This paper presents interesting experimental results that show the importance of condensation of organic vapors in gasoline engine exhaust on preexisting seed. I have two comments on the theoretical interpretation and implications.

Comment 1:

The authors hypothesize that the uptake of organic vapors on ammonium sulfate seed is governed by solubility. This is explicitly stated in page 3471 line 22 "*for a dissolution process that reaches equilibrium with the gas phase, ratio of COM to seed mass is the solubility…*" The primary piece of evidence presented in the manuscript to support this statement is the linear relation between condensed organic mass (COM) and sulfate seed mass (M_s) presented in Figure 2a in the manuscript. However, this interpretation is not accurate. COM is dictated not by solubility, but by equilibration with the gas phase according to: $C_v = C_{v,0} - COM = x \gamma C_{sat}$, where : $x = \frac{COM}{COM + M_s}$ is the mole fraction of the organics (assumed to be equal to the mass fraction for simplicity); C_v and $C_{v,0}$ are the current and original (before condensation) organic concentrations in the gas phase; C_{sat} is the saturation concentration of organics; and γ is the activity coefficient. Here all the organics in the gas phase are represented by a single compound for simplicity (in the same fashion as the authors do). For this simple model, COM is given by the following quadratic equation:

$$COM^2 + (M_s + \gamma C_{sat} - C_{v,0}) COM - C_{v,0} M_s = 0 \qquad (1)$$

So, the ratio COM / M_s should decrease as M_s increases, because the seed becomes more efficient in scavenging the organic vapor and equilibration is achieved at lower COM / M_s ratios. This is shown in Figure 1 below for a hypothetical case. It depicts how the experimental data presented in Figure 2a in the manuscript might be mistaken for exhibiting a linear relation, because the seed concentrations investigated cover a relatively small range.
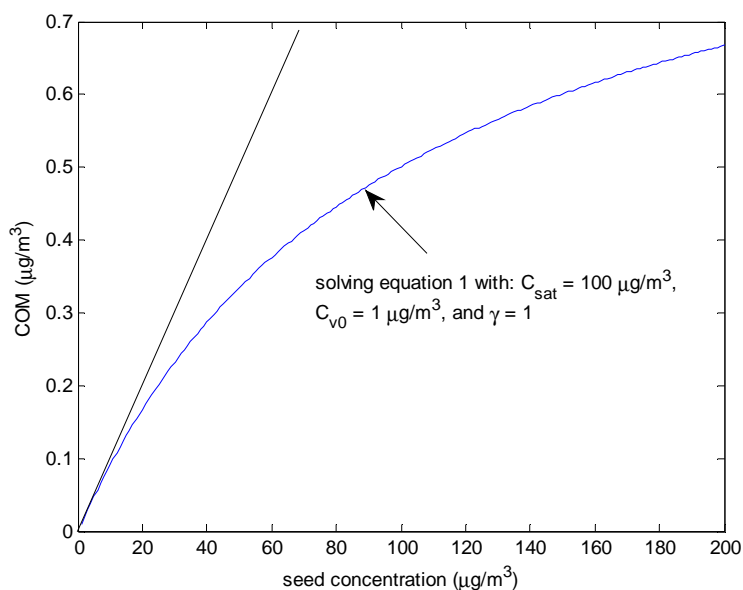
Figure 1

Solubility limits could not have been achieved in the experiments done in this work. Thermodynamic equilibrium of a semi-volatile compound at solubility requires that the activity of this compound in the solution be equal to the activity of the pure compound, i.e.: $x\,\gamma = 1$, in which case the concentration of the compound in the gas phase should be equal to C_{sat}. Since in these experiments, the gas phase exhaust was diluted by several orders of magnitude before introducing the seed, $C_v \ll C_{sat}$, and thus the mole fraction of organics in the particulate phase should be much less than $x_{solubility}$.

Along these lines, the authors make the following statement in the Abstract (and later in the text): "… *solubility decreases … with increased dilution*.." Solubility does not depend on dilution because it is an intensive property. In fact, the behavior observed by the authors and attributed to change in solubility (Figure 3 in the manuscript) is a direct consequence of absorptive partitioning, where a simplified analysis is given by equation (1) above.

Comment 2:

Based on their experimental results, the authors conclude that condensation of organic vapors in engine exhaust onto ambient seeds elevates PM levels by an amount comparable to the original PM in the emissions. This analysis leaves out part of the story. The authors look at partitioning of exhaust semi-volatile vapors on inorganic seed, but do not account for the potential evaporation (partitioning to the gas phase) of semi-volatiles from the original PM. If equilibration for a certain organic compound is originally achieved between the particulate

phase and the gas phase (before introducing the seed), condensation of this compound on the seed will disturb the equilibrium, and will cause evaporation from the original PM. This can potentially offset the increase in PM hypothesized by the authors, i.e. the net result will be a mere transfer of mass from the original PM (as it existed in the CVS tunnel) to the seed.