

High Temperature Sampling System for Real Time Measurement of Solid and Volatile Fractions of Exhaust Particulate Matter

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ABSTRACT

This paper discusses the design and qualification of a High Temperature Sampling System (HTSS), capable of stripping the volatile fraction from a sample flow stream in order to provide for quantification of total, solid and volatile particulate matter (PM) on a near real-time basis. The sampling system, which incorporates a heated diesel oxidation catalyst, is designed for temperatures up to 450°C. The design accounts for molecular diffusion of volatile compounds, solid particles diffusion and reaction kinetics inside one channel of the oxidation catalyst. An overall solid particle loss study in the sampling was performed, and numerical results were compared with experimental data gathered at the West Virginia University Engine and Emissions Research Laboratory (EERL) and West Virginia University's Transportable Heavy-Duty Vehicle Emissions Testing Laboratory (THDVETL). Data indicated that “dry soot” aerosol streams, which were produced at a single engine operating mode on an engine test stand point using a dynamometer were largely unaffected by the sampling system. Important differences were identified in the nucleation mode between steady-state and transient tests - a smaller nucleation mode was observed in the transient operation. Depending upon the test cycle, the particle loss, on count basis, in the HTSS ranged from 10% to 26%. The HTSS was found to be very effective in scrubbing the volatile organic fraction (VOF) from the exhaust stream; hence, resulting in a 96% reduction in total PM count and 52% total PM mass under steady-state mode of engine operation. Under transient operation, the total PM count reduction was 55%, and the total PM mass reduction was 32%. The difference in the untreated sample stream and the treated sample was

evident in the concentration and size distribution of nucleation mode particles. The sampling system represents a viable method for collecting nanoparticles and the larger accumulation mode particles and the opportunity to perform an extensive suite of chemical and morphological analyses.

INTRODUCTION

Exhaust total particulate matter (TPM) is defined as the material collected on a filter from a stream of exhaust that is cooled and diluted by fresh air to a temperature of 47±5°C. The material collected is composed of solid (primarily, carbonaceous soot) and volatile matter (unburned fuel, lubricating oil and sulfur fractions) called volatile organic fraction (VOF). While dry soot is formed in the combustion chamber, VOF is transformed into particles only when the exhaust cools down and super-saturation conditions occur. Particles formed by the homogeneous nucleation of VOF ($D_p < 50$ nm) are smaller in size than those derived from dry soot ($D_p > 50$ nm). Therefore, it is important to characterize them separately due to their different morphology, and associated potential health risks. These factors highlight the importance of being able to differentiate volatile and the non-volatile components of TPM in order to better define the PM formation processes, correlating with the PM characteristics with associated parameters of engine operation, and correlate the volatility of PM emissions with toxicity. Different systems and approaches have been used in the past to analyze the PM concentrations, size distributions, chemical and physical characteristics and toxicity. Oxidation catalyst-based systems are capable of scrubbing the volatile fraction as shown by Kittelson et al. [1] and Khalek[2]. Thermo-denuder systems utilize activated charcoal to absorb the VOF from the

untreated exhaust[3]. Hot dilution method prevents the nucleation of the VOF; hence, analysis of the solid fraction is more readily afforded [4]. The motivation for this work was the need to investigate toxicity of the VOF associated with PM emissions. This paper does not discuss PM-related toxicity, but the design process, and qualification of an oxidation catalyst-based PM sampling system, which is capable of operation at higher flow rates and temperatures than any discussed in published literature. To the best of our abilities, the High Temperature Sampling System (HTSS) design process took into account every potential interaction between PM and the sampling system.

