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Atmospheric
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Discussions

Interactive comment on "Particle concentration and flux dynamics in the atmospheric boundary layer as the indicator of formation mechanism" *by* J. Lauros et al.

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We thank the reviewer for valuable comments.

We have improved the language of the manuscript.

Both reviewers notify, that the higher generation reactions products of organics are more volatile than the first generation products. Therefore we have multiplied the organic reaction rates by 0.5. This has improved the consistence with the observations (Fig. 3).

1)... the conclusions about the nucleation mechanism should also be toned down

We agree and have toned down the conclusions. Now we write: “This result indicates that the organic-induced formation mechanism can be responsible for particle formation at a Boreal forest site.”

2) I miss the comparison between the modeled and measured O3, CO, SO2, NO and NOx concentrations.

The mentioned vapours are inputs. We have used vapour concentrations as input for the model if measured/possible. We have written: “For several other gases like CO, SO_2, NO, NO_x and ozone, measurements from the SMEAR II station were used as inputs through the model simulation duration."

3a) I would expect to have an almost uniform vertical profile of the concentration of particles above 100 nm in diameter

Concentration of large particle is higher in the ABL than above because the conditions prefer growth of particles in the ABL (due to coagulation). Aitken mode particles grow faster to accumulation mode particles in the ABL and the vertical profile changes from uniform to upward decreasing. We agree that the difference between the ABL and the FT is probably not as clear as for smaller particles. If we use a vertically uniform accumulation mode concentration, the Aitken mode particle concentration should be less than 20 % of observed or the particle concentration is overestimated above the ABL in Fig. 3. As we do not have measurements, we have used similar initial profile for all modes for simplicity.

3b) I suggest that you do some sensitivity test by varying the initial SO_2 concentration within and above the ABL.

The lifetime of SO_2 is few days and therefore a uniform distribution is justified. However, ECHAM simulations show that an upward decreasing profile above the ABL (interpolated to Hyytiala) is possible (Risto Makkonen, personal communication) and therefore we have carried out a sensitivity study where SO_2 concentration is multiplied by 0.5

above the ABL.

Now we show the resulted profile in Fig. 3a and continue: “The conclusion does not change even if we assume a uniform SO_2 distribution only within the ABL, and the concentration above the ABL is set to 0.5 of the observed value.”

4) On page 20014 line 12-14 you write “on the selected days change of air masses by horizontal advection most likely prevented the appearance of clear “banana-plot” compared to the model”. [. . .] This you will need to illustrate with modelled air mass trajectories (e.g. HYSPLIT model) and I would also like to see a plot of the measured PM1, PM2.5 or volume concentration in Hyytiälä for the three days. If the volume and mass show large variations with time it indicates that the air masses are of different origin.

Particles are measured up to 1 μm and the number concentrations are shown in Fig. 4. The number concentration of accumulation mode particles does not show any dramatic change but varies (randomly) between $1–6 \times 10^2$ cm^{-3}.

Backward trajectories, calculated for every second hour by HYSPLIT, are very similar. The altitude of trajectory path changes few hundreds of meters between sequential time points. Local sources at the surface (horizontal differences) and altitude of trajectory paths (vertical differences) could explain the variation in background aerosol concentrations but we cannot be sure about this due to limitations of the HYSPLIT data. The resolution of GDAS data used in HYSPLIT is very coarse and therefore our hypothesis cannot be proved by HYSPLIT. Now we have removed the explanation: “Kinetic nucleation (Eq. 1) reproduces the observed particle formation events at the surface even if the observed formation events are not as clear as the simulated ones (Fig. 2a and b).”

5) On page 20007 lines 11-15 you write that the first order mixing parameterization (K-theory), have not succeeded to present turbulent mixing in all atmospheric layers

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and conditions, so higher order models have been developed [. . .] Therefore I suggest that you either referee to studies which clearly state that the K-theory does not work as accurately as the more advanced one-and-half order parameterization or that you perform this model comparison yourselves.

