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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Particle formation and growth at five rural and urban sites" *by* C.-H. Jeong et al.

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Response to Referee 2

The authors thank Referee 2 for the thoughtful comments and suggestions. The responses to your comments are marked in italics.

General comments

This study investigated ultrafine particle (UFP) formation and growth at 5 measurement sites (rural and urban) in south-western Ontario, Canada. The sites were separated

by up to 350 km, which provided an excellent opportunity to investigate the spatial scale of regional nucleation events. Only a limited number of studies of regional nucleation events over comparably large spatial scales can be found in the literature, so this manuscript is a very welcome contribution to ACP. A large amount of quality data has been collected (particle number size distributions, SO2, meteorological data for all 5 sites) and interesting methods of analysis have been applied. In particular, the analysis of the spatial homogeneity of the regional nucleation events is interesting and well presented. However, the manuscript could still be significantly improved by further extension of the data analyses, as explained below. The manuscript is well structured and the results are presented in a clear and concise manner. Therefore, following implementation of the changes suggested below I believe it is suitable for publication in ACP.

Specific comments

The authors clearly define a suitable scheme and classify the nucleation events observed at each of the 5 sites into classes I, II and N. They conclude from air mass back trajectory analyses that class I events are associated with cooler drier air from northern Ontario, class N (non-) events are associated with air masses that have passed over distant industrial regions to the south (e.g., Ohio River Valley), and that class II events are associated with high SO2 levels picked up from nearby industrialized regions (P11637, L 2-6).

However, not enough analysis is presented to support this conclusion. Air mass back trajectories are only shown for 3 days in total (1 day to represent each event class). I would recommend plotting event back trajectories of all class I, II and N nucleation events in a single figure, or combination figure similar to the current Fig. 7. This would allow the likely source regions for each of the nucleation event classes to be properly identified (if indeed specific source regions exist) and would justify the conclusions stated above. Alternatively or perhaps in addition I would recommend including CPF plots for particles in the size range 14-25 nm, if possible separated into class I and

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II days. In a dedicated study of particle formation and growth I am not sure why the authors have only shown CPF plots for particles in the size range 14-100 nm (Fig. 4). CPF plots of nucleation mode particles would also allow new particle source regions to be identified, particularly for the class II events which appear to be related to local anthropogenic sources.

We agree that back trajectory analyses for all events should be conducted to support our conclusions. We will add new plots to show all back trajectories of Class I and Class II (Fig. 7), as well as Class N (Fig. S1) in the Supplementary Material. A sentence will be modified in Section 3.3:

"Comparisons were made of the air mass origin at all five sites during the event days (Fig. 7) and the non-event days (Fig. S1in the Supplementary Material), based on back trajectories obtained. . ."

"Most air masses for the Class I events (Fig. 7a), originated from northern Canada and contained less polluted, cooler, drier air. This is consistent with the finding of Nillson et al. (2001), who observed that nucleation events occurred in arctic and polar air masses that corresponded with cold air advection. In contrast, during the non-event days, the air masses usually came from the south and passed over industrial regions (Fig. S1). Generally, hotter, more humid air masses were associated with higher concentrations of $PM_{2.5}$. Nucleation events could be inhibited by high $PM_{2.5}$ mass concentrations due to the scavenging of condensable vapors by pre-existing particles (Kerminen et al., 2001). The back trajectory analysis (Fig. 7b) for Class II at the near-border sites showed that the air masses passed over industrial areas in Sarnia, Detroit/Windsor, and Ohio, suggesting contributions from anthropogenic emissions."