HIGH TEMPERATURE SAMPLING SYSTEM (HTSS)

Before starting the explanation of the HTSS design, it is important to define the operating parameters. The desired flow rate was set up to 200slpm, and an operating oxidation catalyst (OC) temperature range was 250-450°C. The high sample flow rate was selected to maximize the rate of PM mass collected on the filter media (compared to the commercially available systems, Kittelson et al.[1] and Khalek[2] studies). In order to guarantee an homogeneous flow over the OC, a smooth cone diffuser with 6° angle is used. Further, PM losses in the HTSS were minimized. The stainless steel tubing was insulated to minimize heat losses and the consequent radial temperature gradients in the tubing in an attempt to minimize thermo-phoretic losses. The HTSS was designed to operate over a wide range of temperature and sample rates; thus, affording flexibility of use at different engine operating conditions. To remove the maximum quantity of volatile organic fraction (VOF) the OC need to reach maximum conversion efficiency for specific organic class being targeted. Based on the study of Kittelson et al. [1] and Khalek [2] and the characteristics of the OC used in the HTSS the target temperature was set to 350°C, which was in the in the middle of the designed temperature operating range of the HTSS. Owing to the high flow rate at which the system was designed to operate, it was decided to heat the flow tubes in the sampling system instead of externally heating the oxidation catalyst. However, the oxidation catalyst was well insulated. This approach averted the non-uniform temperature distribution inside the oxidation catalyst. Homogeneity of temperature within the oxidation catalyst allowed for similar catalytic reaction in each channel of the ceramic. In addition, heating the flow stream prior to its entry into the oxidation catalyst ensured optimal vaporization of the VOF in the exhaust. The treated exhaust flow was drawn over a T60 Pall® PM filter. Given the high temperature properties of the T60 filter the treated exhaust sample stream did not need any cooling downstream of the oxidation catalyst; hence, avoiding thermophoretic losses.

HTSS OVERVIEW

The HTSS is comprised of:

1. Primary Dilution Tunnel
2. Heated Line (HL)
3. Oxidation Catalyst
4. Filter Holder
5. Intercooler
6. Mass Flow Controller
7. Pump
8. Secondary Dilution Tunnel
9. HTSS Management System

As shown in [Figure 1](#), a sample from a primary dilution tunnel was extracted by means of a heated line that was used to equilibrate aerosol stream to a constant temperature value. The sample flow was split into two - 5slpm was drawn into a secondary dilution tunnel for PM concentration and size distribution measurements with an Engine Exhaust Particle Sizer (EEPS™), and the remainder was drawn across a filter holder (200slpm). An in-line intercooler was essential to avoid any damage to the Mass Flow Controller (MFC) and the pump. Moreover, the presence of the intercooler guarantees a flow through the MFC with a temperature of 25°C avoiding any flow-rate drift during experiments.

HTSS DESIGN

The main constraint of the HTSS design was to minimize particle loss, and maximize scrubbing of the VOF within the OC. The starting point of the design was to identify parameters of the commercially available OC (see [Table 1](#)).

Table 1. Oxidation Catalyst Main Parameters

Material	Ceramic Monolith
Wash Coat	Zeolite, Alumina
Catalyst	Platinum
Geometry	Square Channels
Overall Length [in]	6
Overall Diameter [in]	5.6
Cell Density [# /m ²]	620,000
Square Channel Wall Thickness [cm]	0.0127
Square Channel Open Width and Height [cm]	0.139

The HTSS was designed for relatively large sample flow rate (200slpm), and the OC was appropriately sized; resulting in an optimal flow rate of 0.02splm per channel. This produced

