Journal of Aerosol Science 69 (2014) 32–47

Contents lists available at ScienceDirect

Journal of Aerosol Science

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

Diesel particle composition after exhaust after-treatment of an off-road diesel engine and modeling of deposition into the human lung

Kati Oravisjärvi [a,*], Mari Pietikäinen [a], Juhani Ruuskanen [b], Seppo Niemi [c,d], Mika Laurén [d], Arto Voutilainen [e,f], Riitta L Keiski [a], Arja Rautio [g]

[a] Department of Process and Environmental Engineering, University of Oulu, P.O. Box 4300, FI-90014 Oulu, Finland
[b] Department of Environmental Science, University of Eastern Finland, P.O. Box 1627, FI-70211 Kuopio, Finland
[c] Department of Electrical Engineering and Energy Technology, University of Vaasa, P.O. Box 700, FI-65101 Vaasa, Finland
[d] Turku University of Applied Sciences, Joukahaisenkatu 3A, FI-20520 Turku, Finland
[e] Department of Applied Physics, University of Eastern Finland, P.O. Box 1627, FI-70211 Kuopio, Finland
[f] Rocsole Ltd., FI-70211 Kuopio, Finland
[g] Centre for Arctic Medicine, Thule Institute, University of Oulu, P.O. Box 7300, FI-90014 Oulu, Finland

ARTICLE INFO

Article history:
Received 17 July 2013
Received in revised form
20 November 2013
Accepted 29 November 2013
Available online 16 December 2013

Keywords:
Nanoparticles
PAH
DPF
SCR
Human lung deposition model
Diesel particles

ABSTRACT

Regional deposition of diesel particles in the human lungs was analyzed and the chemical composition of inhaled particles was investigated. The off-road diesel engine with a diesel particulate filter (DPF) or a selective catalytic reduction (SCR) unit and without any exhaust after-treatment system was used. Around 85–95% of the measured particles were of ultrafine size and 53–84% of those nanoparticles. Over 70% of the deposited particles under 0.1 μm and about 45–70% of the deposited particles from 0.1 to 1 μm reach also the alveolar–interstitial level. Elements analyzed in particles were C, O, Fe, Si, Ti, Na, K, Ca, Mg, Ba, Mn, Zn, Cu, Cl, P, S and N. The proportion of PAHs in the measured particle mass was 0.05% and carcinogenic ones represented 1.3% of the total PAHs. The DPF system removed particles efficiently and up to 99% of the particles were removed. The total number of particles deposited in the lungs was generally lower when DPF was used compared to other setups. These particles contained though the largest variety of elements, which are commonly considered harmful to humans. Therefore it is difficult to conclude, whether exhaust particles from a diesel engine with a DPF unit would be less harmful to human health.

1. Introduction

Air pollution is a major health risk causing cancer and several other health impacts (IARC, 2013a). The particles emitted from off-road engines may be harmful for workers. Epidemiological evidence suggests that a long-term employment in jobs with a substantial diesel exhaust exposure is associated with a 20–50% increase in the risk of lung cancer (Zelikoff, 2000) and there is a need for occupational exposure regulations (Maricq, 2007). In June 2012, the International Agency for Research on Cancer (IARC) classified diesel engine exhaust as carcinogenic to humans (group 1) (IARC, 2013a, 2013b).

* Corresponding author. Tel.: +358 50 543 7323; fax: +358 2 9448 2304.
E-mail address: kati.oravisjarvi@oulu.fi (K. Oravisjärvi).

0021-8502/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.jaerosci.2013.11.008

Especially, transition metals and polycyclic aromatic hydrocarbons (PAHs) contribute to the oxidative capacity of particulate matter (PM) and thereby can cause oxidative stress in biological systems (de Kok et al., 2006; Donaldson et al., 2003; Geiser & Kreyling, 2010). Due to the large surface area of ultrafine particles and the chemical reactivity of particle surfaces, particle emissions may include a wide range of different compounds which can be toxic, mutagenic or carcinogenic (Cohen & Nikula, 1999; Geiser & Kreyling, 2010; Penn et al., 2005). Due to the harmful health effects of particles both particle mass and number should be reduced (Kreyling et al., 2006; Osunsanya et al., 2001; Pope & Dockery, 2006). To understand the role of emission reduction technology and diesel particles in human health, it is important to evaluate the actual doses of diesel particles deposited in the human respiratory tract (Oravisjärvi et al., 2011; Pietikäinen et al., 2009; Rissler et al., 2012).

