



Effect of Particle Size and Moisture on Emission of Organic Carbon from Vehicular Exhaust

Ebin Johnson¹, Sahu S. K^{2*}, Rathod T. D.², Ajmal P. Y.², Tiwari M.², Bhangare R. C.², P. V. Rao¹, Pandit G. G.²

¹National Institute of Technology Warangal, Telangana, India

²Bhabha Atomic Research Centre, Trombay, Mumbai, Maharashtra, India

ABSTRACT

For decades, the concentration of aerosols in the lower atmosphere has increased steadily primarily owing to anthropogenic sources such as industrial activity, vehicular emission, biomass burning, and also to secondary aerosol formation. As a consequence of urbanization, a phenomenal surge has been observed in the vehicular population in India, giving rise to elevated levels of traffic related pollutants like carbon monoxide, nitrogen oxides, hydrocarbons, and particulates in urban centers. These pollutants can have both acute and chronic effects on human health. The vehicular emissions are governed by various factors such as purity of the fuel, capacity and health of vehicle, type of fuel etc. Alongwith others, moisture is one of the major product of vehicular emission. The effect of moisture plays an important role not only in the physical characteristics of aerosols emitted from the vehicular exhaust, but also in the chemical characteristics. The tail pipe emission of automobiles mainly consists of soot (black carbon), some organic compounds, and moisture. The interactions between the organic compounds with or without the presence of moisture need to be effectively studied.

In this study, the effect of size distribution and moisture on the emission of carbon in vehicular exhaust was studied. The size distribution of was carried out using variable configuration cascade impactor (VCCI). Additionally, a denuder of activated silica gel was designed to trap the moisture and to study the effect of moisture. The quantification of carbon was carried out using a TC-TN analyzer. The average emission of organic carbon aerosols from both diesel and petrol vehicles were maximum in the lowest particle size i.e. nucleation mode. Both diesel and petrol exhaust showed tri-modal distribution over the range of studied particle sizes. In both diesel and petrol vehicular exhaust, organic carbon was less in moisture free environment (with denuder) in the accumulation mode.

Keywords: VCCI, TC-TN analyzer, denuder, particle size

I. INTRODUCTION

Vehicular emissions continue to attract the attention of environmentalists and toxicologists not only because greenhouse gases are known to have adverse effects on global climate, but also because many organic compounds found in automobile emissions

have high ozone forming potential or health effects on humans and other living organisms. As the number of in-service vehicles continues to grow around the world, several remedial measures including, stringent emission regulating legislation have been employed to reduce the impact of vehicular emissions on the environment and its ecosystems [1].

Motor vehicles comprise a significant source of atmospheric pollutants. Exhaust emissions from vehicles consist of a hot and complex mix of both gaseous and particle phases. The gaseous emissions include carbon dioxide (CO₂) which plays a major role in global warming. The particles emitted are mostly carbonaceous spherical submicron agglomerates formed as a result of incomplete combustion in the engine and are very often coated with various organic compounds [2-4].

Several adverse health and environmental effects have been attributed to emissions from urban vehicular traffic. Epidemiological studies have linked particulate matter in urban environments with mortality, hospital admission increases and various cardiovascular and respiratory diseases [5-7]. The size distribution of these particles plays an important role as, during inhalation, the smaller particles penetrate deeper into the human respiratory system and are more likely to be retained there, enhancing harmful toxicological effects [8, 9]. A knowledge of the organic compounds and the effect of moisture on particles in vehicle emissions, particularly in relation to different types of vehicle used, is therefore of great importance in the understanding of these adverse effects.

According to diameter, particles can be classified in three modes: nucleation mode (particle diameter <50 nm) Aitken mode (particle diameter between 50 nm to 100 nm) and accumulation mode (particle diameter >100 nm) [10]. Particles emitted from vehicular exhaust are mostly in the all three modes.

The aim of the present study was to measure the organic carbon and total carbon emissions from petrol and diesel vehicles and to draw conclusions regarding any differences in the emissions using the two types of vehicles and any difference in emission due to the moisture conditions. A silica gel denuder designed and was used to trap the moisture and to study effect of

moisture on carbon emission from the vehicular exhaust.

In present study near source, size fractionated aerosols from exhaust of vehicles of different fuel types (Petrol and Diesel) were collected in >21 μm to <0.1 μm size range in twelve stages using a variable configuration cascade impactor (VCCI).

II. MATERIALS AND METHODS

2.1 Collection

Various models of both diesel and petrol vehicles were selected for the study. Each vehicle's exhaust was collected with the help of Variable configuration cascade impactor (VCCI) for a sampling time of 15 minutes. Samples were collected on glass fiber filter papers. Filter papers were desiccated and weighed prior to the experiment. The final weight was also measured to know the loading and mass size fraction of organic compounds from exhaust aerosols. For the second set of experiment, a specially designed denuder was connected before the VCCI to trap the moisture. Deposition of aerosols from a vehicular exhaust in different stages of VCCI is shown in Figure 1.

