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Characterisation of diesel particulate emission from engines using commercial diesel and biofuels

T. Ajtai [a, *], M. Pintér [b], N. Utry [b], G. Kiss-Albert [b], G. Gulyás [c], P. Pusztai [d], R. Puskás [d], Á. Bereczky [e], Gy. Szabados [f], G. Szabó [a], Z. Kónya [d], Z. Bozóki [a]

[a] MTA-SZTE Research Group on Photoacoustic Spectroscopy, Szeged, H-6720, Hungary
[b] Department of Optics and Quantum Electronics, University of Szeged, Szeged, H-6720, Hungary
[c] Hilase Development, Production, Service and Trading Limited Company, Szeged, H-6727, Hungary
[d] Department of Applied and Environmental Chemistry, University of Szeged, Szeged, Rerrich Béla tér 1, 6720, Hungary
[e] Department of Energy Engineering, University of Technology and Economics, Budapest, H-1111, Hungary
[f] KTI-Institute for Transport Sciences, Budapest, H-1119, Hungary

HIGHLIGHTS

- We investigated the diluted exhaust emitted by a diesel engine.
- The engine was operated with various mixtures of diesel and biodiesel fuels.
- Concentration and size distribution strongly depend on engine condition and fuel type.
- The thermal evolution of the emitted particulates was also investigated.
- Changes in concentration and structure were both observed due to thermal treatment.

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ABSTRACT

In this paper, the number concentration and the size distribution of diluted diesel exhaust particulate matter were measured at three different engine operating points in the speed-load range of the engine as follows: 1600 rpm; 50% load, 1900 rpm; 25% load, 1900 rpm; 75% load, adopted from the UN ECE Vehicle Regulation no. 49 (Revision 2) test protocol using pure diesel and biodiesel fuels, as well as their controlled blends. The emitted particulate assembly had lognormal size distribution in the accumulation mode regardless of the engine operational condition and the type of fuel. The total number and volume concentration emitted by the diesel engine decreased with increasing revolution per minute and rated torque in case of all the fuel types. The mixing ratio of the fuels did not linearly affect the total emission but had a minimum at 75% biodiesel content. We also studied the thermal evolution of the emitted particulates using a specially designed thermodenuder (TD) heated at specific temperatures (50 °C, 120 °C, and 250 °C). The first transition, when the temperature was increased from 50 °C to 120 °C resulted in lower number concentrations with small relative shifts of the peak position. However, in case of the second transition, when the temperature reached 250 °C the individual volatile particulates adsorbed onto the surface of soot particles were completely or partly vaporised resulting in lower total number concentrations with a substantial shift in peak position.

1. Introduction

Owning to their climate and health relevance diesel emitted carbonaceous particulates are in the middle of gradually increasing scientific interest nowadays. The highly absorbing carbonaceous aerosol emitted by diesel engines have a positive and direct climatic effect, while particles ejected by i.e. aircraft engines can influence cirrus and contrail formation eventuating in indirect and negative climate forcing respectively (Sekiguchi et al., 2003). According to the latest scientific assessments, regarding its climate impact, carbonaceous particulate matter (CPM), which is the by-product of

* Corresponding author.
E-mail address: ajtai@titan.physx.u-szeged.hu (T. Ajtai).

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incomplete combustion, is the second most important anthropogenic emission. Only the CO_2 gas has larger climatic impact than CPM (Bond et al., 2013). The optical response of aerosol strongly depends on size and chemical composition. Residence time and chemical reactivity also strongly depend on the characteristic diameter of the aerosol especially in the fine size region (PM1). Thus, size determines the climatic effect of ambient particulates both directly and indirectly.

Diesel soot is a dominant source of CPM especially in the fine particle size region (Braun et al., 2005). Due to their large surface area per unit mass and high adsorption capability for toxic substances, diesel soot particles have considerable and adverse health effects. Combustion related carbonaceous particulate matter is considered to be a significant risk factor of cardiopulmonary and cancer mortality (Lloyd and Cackette, 2001). Both the surface area per unit mass and the pulmonary deposition efficiency increases towards smaller particle sizes, therefore, the size of CPM is also a considerable factor of air quality.

