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Breathing easier? The known impacts of biodiesel on air quality

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Abstract

Substantial scientific evidence exists on the negative health effects of exposure to petroleum diesel exhaust. Many view biodiesel as a 'green', more environmentally friendly alternative fuel, especially with respect to measured reductions of particulate matter in tailpipe emissions. Tailpipe emissions data sets from heavy-duty diesel engines comparing diesel and biodiesel fuels provide important information regarding the composition and potential aggregate contribution of particulate matter and other pollutants to regional airsheds. However, exposure – defined in this instance as human contact with tailpipe emissions – is another key link in the chain between emissions and human health effects. Although numerous biodiesel emissions studies exist, biodiesel exposure studies are nearly absent from the literature. This article summarizes the known impacts of biodiesel on air quality and health effects, comparing emissions and exposure research. In light of rapidly changing engine, fuel and exhaust technologies, both emissions and exposure studies are necessary for developing a fuller understanding of the impact of biodiesel on air quality and human health.

Diesel engines as a source of air pollution & associated health effects

Substantial scientific evidence exists on the negative health effects of **exposure** to petroleum diesel exhaust (DE) [1–3]. DE emissions include fine (≤2.5 μm) and ultrafine (≤0.1 μm) **particulate matter** (PM), oxides of nitrogen (NO_x), SO_2, carbonyls and other organic compounds such as polycyclic aromatic hydrocarbons (PAHs). Additionally, large PAHs (greater than five rings) and nitro-PAHs, many with mutagenic and carcinogenic properties, may adsorb onto the high surface area PM [1]. Diesel PM (DPM) also contains small amounts of metals that may pose health risks. The chemical and physical characteristics of DE will vary depending on fuel, engine type, operating conditions and atmospheric transformation processes.

Estimates of the contribution of DPM to ambient PM less than 2.5 μm in size ($PM_{2.5}$) in the USA range from 6% of the total $PM_{2.5}$ inventory to 10–36% in some urban areas in California, Colorado and Arizona [1]. Negative health effects associated with exposure to $PM_{2.5}$ include increased emergency room visits, reduced lung function, exacerbation of asthma, arrhythmia, hypertension and increased mortality rates [4–6]. A comprehensive review of epidemiological *in vivo* and *in vitro* research suggests that exposure to vehicular emissions may be a major environmental factor in US cardiopulmonary mortality and

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morbidity [7]. An integrated review of the epidemiologic literature conducted by Pope *et al.* indicates that low to moderate exposure (5–50 $\mu g/m^3$) to PM $_{2.5}$ (from all sources) results in an exposure–response relationship that is very steep and nearly linear, suggesting there may be no threshold or ‘safe’ level of exposure to PM [6].

Public health scientists are concerned about both cancer and noncancer effects from DE exposure. A recent epidemiological study of underground miners found an increased risk of lung cancer mortality associated with DE exposure after adjusting for other potential confounding factors [8]. A cohort study of railroad workers with occupational exposure to DE indicated elevated lung cancer mortality [9]. US regulatory agencies have determined that DE is a ‘potential occupational carcinogen’ [10] and ‘likely to be carcinogenic to humans by inhalation’ from environmental exposures [1]. Recent studies have associated DPM with cellular oxidative stress and pro-inflammatory responses [11–13]. Increased reactive oxygen species activity or expression of inflammatory cytokines has been reported from *in vitro* studies of DPM and traffic-related PM [11–16]. DPM cardiopulmonary toxicity appears to correlate with its oxidative and inflammatory potential, which is suggested to reside in the polar and PAH fraction of DPM [17]. Transition metals may also play a role in DPM’s oxidative potential [18].

Biodiesel as an alternative to petroleum diesel fuel: a brief overview

Based on the impact to public health, reduction of DE is a desired policy objective. A number of technological risk interventions such as ultralow sulfur diesel fuel, diesel particle filters, oxidative catalysts and improved engine technology have been suggested or implemented to reduce the impact of DE on human health. Onroad heavy-duty diesel (HDD) engines manufactured after 2007 are engineered to yield far cleaner emissions, and **nonroad engine** improvements will be phased in after 2014 [5]. US air quality trends have improved in the last decade, most notably for SO_2, NO_2 and PM_{10}. Ambient 24-h $PM_{2.5}$ levels were 28% lower in 2010 compared with 2001 [19]. However, with HDD engine life expectancy commonly approaching 15–20 years, older HDD models will remain in use for decades. Alternative fuels that can potentially reduce emissions today are of great interest.

