

Diesel particulate matter: risk management strategies for the transportation planning process

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Abstract

Mobile-source diesel exhaust particulate has been estimated to be responsible for more than 70% of the air toxic carcinogenic risk in the Los Angeles region. Diesel particulate emissions vary by engine operating characteristics (e.g., engine load, RPM), and operational characteristics will vary by facility type and congestion levels, thus producing different particulate emission levels. Existing emission factor models capture this effect through a functional relationship where the emissions vary with respect to vehicle speed. Second by second measurements on the US federal test procedure dynamometer driving cycle show PM10 spikes at the hard acceleration points. Transportation plans attempt to reduce congestion and smooth traffic flow. This paper combines available data on variations in diesel particulate emissions with engine operations to provide guidance on how transportation plans can indirectly affect the carcinogenic risk associated with diesel particulate matter from mobile sources.

1 Introduction

National and international agencies responsible for characterizing the possible toxicological risk resulting from air pollutants have identified diesel exhaust as a likely carcinogen [1-6]. California has established a unit risk factor of $3e-4$ excess cancers per lifetime exposure to one microgram of diesel exhaust particulate per cubic meter of air [7]. Diesel exhaust has been suspected of being carcinogenic since at least 1955, when Kotin et al reported on the presence of aromatics in diesel exhaust particulate [8]. Diesel exhaust particulate from mobile sources (including off-road equipment) is estimated to account for 70%



of the total air toxic carcinogenic risk of 1400 excess cancers per million in the South Coast Air Basin [9].

In this paper we develop a conceptual framework for analyzing regional diesel emissions risk. In particular our interest is in examining how changes in the transportation network might impact regionwide risk.

2 What is diesel exhaust

Diesel exhaust is principally a mixture of vapor phase pollutants, particulates with a carbon core; the particulates have condensed hydrocarbon and polynuclear aromatic hydrocarbon (PAH) compounds adsorbed onto their surface [2, 6], condensed droplets of semi-volatile hydrocarbons, sulfates, and trace metallic compounds are also present in both the vapor and particulate phases [1, 4]. Diesel exhaust contains low levels of carbon monoxide and hydrocarbons, and high levels nitrogen oxides and particulate matter [1]. Since the diesel cycle exhibits greater thermal efficiency, carbon dioxide emissions are lower from diesel vehicles per unit of work performed. The gas phase hydrocarbons and particulate matter have both been found to contain toxic and carcinogenic compounds. The principle components of diesel exhaust and their biological impacts are listed in Table 1, adapted from [10].

Table 1: Major constituents of diesel exhaust from [10].

Emissions	Phase		
	Vapor	Particle	Biological Impact
Carbon monoxide	x		Blocks oxygen uptake
Carbon dioxide	x		--
Oxides of nitrogen	x		A respiratory tract irritant and ozone precursor
Oxides of sulfur	x	x	A respiratory tract irritant
Elemental carbon		x	Penetration deep into the lung
Hydrocarbons			Respiratory tract irritants and ozone precursors
Alkanes <= C18	x		Respiratory tract irritants and ozone precursors
Alkenes <= C4	x		Respiratory tract irritants and ozone precursors
Aldehydes	x		Known carcinogens
Hydrocarbons (C14-C35)	x	x	Unknown
Monocyclic aromatic compounds	x	x	Known carcinogens
Polycyclic aromatic compounds (PAH)	x	x	Known carcinogens
Nitro-PAH compounds	x	x	Strong carcinogens

The gas phase of diesel exhaust has typical combustion byproducts such as oxides of nitrogen and sulfur, carbon monoxide, carbon dioxide, and hydrocarbons including PAH compounds [5, 6, 11, 12]. Hydrocarbon



compounds arise from unburned or partially burned fuel and engine oil [13]. Nitro-PAH compounds can also be generated during combustion and through atmospheric chemistry [14-17].