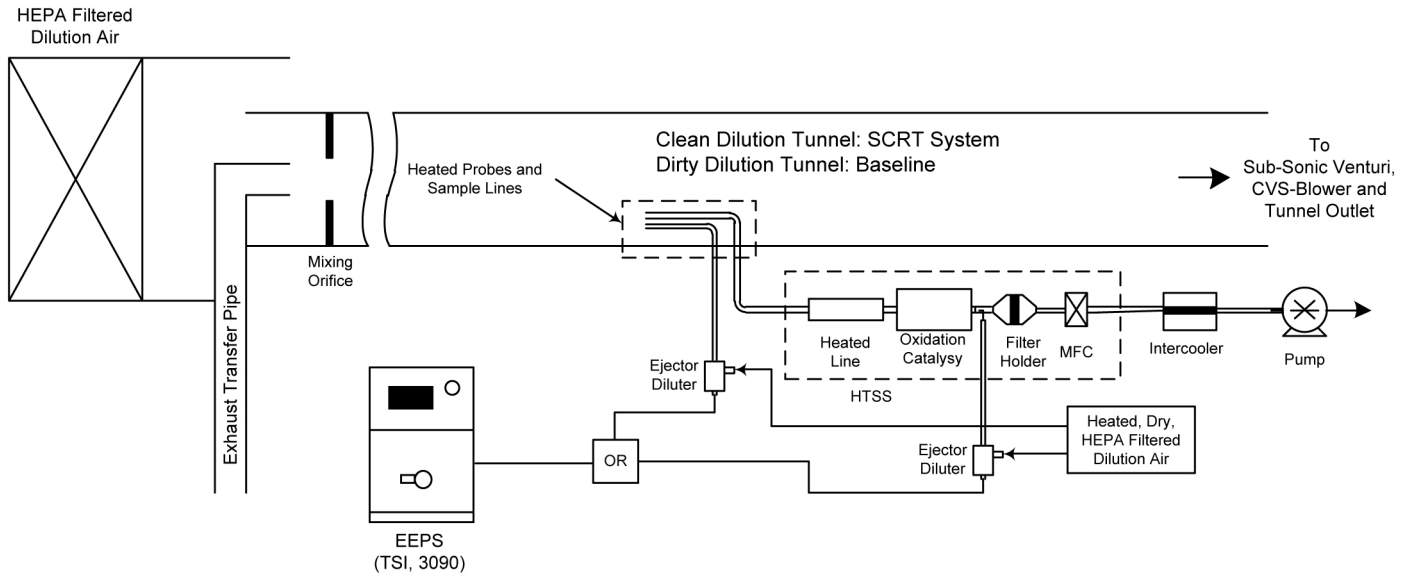


Figure 1. HTSS Schematic

an average residence time of 0.45s, which was between those reported by Kittelson et al.[1] and Khalek[2].

Solid Particle Penetration

Brownian Diffusion

Brownian diffusion is the process by which particles disperse in a concentration gradient. Considering a cross-section of a channel with a defined concentration on the channel axis and zero concentration at the walls, thereby providing the gradient that drives diffusion of particles from the axis to the channel's walls. This is a well-known problem and, even if a closed solution is available, has primarily been solved using the model suggested by Kittelson et al., [1] of Liu [5] for particle deposition in a laminar tube flow, by diffusion. Liu modeled this problem in terms of a dimensionless coefficient:

$$\xi = \frac{DL}{R^2U} \quad (1)$$

Where L is the length of the channel, R is the tube radius, U is the average velocity of the flow and D is the diffusion coefficient. For the work presented herein, half of the hydraulic diameter has been substituted for R . The particle diffusion coefficient is given by the Stokes-Einstein equation [6]:

$$D = \frac{KTC_c}{3\pi\mu d} \quad (2)$$

Where K is the Boltzmann constant, T is the bulk temperature of the flow, μ is the dynamic viscosity and d is the particle diameter. C_c is the Cunningham slip correlation factor and is included when the particle diameter is comparable with the gas mean free path, so as to account for a slip condition between the gas stream and solid particle surface. The expression for the slip factor is given by [7]:

$$C_c = 1 + \frac{2\lambda}{d} \left[1.257 + 0.4 e^{-\frac{1.1d}{2\lambda}} \right] \quad (3)$$

Where λ is the gas mean free path that, for air, is equal to:

$$\lambda = \frac{23.1 T}{P} \quad [\mu m] \quad (4)$$

The Liu relationship between ξ and the particle penetration is:

$$\frac{C_{out}}{C_{in}} = 1 - 2.56 \xi^{\frac{2}{3}} + 1.2\xi + 0.177 \xi^{\frac{4}{3}} \quad (5)$$

With a total HTSS flow rate of 200slpm, the flow in each channel is laminar as shown in Figure 2 below. Penetration was evaluated by Liu formulation at different HTSS temperatures.