Biodiesel, a fuel made from vegetable oil, animal fat or waste grease, has received increased attention in the USA as a renewable and domestic energy source. While relatively recent in the USA, biodiesel has been used in western European countries for at least the last 10–15 years [20]. Biodiesel fuel is biodegradable and has high lubricity characteristics, which may help extend engine life [21]. Hill *et al.* concluded that soy-based biodiesel, compared with diesel, yields 93% more energy than the fossil fuel energy that went into its production, and reduces GHG emissions by 41% [22]. However, biodiesel’s claim as a ‘greener’ fuel is not without controversy. For example, Crutzen *et al.* counter that biodiesel’s impact on GHG emissions is less favorable, as biodiesel agricultural activities increase N_2O emissions, which are more ‘potent’ in warming potential and may offset any CO_2 reductions [23]. Finally, biodiesel and diesel fuel properties differ in ways that impact engine performance. Biodiesel has a higher cetane number, preferable in compression ignition engines. However, biodiesel fuel consumption is typically higher, mainly due to biodiesel’s lower heating value, higher density and higher viscosity [24]. Higher viscosity is also associated with poorer fuel injection atomization, which may affect cold weather performance and emissions [24]. B100 (100% biodiesel) will start to cloud at approximately 32°F and gel at 25°F, limiting B100’s suitability in colder climates [21]. As a result of these properties, biodiesel is added to petroleum diesel in the USA in blends of 5–20% by volume.

Biodiesel: a better alternative for air quality?

Recent data suggest biodiesel may have positive benefits for air quality and human health. Numerous studies have shown that burning biodiesel compared with petroleum diesel reduces PM, CO and total hydrocarbons in tailpipe exhausts [24–30]. The reduction in PM is typically associated with biodiesel's higher oxygen content and lack of aromatic hydrocarbons and sulfur [24,27]. However, researchers have noted increased PM in biodiesel emissions from passenger cars under certain fuel type and engine operating conditions [31–33]. Use of more saturated [31] or oxidized [33] biodiesel blends resulted in higher PM emissions, and cold-start operation also led to increased biodiesel PM in the exhaust [32,33].

Ultimately, multiple factors are critical in understanding the impact of biodiesel on air quality and human health. Although biodiesel blends generally reduce total mass of tailpipe PM compared with petroleum diesel, some studies suggest biodiesel increases particle number concentration or decreases particle size [34,35]. Other researchers have found biodiesel use decreased particle number concentration and decreased particle size [36,37]. With respect to health impact, as particle size decreases, lung deposition efficiency increases [38]; smaller particles, with less total mass, may penetrate deeper into the lung, potentially presenting significant health risks. Evaluating PM composition for organics, PAHs and metals also provides important information on PM toxicity. Many PAHs are known carcinogens and metals play a key role in reactive oxygen species activity associated with inflammatory responses [14,18]. Some researchers have found higher concentrations of metals in biodiesel PM, suggesting higher toxicity [39,40], while others determined similar profiles of metals between fuels [41]. The soluble organic fraction (SOF) of biodiesel PM is typically higher than the SOF of diesel PM [42–44] but the composition and impact on health is unclear. Recent studies of diesel car engine emissions determined biodiesel use decreased total PAH and nitro-PAH concentrations (gas and particle phases) [31] and DPM-associated PAHs [45]. These results suggest that biodiesel's influence on PAHs may be beneficial for human health.

Another approach to understanding the impact of biodiesel on health is evaluating biological responses in cellular and animal models. *In vitro* and *in vivo* studies of biodiesel health effects are limited. Swanson *et al.* reported that biodiesel PM SOF may be a more potent inflammatory stimulant to human airway epithelial cells than diesel PM SOF [46]. Cheung *et al.* determined that the oxidative potential (as measured by the dithiothreitol consumption assay) of both biodiesel and petroleum diesel PM emitted from a car engine were similar, although the biodiesel consumption rate was lower [18]. Jalava *et al.* investigated a wide array of toxicological endpoints (inflammation, cytotoxicity, genotoxicity and oxidative stress) generated by PM emissions from a nonroad engine operated in multiple diesel/biodiesel fuel and engine configurations, with and without a catalyst [41]. In general, the toxicological potency test results between fuel types were either similar or reduced with use of biodiesel compared with diesel [41]. Brito *et al.* reported biodiesel exhaust was more toxic than DE in a mouse model as exposure promoted cardiovascular alterations and pulmonary and systemic inflammation [47]. Assessing differences in toxicological properties between petroleum diesel and biodiesel emissions, and characterizing 'real-world' exposure scenarios to biodiesel exhaust, continue to be important research needs [46,48].

