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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The effects of vehicle emissions and nucleation events on vertical particle concentration profiles around urban office buildings" *by* T. N. Quang et al.

Anonymous Referee #1

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This manuscript reported particle number (PN) concentrations and size distributions, and PM2.5 around three buildings in urban areas of Brisbane, Australia. The vertical profiles of PN and PM2.5 around the three buildings were explored. The results indicated that both PN and PM2.5 concentrations around building envelope were influenced by vehicle emissions and new particle formation. The experiments were well designed and the results were presented logically. However, I have some concerns about the discussion of the nucleation events and their effects on vertical PN profiles.

1. The authors defined "an event where nucleation mode PN concentrations increased during the day time, but the particles did not grow larger during the event period, as

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indicated by a near constant GMD value, was considered a nucleation event". The authors also defined the particles with size <30nm as nucleation mode particles. Based on the definitions, sources could directly emit nucleation mode particles in urban areas such as vehicle emissions, aircraft emissions and ship emissions. Nucleation mode particles could also be formed via gas-to-particle conversion (chemical reactions). Although several nucleation events were observed during the sampling weeks for each building, the authors did not provide detailed discussion on the sources of these nucleation events. For instance, could the nucleation events be caused by direct emissions from vehicles as some studies reported PN emitted from vehicles could be smaller than 20 nm, or ships as they exhausted nanoparticles smaller than 10nm in diameter, or something else? Or if the authors believe that these events were due to secondary formation, can they provide more evidence? Through the whole manuscript, they just observed the increase of nucleation mode PN concentrations on some days but did not give any evidence. Solar radiation and temperature were not enough. The question is how do you differentiate primary emissions from secondary formation of nucleation mode particles?

2. It is hard to believe that new particle formation due to chemical reactions can be apparently observed at roadside sites given that emissions from vehicles are fresh and secondary formation needs time to occur. However, primary emissions of nucleation mode particles are possible at roadside sites such as vehicles and nearby sources. In contrast, at ambient sites (sites away from main emission sources) in urban areas secondary particle formation have been widely reported.

3. In order to differentiate primary emissions from secondary formation of nucleation mode particles in urban areas, it is obviously not sufficient to measure PN and PM2.5 concentrations. Much more chemicals need to be monitored such as gas-phase primary pollutants CO, NO and SO2, and secondary pollutants such as O3 and SO42-. By comparing the time series of these air pollutants with nucleation mode PN, this problem may be resolved.

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4. Apart from local contributions, regional transport is another important factor affecting the secondary formation. New particles could be formed upwind somewhere else and then be transported to the sites. Though the authors stated that regional transport affected the new particle formation at Building B, it is not convincing that this was caused by a nearby industry zone based on wind direction/speed. Backward trajectory analysis is necessary for the discussion of regional transport impact.

Specific comments 1. Experimental

1). sampling locations Figures 1 – 3 did not provide enough information about the sampling sites. It is important to have information on the surrounding environments, especially potential major sources of PN and PM2.5 near the sites. For instance, by checking the Google Earth, I noticed that Brisbane is surrounded by a river. Are there boats on the river? What was the frequency? What fuel was used? Some studies reported that nucleation mode PN emitted from ships was below 10 nm.

2). sampling My understanding is that the rooftop site was continuously monitored while the lower-level sites were switched in the buildings. Clear information needs to be provided on how the sampling was conducted, such as outdoor air was sampled. Where was the sampling inlet? How far was the sampling inlet from the building walls and so on?

3). meteorological data Continuous measurement of meteorological data is critical to understand the new particle formation, if any. By looking at the time series of solar radiation, temperature and winds with pollutants, the possible mechanisms of new particle formation could be explored.

4). Identification of nucleation event There is a contradictory definition on the event. Suggesting revising (4) the new mode shows signs of growth because immediately another definition of “the new mode does not show signs of growth” was given. More critically, the authors did not clarify whether all events were secondary formation or primary emissions or both. The question is how to differentiate primary from secondary

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sources.

2. Results 1) It is not common to say “particle size distribution concentrations”. It may be said either “particle size distribution and concentration” or “nucleation mode, Aitken mode and accumulation mode particle concentration”

2) 1st sentence, para 2, page 1622: It is not obvious for Building B.

3) 2nd sentence, para 2, p 1622: If the traffic flows on the streets showed corresponding peaks, why not show the daily variations of traffic flow?

4) 3rd sentence, para 2, p1622: be careful when the statement of “. . .suggests the occurrence of new particle formation” was given as it could be either primary or secondary emissions.

5) last para, page 1622: how many weekdays were characterised by absent or unclear nucleation events for each site? How do you define unclear nucleation events?

6) 3.2.2 Influence of new particle formation on . . .. . . From figs 8, 9 S5-8, it is hard to believe new particle formation occurred because the PN concentration from 8.5 – 15 nm was almost nil, and no other evidence i.e. O3, CO, NO and SO2 measurement data was given. Also, unlike described in the manuscript, the accumulation-mode PN appeared to have good correlation with nucleation-mode PN in these figs, meaning primary source emissions with different size of particles.

7) 1st and 2nd paras, p1625: it could be true that the N<30 and N<30/N30-300 at the rooftop were higher than those at ground levels. However, the reasons may not be right as the chemistry at rooftop may be totally different from that at roadside. At roadside, chemical reactions for new particle formation in the atmosphere would be very limited due to highly fresh emissions and constrained oxidant concentrations i.e. O3 and OH. Hence, most likely the N<30 was related to direct emission at ground level while the air mass at rooftop could have chemical reactions to form new particles plus vertical diffusion of primary nucleation mode particles from the streets. Also, it should

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be careful that the primary pollutants involving in the new particle formation at rooftop could originate from urban vehicle emissions (not the immediate ones near the sampling sites) and/or from regional transport. To thoroughly understand the mechanisms, solar radiation and wind direction are certainly not sufficient.

8) 1st para, page 1626: could the ships nearby be one of the causes?

9) 2nd para, p1626: the statement on statistical difference in solar radiation was wrong. P=0.36 means there was no difference between the two sites. Similarly, throughout the entire manuscript, there were many statistical descriptions. But when looked at the mean $\pm$ S.D. and p values, sometimes we don't know which one we should trust. For instance, 1.15$\pm$0.35 vs. 1.88$\pm$1.19, large deviations suggest these two values did not have statistical difference. However, the test for this pair was $p<0.001$, suggesting significant difference. Generally mean $\pm$ 95% confidence interval would avoid this confusion.

10) last para, page 1626 and 1st para, page 1627: From Table 3, there was a significant difference in PN between the rooftop and street level at Building B ($p = 0.01$). However, here it said no difference. Is this for 16 Jan 2010 one day? If so, give PN concentrations at both sites and p-value.

11) 3.3 vertical profiles of particle concentrations For Building B, caution should be taken for vertical profile discussion as only two points were measured. We don't know what could happen between these two points.

12) Table 2: it would be helpful if the number of days can be listed for each building when vehicle emissions dominated. Mean $\pm$ SD should be replaced by mean $\pm$ 95% Confidence Interval. It is strange that p-value representation was not consistent. For instance, sometimes $p<0.001$ but sometimes $p = 0.002, 0.006$- both should be $p<0.01$. For $p=0.015$, it should be $p<0.05$.

13) Table 13: same problems as Table 2.

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