We thank our two referees for helpful comments and suggestions. Below we provide responses to each individual comment. The comment is underlined in order to differentiate from the response.

Responses to Referee #1

Comment 1, Abstract, p.3, l.3: Molecular clusters do not form by nucleation, but by molecular interactions. This is named nucleation once it overcomes the nucleation barrier (critical size). Reformulate!

The text in the Abstract has been reformulated as "During an NPF event, particles first form by nucleation and then grow further in size."

Text in the Introduction has also been reformulated with respect to the definition of nucleation.

Comment 2, p.3, l.3: growth by condensation is one part of the whole process. As the Kelvin effect hinders condensation at size ranges especially below 5-10 nm, which is critical for the nucleation to occur, other processes contribute as well that cannot be named condensation: (i) coagulation and coalescence, (ii) dissolution in particle mass or water (Raoult effect) and (iii) reactive attachment (e.g. polymerization). Please reformulate "by the uptake of vapours".

The text "grow further by condensation" is removed. A new sentence "Among various physical and chemical processes contributing to particle growth, condensation by organic vapors has been suggested to be important." is added.

Comment 3, p.3, l.10-11: "suggesting missing atmospheric sulfuric acid sources" is probably a too strong statement as this may be caused by false assumption of sinks too. Common models assume a negligible saturation vapour pressure of sulfuric acid, which is according to literature not exactly true. As vapour pressures are functions of temperature this is expected to vary notably throughout the day. Please add the potentially different sink terms as well. The calculated OH concentration is one of the most critical in this respect, as OH reacts with nearly any species available except a handfull of substances. There might be an additional OH production via the ozonolysis of alkenes and the hydroperoxide channel. This impacts on the simulation approach via a specific assumption during night and early morning as investigated by the co-authors earlier on: E.g. all the monoterpenes are treated in a certain mixture out of three types, α-pinene, β-pinene and limonene which even provides a huge amount of species and reaction speeds. However, the real mixture is more complex with some notable differences in OH production and the individual terpenoid contributions may vary notably throughout the day affecting OH-cycling.

We agree with the comment about sulfuric acid and the overestimation of sink term has been discussed in the result part. The text in the Abstract is now modified as “With the latest Criegee intermediates reaction rates implemented in the chemistry scheme, the model underestimates sulfuric acid concentration by 50%, suggesting either missing sources of atmospheric sulfuric acid or an overestimated sink term.”

The chemistry scheme employed, as has been explained in Section 2.2 (p.9041, l.14-28), takes the full MCM paths for major known organic compounds, including the dominant local biogenic emitted compounds, MBO and monoterpenes. The full paths are available for alpha-pinene, beta-pinene and limonene, which altogether account for approximately 62% of the measured sum of monoterpenes (Ortega et al., 2014). For other monoterpenes and sesquiterpenes, whose full MCM paths are not available, we have included the their first order oxidation reactions. Thus the chemistry scheme does not approximate monoterpenes to consist of only alpha-pinene, beta-pinene and limonene, but indeed there is uncertainties due to unknown reactions. The emission factors used to simulate the monoterpenes emissions are specified for different species including myrcenen, sabinene, limonene, 3-carene, ocimene, alpha-pinene, beta-pinene and other monoterpenes (Harley et al. 2014). So the modeling work has tried to include the most available knowledge related of the oxidation cycle of biogenic organic compounds. Due to limited knowledge in anthropogenic organic compounds, for example the chemistry related to Toluene is omitted, and other unknown organic compounds which react with OH, the modeled OH is indeed not accurate. However, the result is based on the best knowledge by the time of conducting the model simulations.

Comment 4, p.3, l.11ff: The impact of MBO+OH is a very nice result! But it’s hard to understand the specific processes. MBO has got a molar mass of 86 g/mole with a single hydroxyl group that doubles during the reaction with OH. However no organic compound with 5 carbon atoms being a dialcohole will presume a saturation vapour pressure or even partitioning coefficient for the early stage of particle formation. So understanding seems only possible if treating MBO as a marker for BVOCs (oxidized VOCs)+OH. Thus, MBO is the wrong candidate at the right place at the time of interest with a similar bevaiour. This results in multifunctional organic peroxy radicals leading to some kind of polymerization. There are multiple of articles on this point. Do the authors have any suggestion about the involved processes? If so, please name it to focus future investigations!