Biodiesel: comparing emissions versus exposures

While biodiesel emissions studies suggest air quality and health benefits from reductions in PM mass and hydrocarbon concentrations, biodiesel exposure studies are nearly absent from the literature. Exposure is typically defined as contact between a chemical, physical, or biological agent with a target, such as a person [49]. Exposure is considered the intermediate step in the following process, based on a conceptual model of risk outlined by Ott *et al*: sources of pollutants, movement of pollutants, exposure, dose and health effects [49]. Evaluating each step in this conceptual model of risk is critical to understanding biodiesel's overall impact on air quality and health. **Tailpipe emissions studies** help elucidate the first step but it is unclear whether biodiesel tailpipe emission reductions will result in similar exposure reductions in the workplace and near-field environment. A number of diesel **exposure assessments** in various occupational settings have been performed over the past two decades. A review of this literature completed by Pronk *et al.* shows most workers (such as drivers, mechanics and equipment operators) typically experience DPM exposures considerably higher than those experienced by the general public [50]. There is little known about the impact of biodiesel use on exposure in occupational settings.

Certainly, performing tailpipe emissions testing via set protocols has advantages compared with exposure assessment, such as researcher control over environmental variables (e.g., temperature, dilution ratios, wind speed and humidity) and operational variables (e.g., engine model and duty cycle). However, such data may not necessarily reflect emissions from real-world engines, 'stop and go' driving patterns or pollution impacts from nearby traffic. Shah *et al.* used a mobile laboratory to measure on road, 'real-time' petroleum diesel tailpipe emissions from heavy-duty trucks and found that PM, **elemental carbon** (EC) and **organic carbon** (OC) levels were highly variable and strongly dependent on the mode of vehicle operation [51]. Trucks in congested traffic conditions produced higher emissions compared with highway cruise conditions [51]. Other researchers found that the OC/EC ratio of DPM varies, with heavier load conditions increasing EC levels and lighter load/idling conditions increasing OC levels [52]. The real-world variability in engine model and operating modes; the fate, transport and chemical reactions of emissions in the atmosphere; and the impact of changing meteorological conditions, make it difficult to predict occupational or community exposures based on tailpipe emission data sets. All these factors influence exposure and, ultimately, health effects.

On-road studies comparing biodiesel with diesel reveal notable differences compared with tailpipe emissions studies. The tailpipe emissions literature indicates B20 use reduces PM emissions by an average of 10–24% [25,26,29]. An on-road study comparing B20 and diesel emissions from motor graders during various real-world repair activities, such as resurfacing, found PM emissions factors were reduced from 8 to 48% with B20 use [53]. Bugarski *et al.* evaluated the use of biodiesel blends in a 56-horsepower nonroad engine operating under different conditions in a simulated underground environment [54]. Increasing the blend percentage from B50 to B100 generally decreased the EC fraction and consistently increased the OC fraction, suggesting an increase in particle-bound organic fraction of the PM [54].

In short, real-world operation may produce far different biodiesel emissions profiles than predicted by standardized engine test protocols. This adds a layer of complexity in understanding average exposure concentrations in a workplace or community setting. Exposure assessment attempts to quantify the concentrations of pollutants people are actually inhaling. Another benefit is the collection of exposure data associated with in-use engines. Yanowitz and McCormick report that the biodiesel emissions database may not be representative of the current North American fleet as more than 50% of the tested engines were from 1995 or earlier, while over 75% of on-road vehicles were post-1995 models [30].

Exposure assessment studies of biodiesel can also take advantage of well-established industrial hygiene and environmental air monitoring methods. Many of these methods have been developed by regulatory agencies and are easily adaptable for use in the field. For example, EC is considered a 'signature' for DE and its determination via the National Institute of Occupational Safety and Health method 5040 is well accepted [3,55,56]. Exposure assessment is performed by measuring concentrations of air contaminants over time in a worker's breathing zone ('personal sample'), at a specific location within the workplace ('work area sample') or in a nearby location ('near-field sample'). Data can be collected by trained personnel, making it an excellent opportunity for community participation [57] or undergraduate research [58,59]. Results can be compared against existing occupational and environmental health standards to evaluate impact on exposure and health. Common DE exposure scenarios include construction sites, industrial warehouses and underground mines [60].

Biodiesel's impact on exposure in an occupational setting

The author and colleagues evaluated B20 versus petroleum diesel exposure profiles (in-cabin, work area and near-field) at a rural materials recovery facility utilizing heavy-duty nonroad equipment [58,59]. B20 use resulted in significant reductions in $PM_{2.5}$ mass (56–76%), reductions in EC (5–29%) and increases in OC (294–467%) [59]. Concentrations of $PM_{2.5}$ measured during petroleum diesel use were up to four times higher than $PM_{2.5}$ concentrations measured during B20 use [59]. Our study and others demonstrate that workers typically experience much higher exposures to DPM than populations in polluted urban areas [61–65]. Use of B20 as an alternative to petroleum diesel may help reduce workplace concentrations of DPM. Additional biodiesel versus diesel exposure assessment studies in different real-world settings are recommended.

