMurry.

c) Page 27235, line 4: Which diameter is the stated detection limit of 1.2 nm?

d) Page 27236, line 20: The main reason for the ‘failure’ of KK when applied to chamber studies is the fact that it was not originally even designed for such cases, i.e. assumption (1).

e) Page 27245, line 11. This “method of Lovejoy” has been used much earlier widely in the nanomaterials synthesis community, see e.g. paper by Landgrebe and Pratsinis (1990), ‘A discrete sectional model for powder production by gas-phase chemical re-

ACPD
14, C9457–C9460, 2014

action and aerosol coagulation in the free molecular regime.', J. Colloid Interface Sci. 139, 63-86.

f) Page 27245, line 15. What are the loss terms in the simulation (figure 4)? Is self coagulation important in this simulation, at any stage? If a case such as in figure 4 (as an experimental steady state size distribution) would be analyzed, how would one obtain the growth rate as a function of size from the data, which is needed for the analysis?

g) Page 27247, line 8 (and figures 5 and 6): How is the "accurate" formation rate obtained, especially if self-coagulation is important?

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 27233, 2014.