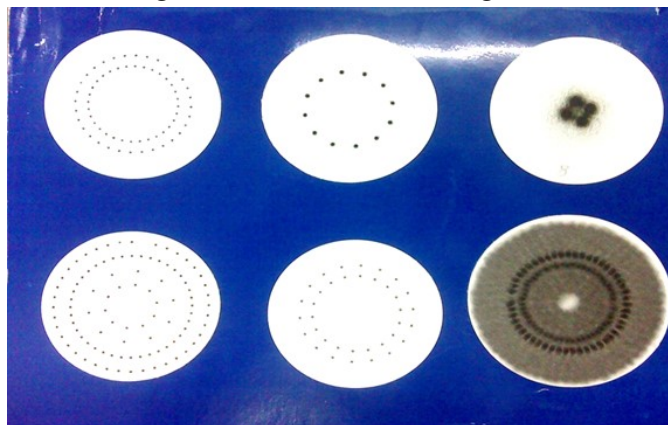


Figure 1. Deposition of aerosols from vehicular exhaust in different stages of VCCI.

1.2 Experimental setup

The experimental setup for determination of particle size distributions of aerosols from vehicular exhaust is shown in the Fig 2. It consists of a funnel shaped pipe

which was kept close to the exhaust pipe of vehicles. It allows all the exhaust to pass through the denuder. A denuder of activated silica gel was used to trap the moisture and to study the effect of moisture on the organic and total carbon from the exhaust. It was followed by a variable configuration cascade impactor (VCCI) for size fraction and Anderson pump to maintain the air flow.

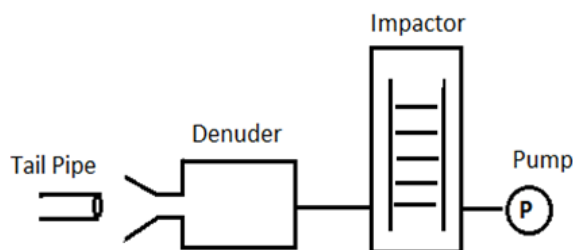


Figure 2. Setup for collection of size fractionated carbonaceous aerosols from vehicular exhaust

2.3 Cascade impactor

Mass size distribution of carbonaceous aerosol from vehicular exhaust was assessed on variable configuration cascade impactor (VCCI) [11]. Glass fiber paper discs (EPM 2000, Whatman) were used as an impaction surface for particles as well as a backup filter in the cascade impactor. Glass fiber paper were dried in an oven at 100 °C for 2 h, kept in a desiccator for 24 h and weighed prior to sampling. Air flow rate of 10 lpm was maintained in the impactor. The size ranges (μm) collected from the different stages of cascade impactor were as follows; >21.3 , 21.3–15.1, 15.1–11.2, 11.2–7.38, 7.38–5.47, 5.47–2.23, 2.23–1.13, 1.13–0.75, 0.75–0.50, 0.50–0.30, 0.30–0.10 and <0.10 . The pressure at the last stage was continuously measured through the course of sampling so that the flow rate and stage cut off can be monitored. Sampling time was 15 minutes for each type of vehicle which passed about 0.15 m^3 volume of air approximately.

2.4 TC-TN analyzer

Total carbon (TC) and total organic carbon (OC) was analyzed with help of TC-TN Analyzer (Primacs SNC-100). According to different temperature profiles, the

total carbon and total organic compounds of samples are directly given by the instrument with the help of standard curves. Standard curves were prepared with EDTA standard. In TC method the sample is automatically introduced into the combustion oven and combusted at given temperature. At the end of the combustion procedure, the gas mixture is directly led to the Non dispersive infra-red (NDIR) detector. NDIR measures the amount of CO_2 gas released after combustion. At 600°C instrument gives the total organic carbon present in the sample and at 1200 °C, it will give total carbon content of the sample.

III. RESULTS AND DISCUSSION

The average emission of organic carbon aerosols from both diesel and petrol vehicles were maximum in the lowest particle size i.e. nucleation mode. Both diesel and petrol exhaust showed tri-modal distribution in different particle sizes. In both diesel and petrol, organic carbon was less in moisture free environment (with denuder) in the accumulation mode.

The total carbon aerosols from both diesel and petrol vehicles were maximum in the lowest range or nucleation mode. The moisture free condition i.e. with denuder condition showed less emission of total carbon than with the denuder.