A useful framework for organizing discussion of particle formulation, which we have interpreted into Figure 1 [18], organizes particulate formation into four phases. The first phase is soot formation in the cylinder from partially burned fuel and oil molecules nucleating and coagulating into primary particles [13, 18-22] and growth of primary particles into agglomerates [13]. Oxidation reduces the diameter of the primary particles even as they form larger agglomerations [13, 21]. The second phase considers transformation by exhaust after-treatment devices -- the presence of particulate traps and oxidation catalysts affects the subsequent phases. The third phase accounts for dilution and cooling of the exhaust as it leaves the exhaust system. Additional particles form during the dilution phase including the nucleation of semi-volatile organics and sulfate [18, 19, 23]. The fourth phase, which accounts for atmospheric transformation of particulates, is of less interest to this study. Baumgard and Johnson cite several studies documenting particle formation downstream of ceramic particulate traps; one of the studies [21], showed that the ratio of semi-volatile particulate mass to that of solid particulate mass was five times higher downstream of a particulate trap. Since the trap removes solid soot particles it is inferred that the hydrocarbons are nucleating downstream from the trap [18].

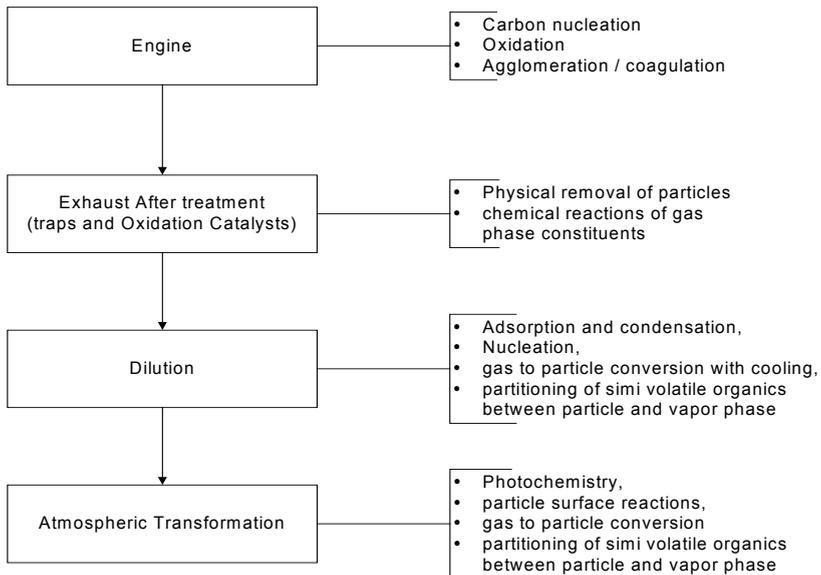


Figure 1: Particle formation/transformation processes adapted from [18].

The diesel exhaust particulate size distribution is bi-modal, having nucleation and accumulation modes [19]. The smallest particles, primary particles, are in



the nucleation mode from approximately 1 to 50 nanometers (0.001 to 0.05 microns), where gas to particle transformations and condensation occur [12, 18, 24-26]. Primary particles of inorganic carbon have a size range of about 10 to 80 nanometers (0.01 to 0.08 micrometers) [27-29]. Nuclei mode particles grow into the accumulation mode size range (80 to 1000 nanometers) through coagulation [30]. The surface area per unit mass does not decrease much as the particles coagulate; thus the bulk of the surface area occurs in the accumulation mode [12]. Figure 2, adapted from [19] depicts diesel particulates with an elemental carbon core coated in adsorbed organics. Primary elemental carbon particles coagulate to form agglomerates with a chain like shape [27]. These clusters have a very high surface area; citing industry data, CARB reports a range from 50-200 m² per gram of particulate [6]; Rothenberg et al report a range of 60-120 m² per gram of particulate but graphically show data ranging from 20-200 m² per gram of particulate [31]. At cooler temperatures the particles are coated with semi-volatile organics that are not present at higher temperatures [31, 32]. At high temperatures, most of the organics are in the gas phase [33] leaving more of the porous surface area of the elemental carbon exposed.

