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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Traffic-originated nanocluster emission exceeds H_2SO_4-driven photochemical new particle formation in an urban area" *by* Miska Olin et al.

Anonymous Referee #3

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General Comments: Olin et al. leverage previously collected nanocluster aerosol (NCA), trace gas (H2SO4, NOx, CO2), meteorological, and particle size distribution data from a month-long sampling campaign in Helsinki, Finland (Hietikko et al., 2018) to propose an updated model of NCA and H2SO4 formation in urban environments (Figure 4). This model is comprised of two pathways for generating NCA and H2SO4, respectively. Olin et al. argue that immediate implications from H2SO4-containing NCA on human health necessitates models including their H2SO4-NCA conceptual model. Generally, speaking none of the highlighted pathways are novel or new. Primary H2SO4 was identified by Arnold et al. (2012), and the direct emission of NCA from vehicles was identified by Rönkkö et al. (2017). To this end, the proposed model,

and ensuing discussions, feel more well suited for a review-type article opposed to a research article. This particularly true as much of the analysis focused on supporting that NCA sourced from traffic and not regional NPF events. This argument seemed redundant to the earlier paper (Hietikko et al., 2018) which the NCA data was sourced from.

The manuscript is generally well written, and the arguments are comprehensible. The manuscript's shortfall is the lack of NCA composition data. The lack of composition data makes the influence of primary H2SO4 to both number and mass concentration of vehicle-emitted NCA unsubstantiated and, from my perspective undermines what would be the major contribution of this manuscript (in respect to the model). The authors adequately demonstrate that NCA is decoupled from NPF events via a series of regression and correlation analyses. However, the reliance on data published in Hietikko et al. (2018) undermines the impact of these observations, as this was the primary focus of the earlier study. In contrast to the earlier study, Olin et al. do provide an off-the-hand annual estimate of traffic-derived NCA in Helsinki using CO2 emission factors. I find that the general applicability of this approach might be questionable. In particular, the authors suggest the pathways (Figure 4) need consideration in regional chemical transport models but provide no clear means to facilitate this implementation. I can imagine that the complexity of this challenge would likely be confounded by varying relationships between NCA and H2SO4 and vehicle emissions (engine types, fuel types, emission standards, etc.) but am not enough of an expert to make specific recommendations. The challenges in implementing the conceptual model in regional chemical transport models is not elaborated on in this manuscript.

Lastly, I felt details pertaining to rationale and underlying assumptions were sometimes lacking in this manuscript. Specific points are made below. Although short manuscripts are ideal, I think this manuscript would greatly benefit from a more robust discussion on why certain decision were made. For instance, the authors opt to utilize CO2/NOx as a proxy for traffic flow when the authors have direct measurements of traffic flow.

Why not just use traffic flow? This was particularly odd as the authors state that background fluctuations in CO2 make it an unreliable proxy for local traffic. I suspect these decisions might relate to the desire of making their observations relatable to chemical transport models and vehicle emission factors. There may be other reasons; however, they are not clearly stated in the manuscript.

Specific Comments: 1) As stated in the general comments, my most significant issue with the manuscript is that the composition of the NCA is not actually measured during the deployment in question. This is important as much of the manuscript relies on the argument that NCA composition is decoupled from secondary H2SO4, and thus, NCA should be compositionally influenced only by primary H2SO4 (Figure 4). To this point, the authors state on line 32, page 2: "the unknown chemical composition of traffic-originated NCA-sized particles…". Although, I do not view the author's conclusion as impossible, or even unlikely, as evidence exists that emissions from motor vehicles contain H2SO4 gas which may contribute NCA emissions (Arnold et al., 2012). Nonetheless, the authors fail to support their argument with results at hand. At present, the evidence suggests NCA formation can occur independent of NPF events in respect to the H2SO4 condensation sink (CS). I appreciate the challenge in measuring the composition of particles on a particle-by-particle basis or the small size range of NCA due to mass constraints; however, pathway 1A (figure 4) is not supported in their data.

