Review of the manuscript NO. acp-2014-95 entitled: "Modeling ultrafine particle growth at a pine forest site influenced by anthropogenic pollution during BEACHON-RoMBAS 2011", by Y. Y. Cui et al.

We thank the reviewers for their comments on our paper. To guide the review process we have copied the reviewer comments in *black italics*. Our responses are in regular blue font. We have responded to all the referee comments and made alterations (in **bold text**) to our paper.

Response to Referee #2:

We thank the reviewer for his/her comments and hope we could respond to them in a satisfactory manner.

Major comment

R2-1) *The scientific approach and applied methods seem valid, and the authors have done a good job in investigating several different aspects that may influence the growth events. However, for a manuscript to be published in ACP it needs to include "substantial new concepts, ideas, methods or data". The authors need to state clearly in the introduction what is novel in their approach compared to the existing literature. The manuscript is reasonably well written in most sections, although some paragraphs need to be more clearly written or require more information.*

We thank the reviewer for recognizing the merits of our study. Following his/her suggestions we have stated more clearly in the introduction what the novelties of our study are compared to previous modeling studies. First, we are investigating the role of anthropogenic pollution on the formation and growth of ultrafine particles in an environment representative of urban-rural interface regions. More and more people live in these areas (e.g. the Colorado Front Range, the Alps), and it is currently unclear how the urban activities are changing the formation patterns of ultrafine aerosols and what the consequences for CCN concentrations are. Ultrafine particle formation events have been rarely investigated in these areas, especially in the terpene-rich forest environments. This is now explained in the manuscript:

"Understanding how forest environments respond to the inflow of pollutants from the nearby cities is of great scientific interest as more and more people live at the forest-urban interfaces. Jung et al. (2013) showed that the inflow of urban air masses could favor the initiation of the burst of nucleation mode particles in an isoprene-rich deciduous forest in Northern Japan. This study will focus on terpene-rich forests which have been comparatively less studies.. The Bio-hydro-atmosphere interactions of Energy, Aerosols, Carbon, H2O, Organics & Nitrogen (BEACHON, Ortega et al., 2014) field program collected long-term measurement of trace gases, aerosols and meteorological parameters at the Manitou Experimental Forest Observatory (MEFO), which is located within the semi-arid ponderosa pine forest in

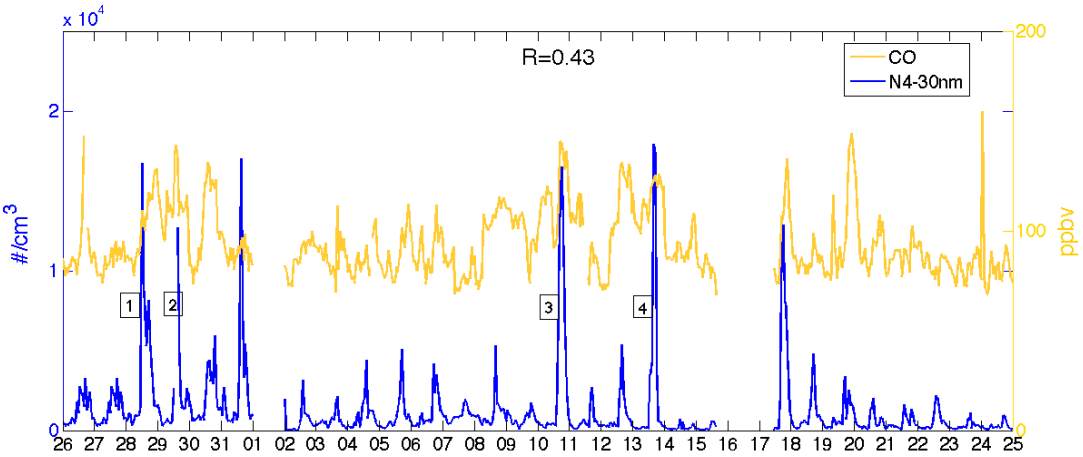
the Colorado Front Range. The site is representative of an urban-rural interface and provides a unique opportunity to study aerosol formation in a monoterpene-rich environment that is periodically influenced by the inflow of anthropogenic pollution from Denver and Colorado Springs (DiGangi et al. 2012; Fry et al. 2013; Ortega et al. 2014).”

In addition, our study uses the online-coupled chemistry-transport model at high spatial resolution (4km) to simulate these episodes. The online models are rarely used to study nucleation events, and they have the advantage of simultaneously treating emissions, chemistry and meteorology, and therefore are better suited for predicting the feedbacks between aerosols and clouds than offline models. This has been added to the new manuscript:

“The online WRF-Chem model is particularly well suited for this study as it simultaneously treats biogenic emissions, chemistry and clouds.”