In diesel engineering technology, the simultaneous reduction of PM and nitrogen oxides (NO_x) is a big challenge, because the strategies for reducing one component may lead to an increase in another. To reduce both PM and NO_x two alternative solutions are required (López et al., 2009; Mooney, 2007). For current and future emission legislation, a variety of exhaust after-treatment devices is essential (McDonald et al., 2004a; Mooney, 2007). For NO_x reduction, urea-based selective catalytic reduction (SCR) is commonly used in on- and off-road engines (López et al., 2009; Maricq, 2007). However, particulate filters (DPF) and diesel oxidation catalysts (DOC) have also become more standard in off-road engines and are already common in motor vehicles (Mooney, 2007). The DPF significantly lowers the PM mass emissions, but its effect on particle number is two-sided. The mass is dominated by the soot accumulation mode, which is efficiently trapped in DPF, but the particulate number can be dominated by nuclei mode particles formed downstream of the DPF (Maricq, 2007), although the DPF seems to be able to remove also ultrafine particles and nanoparticles very effectively from the engine exhaust (Niemi et al., 2009). After-treatment of exhaust gas does not just lower emissions, but it also alters the chemical composition of vehicle exhaust (Maricq, 2007).

The off-road engines contribute about 50% of the total particle emissions from combustion engines. As the lifetime of off-road engines is longer than that of on-road engines, a reduction in the emissions takes more time to become effective. The relative contribution of these engines to the total particle emissions will therefore increase in the future (Burtscher, 2005). The new emissions standard (stage IIIB) for off-road engines introduced the particle limit of 0.025 g/kWh in the beginning of 2011. This is about 90% of the emission reduction relative to the previous standard (stage IIA) (DieselNet, 2013). To achieve this new limit value, it is anticipated that engines will have to be equipped with particulate filters (Mooney, 2007), even if some manufacturers have managed to reduce PM mass sufficiently by injection improvements and combustion chamber optimization.

The aim of this study was to estimate the regional deposition of diesel particles in the human respiratory tract and to investigate the chemical composition of those particles. Deposition of particles was simulated by utilizing the computerized lung deposition model ICRP 66 with an in-house script. The used engine was an off-road diesel engine, which was equipped either with a diesel particulate filter (DPF) or a selective catalytic reduction (SCR) unit and for comparison an engine lacking the after-treatment system was evaluated. Diesel exhaust particles were measured using an Electrical Low Pressure Impactor (ELPI). Size distribution and chemical composition of particles were analyzed with special focus on transition metals and PAH compositions.

2. Experimental

2.1. Experimental setup

This study investigated particle size distributions emitted from an off-road diesel engine. The tested engine was a common-rail (CR) off-road diesel engine with six cylinders, manufactured by Agco Sisu Power Inc. The technology of the used engine was at the EU IIIA level and the rated power was 150 kW/2200 rpm. The diesel engine was originally equipped either with SCR with an injection of an aqueous urea solution, 32.5% (AdBlue, DIN 70070/AUS32) or with an alternative catalyst system having an oxidation catalyst and a diesel particulate filter (DPF) (Johnsson & Matthey/CRT). The test runs were also driven without any after-treatment exhaust system. The test runs were carried out at 75% load at an engine speed of 1500 rpm: this was the peak torque speed.

The used fuel was summer grade diesel fuel oil (Neste Oil Corporation), with ultra-low sulfur content of about 13 mg/kg (13 ppm). The diesel fuel contained 12.7 mg/kg S, 0.01 mg/kg Ca, 0.05 mg/kg Mg, 0.03 mg/kg P and the lubrication oil 4711 mg/kg S, 2491 mg/kg Ca, 3.1 mg/kg Na, 1.5 mg/kg K, 6 mg/kg Mg, 983 mg/kg P. All the test runs were performed in the Internal Combustion Engine Laboratory at Turku University of Applied Sciences, Finland.

2.2. Particle measurements

Particle size distributions were measured using the ELPI measurement system (Dekati Ltd., Finland). The ELPI measurement system yields particulate number concentrations in 12 non-overlapping size bins covering the entire measurement size range from 30 nm to 10 μm (Keskinen et al., 1992; Marjamäki et al., 2000). In order to extend the measurement range to start from 7 nm an extra filter stage was set to ELPI. The size-ranges are represented by geometric means of 0.021, 0.041, 0.077, 0.128, 0.199, 0.306, 0.485, 0.775, 1.23, 1.94, 3.04 and 6.19 μm. The ELPI was also used to collect the particles for a later analysis by their structure and chemical composition. The particle mass was sampled into 12 impaction foils from each measured particle size-range.