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Application of the PMP methodology to the measurement of sub-23 nm solid particles: Calibration procedures, experimental uncertainties, and data correction methods

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ABSTRACT

This paper discusses issues which will appear if the PMP methodology is extended to the measurement of particles with sizes below 23 nm. In addition, the origins of sub-23 nm particles from three different types of engines are discussed. An increased uncertainty was observed for the detection efficiency measurements with a particle number counter (PNC) at particle sizes below 10 nm. The particle concentration reduction factor (PCRF) of a volatile particle remover for 15 nm particles was higher than those for 30, 50 and 100 nm particles because of increasing particle losses. Then, exhaust particle emissions from a gasoline direct injection (DI) passenger car and trucks with and without a diesel particulate filter (DPF) were measured using the PMP methodology with a PNC whose D_{50} value was 2.5 nm (PNC_2.5) and a normal PMP PNC (PNC_23). The fluctuations of the results for PNC_23 and PNC_2.5 were almost the same when the effect of re-nucleation of semi-volatile particles was removed. Different procedures for estimating the PCRF showed varying results, −23% to +41% in the worst case. The profiles of sub-23 nm particles from diesel trucks were similar to those of particles larger than 23 nm. In the case of the gasoline DI car, a peculiar emission corresponding to hydrocarbon nucleation was observed in high-speed operation. Particles emitted from the diesel engines originated from combustion processes in the case of the truck without the DPF and were strongly affected by the DPF conditions in the case of the truck with the DPF.

1. Introduction

In 2013, the World Health Organization (WHO) classified particulate matter (PM) emitted from diesel engines as a group 1A carcinogen (World Health Organization, 2012), and even before then, the toxicological aspects of PM emissions from automobiles had been investigated (Furuta et al., 2008; Nikula, Snipes, Griffith, Henderson, & Mauderly, 1995). The reason for the interest in this PM is that the sizes of PM from diesel engines are usually in the range of 10–100 nm, which are called ultrafine particles (UFPs; Kittelson, 1998), and they are considered to be one of the major elements of PM2.5, which is clearly related to human diseases (Dockery et al., 1993; World Health Organization, 2013). In addition, recent studies have indicated

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that UFPs have a considerably higher toxicity than larger particles (Bakand, Hayes, & Dechsakultorn, 2012; Fujitani et al., 2012; Oberdörster, Oberdörster, & Oberdörster, 2005).

The European Union started a new approach for regulating the particles emitted from diesel engines for light-duty vehicles in 2011 (UNECE Regulation 83, Commission Regulation 692/2008, 2008) and for heavy-duty vehicles in 2013 (UNECE Regulation 49, Commission Regulation 582/2011, 2011), after discussions in the PMP Working Group of the UNECE GRPE (Andersson & Clarke, 2004; Andersson, Giechaskiel, Munoz-Bueno, & Dilara, 2007; Andersson, Mamakos, Martini, & Giechaskiel, 2010; Giechaskiel, Dilara, & Andersson, 2008; Giechaskiel et al., 2012). The new approach counts the particle number (PN) of solid particles. This method has high sensitivity for UFPs emitted from diesel engines compared with the traditional filter weighing method. However, the lower detection limit (D_{50}) of the PN method was set to 23 nm to increase the experimental repeatability by removing the effect of nucleation mode particles whose diameters are usually below 23 nm (Giechaskiel et al., 2012).

In addition to the particles emitted from diesel engines, recently the emissions from gasoline direct injection (DI) engines have also been an issue (Khalek, Bougher, & Jetter, 2010; Mamakos, Steininger, Martini, Dilara, & Drossinos, 2013), and the European Union decided to introduce new regulations from 2014. For the next step, the PMP working group has started discussions about the regulations for solid particles smaller than 23 nm. This is because the particles from gasoline DI engines are considered to contain a higher percentage of particles below 23 nm (Giechaskiel & Martini, 2013). In addition, Rönkkö et al. (2014) pointed out the existence of metals in particles below 7 nm from gasoline DI vehicles. The simplest procedure for measuring sub-23 nm particles is to replace the counter in the PN measurement system with one having a lower D_{50}. However, there are some issues that need to be resolved in this procedure. One is the calibration process, which is particularly important for devices that are used in type approval tests. In addition, establishing a procedure for estimating the total particle concentration reduction factor (PCRF) is also important in evaluating the effect of smaller particles. Another issue is that the characteristics of sub-23 nm particles from engine exhaust are mostly unknown.

In this study, a calibration process was verified using the PMP methodology with a particle number counter (PNC) having a D_{50} value of 2.5 nm. The detection efficiency of the PNC for particles smaller than 10 nm was confirmed. The PCRF of a volatile particle remover (VPR) was also observed with 15 nm particles, in addition to the 30, 50, and 100 nm particles that have conventionally been used to estimate the total PCRF. Then, the exhaust particles from three vehicles were observed to evaluate the repeatability of the measurements and the effect of the total PCRF estimation procedure on the actual results. The tested vehicles were a passenger car with a gasoline direct injection (DI) engine, and two medium-duty trucks; one of the trucks was equipped with a diesel particulate filter (DPF) and the other was not. The effects of renucleation of semi-volatile particles, which has been observed by several researchers (Jonson et al., 2009; Zheng et al., 2011; Zheng et al., 2012), were minimized by using a high dilution. In addition, the profiles of particles below 23 nm were compared with those of other emissions, and their characteristics are discussed.

2. PMP methodology using particle counter with D_{50}=2.5 nm

2.1. Outline of measurement system

Figure 1 shows an outline of the measurement system used for measuring the particles emitted from automobiles. The adopted measurement system (APC, AVL Inc., 2014) meets the requirements of the PMP. Exhaust gas from an automobile was introduced into the measurement system after dilution by a constant volume sampler (CVS; CVS7400, Horiba Inc.).

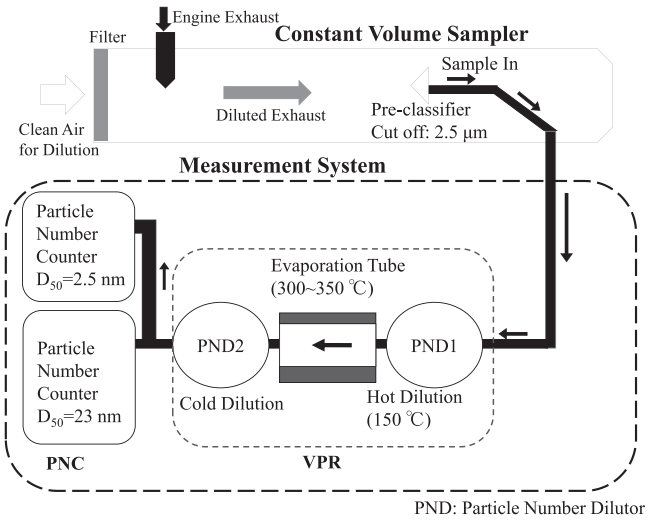

Fig. 1. Outline of tested measurement system.