We have clarified: ". . . which presents mixing more reliably than the earlier utilised first order schemes (Boy et al., 2006) and is easier to utilise in aerosol flux calculations than the nonlocal schemes mentioned above."

6) On page 20015 lines 12-13 you write "in the presented simulations (Fig. 2c and d) we have assumed that 5 % of organic reaction products are able to condense on freshly nucleated particles. How many percent of the organic reaction products are able to condense on the Aitken and accumulation mode particles? [. . .] This seems to be a quite crude assumption to have a fixed fraction which does not vary with the temperature and total organic particle mass.

The same amount (5 %) is able to condense on Aitken and accumulation mode particles. We have rewritten (now in section 3.3): "In the presented simulations we have assumed that 5 % of organic reaction products are able to condense on particles."

Now results from sensitivity studies are shown in Fig. 3b. The assumption may be crude, but presently we do not have any better solution or parametrisation. We have to take this as a suggested improvement and consider it in the future.

7) Which compounds except organic oxidation products grow the particles e.g. NH_3, H_2SO_4, HCl and HNO_3?

The participating vapours are mentioned in section 3.3: "Nucleation is followed by growth, according to the nano-Köhler theory, having sulphuric acid and reaction products of organics, oxidised by OH, as a condensing vapour. Simultaneously water, sulphuric acid and reaction products of organics, oxidated by OH, NO_3 and O_3, participate in the conventional condensational growth of particles."

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8) Another question I have is how the organic reaction products which are able to condense or form nucleation clusters are formed? Are they assumed to form directly after the first oxidation step or do they need to be oxidized several steps?

Now we have tested different reaction rates for organics, see Fig. 3a. We write: "The concentration gradient between the ABL and the residual layer decreases if the nucleating and condensing reaction products of organics are higher generation products, in other words, if the nucleating organic products are formed slower than the first order products (Fig. 3a)."

9) What is the temporal and spatial resolution of the monoterpene emissions calculated with MEGAN? Can you specify the temporal and spatial resolution? How detailed is the forest species composition described?

The information on timestep and resolution can be found in the text. We have (re)written: 1) "In our simulations, the model consists of 52 layers, of which 18 are within the vegetative canopy in the lowest 15 m. The model vegetation consists of pine." 2) "The emission rates depend on leaf temperature and the available solar radiation on sun and shade, calculated separately for every model level." 3) "The meteorology, including atmospheric mixing of the chemical species, is simulated with a 10 second time step and after each 6 steps emissions and chemical reactions, separately for each atmosphere layer, are simulated for 60 seconds."

10) On page 20011 line 14–15 you write that the constant P value used when calculating the organic nucleation rates varies between 1×10^{-4} *and* 2×10^{-4} *cm*$^{-3}$ *depending on the organic reaction products which are participating nucleation. What do you mean by that? And how do you assign realistic values of P which is different for different organic reaction products?*

The value (P is 1–2$\times10^{-4}$ cm^{-3}) depends on the organic products which is participating nucleation. The value varies as the mean concentration of organic products differ from each other.

Now we write: “ The value of P was set to 1–2$\times10^{-4}$ cm^{-3}, depending on the (concentration of) organic reaction products which are participating nucleation: 1$\times10^{-4}$ cm^{-3} for OH oxidation products and 2$\times10^{-4}$ cm^{-3} for O_3 oxidation products.”

11) On page 20012-20013 you describe the new advanced deposition parameterization with several levels inside the canopy. This seems to be an important improvement of the model however you never test the new model against earlier model versions which described the deposition to canopy with a bulk parameterization. Although this is not a requirement from me this would be a useful and good test to do.

We studied only three days while deposition is potentially more important in long term studies. We have had some interest to compare different deposition parameterizations in the future.

12) Do you consider any primary particle emissions?

We do not consider primary emissions yet but this could be an improvement in the future. We have assumed that primary particle emissions are not important in studied conditions.

Now we write: “We consider different formation paths for secondary particles but primary particle emissions are ignored.”

