EXPOSURE TO DIESEL EXHAUST EMISSIONS: IRRITANTS

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Introduction

Engine exhaust emissions are amongst the most complex and most extensively studied air pollutants. They are the most problematic air contaminants in diesel-powered submarines. The two major sources being fugitive engine emissions and re-entrainment of the engine exhaust through the snorkel, commonly referred to by submariners as "getting your own back". Other military platforms such as tanks, armoured personnel carriers, transport vehicles, fixed wing aircraft and helicopters also operate under conditions where personnel may be routinely and directly exposed to engine exhaust emissions.

Over the years various components of diesel exhaust have been singled out for their toxicity. Carbon monoxide was initially identified as the most toxic component and first subject to monitoring on a RN submarine in 1947 (Ellis) with the exclusion of the other exhaust components of lesser known toxicity at the time. With the growing use in commercial and private road vehicles in the post-war period, other exhaust components were targeted for their toxicity.

In 1974 the US Environment Protection Agency (EPA) introduced emission standards for heavy-duty diesel engines which included carbon monoxide and nitrogen oxides in combination with hydrocarbons under the Clean Air Act of 1970. Subsequently, in 1985, separate emission limits were introduced for nitrogen oxides and hydrocarbons and in 1988 diesel particulate emission limits were added (EPA, 2002).

During the same period, diesel exhaust emissions of aldehydes were reported causing acute lachrymatory health effects at low concentration such as 1 -3 ppm (Cernansky 1983, Nightingale et al. 2000). In `1991 the Californian Low Emission Vehicle (LEV) standards, were introduced through the Clean Air Act Amendments (CAAA) of 1990, which addressed emissions of formaldehyde in addition to the above emissions for both petrol and diesel-powered vehicles (CARB, 2016). Although the LEV controls on formaldehyde exhaust emissions for vehicles have been maintained (CARB, 2016), aldehyde exhaust emissions have been consistently omitted from the European Union (EU) vehicle exhaust emission regulations (DieselNet 2019a).

The aim of this paper is to review aldehyde emissions from internal combustion engines and their potential acute health effects within a military occupational environment and in the context of commonly regulated emissions such as particulate matter (PM), carbon monoxide (CO), nitrogen oxides (NO_x) and hydrocarbon emissions and to suggest a strategy for monitoring aldehydes.

Current Combustion Engine Exhaust Emission Controls

US Federal and US EPA emission standards for passenger cars and light duty trucks, Tier 1 standards were introduced from 1991 to 1994 and covered PM (diesel engines only), CO, NO_x, total hydrocarbons (THC) and non-methane hydrocarbons (NMHC) (DieselNet 2019b). These were replaced by Tier 2 regulations over the period 2004 to 2009 and later by Tier 3, standards which were signed in law in 2014. Tier 3 regulations are closely aligned with Californian LEV III standards and include emission controls on formaldehyde (DieselNet 2019c).

EU diesel and petrol engine emission standards for passenger cars, Euro 6, apply to CO, HCs, NO_x and PM but do not include aldehydes (DieselNet 2019c). Similarly, for locomotive engines, EU Stage V, which bear the closest resemblance to submarine engines.

Diesel Exhaust Emissions Monitoring for Occupational Exposure

Many of the new regulations for occupational exposure to diesel exhaust have been prompted by exposures in underground mining where diesel-powered equipment is operated in confined spaces. Here the recent focus has been largely on diesel particulate matter (DPM) in addition to the long-standing hazards of CO_2 , CO, NO_x and SO_2 as well as the recognition of hydrocarbon and aldehyde emissions (DMP 2013, MSHA 2016, Maximilien et al. 2017). In addition to US Federal regulations, some US state authorities have also regulated CO, NO_x and DPM exposures from diesel engine exhaust in underground coal mines (MSHA 2016) following exposure surveys of miners (Stewart et al. 2010, Coble et al. 2010). Likewise, vehicular exhaust exposure monitoring has concentrated on PM, CO and NO_x (Maximilien et al. 2016).