R2-2) *It would be interesting to see the correlations of N4-30 nm with NOx and CO since these are tracers for primary emissions. Furthermore, I think NOx and CO (or at least one of these) should be added to Fig. 3. If NOx or CO is higher during APEs, it is likely that primary aerosol particles contribute to the “inflow of anthropogenic pollutants” and to the APEs.*

We agree that CO and NOx are good tracers of anthropogenic emissions, and high CO is correlated well with N4-30nm during the four PBE events under study here (see figure below). These results are similar to SO2, which we already plotted in Figure 4 and discussed in the paper. As suggested by the reviewer we added the comparison of diurnal profiles of CO between PBE and Non-PBE days to Figure 4. As expected, we find higher values of these variables during PBE days.



However, this doesn’t indicate that the primary emitted aerosols from anthropogenic

sources significantly contribute to N4-30nm. These primary aerosols are typically emitted into larger sizes (e.g. 30-40 nm for the traffic emissions, Brines et al. 2014). For instance, in the WRF-Chem model we assume that the primary aerosol emissions follow a lognormal distribution with a median diameter of 50 nm and a standard deviation of 2nm, thus particles with sizes below 30 nm can only be generated by nucleation.

This is now explained in the manuscript**: "CO levels are also higher during PBE days, which confirms that the MEFO site is influenced by the inflow of anthropogenic pollutants. The contribution to N4-30nm of primary particles transported from the Front Range is however expected to be minor as anthropogenic emissions typically occur in larger size ranges (30-40 nm for traffic, Brines et al. 2014)."**

R2-3) *One issue that needs to be discussed more thoughtfully throughout the manuscript is the increase in CCN concentration during the APE events. For instance on page 5624, lines 9-10, the authors write "that nucleated particles had a few hours to grow before arriving at the measurement site". In the Conclusions section, the authors write "SO2 plumes advected from the Colorado Front Range combined with biogenic monoterpenes significantly affect particle number concentrations and CCN during APEs ". It is very unlikely that aerosol particles, containing large amounts of organics, activate at a supersaturation of 0.5% if they are smaller than 50 nm (see e.g. Fig. 1 in Dusek et al. (2006)). Given the observed growth rates of 2.3 nm hr-1, it would take ~22 hours for the particles to reach a diameter of 50 nm. Therefore, the increase in CCN concentration during the afternoons on APE days cannot be associated with the growth of particles that were nucleated only a few hours away. To me it seems that the higher CCN concentrations on APE days are not a result of the APEs themselves but rather a result of higher monoterpene and SO2 concentrations on APE days (Figs. 3a-b) resulting in enhanced growth of pre-existing particles. In Fig. 3e it is clear that concentrations of particles larger than 70 nm are also higher on APE days than on Non-APE days.*

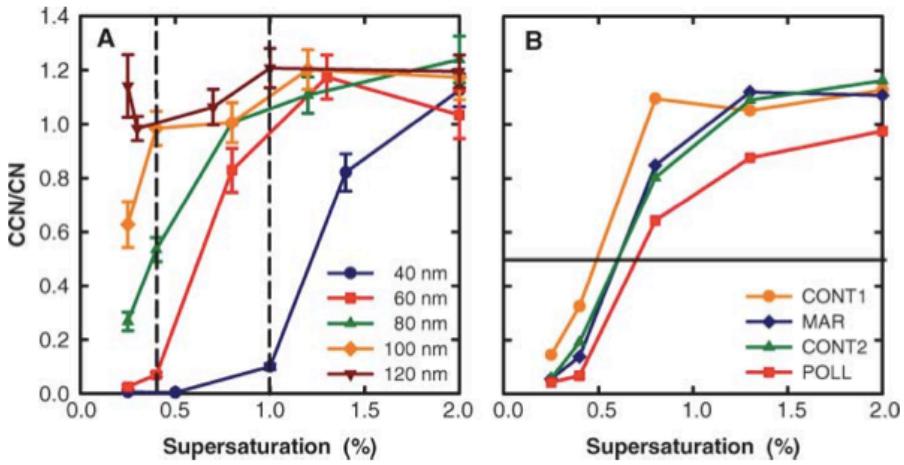


Fig. 1. (**A**) An example of size-resolved 6-hour averaged CCN spectra for particle diameters between 40 and 120 nm. Sixteen individual spectra have been averaged for each diameter. Error bars correspond to 95% confidence intervals of the mean. Vertical lines indicate the *S* values of 0.4 and 1% for which CCN size distributions are derived. CCN/CN ratios that are higher than 1 are due to a small bias in the calibration of the sensing volume, which probably changed slightly during the transport of the instrument to the field site. (**B**) The CCN spectra of particles with d_p = 60 nm are compared for different air mass conditions: CONT1 represents aged industrial pollution, MAR aerosol with Atlantic origin and short transport times over land, CONT2 rural continental aerosol, and POLL urban aerosol after a few hours of aging.

We agree with the reviewer that particles smaller than 50nm are unlikely to activate as CCN at 0.5% SS as shown above (Fig. 1 in Dusek et al. (2006)), and that the increase in CCN at 0.5%SS is not due to local nucleation events, but rather to the growth of pre-existing particles. We have clarified this discussion in the revised manuscript:

"Measured CCN (0.5% SS) number concentrations at the surface are also up to a factor of two higher during afternoon hours on PBE days compared to Non-PBE days (Fig 3d). A sharp increase in CCN is observed in the afternoon, typically three hours after the start of PBEs. It should be noted that only particles larger than ~60nm are likely to activate at 0.5% SS (Dusek et al. 2006). Given the observed growth rates of 2.3 nm hr^{-1} (Table 1), it would take >20 hours for freshly nucleated particles to reach a diameter 60nm. Therefore, the higher CCN concentrations on PBE days are likely the result of the enhanced growth of pre-existing particles."

