Explicit Quantification of In-Cabin Concentrations of School Bus Exhaust and Crankcase Emissions Using Intentional Tracers

Extended Abstract # 100

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INTRODUCTION

Exposure to motor vehicle emissions while traveling on roadways has become the focus of some attention due to data showing concentrations of particulate matter (PM) and other species inside vehicles that are high relative to even urban outdoor environments. Particular emphasis has been given to concentrations of diesel exhaust PM inside school buses that are attributable to the buses’ own exhaust. Previous studies have produced estimates of exhaust PM inside school buses that are attributable to the buses’ own exhaust ranging from 0.2 to more than 20 µg/m³. Most, however, relied on inferential and non-specific methods and did not include emission measurements. As a result, they were unable to definitively resolve bus emissions from those of other sources. Only one study of one bus (yielding the lowest in-cabin exhaust PM concentration estimate), using emission measurements and a fuel-based quantitative tracer, provided bus-specific results.¹

One study that sequentially applied retrofit emission control technologies² found using non-specific optical methods that crankcase vent emissions may be an appreciable contributor to in-cabin mass concentrations. Since crankcase and exhaust PM emissions differ markedly in size and composition, the ability to differentiate between them is important. Building from previous fuel-based tracer work, this study developed a new dual tracer method to quantify in-cabin concentrations of school buses’ exhaust and
crankcase particulate matter (PM). The new method continues the use of an organo-metallic iridium complex dissolved in the fuel, and adds a high molecular weight deuterated alkane tracer in the lube oil. A companion paper describes the carbon speciation methodology and results. On-road emission sampling was used to establish exhaust and crankcase emission rates for both fine particles (PM$_{2.5}$) and particle-bound iridium and deuterated alkane tracer. Samples collected inside the bus and inside a lead vehicle traveling in front of the bus were analyzed for tracers to allow estimation of the contribution of the two tracer-tagged sources.

EXPERIMENTAL

As a complement to a previously planned study of in-cabin exposures and health effects in Seattle school buses, a two-week measurement program was conducted to evaluate methods for quantifying in-cabin source contributions. Two conventional (front engine) school buses (model years 2000 and 2003, equipped with diesel oxidation catalysts) were tested on a residential school bus route. For each bus, in cabin and emissions testing was conducted during one week. Two closed-window in-cabin testing runs of approximately 2-hours duration were conducted on one day; two windows-open runs over the same route were conducted the following day. Following in-cabin testing, emissions samples were collected on portions of the bus route using a portable proportional sampling dilution tunnel. Three sets of exhaust samples and three sets of crankcase vent (road draft tube) samples were collected for each bus.

An organo-metallic coordination complex of iridium (Ir) was dissolved in a small amount of toluene and added to the fuel of each bus to yield an approximate concentration of 10 mg of Ir per gallon of fuel. The lubricating oil tracer was fully deuterated normal hexatriacontane (n-C$_{36}$D$_{74}$). 100 g of this material was dissolved in approximately 20 quarts of new lubricating oil, which was added to the first bus following an oil change, with additional oil added to bring the engine oil up to normal operating levels. This oil was drained from the first bus and used to refill the second bus prior to testing of the second bus.

In-cabin PM$_{2.5}$ samples were collected on 47mm Teflon and quartz filters at a target flowrate of 120 LPM. A glass impactor inlet for the samplers has a nominal cutpoint 2.5 µm at that flow. During sampling runs, four samplers were used with one pair collecting sample over the full duration of the run, and the other pair run with one pair of filters for the forward portion of the route (roughly the 1st hour of the route), and the other on the reverse route (2nd hour). Samples were also collected in a “lead vehicle,” a minivan with windows open traveling approximately three minutes ahead of the bus, to allow determination of background tracer levels and PM$_{2.5}$. PM$_{2.5}$ mass concentrations were determined by gravimetric analysis of the Teflon filters, which were then analyzed for iridium by instrumental neutron activation analysis (INAA). After taking a punch from the quartz filters for organic and elemental carbon analysis, the remainder of the filters were extracted and analyzed by gas chromatography and mass spectroscopy (GC/MS) for the alkane tracer and other species.
RESULTS AND DISCUSSION

Emissions Data

On-road PM$_{2.5}$ exhaust emission rates were typical of buses of this type and age. The first bus’s exhaust emission rate averaged 0.15 g/km over three tests, and the second bus averaged 0.14 g/km. Somewhat surprisingly crankcase PM$_{2.5}$ emissions are a significant fraction of exhaust emissions, averaging 0.03 g/km for bus 1 and 0.10 g/km for bus 2. Figure 1 shows these test results, as based on gravimetric analysis of the Teflon filters. Comparisons between the gravimetric mass and estimated mass calculated from EC and OC analyses of the quartz filters (calculated as EC + 1.2xOC to account for non-carbon elements) indicate that some sample line leaks may have occurred during testing.