2) From my understanding organic vapors, NH3, NOx, and H2SO4, are all precursor gases to new particle formation events (Kerminen et al, 2018). I am not an expert in NPF events and do not have a great sense of the relative occurrence and frequency that these different gases contribute to NPF events. At present, CS is only calculated in respect to H2SO4. I encourage the authors calculate CS in respect to trace gases known to contribute to NPF events. The argument that these are truly primarily emitted particles will be supported by a more robust calculation of CS.

3) Presently, it is unclear about the frequency of NCA events in respect to the CS.

Timeseries data for NCA and CS are included as supplementary figures. The authors' current presentation makes it difficult to distinguish events as there appears to be a lot of covariance between CS and NCA (and other diurnal properties). I encourage the authors to include these panels (as well as traffic flow and solar radiation) as a main figure. Furthermore, it would be nice if the authors provided a more quantitative feel for the frequency that NCA events occur during periods of high CS. There may be better ways to do it, but something along the lines of a running (windowed) Pearson's correlation (defining an R threshold as an event) between CS and NCA would prove helpful in distinguishing these periods.

4) The authors argue that NOx provides a good proxy for traffic flow. This claim is moderately supported by averaged trend data provided in Figure 3. My concerns are twofold. First, although the variance in the profiles appear to co-vary, why not simply correlate NOx and traffic profiles (separating weekends and weekdays). As a side note, I do not understand the decision to take the geometric mean opposed to the arithmetic mean. Second, the authors do not state the significance for using NOx as a proxy for traffic. It is strange to me as the authors have a direct measure of traffic 600 m up the road which they use for justifying NOx as a tracer for traffic.

5) Perhaps I am missing the rationale; however, I think Figure 8 should be SI vs CO2, where SI is not binned into separate groups. If the argument is that CO2 does not have a diurnal profile, then the slope will be $\sim$0.

6) The abstract was lacking quantitative finding. I find that statements such as, "frequently correlated" (line 6, Page 1), to be extremely vague and really should not be in abstracts.

7) There is no statement on the availability of underlying data (https://www.atmospheric-chemistry-and-physics.net/about/data_policy.html).

8) The abstract makes mention of health effects. However, this was not a major finding in this work. Although fine to mention in the introduction and conclusion, it should not

be included in the abstract.

9) From my perspective, this is optional. The authors propose a conceptual model showing sink processes for NCA. I am generally interested in the loss rates for NCA in respect to the observed size distributions (i.e., coagulation rates). This would add a distinguishing feature from analysis presented in Hietikko et al. (2018). The authors would probably need to make assumptions about air parcels not mixing; however, this would provide an upper-bound estimate for NCA lifetime. I think this may have implications towards the authors earlier comments about potential health effects.

10) The authors did not outline how CS was calculated. Please include in the methods.

11) It is unclear what the authors mean by "weighting factor" as the authors regression analysis was not outlined in the methods. I am assuming the authors just average everything in a given bin.

12) The authors choice for bin widths (all regressions) may be justified but appear arbitrary without presenting a rationale. Personally, I think the data underlying regressions in Figures 6c,d Figure 7, and Figure 8 should be shown and not binned. If the data density is too high (graphical representation), the authors could possibly facet the different NOx and SI bins.

13) Figures 6c,d-8 should have confidence intervals for the intercepts so readers can evaluate the robustness in the intercepts (really relevant to 6c).

14) Line 18, Page 9: How many instances of NCA events occurred during cloudy weather? Was this the only time? This could be highlighted on the timeseries mentioned in main comment 3.

15) Line 2, Page 10: I am not familiar with momentary concentrations.

16) Line 4, Page 10: The use of relating NCA/H2SO4 to CO2 appears be creating an annual estimate of NCA. I did not think this was clearly articulated. Furthermore, on line 28, Page 5, the authors state: "because traffic does not cause a clear signal on

the measured CO2 concentration due to a high and varying CO2 background level, the NOx concentration was selected to represent the traffic-originated emissions overall."

Technical Comments: 1) Line 21, Page 2: "an evidence" should just be "evidence"

2) Line 10, Page 17: two doi's

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