

This study investigates the effect of fuel ethanol content on primary particulate emissions and subsequent secondary aerosol formation potential of vehicle exhaust. The authors observed a decrease in PM loadings in both primary emissions and secondary production when ethanol fuel is used during the New European Driving Cycle (NEDC) by a flex-fuel vehicle. Compared with the initial submission, the authors have significantly improved the introduction section, especially clearly defined the scope and goal of the current study. A few issues, as listed below, remain to be resolved prior to publication on ACP.

General comments

This issue has been brought out earlier yet remains elusive in the current version of the manuscript. The SOA formation potential of ethanol is significantly lower than other common anthropogenic precursors such as aromatics and along-chain alkanes considering its high vapor pressure and small molecular size. As a result, it is not surprising that substitution of regular fuel materials with equal mass of ethanol leads to a reduction in PM emissions. The key question here is essentially the energy production efficiency of the ethanol substituted fuel. The authors are suggested to normalize the reported values for primary and secondary emissions either by the total fuel mass consumed or by the total energy produced during one NEDC driving cycle. For example, a unit of microgram per cubic meter particles per gram fuel consumed would be more appropriate and illustrative to evaluate the effect of ethanol content in the fuel on the PM emissions.

The authors report the primary emissions being 0.45, 0.25, and 0.15 mg m⁻³ for the E10, E85, and E100 fuel, respectively, over one driving cycle. It is necessary to give the uncertainty estimations associated with the measurements. Particularly, the authors need to measure the particle wall loss rate using inert aerosols (e.g., ammonium sulfate particles or black carbon particles) under identical experimental conditions and apply the loss rate to the measured overall primary emissions. Additionally, the authors are encouraged to discuss the uncertainties in the measured total hydrocarbon concentrations due to vapor wall losses.

The SOA yields from benzene, toluene, and xylene, and some alkanes under both low and high NO_x conditions have been recently corrected by accounting for the impact of vapor wall losses (Zhang et al., PNAS, 2014). The authors are suggested to update the SOA yields values in Section 3.5, which could potentially improve the extent of agreement between predicted and measured SOA mass.

Minor comments

- Page 7, Line 25: A brief description on the gas-phase measurements of CO, NO_x, and ammonia needs to be given.
- Page 9, Line 13: The unit 'mg/km' is inconsistent with the unit given in Figure 2. On the other hand, the concentration unit for Figure 1 is 'mg/km'.
- Page 10, Line 23: It is interesting that the oxidized fraction of particulate organics remains the same with and without the PAM chamber. Is this because the OH-reactive small hydrocarbons consumed most of the OH radicals but did not result in organic aerosol formation due to the high volatility of oxidation products?
- Page 11, Line 23: Inaccurate statement. Small particles (e.g., $D_p < 100$ nm) exhibit high deposition rate due to molecular diffusion. But the wall deposition rate of large particles (e.g. $D_p > 300$ nm) is equally high due to the gravity induced deposition and high inertia.
- Page 16, Line 15: I am not sure if wall loss plays a role here. Are the authors indicating that the wall loss rates E100 emissions are significantly different from the E10 and E85 emissions?