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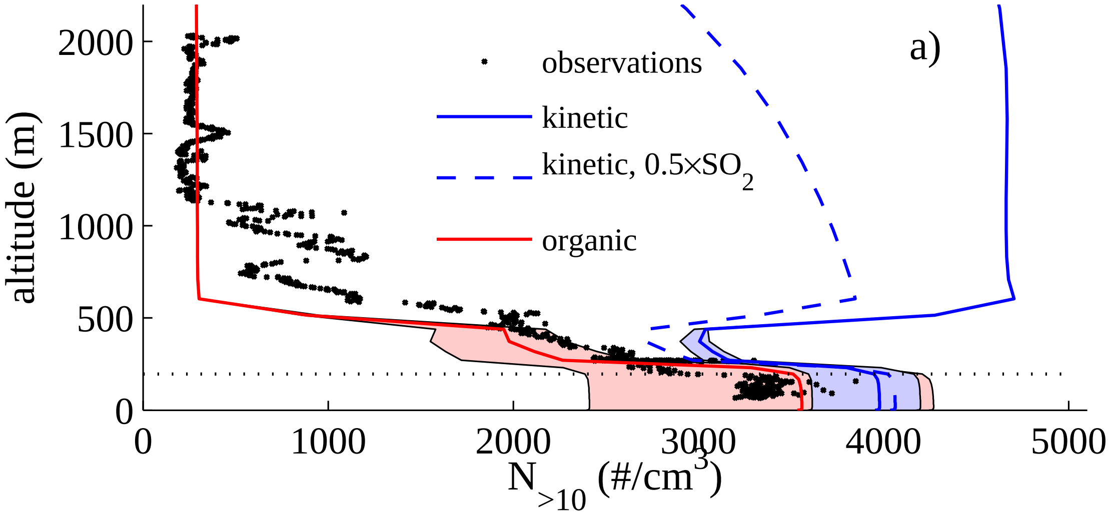

Fig. 1. ...3a) The shaded areas show the range of concentration when the reaction rate of organics is multiplied by 0.2–1...

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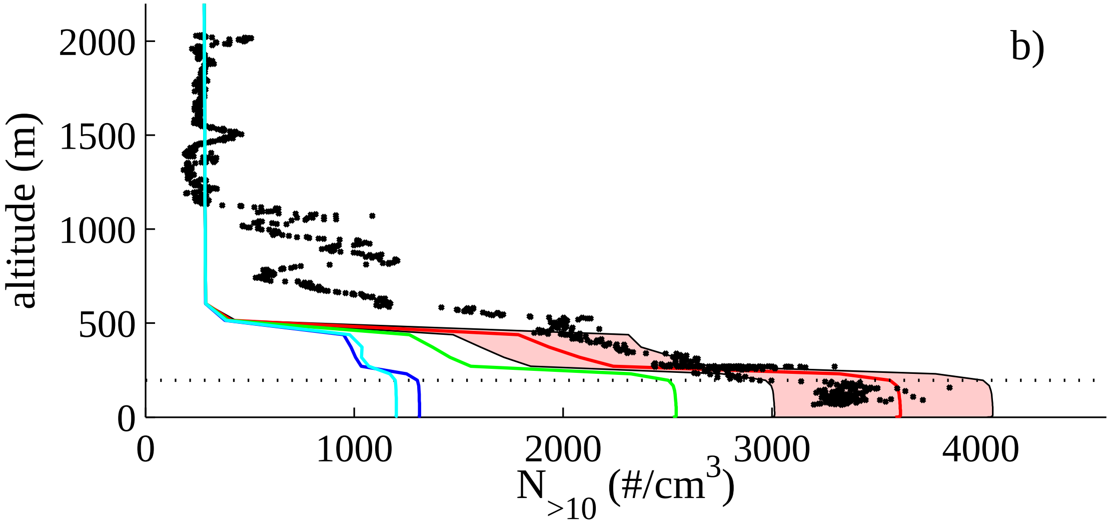

Fig. 2. 3b) The shaded areas show the range of particle concentration when 2–10 \% of organic vapours can condense on particles (red)...

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