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**Condensational uptake of semivolatile organic
compounds in gasoline engine exhaust onto
pre-existing inorganic particles” by S.-M. Li et al.**

S.-M. Li et al.

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Response to the comments by Anonymous Referee #2

The original comments by the reviewer are given in brackets.

[1/ General Comments The paper describes experiments of interaction between organic gaseous emission of a gasoline (LD-SI) engine with pre-existing particles and the consequent formation of OA. Tests were done with diluted filtered exhaust and in-

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organic, laboratory-generated seed particles. Author's observations also lead to the conclusion that dissolution is the main reason for the uptake (amount of condensed organic mass is proportional to the seed particle mass).

The authors add new an interesting questions to the already challenging matter of SOA formation. By suggesting a new path to the formation of OA, their conclusions are lighting an argument which might open further studies with implications in the emission inventories and vehicle emission regulation as well as in sampling procedures described in the legislative protocols.

This paper shows the importance of opening a discussion on the effects of mobile emissions' sources when the exhaust gets in contact with atmospheric air at the exit of the tail pipe and expands. That concerns spark ignition vehicles, but not only. Dynamic conditions -cooling and dilution of the exhaust gas phase- may also introduce further questions about the influence of the ambient temperature and relative humidity in the exhaust evolution when emitted into the atmosphere.]

We agree. More experiments need to be conducted to probe the effect of T and RH on the condensation and evaporation processes.

[In any case, regardless the importance of the topic discussed in this paper, before concluding the significance of these findings in the regulation of mobile sources' emissions, further considerations and experiments should be necessary (i.e., use diluted non-filtered exhaust in the experiments, extend the study to other engines and fuels as well as different driving conditions). Revision of the accuracy of the emission databases or questioning the emission control policies need more than some indicators of their incorrectness.]

We agree that we need to conduct more experiments in light of the present findings, to help narrow down aspects of the processes not addressed in the current paper.

[2/ Specific Comments]

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[3463- line 22-24; The fact that the exhaust composition from spark ignition gasoline engines is very different to the diesel exhaust, makes this statement inappropriate. One of the negative aspects of diesel exhaust if compared with gasoline is that more nitrogen oxides (NO_x) and soot are released, while gasoline vehicles may have higher emissions of HC and CO. The drastically different composition of the exhaust makes, at least, questionable the similarity in the dilution effect and behaviour of different exhaust types once in contact to the air and close to the tailpipe. Aftertreatment devices have made those differences smaller but still the engine described here is not one of the latest generations.]

The text has been revised to take into account this difference.

[3466- line 7-8; This sentence is very imprecise. Description of the engine test requires more details. Time and length of the experiments should appear in the table. Some values for “Exposure time” are missing in Table 2/ pg 3483: days 19, 20 and 24th of April (?) The driving conditions, cold/hot start and settings of the engine greatly determine the exhaust composition. Most of the volatile hydrocarbons are emitted immediately after the engine starts running, until the catalyst is active and fully efficient. So experiments which have been done in very different starting conditions may lead to erroneous interpretation. May be the engine still hot when the repetition starts?]

We have added the durations for each set of experiments in Table 1. All experiments were run after the engine has warmed up and the temperature of the catalyst reached a steady state making the catalyst active.

Table 2 now has all the exposure times completed.

[3466- line 9-10; The authors study the formation of COM with known laboratory generated seed and they used diluted filtered exhaust, meaning exhaust gas phase free of the primary exhaust particulate. In view of the content of sulphur in the fuel used in the experiment, it should be very revealing to do also the experiments with diluted –non filtered- exhaust. HR-ToF-AMS could be used also in these conditions and it should

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consent to have a more realistic estimation of what happens when the vehicle exhaust (gas phase + particle phase) reaches the open air and meets seeds particles.]

We agree in principle, particularly if one could detect the chemical composition of the primary particles online. However, the AMS was thought not capable of detecting the primary particles because of their sizes (AMS effective particle size range is from 50 nm to 600 nm vacuum aerodynamic diameter, versus the 10-15 nm diameter for the primary particles). It may be able to detect the upper tail of the gasoline particles; this remains to be conducted. We did find a recent publication by Mohr et al. (2009) who reported AMS organic mass spectrum for particles from a gasoline engine emissions. We have included this reported result in our discussions.

Our goal was to specifically look into the process of organic vapour condensation onto existing particles. The presence of primary particles in such experiments would not have produced noticeably different results on the condensation. We have now given some detailed replies to the other reviewer who also has raised a similar issue. Essentially, the primary particles are expected to evaporate upon dilution, but the amounts of evaporated mass from these primary particles would not be large enough to make a difference to the COM.