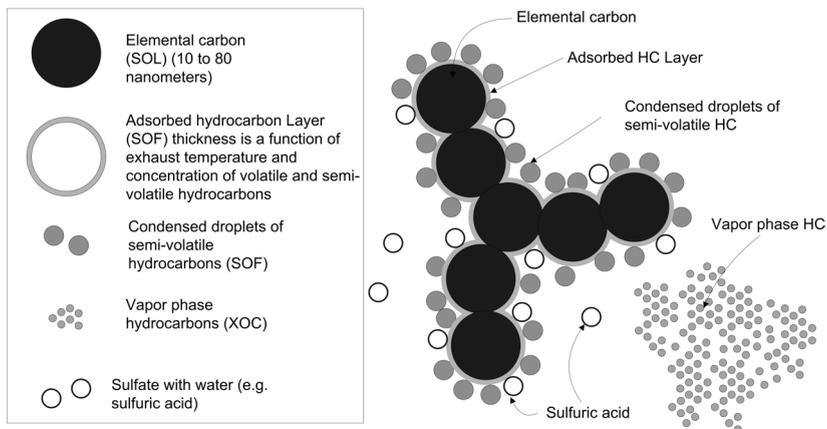


Figure 2: Schematic representation of diesel exhaust particles and vapor phase compounds adapted from [19].

The composition of the particulate matter varies with engine operating conditions and the type of exhaust after-treatment. A study of eleven trucks and busses reported that elemental carbon and organic carbon account for about 73% to 83% of the particulate mass without any after-treatment [34]. The presence of a particulate trap will alter the ratio of the elemental carbon mass to organic carbon mass; for example in the above study the traps were found to be about 95% effective, and their installation cut the elemental / organic carbon ratio from about 50% to about 5% [34]. The organic carbon portion of the particulate is referred to as the soluble organic fraction (SOF) because it can be extracted with

organic solvents. The remaining solid core is referenced as SOL in the literature. Gas phase organics have been referred to as XOC to distinguish them from those already condensed onto particles. Condensed sulfate is also present [19, 21]. Consistent with the results of Kishi et al reported above, engines running at low load typically produce fewer, but larger, particles with a higher organic fraction, while higher load, hotter, operation leads to an increased number of small particles with less associated organics [6]. Sulfate is emitted primarily in the form sulfuric acid (SO_4^{2-} or H_2SO_4), from the combination of SO_3 and water [1, 35, 36]. Sulfur is oxidized to SO_2 during combustion, but then requires a catalyst to convert it to the intermediate SO_3 form [35]; elemental carbon can serve as that catalyst though the reaction is slow [36]. Oxidation catalysts, used for hydrocarbon and carbon monoxide control on many light-duty diesel and newer heavy-duty diesel vehicles are also very effective catalysts for the SO_2 to SO_3 conversion making low sulfur diesel fuel critical to the implementation of clean diesel engine technologies [37]. Fuel quality also affects particulate mass emission rates. A comparison of emissions and mutability between diesel #2 and a high quality Fischer-Tropsch diesel showed a reduction in total particulate mass and mutagenicity of the SOF associated with a Fischer-Tropsch diesel blend having low aromatic content, low sulfur content, and a high cetane number relative to diesel #2 [38].

3 Carcinogenic mechanism

Increased risk of lung cancer with exposure to diesel exhaust is a primary concern. Mutagenic, acute and chronic effects are also suspected, the preponderance of the concern is with lung cancer, for example the epidemiological studies by Garshick and his colleagues [39-42]. The biological mechanism has not been identified but three broad hypotheses were reported by the California Air Resources Board [6]:

- That tumors result from carcinogenic compounds adsorbed onto particulate surfaces (e.g., the SOF). This mechanism would likely lack a dose/response threshold,
- That lung DNA is oxidized by something in the diesel exhaust other than particle induced inflammation (e.g., the XOF), and
- That the particulates are responsible for the carcinogenicity (e.g., the SOL).

From the first study in 1955, animal studies have shown that the soluble organic fraction is carcinogenic [8]. In studies exposing rats to only the gas phase of diesel exhaust, carcinogenicity has not been observed [4]. This result is consistent with work that found 90% of the Ames assay mutagenicity of diesel exhaust samples was associated with the particulate phase; and the bulk of that being from nitro-PAH compounds [43]. The elemental carbon core of diesel particulate is similar to carbon black. When exposed to diesel exhaust particulate, rats have shown greater susceptibility to lung tumors than mice or hamsters, with hamsters being resistant to lung tumors [10]. Studies have been performed that checked if biologically inert carbon black and titanium dioxide



have different dose/response curves than diesel exhaust in rats; no differences in the dose/response curves have been observed [44, 45].

