

Measurements of Dioxin and Furan Emission Factors From Heavy-Duty Diesel Vehicles In The United States

Alan W. Gertler, John C. Sagebiel, and William A. Dippel, Energy and Environmental Engineering Center, Desert Research Institute, Reno, Nevada 89506 USA

ABSTRACT

This paper describes the results of a study of the on-road emissions of dioxins and furans from mobile sources. This work was done in response to the US EPA's draft Dioxin Reassessment document that used data from sources outside the US to estimate an emission factor for the US fleet. The EPA estimate for dioxin emissions from the heavy-duty fleet was 0.8 ng/mile expressed in terms of TEQ (toxicity equivalents). The objective of this work was to measure on-road chlorinated dioxin and furan emission factors from in-use vehicles operating in the US with particular emphasis on heavy-duty vehicles. The experimental approach was to measure emissions the Fort McHenry Tunnel, Baltimore, Maryland. All air entering and leaving the tunnel was sampled for concentrations of dioxins and furans (during ten sampling periods of 24 hours each). The difference between the mass of material entering and the mass of material leaving the tunnel was taken to be the amount produced by the vehicles in transit. These measurements were combined with information on vehicle counts and tunnel length to determine an average emission factors. The study was conducted from October 25 to November 6, 1995. The average heavy-duty diesel emission factor determined in this study was 0.28 ng TEQ/mile.

INTRODUCTION

In a recent draft Dioxin Reassessment document,¹ the US EPA reports estimated dioxin and furan emission factors from mobile sources. The EPA estimate for dioxin emissions from the heavy-duty fleet was 0.8 ng/mile expressed in terms of TEQ or Toxicity Equivalents (a set of factors intended to adjust concentrations of various chlorinated dioxins and furans based on relative toxicity). This estimate was primarily based on studies conducted outside the US, including one on-road study done in a tunnel in Norway.² While there is little doubt that motor vehicles are sources of dioxins and furans, the magnitude of these emissions is uncertain. The application of the Norwegian results, which were confounded by a light-duty fleet operating on leaded gasoline, to the US fleet has also been criticized³ and the US EPA has indicated additional research is needed.

The primary objective of this work was to measure on-road emission factors for chlorinated dioxins and furans from in-use vehicles operating in the US. The approach taken was to measure mobile source emissions in a tunnel — the same methodology as was previously applied to measure regulated gaseous emissions from mobile sources.^{4,5}

EXPERIMENTAL METHODS

The Fort McHenry Tunnel is a four-bore tunnel, two lanes per bore, carrying Interstate 95 east-west under the Baltimore Harbor. This study was performed in Bores 3 and 4, the eastbound bores

Dioxin '97, Indianapolis, Indiana, USA

(length 2174 meters). Light-duty vehicles are allowed in both bores, while trucks are directed into Bore 4, the right-hand bore. The fleet in Bore 3 generally contained less than 2% heavy-duty diesel vehicles, while Bore 4 contained on average 24 to 25% heavy-duty diesel vehicles during the course of this experiment. Posted speed was 50 mi/hr in the tunnel, 55 outside. Traffic flowed freely except for sporadic light braking/slowdown at the exit at rush hour.

Sampling stations were set up at six locations: one each at the supply (air intake) for the ventilation air at the west and east ventilation buildings, one each on the catwalk in bores 3 and 4 at the west (entrance) end of the tunnel, and one each on the catwalk in bores 3 and 4 at the east (exit) end of the tunnel. Each station consisted of a high volume dioxin sampler and a sampler for particles less than 10 μm in aerodynamic diameter (PM_{10}) and a propeller anemometer for air flow (except the ventilation buildings). A video camera was placed at each exit station and video tapes from the cameras were used to determine vehicle counts and traffic composition.

Following sampling, the filters and PUF media were shipped to the analytical lab for analysis. The general sampling and analysis approach followed was that of US EPA Method TO-9,⁶ but employing the analytical improvements of Method 8290.⁷

RUN DESCRIPTIONS

A total of 15 runs during the 10 sampling periods were performed in the two bores (Table 1). There were 5 daytime experiments performed in Bores 3 and 4 (10 runs total) and 5 nighttime runs performed only in Bore 4 (5 total runs). Day runs commenced at 0600 and ended at approximately 1800. Night runs began at 1800 and ended at approximately 0600 the next day. End times are approximate since time was required to change out the sample media for the dioxin samplers. No speed data were recorded as part of this study. Based on previous Fort McHenry work speeds were on the order of 50 mi/hr with the entering traffic slightly higher and the exiting (uphill) traffic slightly slower. Bore 3 contained, on average 1.9% heavy-duty vehicles, while Bore 4 contained 24.2% heavy-duty vehicles. The fraction of heavy-duty vehicles in Bore 4 was similar for the day and night periods — 24.0% and 24.8%, respectively. Daytime vehicle counts in Bore 4 were 2.25 times greater than the nighttime counts. Bore 4 and Bore 3 daytime vehicle counts were, on average, within 10% of each other.

RESULTS AND CONCLUSIONS

The results of the chemical analyses were tabulated, validated, and emission factors were calculated for each run period using the methodology described by Pierson et al.⁵ The outlet, inlet, and vent concentrations in the Bore 3 samples were similar; therefore, emission factors could not be estimated. The outlet concentrations of all species in the Bore 4 samples were, on average, a factor of 2.0 to 4.5 greater than those from the inlet and vents and emission factors could be calculated for these runs. Since LD emission factors could not be estimated from the Bore 3 measurements, we could not separate the LD component from the Bore 4 results to obtain HDD emission factors. Given the large fraction of HDD vehicles in Bore 4 (24 to 25%) and the assumption that HDD dioxin and furan emissions were significantly greater than LD dioxin and furan emissions, all observed emissions in Bore 4 were attributed to the HDD fleet. The result was therefore an upper bound for the actual emission factor in the tunnel.