The oxidation products of MBO included in the lump sums for aerosol simulation have molar mass range 135 to 180 g mol^{-1} and most of them have 5 carbons. We agree that it is possible that MBO is the wrong candidate at the right place and we have thus stated in the Conclusion (p.9052, l.24-25) that “The compounds (responsible for the particle growth) should have a similar daily pattern and concentration level as the OH oxidation products of MBO”. We do not have any concrete idea about the possible reaction candidates for MBO. Two experimental works by Zhang et al., 2012 and 2014 showed possible evidence of the role of MBO in SOA formation at the modeling site, Manitou Experiment Forest Observatory. Further work is needed to specify the responsible organics (direct emitted or reaction products from MBO) which contribute significant to the growth of particles during daytime at this station. This would need further experimental and theoretical work and is not in the frame of this manuscript.

Comment 5 Model validation for meteorology and chemistry, p.13/14: The model SOSAA seems to underestimate the mixing layer height notably. This is not a local phenomenon but applies at different sites in a similar way. Could the authors briefly indicate about the magnitude of the impact of this on the calculated results e.g. by taking measurements instead of calculations with interpolation in between the observations? This would cause a different dilution and different deposition rates.

The boundary layer height in the model is not used to calculate the turbulent mixing or dilution of scalar quantities. The boundary layer height represented is a diagnostic parameter calculated from the structure of turbulence in the model, and changing it will not change the properties of turbulence in the model. In other words in the model turbulent mixing governs BL height, not another way around. However, we agree that higher BL heights could dilute the concentrations of emitted organic compounds significant, but with our model setup it would not be possible to investigate this topic.

Comment 6, p.15: Different timescales for mean daily pattern of compounds seems very critical. If possible the same time frame should be applied for all important gases as small changes sometimes have notable consequences, hiding important features. The uncertainty range is as large as always. How well the model performs if the upper concentrations of OH and VOCs are assumed (read-in)? Is that capable in explaining the deviations between model and observations?

The measurements for VOCs, OH, sulfuric acid and NO_2 photolysis rate indeed cover different time period in August 2010. Figure 4, 6 and 7 in the manuscript are reproduced as mean diurnal profiles for period 13-14 and 16-23 August 2010, when measurements are available for all five quantities. The mean diurnal profiles do not changed qualitatively for NO_2 photolysis rate, concentrations of OH, H_2SO_4 and monoterpenes. However, the new diurnal profile for MBO indicates that the modeled MBO is actually underestimated by 20% to 25%. This may partly explain the overestimated OH concentration in the afternoon in addition to the previously stated overestimation in photolysis. The discussion related to MBO and OH has been modified in Section 3.2 according to the new plots.

We have considered reading in the measurements to the model for constraining the chemistry. However, it is not done because 1) the measurements coverage and frequency various across different gas species; 2) As a column model, input measurements are expected to be applied at least throughout the boundary layer. Otherwise, perturbing only one layer with the measurements would cause extra dilution or transport during the meteorology simulation, which are not true and may exert influence to other modeled species.

Comment 7, p.16 and Fig.6: As OH sometimes compensates missing production and sink terms due to the multitude of connections at reasonable photolysis rates this may explain the morning and early midday behaviour of OH. What happened during the afternoon, i.e. cloudy sky, differences between the different days of averaging? This is indicated in Fig.7 and the photolysis rate of NO_2 as well. Please provide more info on this.

As indicated by Figure 1 (see below), only August 13 is a clear cloudless day in period 13- 14 and 16-23 August 2010, during which the OH and NO_2 photolysis rate averages are made. Cumulus clouds developed during afternoons as indicated by the fluctuations in measured photolysis rate.

We tried to include the cloudiness condition by scaling the clear sky actinic flux spectrum with the ratio of measured to TUV modeled clear sky photolysis rate of NO_2. The modeled photolysis rate of NO_2 is within the measurement uncertainty of 10% - 20% (Seroji et al., 2004). Though the modeled NO_2 photolysis rate is within measurement uncertainty of 10% to 20%, it is still possible that the photolysis rate is indeed overestimated in the cloudy afternoon, as can be seen in Figure 7 in manuscript. The scaling method may not work well enough that in the cloudy afternoon, photolysis rates of NO_2 and the photolysis production of OH is overestimated.