The UK, Health and safety Executive (HSE 2012) has recognised the hazardous nature of formaldehyde in engine exhaust emissions and has recommended avoidance of exposure to engine exhaust where possible, or at least, has advocated the control of such exposures. To this end carbon dioxide has been proposed as a conveniently monitored surrogate for diesel exhaust exposure and risk assessment in the work place in addition to visual assessment of smoke and soot together with a subjective assessment of irritancy.

The need to monitor aldehydes may have been reduced in priority over time, since it has been claimed that significant progress has been made in reducing the exhaust emissions of aldehydes (formaldehyde and acetaldehyde). This has been supported by comparing emission data from heavy diesel engines using 2004 and 2007 technologies (Liu et al. 2010, Khalek et al. 2011). A study published in 2001 (Lloyd and Cackette) showed that low volatile carbonyls had, at the time, constituted the largest fraction of gas-phase organic compounds emitted from a medium-duty diesel truck engine with acetaldehyde and formaldehyde predominating.

Aldehyde Emissions from Engine Exhausts

While aldehyde emissions from engine exhausts may have declined with advanced engine management technology in modern, well-maintained engines, because of the nature of the combustion process, the operating conditions of the engines and engine wear, aldehyde emissions can still be problematic. For example, controlled human exposure chamber studies of diluted exhaust emissions from a Volkswagen Passat diesel car engine, 81 kW in idle mode, measured formaldehyde at 0.40 mg m⁻³ (0.33 ppm) and acetaldehyde at 0.20 mg.m⁻³ (0.1 ppm) (Xu et al. 2013, Wierbicka et al. 2014). By comparison the NO_x concentration was 1.3 ppm (8h avg.) and DPM (< 1 μ m dia.) averaged ~300 μ g.m⁻³. The authors concluded that throat and eye irritations were primarily due to aldehydes, consistent with previous findings (Ceransky 1983, Rudell et al. 1996).

Formaldehyde has been previously shown to produce these symptoms at concentrations in the range $0.16 - 0.54 \text{ m gm}^{-3}$ (0.13 – 0.44 ppm) both from exhaust exposure studies (Rudell et al. 1994; Wilhelmsson et al. 1992); and as formaldehyde alone, where the minimal eye irritation was reported at 0.4 - 0.6 mg m⁻³ (0.3 - 0.5 ppm) for daily 4 h exposures (Lang 2008). In addition to formaldehyde and acetaldehyde, acrolein is also known to be present in diesel engine exhaust (Ceransky 1983) and has been reported to cause eye irritation at a concentration of 0.2 mg m⁻³ (0.1 ppm) (Dwivedi et al. 2015).

Sawant et al. (2008) carried out controlled human exposure trials where the concentration of diesel exhaust was adjusted to 100 μ g.m⁻³ DPM, using an International 444E, 7.27 L turbo-charged V-8 diesel engine. At this dilution formaldehyde, acetaldehyde and acrolein were measured at 0.053 – 0.074 mg.m⁻³ (0.043 – 0.060 ppm), 0.020 – 0.028 mg.m⁻³ (0.011 – 0.016 ppm) and 0.0009 – 0.0043 mg.m⁻³ (0.0004 – 0.002 ppm) respectively. After exposure for 2 h, no adverse effects were observed in terms of loss of lung function for the 11 subjects including 7 asthmatics however, symptoms of nasal and eye irritation was not investigated.

In terms of exposure limits, the American Conference of Occupational Hygienists (ACGIH 2017) has established a Short-Term Exposure Limit (STEL) of 0.36 mg.m⁻³ (0.3 ppm) and an 8 h Threshold Limit Value – Time-Weighted Average (TLV-TWA) of 0.12 mg.m⁻³ (0.1 ppm) for formaldehyde which are consistent with earlier sensory response findings (*vide supra*). Currently the ACGIH maximum occupational exposure limit (TLV-ceiling) for acetaldehyde is 45 mg.m⁻³ (25 ppm) and for acrolein it is 0.23 mg.m⁻³ (0.1 ppm). Hence on the basis of the exhaust exposure concentrations reported above, formaldehyde and acrolein are most likely to be major gaseous irritants in engine exhaust emissions.