The average observed TEQ emission factor for the three valid day/day runs was 0.27 ± 0.02 ng-TEQ/veh-mi, where the reported repeatability was one standard deviation about the mean. For

TRANSPORT AND FATE

the four valid night/night runs, the result was 0.29 ± 0.18 ng-TEQ/veh-mi. For the limited range of vehicle operating conditions (speed and acceleration) and type (interstate trucks), the average HDD emission factor for the combined runs was 0.28 ± 0.13 ng-TEQ/veh-mi.

These results are lower than the EPA estimate of 0.8 ng-TEQ/mile. Possible explanations for the difference may be because the EPA estimate is based in part on a Norwegian study, where:

- The heavy-duty diesel fraction in the Norwegian study was between 3 and 15% of the total fleet and the results were extrapolated to 100% heavy-duty diesel.
- The light-duty fleet in the Norwegian study was operating using leaded fuel, a source of dioxins and furans.
- There are likely to be technology and fuel differences between Norwegian and US heavy-duty diesel vehicles.
- It is possible there were differences in load on the vehicles in the two studies.

Emission profiles were also compared with the results of German dynamometer tests⁸ (Figure 1). Given the differences in the tests, the results were in good agreement.

PM₁₀ emission factors were also estimated as part of this work. The observed heavy-duty diesel emission factor of 0.32 ± 0.11 g/mile was lower than the 0.54 ± 0.12 g/mile observed in a study in the Fort McHenry Tunnel in 1993.⁹ Although the results agree to within the experimental uncertainty, possible reasons for the apparent difference may be due to the shorter run periods (1-hr.) and the dominance of 5 high emission factor runs in the 1993 study.

CMB modeling¹⁰ was conducted on the Bore 4 outlet PM₁₀ filters. Resuspended road dust was found to account for 15.5 ± 3.3 % of the measured mass. The contribution from resuspended road dust to the observed dioxin and furan emission factors was estimated to be approximately 4%, calculated by incorporating the measured concentrations of dioxins and furans in collected road dust. Results of the inorganic analyses were also used to determine the impact of ambient PM₁₀ chlorine levels on dioxin and furan mass emission factors. An analysis of this data indicated there was no correlation between ambient PM₁₀ chlorine and dioxin and furan emissions.

LITERATURE CITED

1. EPA, *Estimating Exposure to Dioxin-Like Compounds, Volume I: Executive Summary*; U.S. Environmental Protection Agency. U.S. Government Printing Office, Washington, DC, 1994, EPA/600/6-88/005C5.
2. Oehme, M.; Larssen, S.; Brevik, E.M. *Chemosphere* **1991**, 23, 1699-1708.
3. Unsworth, J.F. *Comments on U.S. E.P.A. Draft Reassessment of Dioxins: Exposure From Heavy-Duty Diesel Trucks*, 1994, Report no. TRCP.3689R submitted to US EPA.
4. Pierson, W.R.; Gertler, A.W.; Robinson, N.F.; Sagebiel, J.C.; Zielinska, B.; Bishop, G.A.; Stedman, D.H.; Zweidinger, R.B.; Ray, W.D. *Atmos. Environ.* **1996**, 30, 2233-2256.
5. Pierson, W.R.; Brachaczek, W.W. *Aerosol Sci. Technol.* **1983**, 2, 1-40.
6. EPA, *Compendium of Methods for the Determination of Toxic Organic Compounds in Ambient Air*. Environmental Protection Agency. U.S. Government Printing Office, Washington, DC, 1988, EPA Document EPA/600/4-89/017.
7. EPA. *Method 8290: Polychlorinated Dibenzodioxins (PCDDs) and Polychlorinated Dibenzo-furans (PCDFs) by High-Resolution Gas Chromatography/High-Resolution Mass*

Dioxin '97, Indianapolis, Indiana, USA

Spectrometry (HRGC/HRMS). Environmental Protection Agency. U.S. Government Printing Office, Washington, DC, 1994. Revision 0, September 1994.

8. Hagenmaier, H.; Dawidowsky, N.; Weverrus, U.; Hutzinger, O.; Schwind, K.H.; Thorne, H.; Essers, U.; Buhler, U.; Greiner, R. In *Emission of Polyhalogenated Dibenzodioxins and Dibenzofurans from Combustion - Engines. Short Papers, Volume 2*. Presented at Dioxin '90, 10th International Symposium on Chlorinated Dioxins and Related Compounds; Bayreuth, Germany, September, 1990.
9. Gertler, A.W.; Wittorff, D.N.; Zielinska, B.; Chow, J.C. In *Proceedings of the A&WMA International Conference on Particulate Matter: Health and Regulatory Issues*, Pittsburgh, PA, 4-6 April 1995.
10. Watson, J.G.; Robinson, N.F.; Chow, J.C.; Henry, R.C.; Kim, B.M.; Pace, T.G.; Meyer, E.L.; Nguyen, Q. *Environmental Software*, 1990, 5, 38-49.

Table 1. Summary of experimental periods, vehicle counts.

	Bore 3	Bore 4	Bore 4
Period	Day/Day	Day/Day	Night/Night
Start Time	0600	0600	1800
End Time	1800	1800	0600
# Runs	5	5	5
Total light-duty	147,286	107,918	47,426
Total heavy-duty	2,875	34,096	15,642
Total vehicles	150,161	142,014	63,068
Fraction heavy-duty	1.9%	24.0%	24.8%

Figure 1. Comparison of homologue profiles from this study with those of Hagenmaier et al.⁸

