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Geoscientific Model Development Discussions

Interactive comment on "An aerosol dynamics model for simulating particle formation and growth in a mixed flow chamber" *by* M. Vesterinen et al.

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I do not believe that this paper should be published in its present form; it may be publishable after extensive revisions. Ultimately, the conclusions are rather vague and not well justified. As a result, there is nothing for the reader to take away from it. There are a number of significant problems.

(1) The model does not seem to fit the data very well in that none of the cases tested can reproduce the large initial growth rate and all of the cases give similar results for the later condensation sinks and particle numbers. So the experiments are not sensitive to the value of 'gamma' but are sensitive to something that has not been investigated in the model. It is hardly news that it is hard to design an experiment that gives good data on nucleation rates. So what do we learn from this?

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The authors suggest that a second product might produce results that are significantly different from increasing the yield of the one product considered; but I do not understand why this would be. The total volume formed implies some yield, so why does using that yield not produce the observed growth? Is it because the yield depends on aerosol mass (e.g., Odum at al.)? In that case, why not include a second, semi-volatile product and treat it using the Odum model?

Equation 2 is for an irreversible reaction. Perhaps the condensing species has a non-negligible vapor pressure. Would that make the model work better?

(2) It is not clear how coagulation is treated; this may have a significant effect on the results. For instance, high nucleation rates combined with high coagulation rates could produce a rapid increase in surface area while limiting the peak number concentration. That might explain the poor fit in Figure 4. The authors only say that they calculate coagulation rates by the method of Fuchs and Sutugin. Do they account for van der Waals forces? Those are known to greatly increase coagulation rate constants.

The data in Figure 2 suggest that coagulation is not properly accounted for. Figure 2(b) is puzzling and, I think, impossible. One expects beta to pass through a minimum at sizes of a few hundred nm, with diffusive deposition dominating at small sizes and settling and impaction dominating at large sizes. The graph implies that the largest particles have a lifetime in the chamber of three years; gravitational settling is surely much faster than that. The authors imply that measured losses are excessive at very small sizes. Coagulation could explain these results since it causes the size distribution to shift to larger sizes.

It should be possible to separate the two effects by fitting the evolution of both number and volume distributions, as done by Verheggan and Mozurkewich. At a minimum, the derived wall loss rate constants should be used to compute the change in integrated aerosol volume and this should be compared to the actual change in volume; if the volume loss is overestimated, it can be concluded that coagulation is significantly affecting

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the number concentration.

There are also some technical issues with Figure 2. What is the particle number concentration? Figure 2(a) is not labeled correctly; the vertical axis must be a derivative (dN/dDp, or dN/dln(Dp), or dN/ dlog(Dp)). If it is the first, the units are wrong.

Equation (5) could do with more explanation. It seems an unusual form and perhaps overly complex. Why this form? The flow contribution to beta is not included in equation (5) since there is no additive, linear term, and does not seem to be included in the results in Figure 2(b). That suggest that the data in Figure 2(a) have been somehow corrected for the flow term; but the text says nothing about how this was done. Explanation is needed.

(3) Other than for the exponent in the nucleation expression, no sensitivity analysis is provided. As a result, it is not clear which parameters are well determined and whether the assumptions are well justified.

(4) The amount of hydrocarbon consumed appears to be poorly determined, which makes the fitted yield (the only parameter to which the model is sensitive?) highly uncertain. Some examination of the effect of a large error in the change in [HC] should be included.

(5) The authors suggest that the fact that the model gives oscillations, as in the data, is highly significant. What causes the oscillations? It is not clear if the oscillations are due to anything well defined or if they are due to poorly defined conditions in the reactor.

Is plug flow assumed in the reactor? Is this assumption justified? Does it affect the results? Plug flow, or even laminar flow, is very difficult to obtain in large volumes (there will be radial convection induced by geometrical asymmetries and/or temperature gradients). The flows here are far to small for well developed turbulent flow. So unless strong mixing is forced (e.g., a CSTR), the flow regime is likely to be undefined. In that case, erratic behavior, such as nucleation bursts, are far more likely than in a well

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defined flow and it may be the flow variations that are causing the oscillations.

Technical comments and corrections:

Page 392, just after equation (2): “Different values for were tested (from 0 to 3)” - ‘gamma’ is missing.

Page 396, just after equation (7): “The factor describes the carrier gas flow“ - ‘beta’ is missing.

Page 396, just after equation (8): “is volume of the particle“ - ‘Vj’ is missing.

page 400: “All the model runs were initialized with a background aerosol consisting of one log- normal mode (dg = 7.5 nm, s = 1.01 and N = 100 cm-3)”. But there was nothing said about seed particles in the experimental section. I am guessing that this is too little aerosol to matter physically and that it was included in the model for some numerical reason. But I should not have to guess. Why was this done? Do the parameters chosen affect the results?

The symbol ‘beta’ seems to serve multiple purposes. This can get confusing.

I don’t think that the parameter ‘sigma’ for the size distributions was defined anywhere. There are several choices (std. dev., ln(std. dev.), log(std. dev.)).

In figure 7, one has to guess at the values of the tick marks on the vertical axis since only one is labeled.

Interactive comment on Geosci. Model Dev. Discuss., 4, 385, 2011.