Journal of Aerosol Science 48 (2012) 18–33

Contents lists available at SciVerse ScienceDirect

Journal of Aerosol Science

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

Experimental determination of deposition of diesel exhaust particles in the human respiratory tract

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ARTICLE INFO

Article history:
Received 19 October 2011
Received in revised form
16 January 2012
Accepted 17 January 2012
Available online 26 January 2012

Keywords:
Diesel exhaust
Particle
Agglomerates
Lung deposition
Dose
Surface area

ABSTRACT

Diesel emissions are a major contributor to combustion-generated airborne ambient particles. To understand the role of diesel particulate emissions on health effects, it is important to predict the actual particulate dose deposited in the human respiratory tract, with respect to number, surface area and mass. This is complicated by the agglomerate nature of some of these particles. In this study the respiratory tract deposition fraction in the size range 10–500 nm, was determined for 10 healthy volunteers during both idling and transient engine running conditions of a heavy duty diesel engine. The aerosol was characterized with respect to both chemical and physical properties including size resolved particle effective density. The dominating part of the emitted particles had an agglomerate structure. For those formed during transient running conditions, the relationship between particle mass and mobility diameter could be described by a power law function. This was not the case during idling, most likely because of volatile compounds condensing on the agglomerates. The respiratory tract particle deposition revealed large intra-subject variability with some subjects receiving a dose that was twice as high as that of others, when exposed to the same particle concentration. Associations were found between total deposited fractions (TDF), and breathing pattern. There was a difference between the idling and transient cycle with TDF being higher with respect to number during idling. The measured size-dependent deposition fraction of the agglomerated exhaust particles from both running conditions was nearly identical and closely resembled that of spherical hydrophobic particles, if plotted as a function of mobility diameter. Thus, for the size range covered, the mobility diameter could well describe the diameter-dependent particle respiratory tract deposition probability, regardless of the agglomeration state of the particles. Whilst mobility diameter well describes the deposition fraction, more information about particle characteristics is needed to convert this to volume equivalent diameter or estimate dose with respect to surface area or mass. A methodology is presented and applied to calculate deposited dose by surface area and mass of agglomerated particles. The methodology may be useful in similar studies estimating dose to the lung, deposition onto cell cultures and in animal studies.

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doi:10.1016/j.jaerosci.2012.01.005

Nomenclature

a_k,b_k, c_k, d_k	Fitted parameters for describing measured deposition fraction (Eq. (7))
APM	aerosol particle mass analyzer
B	mechanical mobility
C_c	Cunningham correction term
C_{ex}	exhaled particle concentration
C_{in}	inhaled particle concentration
CPC	condensation particle counter
D	particle diffusivity
d	particle diameter
DEP	diesel exhaust particles
$DF(d)$	size-resolved deposited fraction in the respiratory tract
DF_{equip}	instrumental particle losses in RESPI
DMA	differential mobility analyzer
d_{me}	particle electrical mobility equivalent diameter
d_{pp}	primary particle diameter
EC	elemental carbon
f	breathing frequency
FVC	forced vital capacity
GMD	geometric mean diameter
k	Boltzmann constant
K	parameter fitted describing the mass–mobility relation of the particles (Eq. (3))
m	particle mass
m_{agg}	mass of individual agglomerates
MV	minute volume
N	particle number
N_{agg}	number of agglomerates
N_{pp}	number of primary particles
OC	organic carbon
PAH	polycyclic aromatic hydrocarbon
Q	inhaled volume flow
s.d.	standard deviation
SA	surface area
SA_{agg}	surface area of individual agglomerates
SA_{sph}	surface area of spherical particles
SMPS	scanning mobility particle sizer
t	exposure time
T	temperature
TC	total carbon
TD	thermo-denuder
TDF_m	total deposited fraction in the respiratory tract with respect to particle mass
TDF_N	total deposited fraction in the respiratory tract with respect to particle number
TDF_{SA}	total deposited fraction in the respiratory tract with respect to particle surface area
TEM	transmission electron microscopy
TEOM	tapered element oscillating microbalance instrument
V_D	instrumental dead space
V_T	tidal volume
η	kinematic viscosity of air
ρ_{eff}	effective density
ε_m	mass–mobility exponent
ρ_{pp}	density of the primary particles
σ_g	geometric standard deviation

1. Introduction

Air pollution has been associated with a variety of adverse health effects (Pope & Dockery, 2006) and is believed to be one of the major causes of premature deaths worldwide (Lopez et al., 2006). Particles emitted from combustion processes, whereof diesel engines are a major contributor, are among the most common emissions in populated areas. Epidemiological and toxicological studies have shown associations between diesel exhaust and adverse health effects (e.g. Hart et al., 2009; Mills et al., 2007; Sydbom et al., 2001). Experimental exposure studies of humans have revealed that exposure to dilute diesel exhaust induces a pronounced neutrophilic airway inflammation along with negative cardiovascular effects in terms of reduced vasomotor function and impaired endogenous fibrinolysis (Salvi et al., 1999; Mills et al. 2005; Mills et al., 2007; Barath et al., 2010). Whilst recent research efforts have provided a basis for better understanding of exposure, deposition, uptake and kinetics, the mechanisms behind the health effects are far from fully understood.

The exhaust from diesel engines includes several air pollution components such as ultrafine soot particles, smaller nucleation mode particles, nitrogen oxides, a range of organic compounds including polycyclic aromatic hydrocarbons (PAHs) and oxy-PAHs, and metal emissions, all of which are believed to play an important role in the observed health effects caused by ambient aerosols (Sydbom et al., 2001; Maricq, 2007). A key link between exposure to diesel emissions and the health response is the respiratory tract deposition of inhaled particles. So far, respiratory tract deposition has predominantly been investigated for spherical, hydrophobic and monodisperse particles, and only few experimental studies have investigated the deposition of real-world aerosols, such as combustion exhaust particles, which may be agglomerated, non-hydrophobic and almost always polydisperse (Maricq & Xu, 2004; Rissler et al., 2005, 2006). Furthermore, more lung deposition data are needed as the large intra-subject variability is not captured by models (Hofmann, 2011).

Measurements of respiratory tract deposition of aerosols emitted during combustion, other than tobacco smoke, are scarce (Löndahl et al., 2008, 2009; Morawska et al., 2005). Löndahl et al. (2008, 2009) have shown that hygroscopicity indeed alters particle lung deposition substantially. Morawska et al. (2005) examined deposition of particles from diesel and petrol combustion in 14 healthy non-smoking young adults. The deposition partly deviated from model predictions and hygroscopicity was suggested as the main explanation. Another suggested explanation was particle morphology. The hygroscopic growth, chemical composition or effective density of the particles was, however, not provided in that study.