In addition to the acute sensory effects, the three aldehydes also exhibit chronic health effects. Formaldehyde was initially classified as a probable carcinogen by the US EPA (1987) under conditions of prolonged exposure or high concentrations. In 2004, the International Agency for Cancer (IARC) classified formaldehyde as a carcinogen to humans (IARC 2004). The National Institute for Occupational Safety and Health (NIOSH) has recommended that acetaldehyde be considered as a potential carcinogen while the evidence for the carcinogenicity of acrolein is incomplete (NIOSH 2020) although it has been reported that acrolein causes DNA damage and inhibits DNA repair (Wang et al. 2012).

As for the other gaseous emissions from engine exhaust, human sensitivity to nitrogen dioxide is considerably greater that of carbon monoxide and comparable to formaldehyde and acrolein, for example, the CO TLV-TWA (ACGIH 2019) is 25 ppm (29 mg m⁻³) while the NO₂ TLV-TWA (ACGIH 2019) is 0.4 mg m⁻³ (0.2 ppm). Carbon monoxide is odourless but humans can detect the odour of NO₂ at low concentrations. Earlier studies found that at a NO₂ concentration of 0.23 mg. m⁻³ (0.12 ppm), 3 of 9 subjects perceived the odour immediately, and 8 of 13 detected a threshold concentration of 0.41 mg.m⁻³ (0.22 ppm) (Henschler et al. 1960). At a higher concentration, 0.79 mg. m⁻³ (0.42 ppm), 8 of 8 subjects recognized the odour (Henschler et al. 1960). Feldman (1974) reported that 26 of 28 subjects perceived NO₂ odour at concentrations of 0.2 mg.m⁻³ (0.11 ppm). At slightly higher concentrations, 0.9 to 8 mg.m⁻³ (0.5 to 4 ppm), a study by Kerr et al (1978, 1979) found that some asthmatics experienced slight burning of the eyes sensation, slight headache and tightness of the chest after exposure for 2 h. Thus the no-adverse-effect level (NOEL) for short-term exposure is considered to be 0.94 mg.m⁻³ (0.5 ppm) (NRC 2012) while the occupational TLV-TWA has been set at 0.37 mg.m⁻³ (0.2 ppm) (ACGIH 2019).

Although a plethora of hydrocarbons have been reported in diesel exhaust emissions, the health effects of these compounds are not immediate, at the concentrations found in the exhaust emissions, but are mostly chronic particularly in the case of aromatic hydrocarbons and polycyclic aromatic hydrocarbons (PAHs), although the more volatile hydrocarbons may contribute to the exhaust odour (Khalek et al. 2011).

Conditions for the Generation of Aldehyde Emissions.

Studies of carbonyl exhaust emissions from diesel engines published in 1962 (Linell and Scott) indicate that peak concentrations of formaldehyde and acrolein are generated at high engine loads and high engine speeds and also at high loads and low engine speeds (Figs. 1a, 1b). To a lesser extent,

idling speeds, with no load, are also likely to generate elevated formaldehyde and acrolein concentrations compared to average operating conditions. Understandably, engine combustion appears to be optimised for mid-range engine speeds and loads representing typical operating conditions.

Carbonyl emissions from a more modern engine (Pang et al. 2006) also show elevated carbonyl concentrations at both low and high engine loads but the effects are less pronounced compared with the older engine (Fig.2). Thus there is considerable variability in the data, for example Figure 2b shows an anomalous behaviour in formaldehyde concentrations compared with the other carbonyl compounds. In this case, formaldehyde concentrations decline at engine speeds >1600 rpm whereas the other carbonyl concentrations have increased in concentration. Variations in exhaust emissions, including formaldehyde, have been observed from engine to engine with respect to engine load and speed (Chin et al. 2012) making it difficult to predict conditions for maximum aldehyde emissions with certainty.

In terms of occupational exposure, engine idling conditions are more likely to be problematic when the vehicle is stationary and there is little exhaust dilution from airflow compared with high engine loads when the vehicle is in motion. In the case of submarine and generator engines however, the diesel engines are operated at constant speed and constant load. Here, engine starts are most likely to generate emissions due to partially combusted fuel before stable operating conditions are achieved. The submarine situation is unique in that engines cause two problems, fugitive emissions into the engine room and exhaust entrainment through the snorkel into the submarine ventilation system.