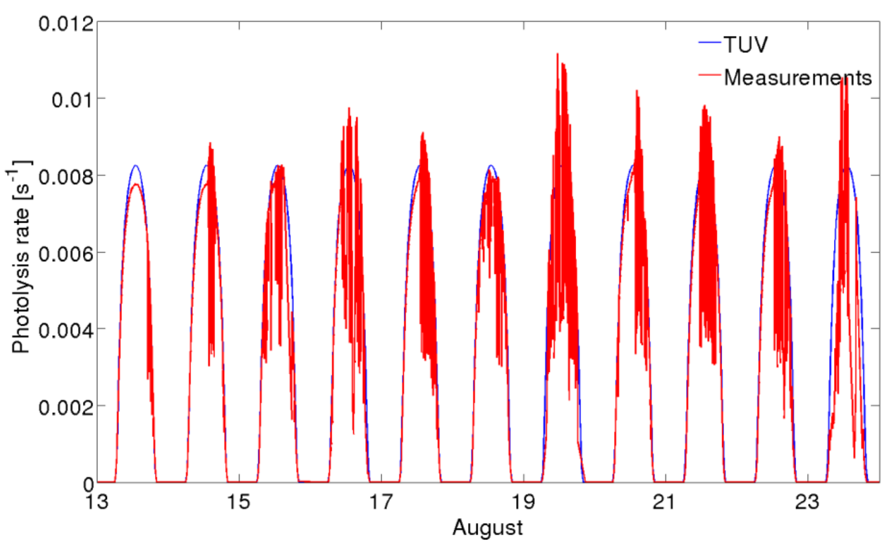

Figure 1. The measured and modeled clear sky photolysis rate of NO_2 from August 13 to 23.

Comment 8, p.32, Table 1: Please note, there is NO organic condensation but partitioning in the atmosphere as there is always organic material present casing subsidence below the saturation levels. Please modify the expression "organic condensing vapor" and replace it by "organic vapor type assumed" or something similar.

We disagree with the referee at this point. If the partial vapor pressure of an organic vapor in the atmosphere exceeds its saturation vapor pressure, the organic vapor will condense onto particle phase. Such condensation process is a main pathway for particle growth.

Comment 9, p.40: How well the usually taken 3-component assumption (α-pinene, β-pinene and limonene) matches with the plots shown? Is the simplified assumption made elsewhere justified or not? Please provide a brief statement. Regarding the plot only parts of it are informative. Could you provide more information shortly: Which kind of species are summarized in here, i.e. the stable ones or stable and radical products? Otherwise skip that plot.

A table summarizing the species included in Vap I, II, and III oxidized by OH, NO_3 and Ozone has been added to the manuscript (Table 2) to provide a better overview of which compounds were considered for the growth of the particles in this study.

Responses to Referee #2

Comment 1: The daytime MT and MBO concentrations were described by emissions from MEGAN and extended MCM photochemistry. The diurnal cycles of the precursor VOC were also described qualitatively quite well. However with too high overall conc. of MBO (factor 2-1.5) and very high nighttime concentrations of MT. The proposed explanation for the latter is a too high night time temperature predicted of the model. But this hypothesis could be tested by testing the T-dependence of the main emissions in the MEGAN emission algorithm. I suggest to do that in order to convince the readers that this is indeed the explanation.

Sensitivity study of the temperature dependence in MEGAN algorithm has been conducted for total monoterpene emission rates with the stand-alone MEGAN, in order to see clearly the dependence. The averaged diurnal profiles of temperature from the measurements and from the model (shown in Figure 1 in the manuscript) are used as the input for a one day simulation. The results from the sensitivity study, as indicated in Figure 2, show that the emission rates of total monoterpenes are higher by almost 100% with the higher modeled temperature during the night. The increased emission rates should explain for the overestimated monoterpenes concentration during the night.

“Sensitivity studies have been conducted for the response of total monoterpene emission rate to temperature. An increase of five Celsius degrees in the night may increase the emission rates by 80% to 100%.” is added to the second paragraph of Section 3.2 in the manuscript.