4 Emission rates

The literature on diesel exhaust particulate emission rates can be divided into two groups: (1) the modeling of emissions, and (2) research on emission controls and measurements.

There are two regulatory emission factor models in the United States. The US EPA MOBILE model incorporated exhaust particulate emissions with MOBILE 6.1 via inclusion of the algorithm from the older PART5 model [46]. California has a separate model EMFAC 2002, which also estimates exhaust particulate rates [47]. For exhaust particulates, MOBILE 6.1 does not adjust the emission rate to account for changes in vehicle operation [48]. EMFAC 2002 uses an exponential 2nd order polynomial to adjust the emissions as a function of speed with nearly identical functions for all diesel powered vehicles. In addition to these regulatory models, Singh et al have developed a model for estimating particulate emissions from European vehicles [49-51], and a related model for US vehicles [52]. The two models also relate emissions to vehicle operation drawing on earlier fuel consumption and particulate emission data [53]. The European model also extends the relationship to look at modal profiles near intersections. The heavy-duty diesel vehicle speed correction factors from these four models are shown in Figure 3. None of these models have published a derivation of their speed/emission relationships.

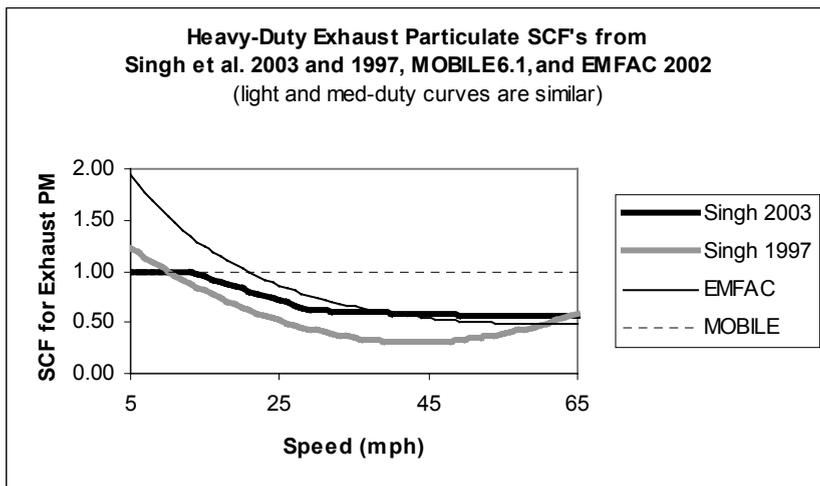


Figure 3: Heavy-duty vehicle speed correction factors.

Most model data is based on laboratory dynamometer testing of engines and vehicles over driving cycles that attempt to capture the range of expected

operation conditions. For example, the UDDS [54] and 13 Mode Test [55] are used in MOBILE; the European Regulation 49 Test is the same test cycle as the 13 Mode Test [56]. The resulting emissions data are representative of aggregate vehicle activity and can not tell us much about what types of roadway investments will tend to perturb this aggregate emission rates either up or down [57]. The emission testing literature provides some insights that the aggregated emissions modelling literature does not provide.

There is qualitative evidence that particulate emissions are reduced as traffic flow is smoothed. Different dynamometer cycles have been developed to specifically represent slow moving stop and go truck creeping in and around freight yards and loading docks, operation on city streets, and operation on highways. The California Air Resources Board developed a cycle with an idle mode, yard mode, transient mode (representing stop and go driving conditions on streets), and a cruise mode (representing freeway driving) [58]. As the test progresses through the modes, from creep mode to transient mode and finally to cruise mode, it qualitatively becomes smoother. Testing of a 400 hp Mack tractor resulted in measured emissions of 1.061 g/mile for the creep mode, 0.561 g/mile for the transient mode, and 0.138 g/mile for the cruise mode, averaged over 3 tests [58]. West Virginia University has developed dynamometer cycles representing a variety of operation conditions, and which support the concept that smoother flow reduces emissions. Their results indicate that cycles with low speeds and lots of idle produced more particulate emissions [59].