3.1 Mass size distribution of OC aerosols from diesel vehicles

The distribution of mass concentration with particle size (aerodynamic diameter) of the aerosols from exhaust of diesel vehicles is shown in Fig.3. The average total mass concentrations of organic carbon in all the size fractions were found to be 68.32 mg/m^3 without denuder and 81.99 mg/m^3 with denuder. A multi modal mass size distribution was observed in which a maximum mass concentration was seen in $<1 \mu\text{m}$ in particle size in both with and without denuder cases. The mass percentage of organic carbon particles

was 56% in without denuder case and 78% in with denuder case.

The mass percentage of organic carbon aerosols having particle size $<11.2\mu\text{m}$ (respirable size) [11] of the total organic carbon aerosols was found to be 77% in without denuder and 74% in with denuder case, which specifies that major part of organic carbon mass emitted from the diesel vehicles is capable of getting into respiratory track. The mass percentage of fine particles ($<2.23\mu\text{m}$ aerodynamic diameter) [11], which are capable of deep pulmonary infiltration and alveolar deposition, was found to be 46% and 56% in without and with denuder cases respectively. The contribution of ultrafine particles (aerodynamic diameter $< 0.1 \mu\text{m}$) to the total organic carbon mass emitted as exhaust was around 30% in without denuder and 28% in with denuder case.

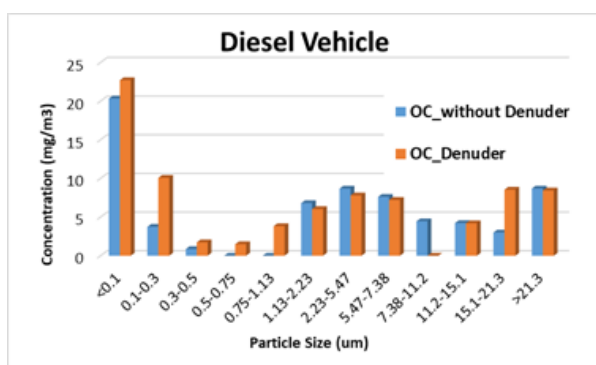


Figure 3. Mass size distribution of OC aerosol from diesel vehicles

3.2 Mass size distribution of OC aerosol from exhaust of petrol vehicles

It is well understood from the literatures that aerosols from vehicular exhaust have a tri-modal mass size distribution. The fine mode is about $0.1 \mu\text{m}$ (also referred to as submicron mode, ultrafine mode, vaporization mode, condensation mode, etc.) while the coarse mode is often larger than $1 \mu\text{m}$ (also referred to as super micron mode, coarse mode, residual mode, etc.). The mass size distribution of OC aerosol generated from exhaust of petrol vehicle in present study is shown in Fig 4. The distribution pattern was found multimodal and maximum concentration was at size $<1 \mu\text{m}$. A third mode was

found in coarser size range i.e. 11.2 to $15.1 \mu\text{m}$. The mass percentage of organic carbon particles was 68% in without denuder case and 73% in with denuder case.

The average total mass concentration of the organic carbon aerosols at the sampling point of setup was observed 105.42 mg/m^3 in without denuder (with moisture) case and 100.53 mg/m^3 in with denuder case. The mass percentage of particle having aerodynamic diameter $<11.2\mu\text{m}$ to the total organic carbon aerosols was 74% and 81% in without denuder and with denuder cases respectively, which indicates that a major part of OC mass emitted from the exhaust is going to respiratory track. The mass percentage of fine particle to the total particle mass was found 53% and 60% in with moisture and without moisture case respectively. The contribution of ultrafine particle i.e. particle having aerodynamic diameter $< 0.1 \mu\text{m}$ to the total mass emitted was found to be 19% with moisture and 25% in without moisture.

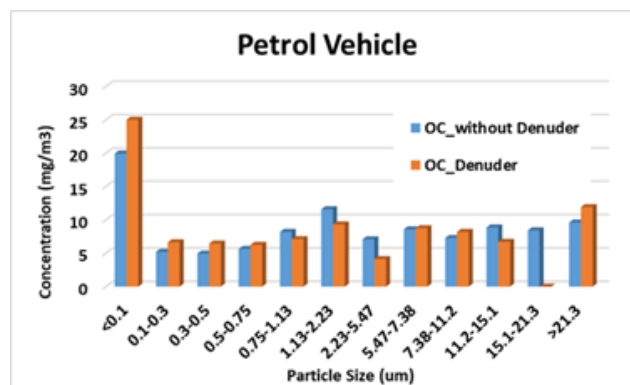


Figure 4. Mass size distribution of OC aerosol from petrol vehicles

3.3 Mass size distribution of total carbon aerosol from exhaust of diesel vehicles

The average total mass concentration for TC aerosols from exhaust of diesel vehicles was observed to be 121.83 mg/m^3 in without denuder (with moisture) case and 105.34 mg/m^3 in with denuder (without moisture). The mass size distribution of carbon aerosol generated from exhaust is represented in Fig.5. Maximum mass concentration was found in size

