review Comment on acp-2020-1282

Anonymous Referee #1

Referee comment on "Measurement report: The influence of traffic and new particle formation on the size distribution of 1–800 nm particles in Helsinki – a street canyon and an urban background station comparison" by Magdalena Okuljar et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1282-RC1, 2021

The manuscript by Okuljar et al builds on the legacy of urban particle and gas measurements in the city of Helsinki, the novelty being 1-3 nm particle measurements with simultaneous measurements of primarily new particle formation (NPF) vs. primarily traffic gas phase tracers, sulphuric acid (SA), and NOx, respectively. This combination of measurements, especially under the aspect of two collocated sites, one traffic the other urban background, is unique and yields insight into important processes that in turn are critical for future measures concerning air quality and human health. Thus, I recommend to consider this work for publication.

In my opinion, this work and the presented partitioning approach could further increase their impact by a more quantitative presentation of the key results together with the uncertainties and how the latter could challenge the most important findings here, e.g. that during daytime the direct traffic source of 1-3 nm particles can be minor (vs. the NPF source). It would be much appreciated if you could provide more detailed suggestions on how to adopt your regression approach (or the findings from it in your study) to other sites or other tracers to help address the knowledge gaps and uncertainties present in current generation air quality models. E.g. as of now the reader does not know whether your findings (e.g. the one summarized above) is in line or challenges current approaches on estimating total N of very small particles in urban areas. I provide some detailed comments below, and I’m looking forward to reading your work in the future.

General method:

In the introductions you report that also SA has a indirect and direct traffic source (L53&54), however in L209 you describe your usage of SA as a NPF tracer in your correlation/partitioning method (vs. NOx being the traffic tracer). Only much later in your case study description we learn that based on your observations SA seems to be much less correlated to traffic than NOx and sub 3 nm (street site). You acknowledge this uncertainty in L417 and state that it could lead to an underestimation of the traffic contribution to sub 3 nm N in your method. Could you investigate this error in a more quantitative way, how strong is the correlation between SA and NOx and does it vary as a function of traffic rate? By how much do you think you might underestimate the traffic 3 nm particle source due to this effect and does this underestimation vary under different
conditions? Could this uncertainty challenge your findings of only small traffic sources of sub 3 nm N during NPF?

You use the fitting coefficients for your partitioning model from correlations based on arbitrarily chosen background conditions, i.e. times when either NOx was low (for the sub 3 nm N vs. SA regression), or when SA was low (for the sub 3 nm vs NOx regression). If then applied to all data, how do you deal with measurements from times when NOx, or SA, were not low/or high. The regression coefficients might vary in the intermediate regimes (based on Fig. 9 and SI figure).

**Detailed minor comments:**

L44: why should the adverse health effects of nanoparticles change the number of deaths. Please rephrase and specify why you think that the modeled numbers are biased low, and how this ties to your study.

L54: I suggest to reorganize wording, put „emitted”, e.g.before “(Arnold)”

L58 I suggest to remove “on the other hand”. I don’t see why the following statement is somewhat contra- the statement before L58

L64: it could be helpful to give an example for particle sources that are specific to traffic activities vs. traffic emissions, maybe even before this line. Readers might think this is interchangeable.

L67-68: this sounds over-generalized, and is not very helpful for the reader. The general implications for traffic and traffic related emissions on human health are not ambiguous, don’t you agree?

L76: I suggest to use plural for „distribution”

L85: Wouldn’t it be useful to now the measurement heights and also the heights and distances to the closest roads, for both measurement station, respectively?

L150, correction described in limited detail. I suggest to describe more details of the procedure and the amount and variability of the resulting correction (could go in SI. At minimum the reader should know why this correction is important, how it changes the measurements and how certain you are about the correction result.

L199, please specify what type of measurement the 50% uncertainty number refers to: an instantaneous measurement? an hourly average concentration? Is this significant for the purpose of your study?

L228, doesn’t this „decreasing trend with size” also apply to the night period? (this might be obscured by log plot). Looking at Figure 2, It also appears that the <100 nm fraction has a much smaller relative contribution to total N as compared to the 3 other time periods (noon, afternoon, night). That seems surprising to me, given that the morning rush hour related to 3 nm N as you describe in the following lines. (e.g. in L279 you say that “the diurnal variation of nucleation mode particle concentration is similar to that of sub 3nm particles...”; or looking at fig 4a, the largest N is observed for 3-25 nm around 05 to 08 local time).

Fig2 – interestingly at the street site, the <100 nm fraction is the smallest in the morning. I suspect that the axis label should read nm, please correct if wrong.
Fig. 3, It would be helpful to know the difference in traffic density (not necessarily in real time from the time of measurement, but maybe from previous studies) between the two sites to interpret the significance of differences between blue and red lines.

Given that your sample size is not very large, how much do these figures change (especially plot c) if you limit your comparison to periods where all instruments at all sites were running simultaneously (i.e. around 100 or so hours for both sites). In the current version you subtract median statistics from a sample group “background” that is much larger (in sample size) than the sample group “street”. How sensitive are the described and discussed patterns to meteorology etc. which might be different between the two sample groups.

L350, here your discussion could improve by a more quantitative description of the background conditions and confounding parameters, rather than stating “traffic volumes are lower”, or “could be a plume from e.g. a ship”. Can’t you use your data and other data from Helsinki to investigate if this is a feasible explanation (wind direction, time of day).

L337: While I can follow your observation of more peaks in N <3 nm as compared to SA at the street site, it would be much more informative to describe and analyze this in a more quantitative way. This seems to be an important result of your study, it could be “fleshed out” by more statistics and significance testing. Is really the majority of N not originating from SA (L337)? It seems to me that the largest peaks in N are accompanied by largest peaks in SA and that the peaks in N without a counterpart in SA are rather minor peaks. It’s hard to judge by only looking at figures (thus my suggestion above).

L376, I suggest to rephrase “number of studied points” to sample size

Table 3, did you transform WD values before correlation? If so please state how in a footnote. If not, this correlation might not be useful.

L389, do you mean that bins were chosen to achieve similar classification sizes? What are “enough data points”? Maybe just use group NOx low (n = xxx), vs. group SA low (n = yyy), with bin cut offs of *NOx < ... to clarify this in a non-wordy fashion.

L391, not only is the slope of sub 3nm vs NOx much smaller at the background site, your plot 9d also shows a much more varying slope as a function of SA at the background vs. the street site. Do you agree? Could this be addressed in your discussions following L391. For example, for the yellow and red dot clouds, the fits might be similar in slope for street vs background.

L418, Couldn’t you study this possible relationship running a cross correlation analysis on your data? This ties to your discussion in line 467 about the possible traffic source of SA precursors.

Fig 10, I wonder if you would achieve a better fit for the background station, only using SA as a predictor.