Particulate emissions are correlated with high power operation and sudden increases in power, but differentiating the relative importance of each effect is difficult because transients also coincide with high power events [60]. Studies on the repeatability of emission measurements between laboratories have demonstrated that the relative aggressiveness of the vehicle operation on a test cycle can account for large changes in particulate emission rates [61]. A non-aggressive driver will have fewer instances of high power operation and fewer large power transients than an aggressive driver. Work done to establish the validity of fast response particulate measurement instruments has also shown the relation between high power transients and increases in emissions. Figure 4 adapted from [62], corroborates that hard accelerations result in spikes of diesel particulate.

The carcinogenicity of the particulate emissions likely varies with the operational conditions. It was shown earlier that low engine temperatures, corresponding to low load, tends to increase mass of the soluble organic fraction of the particulate relative to that of the carbon core by increasing the thickness of the adsorbed hydrocarbon layer [32]. The relative mutagenic potential of diesel particulate extracts from an engine operating at a variety of speed and torque combinations was studied via Ames tests [38]. These data show that the highest mutagenic effects occur with intermediate engine loads, followed by high load, with low engine loads having much lower mutagenic effects. However, the fuel injector type used in the test was one that has problems atomizing fuel at low injection pressures, and what was thought to be unburned fuel stained the sample filters [38] during the low engine loading tests; this may discredit the low load



results. It would be reasonable to anticipate that mutagenic effects would decrease as engine load increased because the higher temperatures associated with higher engine loads should lead to more complete oxidation of the organics.

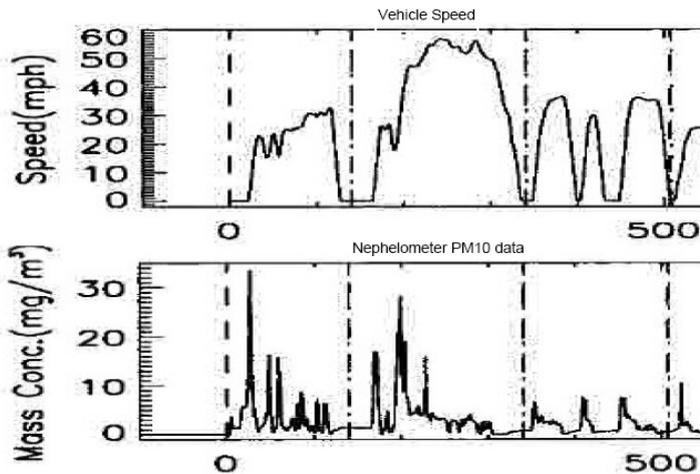


Figure 4: Fast response data for a 1996 Dodge diesel pick-up adapted from [62] showing spikes in particulate emissions coinciding with hard accelerations. Horizontal axes is time measured in seconds.

5 Discussion

The SOF of diesel exhaust particulate contains carcinogenic compounds, and lung cancer has been observed in epidemiological studies. Laboratory tests have shown that diesel exhaust and diesel exhaust particulate is carcinogenic in animal exposure studies, but no carcinogenic effects have been observed in animal tests using only the gas phase of diesel exhaust. It is therefore reasonable to focus on how changes in the transportation system would impact the quantity and properties of the SOF when assessing carcinogenic risk from transportation investments. Smoothing of traffic flow reduces particulate emissions. It is not clear if high engine loads, large transients in load, or some interaction between the two are responsible for higher particulate emissions rates observed. If carcinogenic impacts are correlated with less-oxidized soluble organics then increasing network speeds might reduce the carcinogenic potential per unit mass of particulate emissions. The one study that looked at mutagenic potential vs. engine conditions was inconclusive as the low load samples appeared to have been sprayed with un-combusted fuel. Transportation investments that target traffic smoothing projects without substantially increasing average speed are likely to be the best strategy for minimizing exposure.

If soluble organics are the principle cause of concern, then particulate count and size distributions are not a significant issue in assessing risk as the surface area per gram of particulate does not decrease as particles coagulate. Mass based

emission data may therefore be reasonable surrogates for risk given the lack of information about how carcinogenic potential varies with engine operation.

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