[3467- line 27-29; Description of the experiments and data analysis are not very detailed. I fully agree with comment of anonymous referee #1 in page C781 that the AMS data should be included in this paper, at least as supplementary material. They are extremely important to support some of the conclusions reached in this paper and, particularly regarding COM.]

We agree. We have now included an AMS mass spectrum, which is quite typical from all experiments, in the revision (see new Figure 6). This figure answers a lot of questions of Reviewer #1. Text has been revised to include a discussion of the implications of the mass spectrum in light of ambient observations.

[3469- line 17-21; The effect described here is not the scope of the protocol for sam-

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pling vehicle emissions. Filtering the air coming into the dilution tunnel is a requirement for the reproducibility and repeatability of the methodology.]

We have revised the text to make clear the statement that such a processing is not considered in current standard testing methods.

[3/ Technical Comments]

[3469- line 19; Should say “are” instead of “is”]

Error corrected.

[3482- Table 1; Values for total mass (in $\mu\text{g}/\text{m}^3$) for 24th of April are very doubtful. In general the intra-day repetitions for this mode are not very good in terms of Total Mass. Same for the values of 01-May and Mode 2. May be due to the fact that the 2nd experiment was done when the engine was still hot (warm) from the previous experiment?]

Yes. In fact in all experiments the engine was hot and so it should not be an issue. In both cases of Apr 24 and May 1, there was a process of adding more fuel which may have cooled down the fuel temperature somewhat, and thus resulting in the changes in the total primary particle mass. The conclusions of the paper, however, are not affected by these temporary fluctuations.

[3483- Table 2; There are no values for the “Exposure Time” referred to days 19, 20 and 24th of April (?). It should be useful to see in this table again the engine mode (in table 1) as well as the seed particles composition.]

Exposure time values are now added to Table 2 for the three days.

[3485- Figure 2a; It would be better a bigger type for characters and values inside the graph and axis (smaller than graph 2b). Same for 3 and 4 3489-3492- Figures 5&6; It should useful to specify in the legend the domain covered (eastern North America) and grid space 15km.]

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The figure is now relabelled Figure 2 and remade. Figures 3 and 4 (new Figures 4 and 5) are now remade to make them more obvious. Figure captions for Figure 5 and 6 (now Figure 7 and 8) are revised to indicate that domain and grid.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 3461, 2011.

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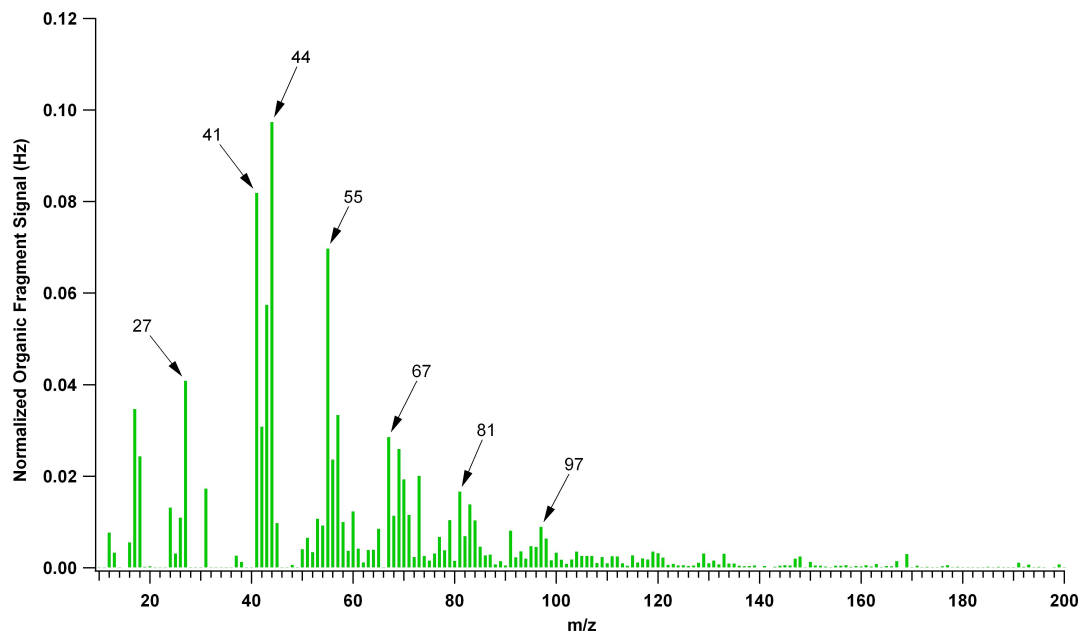


Fig. 1. new Figure 6

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