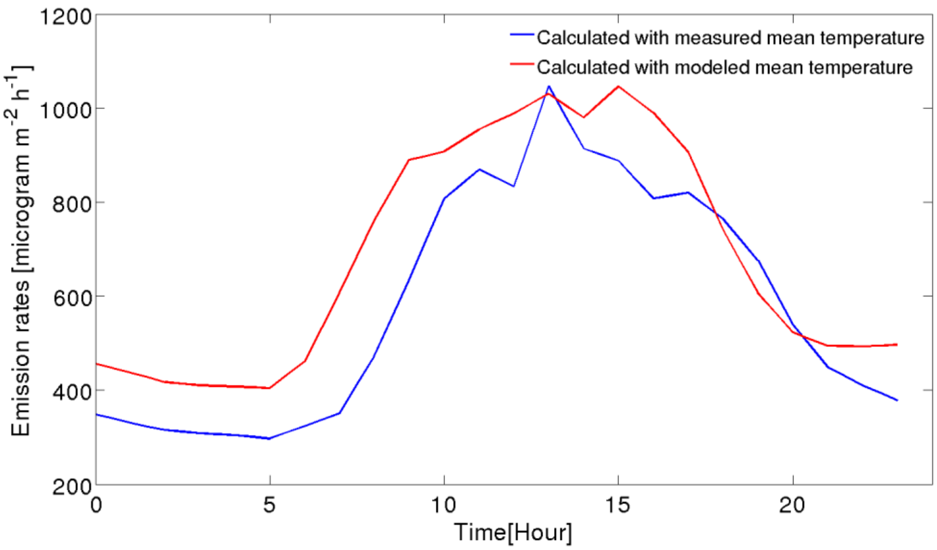

Figure 2. MEGAN simulated total monoterpenes emission rates based on the measured and modeled mean temperature shown in Figure 1 in the manuscript.

Comment 2: Amazingly the model fails substantially in predicting the daytime sulfuric acid concentrations and the afternoon OH concentrations. The argument that a JNO2, too low by about 20% around e.g.16:00-17:00h in the model compared to the measurement leads to a factor of two too low OH concentrations at that time period seems not too convincing to me. The question arises is if the model has missing OH sinks, and if these are organic vapors which are oxidized. How would this affect the predicted aerosol dynamics. I suggest to discuss this point in more detail in the manuscript.

We reanalyzed the situation and concluded that the major reason for the overestimation in OH should due to missing sinks. Previous studies by Nakashima et al. (2014) suggested a missing OH reactivity of 29.5% at MEFO based on measurements. Missing OH reactivity is also found for other forest environment. For example Mogensen et al. (2011) concluded that more than 50% of OH reactivity is mission for a boreal forest site in southern Finland.

Diurnal profiles of all gas species are updated that the averages are based on the same period according the request in Comment 6 by Referee 1. The new averaged diurnal profile of MBO shows slight underestimation instead of overestimation. The underestimation in MBO may also lead to overestimation in OH. Finally, we cannot eliminate the possibility of overestimated photolysis production of OH in the afternoon, as indicated by the overestimated NO_2 photolysis rate in Figure 7 in manuscript.

In case there are highly reactive compounds emitted by the forest which are not included in the model and not have been identified, there is a high chance that the reaction products of these compounds will also contribute to the growth and formation of particles. Maybe they could be also have a similar pattern as MBO and would explain the Comment 4 from Referee 1 related to this topic. However, in case the missing organics are reaction products from the organics already included but not handled explicit in MCM-chemistry, our assumptions for the condensing vapors would hold.

Comment 3: The too low H2SO4 concentrations were compensated by increasing the kinetic coefficient K in the nucleation parametrization. How critical is the adjusting of K in context of too low prediction of H2SO4 ?

Sensitivity studies of nucleation coefficient has been conducted in the same way as described in Zhou et al. (2014). The total number concentrations of particles between 15nm and 200nm are shown for the measurements and model simulations with different kinetic nucleation coefficients. The coefficient k = 5e-21 $cm^{-3}s^{-1}$ is used for the simulation that is presented in the manuscript (Line II in Figure 3 below). From Figure 3 it can be seen that doubling the nucleation coefficient approximately increases the total number concentration by 40% to 50%. Figure 4 depicts the averaged one-day number size distributions based on different nucleation coefficients. Figure 5b is the same as the plot of Experiment III in Figure 9 in the manuscript. Figure 5a is too low in concentration compared to the DMPS measurements while Figure 5c gives too high concentration compared to measurements.