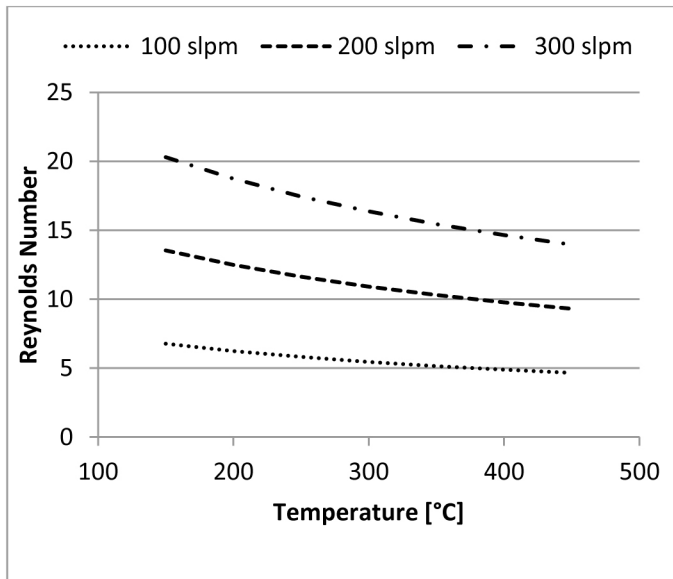


Figure 2. Reynolds Number Analysis for different HTSS operation points

Figure 3 represents the curves of Solid Particle Penetration for various temperatures with a flow rate of 200slpm. This parameter is identified as the ratio the number of particle passing through the OC and over the number of particle entering the brick. This parameter can be predicted with the described model for any particle size. It is evident that particle penetration has an inverse functional dependence with temperature, due to the higher diffusion coefficients. It is important to note that temperature does not strongly affect penetration.

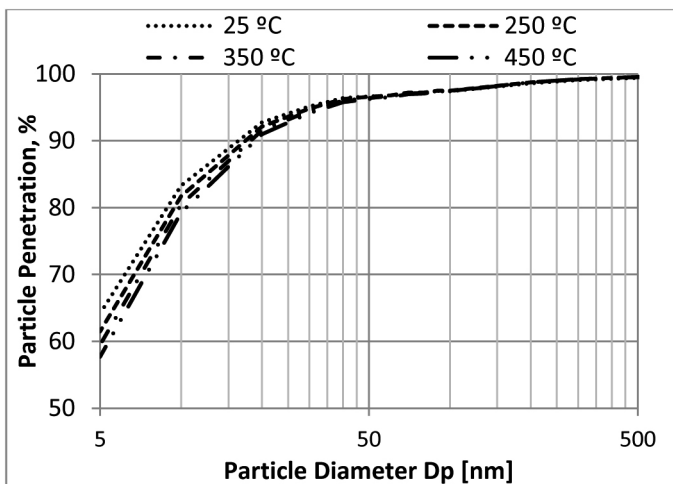


Figure 3. Solid Particle Penetration for different Temperatures with 200slpm

It was decided to perform the particle loss study with actual exhaust stream over steady-state and transient operation modes. Needless to say, evaluation of PM loss in the HTSS at high temperatures was difficult because of the chemical reactions occurring in the OC. It was decided to perform the particle loss study with the HL off, leading to a temperature flow of 25°C. Figure 3 helped in this decision, it is evident the effect of temperature on particles losses is negligible; hence the results of particle loss study at 25°C are not going to be different from those at higher temperatures.

Inertial Impaction

Johnson [8] studied the upstream edge of the ceramic wall between adjacent channels as a site of inertial impaction in Diesel catalytic converters basing his work on Langmuir's study. It was reported that significant deposition occurs for Stokes numbers of 0.6 and above. The Stokes number for flow around a flat ribbon of width 2w and an upstream velocity U is defined as:

$$Stk = \frac{\rho_p d^2 U C_c}{18 w \mu} \quad (6)$$

In the OC the Stokes number for the largest particle diameter (1µm) is much smaller than 0.6 so inertial impaction deposition is not expected.

Thermophoresis

Thermophoresis is the motion of particles due to a temperature gradient; in particular particles experience a net force in the direction of decreasing temperature. In this case the presence of a heated line and the good insulation over the oxidation catalyst and tubes reduced the likelihood of thermophoretic losses.

Gravitational Sedimentation

For Diesel PM, gravitational sedimentation was not expected.

Overall Losses

By applying the same loss analysis to the heated line and other tubes of the HTSS, it is possible to obtain an overall Solid Particle Penetration, as shown in Figure 4.