From the comparison of our best simulation (Nucleation-bsoa) and the default WRF-Chem configuration (Ref-8bins) which doesn't account for sub-40nm particles, it appears that sub-40nm particles and their growth to larger sizes (>60nm) that can activate at 0.5% SS significantly contributes to CCN. This was also added to the revised paper:

"The Nucleation-bsoa simulation reproduces more accurately CCN (0.5% SS) concentrations than the Ref-8bins run, especially during PBE days. This difference suggests that sub-40nm particles and their growth to larger sizes (>60nm) that can activate at 0.5% SS significantly contributes to CCN."

The conclusion was updated to read:

"Considerable differences between PBEs and Non-PBEs indicate that pollution plumes rich in SO2 and primary particles that were advected from the Colorado Front Range combined as well as the enhanced biogenic monoterpenes concentrations significantly affect particle number concentrations and CCN during PBEs."

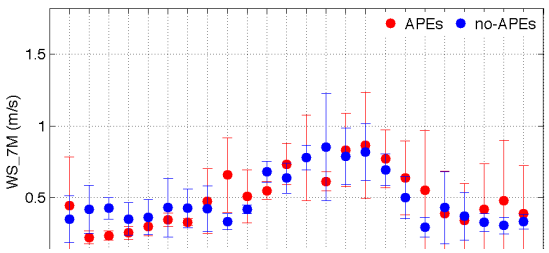
R2-4) *Page 5623, line 28 to page 5624, line 3: This paragraph needs to be more clearly written. On page 5623, line 28 it is written: "Jung et al. (2013) compared APE burst time and particle number size distribution at an urban site and a forest site, and found that late APE burst time and broader particle number size distributions were observed at the forest site than at the urban site. Figure S1 shows similar results for this study." To me it is not clear how you can observe in Fig. S1 what Jung observed. What time periods are the different size distributions in Fig. S1 averaged over? Do these size distributions represent the beginning of the APE events or the whole events? The following sentence: "These characteristics imply that several hours are needed for urban plumes to reach the site and that new particle formation is happening most likely hours away from the site". Are these conclusion drawn from Fig. S1?*

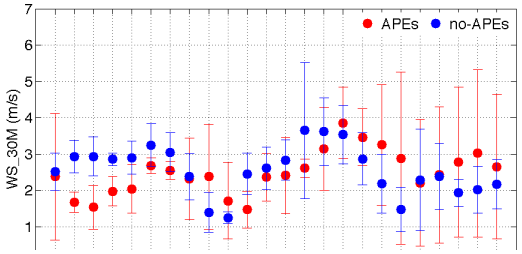
We agree with the reviewer that the comparison with Jung et al., 2013 was misleading, and we have simplified and clarified the manuscript to read:

"The PBEs at the MEFO site typically started around noon and early afternoon (10:20–15:00 MST, Table 1) following a shift in wind directions generally to the east (Fig 5a). This late onset time was reported for other forest sites (Jung et al. 2013)."

"The observed number size distribution during these PBE days (Fig S4) shows a relatively broad distribution similar to previous study performed in an anthropogenically-influenced forest (Jung et al. 2013). Figure S4 also shows the absence of particles smaller than 5nm. Especially in August (Fig. 7a), particles smaller than 10 nm were almost never observed, suggesting that nucleation likely occurred in upwind areas or in the free troposphere, and that freshly nucleated particles grew for several hours before reaching the measurement site."

R2-5) *Page 5622, lines 22-23: "Significantly higher mean values are observed during APE days for both monoterpenes and SO2". The authors link the higher SO2 concentrations to "anthropogenic inflow" and on next page to air masses from industrial sources located in the Colorado Springs area, but what is the reason for the higher monoterpene concentrations when advection is from these areas?*





We checked the wind speed at 2m, 7m, 16m, and 30m, we found during APE days, nighttime wind speeds were lower than non-APE days, which could accumulated more monoterpene during APE days.

Minor comment

R2-6) *Page 5629, lines 22-23: "clear difference in the relative abundance of sulfate during the APE (63%)...". Was this the case also for other APE days than 10 August?*

Unfortunately we didn't have measurements to verify if this was also the case for other APE days.

R2-7) *Page 5617, line 20: What instrument was used to measure the CCN concentrations?*

We added it to the revised manuscript. " **Size resolved CCN measurements made with TSI 3071 DMA followed by DMT CCNC and TSI 3010 CPC (Levin et al. 2012, 2014)**".

R2-8) *Larger fonts are needed in Figs. 5, 6, 7, and 9.*

Thanks, Done.

R2-9) *Page 5630, line 12: should it be 10-6?*

Thanks for catching this error. We have corrected it in the new manuscript.