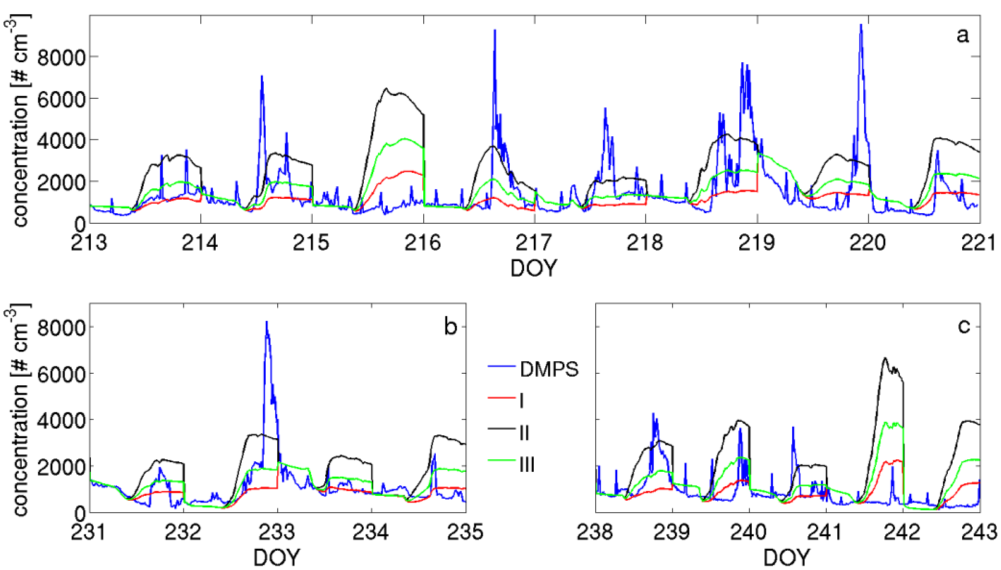

Figure 3. Total number concentration of particles between 15 nm and 20nm from a) 1 to 8 August (DOY 213 – 221), b) 19 to 22 August (DOY 231 – 235) and c) 26 to 30 August (DOY 238 – 243). The time series are based on the DMPS measurements, model simulation with kinetic nucleation coefficient k = 2.5e-21 molecules $cm^{-3}s^{-1}$ (I), k = 1e-20 molecules $cm^{-3}s^{-1}$ (II) and k = 5e-21 molecules $cm^{-3}s^{-1}$ (III).

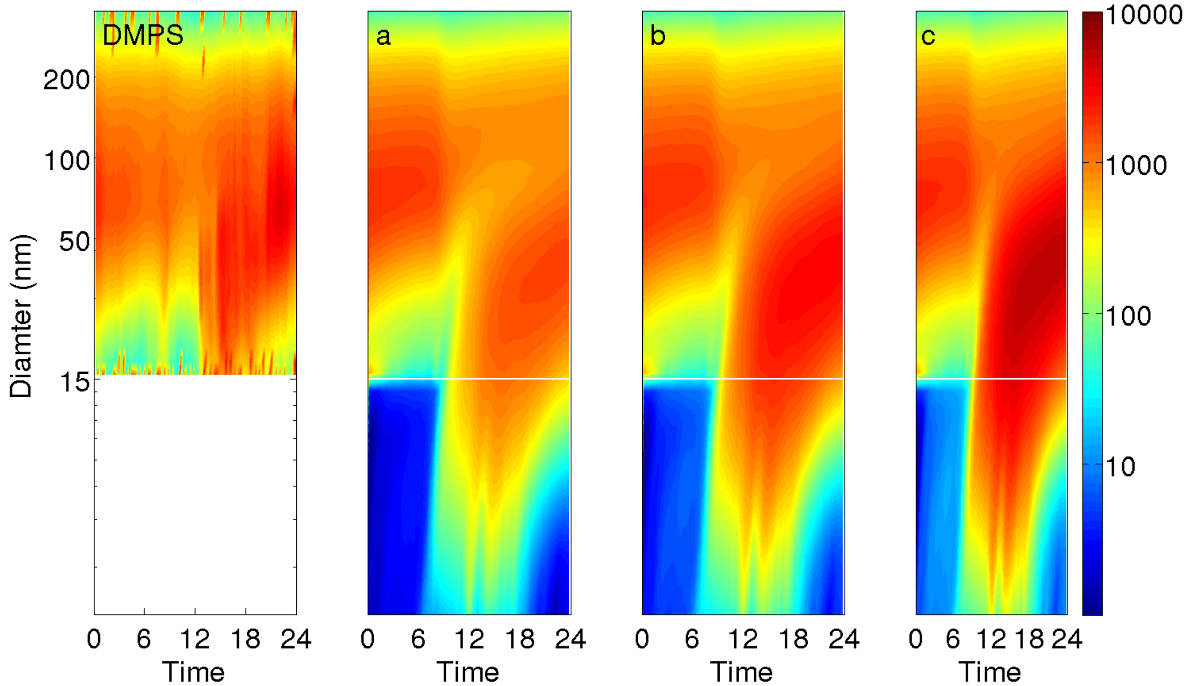

Figure 4. Averaged one-day number size distribution based on the DMPS measurements and model simulation with kinetic nucleation coefficient k = 2.5e-21 molecules $cm^{-3}s^{-1}$ (a), k = 5e-21 molecules $cm^{-3}s^{-1}$ and k = 1e-20 molecules $cm^{-3}s^{-1}$ (c). The concentration unit is molecules cm^{-3}.

