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Interactive comment on "Emissions of organic aerosol mass, black carbon, particle number, and regulated and unregulated gases from scooters and light and heavy duty vehicles with different fuels" *by* R. Chirico et al.

Anonymous Referee #1

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This paper presents the results of chassis dynamometer emission testing of 7 different on-road vehicles spanning vehicle classes (2 wheel/4 wheel, heavy duty/light duty), engine types (2-stroke and 4-stroke spark ignition and compression ignition) and emission control regimes (several Euro specifications for the different vehicle types) operating on several drive cycles and fuels (biodiesel blends). Measurements were taken from raw tailpipe emissions and after one to three stages of dilution in a Constant Volume Sampler and ejector dilutors. Measurements were collected with a range of on-line gas- and particle instrumentation including an HR-TOF-AMS, MAAP, CPC, REMPI-TOF-MS,

FTIR along with a standard vehicle emission gas bench (CO2, CO, HC, NOx). Measured species include non-refractory aerosol species, BC, particle number (>7 nm), mono and poly-cyclic hydrocarbons, several nitrogen-containing compounds and aldehydes. The vehicles were operated under a number of conditions including urban drive cycles and transient and steady state speed steps. Data were analyzed to provide distance-based emission factors representing operation under different driving conditions and speeds. The authors highlight the large difference between emission factors of the different engine technologies represented, and their generally less substantial variation with varying speed/operating mode.

A substantial suite of instrumentation was applied during these measurements, and so the data does represent a detailed characterization of a varied selection of on-road vehicles. However, for several reasons I don't find the manuscript appropriate for publication in ACP. First, I question whether ACP is the appropriate venue for what is very plainly an emission characterization study. Apart from this, I have serious issues with the presentation of data and analysis, both in terms of depth and organization, and some technical issues, which would lead me to suggest not publishing this paper. Finally, a number of publications have come out of this group presenting vehicle emission measurements collected at the same facility of some of the same types of vehicles. The relationship between this work and these other studies is not made clear, nor is what unique contribution (apart from being a study of a number of different vehicle types at once) this paper makes. I did not learn anything new from reading this paper – that 2-stroke engines emit lots of OA and unburned fuel is not new, nor is that higher loads and speeds tend to lead to soot formation in diesel engines. That said, it is good for these data to be published in the archival literature as there is large variation between vehicles even of the same time, and so a contribution of this work would be to increase the observations available to those developing emission inventories and studying emission control approaches. Therefore, I would encourage the authors to work on improving the presentation of this work for resubmission, either in this journal or another.

ACPD
14, C5356–C5362, 2014

I have issues with much of the presentation in the paper. The introduction is rambling and includes excess details on instruments and methods, especially considering the lack of detail in actually discussing the results of measurements in the results section of the paper. For example, three paragraphs are spent describing the operation of the AMS, but the only way AMS data are presented is as organic aerosol mass, which doesn't even scratch the surface of the capability of the instrument and isn't an especially useful way to use this instrument – why not just use filters to capture integrated OC concentrations? The figures in the paper contribute little to the paper. Figures 3 through 6 are essentially massive 'data dumps' – each of them is essentially illegible unless I blew them up to full size on my monitor (especially Figures 4 and 5) and it's not clear what point can be made with these other than generally that emissions from different vehicle classes differ by orders of magnitude. It is not at all clear to me what a reader could expect to extract from these figures. For example, plotting the emission factors for 10 different gas phase compounds the emission factors of which vary over 9 orders of magnitude is not helpful (Fig. 5)? What understanding does this figure help the reader reach? I would expect more nuanced and detailed analysis that help illustrate the links between vehicle type/activity/etc. and emission factors. These figures are basically just the data in the supplemental tables in plot form. Figures should only be included if they somehow help make a specific point.

The paper presents the simultaneous measurement of the disparate vehicle population measured here as its main novelty. While engine type/technology/size/operation are key determinants of vehicle emissions, often the largest source of uncertainty in emissions and inventories is inter-vehicle variability. These measurements do not address this directly as they only measure one vehicle of each type, however the paper does very little to compare the results of his study with other studies, aside from limited and scattered comparison with literature values, which mainly concentrate on ratios (e.g. OA/BC) and on relative differences between vehicle classes or emission control specifications. The paper doesn't compare results with quantities measured from larger samples of similar vehicles in Europe or the US.

ACPD

14, C5356–C5362, 2014

Apart from these overarching concerns about the suitability and presentation of this paper, here I list several other specific concerns:

-As noted above, no analysis of AMS data is included in the paper (e.g. of OA spectra or size distributions). This is of course a useful set of data that resulted from these tests, but also provide indication of whether the AMS was fully capturing the OA, especially from low-emitting vehicles (e.g the spark ignition vehicles, which may emit substantial particle mass below the ∼70nm lower transmission range of the standard AMS aerodynamic lens. Examination of the particle time of flight data could allay concerns of this, and might also be useful information to publish. In addition, no discussion of AMS collection efficiency is included in the manuscript – this can have a factor of 2 or more impact on PM mass, especially in BC-dominated regimes where particle bounce may be a concern.

