Atmospheric Environment 86 (2014) 212–219

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Detailed diesel exhaust characteristics including particle surface area and lung deposited dose for better understanding of health effects in human chamber exposure studies

Aneta Wierzbicka [a,*], Patrik T. Nilsson [a], Jenny Rissler [a], Gerd Sallsten [b], Yiyi Xu [c], Joakim H. Pagels [a], Maria Albin [c], Kai Österberg [c,d], Bo Strandberg [b], Axel Eriksson [e], Mats Bohgard [a], Kerstin Bergemalm-Rynell [b], Anders Gudmundsson [a]

[a] Ergonomics and Aerosol Technology, Lund University, Box 118, SE-221 00 Lund, Sweden
[b] Department of Occupational and Environmental Medicine, Sahlgrenska University Hospital and Academy, University of Gothenburg, Box 414, SE-405 30 Gothenburg, Sweden
[c] Division of Occupational and Environmental Medicine, Lund University, SE 221-85 Lund, Sweden
[d] Department of Psychology, Lund University, Box 213, 221 00 Lund, Sweden
[e] Division of Nuclear Physics, Lund University, Box 118, SE-221 00 Lund, Sweden

HIGHLIGHTS

- DE properties vary significantly under the same DEP mass concentration exposure.
- Reporting detail DE characteristics is needed to explain observed health effects.
- DEP agglomerated structure has to be accounted for to not underestimate lung dose by surface.
- Use of size dependent effective density prevents overestimation of lung mass dose.
- Gas phase components known for carcinogenic and irritation effect should be reported.

ARTICLE INFO

Article history:
Received 15 July 2013
Received in revised form
23 October 2013
Accepted 11 November 2013
Available online 4 December 2013

Keywords:
Diesel exhaust characteristics
Human laboratory exposure
Particle surface area
Lung deposited dose

ABSTRACT

Several diesel exhaust (DE) characteristics, comprising both particle and gas phase, recognized as important when linking with health effects, are not reported in human chamber exposure studies. In order to understand effects of DE on humans there is a need for better characterization of DE when performing exposure studies. The aim of this study was to determine and quantify detailed DE characteristics during human chamber exposure. Additionally to compare to reported DE properties in conducted human exposures. A wide battery of particle and gas phase measurement techniques have been used to provide detailed DE characteristics including the DE particles (DEP) surface area, fraction and dose deposited in the lungs, chemical composition of both particle and gas phase such as NO, NO_2, CO, CO_2, volatile organic compounds (including aldehydes, benzene, toluene) and polycyclic aromatic hydrocarbons (PAHs). Eyes, nose and throat irritation effects were determined. Exposure conditions with PM_1 (<1 μm) mass concentration 280 μg m^{-3}, number concentration 4×10^5 cm^{-3} and elemental to total carbon fraction of 82% were generated from a diesel vehicle at idling. When estimating the lung deposited dose it was found that using the size dependent effective density (in contrast to assuming unity density) reduced the estimated respiratory dose by 132% by mass. Accounting for agglomerated structure of DEP prevented underestimation of lung deposited dose by surface area by 37% in comparison to assuming spherical particles. Comparison of DE characteristics reported in conducted chamber exposures showed that DE properties vary to a great extent under the same DEP mass concentration and engine load. This highlights the need for detailed and standardized approach for measuring and reporting of DE properties. Eyes irritation effects, most probably caused by aldehydes in the gas phase, as well as nose irritation were observed at exposure levels below current occupational exposure limit values

* Corresponding author. Tel.: +46 704946658; fax: +46 462224431.
E-mail address: aneta.wierzbicka@design.lth.se (A. Wierzbicka).

1352-2310
http://dx.doi.org/10.1016/j.atmosenv.2013.11.025

given for exhaust fumes. Reporting detailed DE characteristics that include DEP properties (such as mass and number concentration, size resolved information, surface area, chemical composition, lung deposited dose by number, mass and surface) and detailed gas phase including components known for their carcinogenic and irritation effect (e.g. aldehydes, benzene, PAHs) can help in determination of key parameters responsible for observed health effects and comparison of chamber exposure studies.

1. Introduction

Elevated exposures to diesel exhaust (DE) have been linked with variety of health outcomes including irritant effects, respiratory symptoms, immunologic, lung inflammatory, cardiovascular effects and lung cancer (Ris, 2007). Recently, DE was classified as a Group 1 carcinogen by the International Agency for Research on Cancer (Benbrahim-Tallaa et al., 2012). Levels of occupational exposure to DE are usually higher than the general population.

DE comprises a complex mixture of particle and gaseous components which vary depending on engine type, fuel characteristics, operating load and presence of emission abatement techniques. During chamber exposure studies DE characteristics can be influenced by generation and dilution systems used.