Figure 1a. Formaldehyde emissions from diesel exhaust dependence on engine speed and engine load (Linell and Scott 1962), 7 L, 6 cylinder engine.



Figure 1b. Acrolein emissions from diesel exhaust dependence on engine speed and engine load (Linell and Scott 1962), 7 L, 6 cylinder engine.



<u>Figure 2a.</u> Diesel exhaust carbonyl emissions at various loads and constant engine speed of 1800 rpm for a Commins-4B diesel engine 4 cyl, 3.9 L, 105 - 140 hp (Pang et al 2006).



<u>Figure 2b.</u> Diesel exhaust carbonyl emissions at a constant full load and varying engine speeds for a Commins-4B diesel engine 4 cyl, 3.9 L, 105 - 140 hp (Pang et al 2006).

In addition, some studies have claimed that gasoline bio-fuels tend to produce higher acetaldehyde emissions than hydrocarbon based fuels but only when they contain ethanol whereas formaldehyde emissions tend to be lower for bio-diesel, which do not contain ethanol (Pang et al. 2006, Chin et al. 2012). It has been widely accepted that ethanol contributes to acetaldehyde formation while aliphatic hydrocarbons contribute to formaldehyde formation. (Pang et al. 2006, Chin et al. 2012). Zarante et al. (2010) found no difference in formaldehyde and acetaldehyde emissions between hydrocarbon and bio-diesel containing up to 35 percent castor oil fatty acid methyl esters[#] in hydrocarbon diesel fuel while Liu et al. (2009) found 3-6 fold increases in formaldehyde and acrolein with increasing palm oil fatty acid methyl ester content from 0 - 100 percent. However, there was no consistent increase in acetaldehyde emissions with increasing methyl ester content.

Monitoring and analysis of aldehyde emissions from engine exhausts.

Due to the reactive nature of low molecular weight aldehydes (eg. formaldehyde, acetaldehyde and acrolein), traditional gas sampling techniques such as sorbent tubes, polyester (Tedlar[®]) bags and stainless steel passivated (SUMA) canisters are not ideally suited. The gold standard for this appears to be derivatisation with 2,4-dinitrophenyl hydrazine (DNPH) in the form of a solution (eg. 20 ml) added to a Tedlar bag containing a sample of exhaust gas (eg.10 l) (Roy 2007) or on a substrate in pre-prepared cartridges eg. Waters DNPH coated cartridges, Sep-Pak[®] DNPH-Silica (WAT 037500) (Benvenuti 2007, CPCB 2010). The DNPH forms a stable, non-volatile hydrazone with the aldehydes, in a quantitative reaction, which can later be extracted with acetonitrile and analysed by high performance liquid chromatography (HPLC) using an ultraviolet detector (CPCB 2010).

[#] Vegetable oil based bio-fuels are derive from the transesterification of the vegetable oil producing a more volatile product in the form of fatty acid methyl (or ethyl) esters (Keera et al. 2011).

Strategy for monitoring aldehyde emissions.

Many of the significant engine exhaust emissions can be monitored in real-time, for example DPM, hydrocarbons, CO_2 , CO and NO_x . Aldehydes, such as formaldehyde, acetaldehyde and acrolein require a more complex sampling and retrospective analytical procedure. Hence, if NO_x concentrations appear to be below the NOEL and exposed personnel are reporting nasal and eye irritations, the presence of the above 3 aldehydes should be investigated, particularly if old engines are involved and these effects are experienced at idling speeds or under high load conditions.

Conclusions

The first indication of occupational exposure to aldehydes from engine exhaust emissions are manifested as irritation to the eyes. It is most likely to occur with an old engine at idling speed and higher speeds under heavy load conditions. Under these circumstances it may be prudent to monitor formaldehyde, acetaldehyde and acrolein in addition to the usual emissions such as hydrocarbons, CO, NO_x and DPM. Addressing such acute effects is of paramount importance as these symptoms may impact on the performance and safety of personnel.

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