-The schematic shows that PM filters were collected, and given that this paper is presenting mostly full test-cycle average emission data, it is curious that none of this is discussed. For example, with OC/EC analyses, such filters could help constrain AMS CE and BC mass absorption cross-section, for which an assumed value of 6.6 m2/g is used without discussion. Filter analysis could help test for mass closure of the aerosol analyses by realtime instruments.

-Distance based emission factors are given, but fuel consumption (enabled by either direct fuel use measurements or estimation of fuel consumption via a carbon balance on the carbon-containing exhaust constituents) was not calculated. Given the work of Rob Harley which has shown that fuel-based emission factors are often less sensitive to operating conditions and thus a more consistent way in which to represent fleet-wide emissions, and also that fuel use data are often more available than vehicle travel data, this is an oversight. Calculation of real-time fuel consumption is fairly trivial using the carbon balance method. Finally, the general discussion of emissions from the different vehicles is imprecise. For example, the abstract states that the 'scooters were the main emitters of aromatic compounds', but doesn't specify on what basis (e.g. per

ACPD

14, C5356–C5362, 2014

distance, during a full test cycle, per fuel, etc.). I understand that in this case these vehicles will have the highest emission factors on whatever basis, but in some cases (e.g. comparing diesel and gasoline vehicles) this may be a critical distinction.

Specific points: P16604, L16: The discussion of the dilution correction methodology is very confusing, and I think incorrect. Here it is stated that 'background CO2 concentrations' were obtained by measuring diluted and tailpipe measurements and calculating them, while a few lines later it states that these were measured before each experiment. Also, the flows are mentioned and potentially used in these calculations, but there is no discussion of if/how they were measured.

P16607, L6: This statement is tautological – emissions are lower because concentrations are lower. This is unneeded and it has long been known that uncontrolled diesel engines emit much more PM than gasoline engines.

P16607, L20: The higher OA concentrations during low-speed, urban cycles with large transients are not at all surprising – PM emissions are associated with higher engine load, which is associated with accelerations. The exhaust temperature and catalyst activity may be a factor, but catalysts are not made to remove OA, which are likely due to larger loss of lubricating oil under high load conditions due to blow by in the engine.

P16608, L12: There are more than two possible explanations. Couldn't it just be that the engine is at higher load at higher speeds and so emits more OA?

P16609, L11: The 'volatility of the SVOCs' is an intrinsic property of the emissions, and so isn't affected by the dilution ratio. The phase partitioning of these SVOCs is a function of dilution. However, various work (e.g. (Grieshop et al. 2009; Saleh et al. 2011, 2013) has shown that equilibration time for these particles is substantially longer than they have in this dilution system. In the case of the Lipsky and Robinson, 2006 paper – these materials were those captured on quartz filters, which are sampled over a longer period and reside on the filter material, thus have time to equilibrate with the sampled gas stream.

ACPD

14, C5356–C5362, 2014

P16610, L8: Diesels at high load make soot due to incomplete evaporation of poorly atomized fuel, so while there is a very local high fuel-air ratio, this is not true in the sense it is usually used (average fuel/air in the combustion chamber). Again, this is a well understood source of BC from diesels.

P16611, L12: Work from these researchers should be referenced and discussed if it is mentioned.

P16613, L8: Given the huge influence of nucleation mode semi-volatile particles on particle number, it seems as though the values for PN emissions (at least for light duty vehicles) should be published with a more clear caveat in place that these may not be representative of actual number emissions due to the sensitivity of number emissions to dilution conditions (which in a CVS are not 'realistic'). In addition, given how large the contribution to the nuclei mode is to PN, I am curious how much of the overall mass is being captured by the AMS given its transmission window. The controls used by the PMP protocol (heated dilutor, thermodenuder and high cut-point CPC) are either not or inconsistently applied, and this should be made clear.

P 16613, L22-27: This section of text is repeated on the next page. There are several instances of repeated text in Section 3.2.1.

P16615, L28: CO2 emissions are essentially a proxy for fuel combustion. This should be discussed and as noted also providing fuel-based emission factors and/or fuel consumption values would be helpful.

P16619, L4-5: This is not a useful point or comparison. This ratio will vary widely with source and the aromatic composition of fuels. The ratio from source measurements is typically not used to 'calibrate' the photochemical clock, rather the evolution from a source region (e.g. downwind from a urban area) is used.

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ACPD

14, C5356–C5362, 2014

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