Future perspective

While our exposure studies indicated B20 use reduced PM and EC concentrations, OC concentrations significantly increased, although they were comparable with OC levels reported in diesel exposure assessments [55,64,65]. More detailed chemical characterization of the OC fraction and determination of metals in real-world biodiesel PM are recommended. Additional *in vivo* and *in vitro* research on real-world biodiesel PM is also warranted to understand the relationship between exposure and health effects.

Both tailpipe emissions data and exposure data are less clear regarding the impact of biodiesel on other air toxics of importance, such as formaldehyde, acetaldehyde, benzene and others. LaPuerta *et al.* indicated no real trend in the biodiesel emissions literature regarding aldehydes and other compounds [27]. Di *et al.* determined increasing the biodiesel blend percentage decreased emissions of formaldehyde, 1,3-butadiene, toluene, xylene and increased acetaldehyde and benzene emissions [28]. Fontaras *et al.* determined a significant increase in carbonyls (including formaldehyde, acetaldehyde and acrolein) in tailpipe emissions during B100 fueling [32]. Traviss *et al.* reported lower formaldehyde and acetaldehyde exposure concentrations after equipment switched to B20 [58]. Finally, biodiesel use is generally considered to increase NO_x emissions [27,28]. NO_x emissions and exposures can have negative health effects, as well as play key roles in ground-level ozone formation processes. However, other researchers have shown that NO_x emissions are highly variable between engine types and there may be no net increase in NO_x up to a B20 blend [26,30]. More research on the impact of biodiesel on NO_x emissions and real world exposure profiles would be beneficial.

Existing tailpipe emission and exposure studies indicate that the use of biodiesel blends reduces PM mass concentration compared with conventional petroleum diesel. As negative health effects associated with $PM_{2.5}$ exposure are well established, biodiesel's impact on reducing PM is an important benefit. However, as fuel feedstocks, blend percentages and engine after-treatment technologies continue to change with competing regulatory mandates and economic forces, ongoing research is needed for both diesel/biodiesel emissions and exposures. This research should include characterization of the physical, chemical and biological properties of biodiesel particles and gases in laboratory and real-world settings. Ultimately, both emissions and exposure studies are necessary for developing a fuller understanding of the potential impact of biodiesel on air quality and human health.

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Key terms

Exposure	Contact between a chemical, physical or biological agent and a person
Particulate matter	Complex mixture of tiny (micrometer to nanometer scale) particles and liquid droplets suspended in air, with composition depending on the source. Particulate matter can be made up of hundreds of different chemicals
Nonroad engines	Diesel engines used in a wide range of applications such as logging, construction, agriculture and other industrial uses
Tailpipe emissions studies	Emissions collected via engine dynamometer studies. These studies follow a specific protocol (such as the US EPA Federal Test Procedure at Code of Federal Regulations, Title 40 part 86 subpart N), including how the engine is operated (its load and speed for different time periods, and how emissions are measured)
Exposure assessment	Determination of the magnitude, variability and duration of an exposure
Elemental carbon	The insoluble carbon fraction of soot
Organic carbon	The particle-bound, soluble organic fraction of soot

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Executive summary

Diesel engines as a source of air pollution & associated health effects

- Exposure to the extremely tiny particles and gaseous pollutants generated by the combustion of petroleum diesel fuel is associated with a wide array of negative human health effects.

Biodiesel: a better alternative for air quality?

- Studies have shown use of biodiesel blends in diesel engines reduces tailpipe emissions of particulate matter, total hydrocarbons and total polycyclic aromatic hydrocarbons. However, other studies have reported that biodiesel combustion reduces particle size, changes particle-associated metal composition and increases or decreases concentrations of certain air toxics. Toxicological studies are limited.

Biodiesel: comparing emissions versus exposures

- The real-world variability in engines and driving cycles, fate and transport processes, and other factors make it difficult to predict exposure to biodiesel based solely on emissions data sets. Exposure assessment techniques can provide important data connecting sources of emissions to health effects.

Biodiesel’s impact on exposure in an occupational setting

- While there have been few biodiesel exposure studies performed, recent research supports that biodiesel use in heavy-duty equipment reduces occupational exposure to fine particulate matter.

Future perspective

- As biodiesel fuel feedstocks, blend percentages and engine after treatment technologies rapidly change, both emissions and exposure research are needed to develop a fuller understanding of biodiesel’s impact on air quality and human health.