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Interactive comment on “Multiple daytime nucleation events in semi-clean savannah and industrial environments in South Africa: implications of the driving factors” by A. Hirsikko et al.

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Review of “Multiple daytime nucleation events in semi-clean savannah and industrial environments in South Africa: implications of the driving factors”

This paper explores days where there are more than one nucleation event during the day at two sites in South Africa. The authors attempt to filter out days where the air mass clearly changed during events. The frequency of the multiple nucleation events per day at these sites is interesting, but the evidence for the driving factors of the

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Interactive Discussion

Discussion Paper

2nd event is weak (or non-existent) and there does not appear to be any discussion on the “implications of the driving factors” as the title suggests. There is some relatively straightforward analysis that can be done to further explore the driving factors behind the 2nd events.

I feel that the paper in its present state is incomplete and can be significantly more informative once some straightforward additional analysis is done.

General comments:

-The authors claim that organics are likely a primary driver for the 2nd nucleation events since none of the measured factors seem to clearly indicate their importance in initiating these 2nd nucleation events. Organics have indeed been found to be important in nucleation and the early growth of freshly nucleated particles, but its not obvious from what is presented that organics are playing a stronger role in the 2nd events compared to the 1st events. Given the data and instruments presented in this paper, I think it should be relatively straightforward to determine if concentrations of condensable low-volatility organics are higher during the 2nd event than the 1st. (1) The authors could determine the maximum growth rate possible from H₂SO₄ alone and compare this to the growth rates during the 1st and 2nd events. If the excess growth rate (above the maximum H₂SO₄ growth rate) for the 2nd events are clearly higher than the 1st events, then this is clear evidence for the increased importance of organics during the 2nd event. (2) The authors could calculate the maximum possible increase in the total aerosol volume with time due to H₂SO₄ alone and compare this to the actual change in total aerosol volume (integrated over the DMPS size distribution and corrected by the change in BL height). As with the growth rates, you can see if the excess dV/dt is higher during the 2nd events than the 1st event. Due to the need for correcting for BL-height changes, using growth rates might be more straightforward than dV/dt.

If the above evidence for the role of organics in the 2nd nucleation events can be shown, the paper will be significantly stronger and will not need to rely on speculation

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on the drivers for the 2nd nucleation events.

-Its not clear where the “implications of the driving factors” (as mentioned in the title) are in the discussion. What are the implications of the driving factors (e.g. Would you expect multiple nucleation to be present in other locations based on locations where the timing of the driving factors might be similar? Might CCN production be different in these locations than in locations where only 1 nucleation event occurs?).

Specific comments:

P26034 L11: Why are SO₂ concentrations in surface layer just before sunrise so much lower than SO₂ concentrations in the residual layer. Presumably the two layers had similar concentrations when they became decoupled overnight. Is this because of fast dry deposition of SO₂ in the surface layer overnight, or does the presence of a nocturnal jet cause different wind directions in the surface layer and the residual layers (and thus a different air-mass history). Would you expect low-volatility organic precursors to be in higher concentrations in the surface or residual layers?

P26035 L21: The decrease in CS between nucleation events is not obvious in Figures 1 and 2.

Figure 3, P26036 L5-6 and L16-19, and P26037 L2-3: Growth of the first nucleation mode isn't suppressed, it just disappears entirely! Within the span of about 20 minutes (maybe less), the nucleation-mode dN/dlogDp changes from several 1000 cm⁻³ to around 100 cm⁻³ (more than a 10x decrease). This could not have been caused by the BL lifting (not a 10x loss at least) or by coagulational losses (the condensation sink timescale is about 1 hour according to Figure 3, so the coagulation sink timescale for ~10 nm particles will be something on the order of 10 hours or more). The only reason for this fast decrease in the nucleation-mode concentration that I can think of is a switch to an air mass where the first nucleation event was not occurring. This may be a clue to the reason for the 2nd events in Botsalano. It is probably incorrect to say that the growth of the 1st nucleation event is “suppressed”.

Interactive
Comment

Figure 3: The H₂SO₄ concentration goes to 0 shortly after the 2nd nucleation event starts. How does this happen if SO₂ and Glob are non-zero, CS does not go to infinity, and you are using the Peteja method for estimating [H₂SO₄]?

P26036 L14-16: Figure 3 shows neither an increasing H₂SO₄ concentration (it decreases to 0 as stated in the previous point) nor a decreasing CS for the 2nd nucleation event. Though I suppose this is what you are saying in the last sentence of this paragraph.

P26037 L1: “back-ground” should be “background”.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 26029, 2012.

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