<u>Comment 4: The explanations why the model fails in the sulfuric acid concentrations fall a little too short. How important is the H2SO4 production from OH? You overestimate OH by 100% in the afternoon, so the missing term might be really huge. Is that realistic? Could it be that simply the SO_2 input is too low? I suggest also here more explanation why the model prediction fails.</u>

The underestimated H_2SO_4 concentration is not due to too low SO_2, because the measured SO_2 is taken as model input. The main production of H_2SO_4 is via OH, while a minor production source due to Criegee Intermediates. Besides the missing source terms, overestimated sink term is another reason for the underestimation in H_2SO_4 concentration. Taking into account of the instrument uncertainty between 30% and 60%, the missing sulfuric acid term may not be that huge as seen in the figure. Similar study carried at the boreal forest environment in Finland (Zhou et al., 2014) has indicated comparable level of missing sulfuric acid sources. The discussion related to sulfuric acid in Section 3.2 has been modified for more detailed explanation for underestimated sulfuric acid.

<u>Comment 5: The organic contribution to growth is parameterized by using the first</u>

generations of stable vapors from MBO and MT generated by the oxidants OH, O3, and NO3. Vapor pressures where then attributed to the vapors, and the effect of MT and MBO alone and of both MBO and MT together was studied. MBO and MT vapors are needed to predict the observed size distributions and the agreement between prediction and observation is not too bad. Nevertheless I wonder why the first generation vapors are used as a measure. It is well known that with exception of ELVOC from ozonolysis the vapor pressures of those products are way too high to explain growth and SOA formation. Moreover during daytime first generation products can be oxidized further by OH. How such an ageing process would influence the results?

We agree that the first generation products may be too light and too volatile to contribute to particle growth. For this reason, the first stable oxidation products are used as the assumed organic vapors to contribute to particle growth. These first stable products may thus be n^{th} generation oxidation products with relatively higher molecular weight (molar weight up to 290 g/mol).

The aging process is the major and important process related to the aerosol particle growth. The particle phase chemistry model from the model ADCHAM (Pontus et al. 2014) will be added to SOSAA in the next phase. Meanwhile we are improving the model to use molecule specific vapor pressures calculated by different methods (SIMPOL and/or Nannoolal). Updated chemistry related to extreme low volatility organic compounds are under implementation too. However, the new code is still in the testing phase and not ready for this manuscript.

Comment 6: The authors derive limits for the vapor pressures to match the observations and suggest in the Conclusion section that the condensing vapors should have vapors pressures as low as 10ˆ6 cm-3. The author should discuss in how far the vapor pressures attributed to VapI, VapII, and VapIII match the lumped compound classes. And what can be concluded from such a comparison.

The range of these vapor pressures has been already investigated in earlier studies (e.g. Boy et al., 2006). The method here enables to study the growth without using explicit saturation vapor pressures of the single organic molecules. The explicit saturation vapor pressures are still highly uncertain. However, we agree that this method simplifies the condensation and can only represent approximated growth.

Comment 7, p9039, l13: The tower on the measurement site was not introduced before.

The text has been modified.

Comment 8, p9040, l3: use “differential mobility analyzer” instead of “differential particle counter”

The text has been modified.

Comment 9, p9044, l25ff: Does such a to flat diurnal temperature profile influence the vertical transport? If so, what does that mean for the model observations?

The temperature profile affects vertical mixing through creating or suppressing turbulence through buoyancy. For this mechanism the vertical profile of temperature is important. However, it is not clear whether the discrepancy in temperature causes other discrepancies in the model results. The failure of the model in reproducing all observed phenomena indeed indicates possible influences in reproducing the vertical transport, but the feedbacks are not obvious.

Comment 10, p9045, l14: I suggest to use either “mast” or “tower” throughout the manuscript.

‘Mast’ is now used though out the manuscript.

Comment 11, p9045, l18: I don’t understand point (2), are suggesting that the two different temperature measurements were potentially off by several degrees?

Agree. Possible cause of the large difference in nighttime temperature between mast and sounding measurements may be that one of the measurement instruments has less adequate radiation protection or ventilation compared to the other. But this should have very minor contribution to the difference. The main difference should due to the point 1 and 3.

Text “ (least likely and only has minor contribution to the difference) is added to point 2”.

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