The DE particles (DEP) are almost exclusively found in the submicrometer fraction (<1 μm). Two size modes can typically be distinguished, an accumulation mode and a nucleation mode (Burtscher, 2005). The volatile material in the hot exhaust can during dilution and cooling condense on the solid soot particles (accumulation mode) and/or through nucleation form new particles (nucleation mode). In general the chemical composition of DEP comprise elemental carbon, organic carbon, sulphur compounds and trace metals from lubricating oil and engine wear. Organic substances identified as adsorbed onto particles include C14–35 hydrocarbons (Ris, 2007). The oxygen to carbon (O:C) ratio of the particulate organic fraction of fresh DEP is typically low <0.1 (Aiken et al., 2008).

In majority of human chamber exposure studies, conducted by research groups in Umeå University (Sweden), University of Edinburgh (UK), University of Washington (USA), Los Amigos Research and Educational Institute (USA), US EPA and University of North Carolina (USA), University of British Colombia (Canada) and Rutgers the State University of New Jersey (USA) (summarized in a review by Hesterberg et al., 2010 and in Appendix A), mass concentration of DEP ranged between 60 and 300 $\mu g\ m^{-3}$ and was frequently treated as the major exposure metrics. Apart from the particle mass concentration, the following characteristics have been indicated as important when linking with health effects: particle size, number concentration, surface area, chemical composition, solubility, volatile and non-volatile mass fraction, organic compounds including PAHs, soot core and metals (Giechaskiel et al., 2009; Oberdorster et al., 2005). Even if consensus on the most appropriate metrics has not been reached, correlation of particle surface area to inflammatory response has been recognized (Donaldson et al., 2001; Oberdorster et al., 2005). However particle surface area is not reported in published chamber exposure studies with one exception (Rissler et al., 2012). Respiratory tract deposition is an important link between exposure to DE and observed health effects but lung deposited dose is not reported in the exposure studies with one exception for one type of engine used in Umeå University studies (Rissler et al., 2012). In general DEP properties are not well characterized in chamber exposure studies.

DE consist of a mixture of gaseous components and among them, the aldehydes (formaldehyde, acetaldehyde and acrolein), benzene, 1.3 butadiene, PAHs and nitro-PAHs are important due to their potential carcinogenic effects (Benbrahim-Tallaa et al., 2012). Aldehydes, alkanes, alkenes, oxides of sulphur and nitrogen, are known to induce respiratory tract irritation given sufficient exposure. Gas phase characteristics specifically those having potential carcinogenic or irritation effects, as listed above, are not well characterized in the human exposure studies.

There is a need to pinpoint species-specific or combination of few DE characteristics that would explain observed health effects. To accomplish it detail DE characteristics should be reported as so far such trends are not easy to distinguish.

The aim of this study is to provide detailed knowledge on DE characteristics, indicated as important for health effects assessment. Additionally to compare to DE properties in conducted human exposures as well as highlight the range of DE characteristics that can be determined with known methods and instrumentation. In contrast to most of the human exposure studies we determined DEP surface area, size dependent effective density, fraction and dose of DEP deposited in the lungs as well as chemical composition of both gas and particle phase. These are the DE properties, which were indicated as important to quantify in order to understand the effects of DE on humans. An additional objective was to determine eyes, nose and throat irritation effects assessed via self-rating questionnaires and medical assessment.

This work is a part of a larger DINO project "Health effects of combined exposure to diesel and noise", that aimed to determine influence of combined exposure to DEP and traffic noise on human health. Health effects of DE exposure in DINO study were reported by Xu et al. (2013) and concluded that short-term exposure to DE at ~300 $\mu g\ m^{-3}$ caused temporary decline in peak expiratory flow in healthy subjects and the increase in leukocyte cell counts in peripheral blood indicated a systemic inflammatory responses.

2. Materials and methods

2.1. General methodology

In a laboratory chamber eighteen healthy volunteers (9 men and 9 women), all non-smokers of ages 40–66 (mean 51 years) were exposed twice to DE with high DEP concentration (~300 $\mu g\ m^{-3}$) and twice to filtered air (FA) with low particle concentration (~2 $\mu g\ m^{-3}$). The data presented here were extracted from the DINO project with four exposure conditions: 1) Reference exposure: FA and low traffic noise (46 dB(A)), 2) Diesel exposure: DE and low traffic noise, 3) Noise exposure: FA and high traffic noise (75 dB(A)), 4) Diesel and noise exposure: DE and high traffic noise.

In this study exposures were merged into two groups: 1) DE exposure that comprised of diesel and diesel and noise exposures and 2) FA exposure comprised of reference and noise exposures.

Test subjects (three at the one exposure, relaxed sitting) were exposed for three hours to each exposure scenario with at least one week interval between each scenario. The study was approved by the Regional Ethical Review Board.

The methodology used in DINO study is described in Wierzbicka et al. (2011).