Figure 1. Exhaust and Crankcase PM Emission Rates

Emission ratios of PM to tracer mass are shown in Figure 2 based on single filter analyses. For school bus tailpipe exhaust emissions, the gravimetric mass results from Teflon filters (DPMsb) are divided by the INAA iridium analyses of the same filters (Ir). For crankcase vent emissions, the estimated carbonaceous mass from quartz filter samples (calculated as EC + 1.2xOC to account for non-carbon elements) is divided by the GC/MS deuterated alkane results for the same filters. These results for single filter analyses yield mass ratios that can be used to estimate in-cabin concentrations of exhaust and crankcase vent PM concentrations. Some inconsistencies between gravimetric mass and carbonaceous mass estimates were observed for some source sample pairs, which is tentatively attributed to sample line leaks during on-road operations. A gravimetric to carbonaceous mass ratio for crankcase samples was calculated using sample pairs that did
not appear compromised. This ratio (1.04 ± 0.08) was applied to the carbonaceous mass:d-alkane ratios to calculate the total crankcase PM (PMck) to d-alkane ratio. The DPMsb:Ir ratio was determined directly from single filter analyses of Teflon exhaust samples. The ratios of iridium to d-alkane in tailpipe exhaust samples were more than 1000 times higher than these ratios in crankcase samples. As a result, given adequate sensitivity of in-cabin measurements for these tracers, they provide explicit markers from which the PM contributions of the corresponding source can be determined.

Figure 2. Ratios of PM to Tracers in Exhaust and Crankcase Emissions

In-Bus Tracer Measurements and Inferred Source Contributions

Using in-bus measurements of Ir and d-alkane tracer and the mass ratios of emissions to tracers, the contribution of each bus’s exhaust and crankcase emissions to in-cabin concentrations was estimated. Figure 3 shows the PM$_{2.5}$, EC, OC, and estimated DPMsb and PMck contributions for each set of 2-hour samples during bus 1 testing, and Figure 4 shows these results for bus 2 testing. For each, closed-window runs (designated C1 and C2 were followed by open-window runs. In all cases, lead vehicle tracer data were near the detection limits and are not shown. In cases where gravimetric mass is substantially below the estimated carbonaceous mass (EC + 1.2xOC), leaks in the Teflon filter sampling train are suspected. With windows closed, a relatively large fraction of in-bus PM$_{2.5}$ is contributed by crankcase emissions, ranging from about 9 to as high as 21 µg/m$^3$. A substantially smaller amount is contributed by exhaust, ranging from about 0.2 to 2 µg/m$^3$. Windows-open testing showed combined exhaust and crankcase contributions to drop to about 0.5 µg/m$^3$ or less for bus 1. For bus 2, windows-open crankcase contributions were approximately 2-3 µg/m$^3$, while exhaust contributions were about 5 times lower. Overall, PM$_{2.5}$ concentrations in both the buses and the lead vehicle
were low relative to in-vehicle measurements in other areas. In our previous study\(^1\), we observed in-vehicle PM\(_{2.5}\) concentrations ranging from 39 to 238 \(\mu\)g/m\(^3\).

**Figure 3.** PM\(_{2.5}\), Carbon, and Tracer Results for Bus 1 Runs

**Figure 4.** PM\(_{2.5}\), Carbon, and Tracer Results for Bus 2 Runs
SUMMARY

We utilized a dual tracer method to estimate the in-cabin concentrations of PM$_{2.5}$ from school bus exhaust and crankcase vent emissions. This approach to source attribution proved successful and powerful. Despite some sampling and analysis problems, and the limited number of tests, a number of observations can be drawn from this work:

- The sensitivity and specificity of the iridium tracer method observed in our previous single-bus study was confirmed, achieving a detection limit of approximately 0.002 µg/m$^3$ for DPM$_{sb}$.

- The use of a deuterated alkane lube oil tracer with appropriate sampling and analysis methods can also provide a specific and sensitive tracer for crankcase emissions. As applied here, the detection limit for PM$_{ck}$ was approximately 0.03 µg/m$^3$.

- Crankcase emissions can be a significant fraction of total bus PM emissions.

- In the two buses tested, in-cabin PM mass attributable to crankcase emissions appears to be consistently higher than that attributable to exhaust, with closed-window conditions leading to higher contributions of both.

- Exhaust contributions observed are consistent with or slightly higher than those we found in the previous iridium study, and again are substantially lower than estimates reported in studies using non-specific markers to estimate DPM$_{sb}$ concentrations.

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REFERENCES