Diesel exhaust is composed of versatile mixtures of abundant composites found in various mixing states. Diesel soot originates from versatile sources having different physicochemical features. Generally, diesel soot particles show high variety in chemical composition, size as well as in morphology and microstructure. The characteristic parameter set of diesel emitted aerosol strongly depends on the type of fuel and on the operational condition of the engine as well. Moreover, the chemical composition of the exhausted particles is also strongly affected by the actual sampling conditions. It is because the equilibrium state of vapour-particle partitioning is mainly governed by thermal energy, resulting in different vapour particle ratios at different temperatures. Owning to the dynamic nature of vapour-particle partitioning in the turbulent reactive plume of the engine tailpipe system, representative sampling and accurate measurement of the exhausted particulate is one of the major challenges in this field. Sampling directly from the tailpipe provides useful data regarding the operational conditions of the engine, however it only delivers limited climate and health relevant information. In order to ensure measurement conditions, which are more representative of real ambient processes, a specially designed dilution chamber has to be applied. That way the temperature of the exhaust gas and the concentration of the raw exhaust are reduced close to ambient temperature and to a level required by the applied measurement system, respectively. This way, the initial dilution effect on particle formation can be accurately investigated. To investigate the ambient effects on the chemophysical properties of the diesel particulates, specially designed reaction or smoke chamber experiments are required. Modern diesel engines are all equipped with after treatment devices like EGR (Exhaust Gas Recirculation) and DPF (Diesel Particulate Filter) units. However these after treatment devices do not only simply reduce the amount of emission but also change the characteristics of the emitted particles. Thus, the investigation of raw (engine-out) emission is most essential for fuel and engine development, and also important in order to better understand the masking phenomena of these devices.

When engine exhaust encounters cool ambient conditions, vapour species with high molecular weights are to condensate onto non-volatile soot particles by heterogeneous condensation or form new individual volatile particles through nucleated condensation. Both of these phenomena take place until the thermally driven equilibrium state is reached at a given temperature. At this dynamic equilibrium the residual molecules remain in the gaseous phase. These processes modify the chemical composition and the related mixing state of the freshly emitted diesel particulates, masking their number concentration and size distribution as well. The volatile to non-volatile ratio, the rate of individual volatile particulates with a homogeneous and spherical geometry to the particles composed of the non-volatile soot fractal aggregates and the condensed volatile material with core-shell mixing geometry are essential both from the prospective of climate relevance and risks to human health (Cheng, 2013). The mixing state and also the mixing geometry of the volatile to non-volatile particulate structures can be classified by the different thermal stabilities of the composites (Burtscher et al., 2001). According to this approach volatile material can be removed from the exhaust by a thermodenuder heated at a given temperature. The evaporated substances are subsequently absorbed by an activated carbon filter. Following that it becomes possible to investigate the remaining particles with modified number concentration and size distribution.

Due to gradually increasing scientific interest, the real-time investigation of the number concentration and size distribution of diesel emitted particulates has become an intensively studied issue nowadays. Most of the studies are focussing on diesel particulate size distribution measured directly at the exhaust pipe that is crucial from the point of view of fuel and engine development, however resulting in limited climate and health relevant information. Many of the size distribution measurements carried out under real ambient conditions that focus on traffic related aerosols are also available in the literature. Although, the ambient factors and the cross sensitivity of other sources can reduce the reliability and the general validity of the source related information obtained from these studies. The climate and health relevant source related emission properties of the diesel particulates can be modelled and investigated under controlled laboratory circumstances using a standard dilution chamber at atmospheric pressure and temperature (Burtscher, 2005). There are a great number of source related studies executed under well controlled laboratory conditions in a specially designed dilution chamber available in literature. Furthermore, size distribution measurements aiming to investigate the dynamic changes in the volatile-non-volatile ratio have also been intensively studied. Most of them are focussing on emission characteristics using specific fuel types. Moreover, although the biodiesel content in the diesel fuel is specifically regulated in all EU countries, the effect of the diesel to biodiesel ratio on emission and on volatility have also been investigated, but the number of comprehensive studies using the same measurement condition, engine type and experimental setup are still limited (Popovicheva et al., 2014; Uhrner et al., 2011; Rothenbacher et al., 2008; Bukowiecki et al., 2012; Liu et al., 2007; Abdul-Khalek et al., 1998; Ahlvik et al., 1998; Trapel et al., 2005; Mathis et al., 2004; Steiner et al., 2013).

The ratio of biodiesel to conventional diesel in the total fuel blend can influence both the total number and volume concentration and distribution of the particle emission. Therefore, investigating these parameters in the function of biodiesel content and volatility is extremely important from the point of view of engine and fuel development too. Therefore, the detailed investigation of the size distribution and the volatility of diesel emitted particulates as a function of biodiesel content of the applied fuel are timely and important issues in the fields of climate and air quality research. Hence the aim of this work is to evaluate and characterize the effect of fuel composition on diesel particulate emission at different engine loads and volatility test conditions in order to better understand the characteristics of particulate emissions from vehicle engines.

In this paper, we are studying the number concentration and size distribution of diesel particulate matter in the function of the operational conditions of the engine using diesel and biodiesel fuels as well as their controlled mixtures. We also study the size distribution of the emitted particulates in the function of thermal stability under various measurement conditions.