Showing CPF plots for particle number in the size range 14-25 nm is a good idea to identify local sources for nucleation mode particles. However, in these areas nucleation mode particles were strongly affected by both regional and local sources. In general, CPF is not suitable for finding regional sources. In the manuscript (Sec. 3.1) the CPF

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for the 14-100 nm particles was used to provide a brief description of total particle number concentrations with size distributions in the 14-100nm size range at the five sites. In our revised manuscript CPF for particle number concentrations (14-25 nm) during the Class II event days will be shown in Fig. S2(Supplementary Material). The CPF plots at the near-border sites were embedded into a map to support the identification of likely sources. In Section 3.4 these sentences will be added:

"During all Class II event days CPF plots for particle number concentrations in the 14 nm to 25 nm size range at the near-border sites were depicted in Fig. S2 (see Supplementary Material). Strong contributions from the southwest and the northwest were observed for Harrow, consistent with the directions of U.S. and Canadian industrial sources. The CPF plots for Ridgetown and Bear Creek point the north and the northwest sectors indicating oil refinery facilities and a power plant near Sarnia."

P 11616, L 20 (abstract) and P 11638, L 2 (conclusions): I am not sure on what basis this claim is made. It is suggested that class II events occur in anthropogenic SO_2 plumes and that class I events are possibly related to biogenic emissions from the north. Does particle formation in southern Ontario appear to be more related to anthropogenic rather than biogenic emissions because class II events occurred more frequently than class I events? If so the claim is dubious because class I events appear almost as common as class II events at the 3 near-border sites. This point needs to be discussed further in the text if it is to be presented as one of the main findings of the study in the abstract and conclusion.

We agree that while the class II events were clearly anthropogenic, the class I events may have been anthropogenic or biogenic in origin. Thus we will modify the sentences in the abstract and conclusion sections:

Abstract: "Local short-lived nucleation events at the three near-border sites during this summer three-week campaign were associated with high SO_2, which likely originated from U.S. and Canadian industrial sources. Hence, particle formation in southwestern

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Ontario appears to often be related to anthropogenic gaseous emissions but biogenic emissions at times also contribute."

Conclusions: "Many short-lived strong particle formation events observed during this summer three-week campaign were associated with local industrial sources including trans-border air flows from the south and south-west, and with elevated SO_2. A regional nucleation event associated with unpolluted air suggested that biogenic emissions can also contribute to particle formation and growth at the three near-border sites in south-western Ontario. Hence, particle formation and growth in southwestern Ontario are in general related to anthropogenic emissions as well as biogenic emissions."

P 11621, L 3: Comparison of what variable, concentration? Has the data presented in this manuscript (e.g. Table 2) been corrected for this difference?

We will modify the sentence in Section 2.2:

"Comparison of particle number concentrations measured by the SMPSs used for the Toronto and Egbert sites showed an excellent correlation (r^2 = 0.99), although the SMPS used for Toronto tended to report values 18

We did not correct the difference between two SMPS and we will add this sentence for clarification:

"The SMPS data were not corrected for this difference."

P 11621, L 8: At all sites? This can be seen from Table 3 but it should be outlined explicitly in the methods section as well.

The sentence will be modified for clarification:

"Continuous measurements of $PM_{2.5}$ and meteorological parameters were also performed at all sites. SO_2 concentrations were also available except for the Ridgetown site."

P 11627, L 13; P11629, L24: What is Environment Canada's determination of a lake

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breeze event? Can the authors provide a reference that explains this for readers not familiar with such events?

We will modify the sentence in Section 3.2:

"The abrupt change of wind direction at around 11:00 am was consistent with a lake breeze event observed by Environment Canada."

We will also add a paragraph in Section 2.2 to provide a brief description of lake breeze and corresponding references:

"Due to their proximity to the Great Lakes, air quality in the most sampling site can be impacted by meteorology associated with the lakes, such as lake and land breezes (Hastie et al., 1999; Makar et al., 2010). During the BAQS-Met 2007 campaign, Environment Canada conducted analysis of lake breeze events using comprehensive measurements from both land and lake mesonet sites to assess the importance of the chemical and dynamical influences of the Great Lakes on regional air quality (Makar et al., 2010). The effect of lake breezes on particle number concentrations was also investigated for Harrow, Ridgetown and Bear Creek."

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 11615, 2010.