<0.1 μm which contributed to 24% of total mass emitted in all size ranges. This indicates that mass of TC is mostly concentrated in fine particulate size range. The mass percentage of particles having aerodynamic diameter < 11.2 μm to the total mass was found to be 79%, which shows that a major part of TC emitted from exhaust can reach to the respiratory tract. The mass percentage of particles having < 2.23 μm aerodynamic diameter to the total carbon mass was found 53%. The contribution of particle having aerodynamic diameter < 0.1 μm to the total mass emitted as exhaust was found 24%. Mass concentration of TC in with denuder condition was found 79%, 61%, 23% in < 11.2 μm , < 2.23 μm and <0.1 μm sizes respectively.

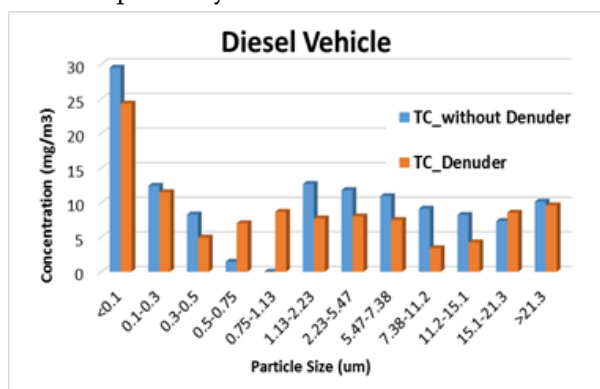


Figure 5. Mass size distribution of TC aerosol from diesel vehicles

3.4 Mass size distribution of total carbon aerosol from exhaust of petrol vehicles

Petrol vehicles are mostly preferred as personal vehicles than diesel vehicles by the people. Total mass concentration emitted from exhaust of petrol vehicles was found to be 156.19mg/m³ at the sampling point of setup. The mass size distribution of total carbon aerosols is represented in Fig.6. The mass percentage of particles having aerodynamic diameter < 11.2 μm (respirable size) to the total mass was found 79%. The mass percentage of particles having < 2.23 μm aerodynamic diameter to the total particle mass was calculated as 56%. The contribution of particles having aerodynamic diameter < 0.1 μm to the total carbon mass emitted as exhaust was found 21%. Mass

concentration of total carbon in the with denuder condition was found 78%, 57%, 19% in < 11.2 μm , < 2.23 μm and <0.1 μm sizes respectively.

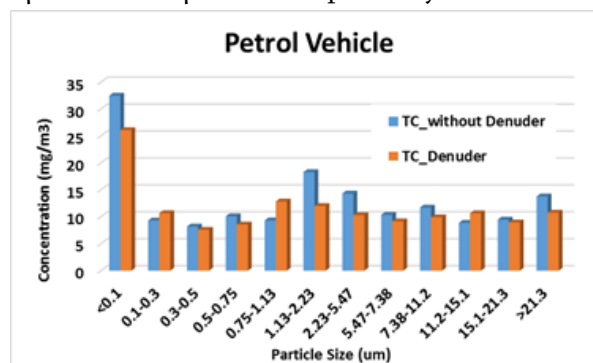


Figure 6. Mass size distribution of TC aerosol from petrol vehicles

IV. CONCLUSION

Results show that in majority of size ranges, the carbon emissions from both Petrol and diesel vehicles were reduced when the moisture from the exhaust was trapped. The reason may be that some carbonaceous aerosols were dissolved in moisture and were removed along with it. It has been also noted that other than nucleation mode most carbonaceous aerosols are accumulated in the size range 1.13-5.47 μm in the accumulation mode. In nucleation mode petrol vehicle is having less mass concentration than diesel vehicles, while in accumulation mode petrol vehicle is having more mass concentration than in diesel vehicles and in coarse mode mass concentration in both diesel and petrol vehicles was found to be almost same. However, the differences are not always statistically significant owing to the large variation in emissions from different vehicles powered by the same type of fuel.

V. REFERENCES

- [1]. M. C. Lim and G. Ayoko. 2006. Atmospheric Environment, 40(17), 3111-3122.
- [2]. L. M. Hildemann and G. R. Markowski. 1991. Environmental Science & Technology, 25(4), 744-759.

- [3]. Z. D. Ristovski and L. Morawska et al. 1998. Environmental Science & Technology, 32(24), 3845-3852.
- [4]. M. J. Kleeman and J. J. Schauer et al. 2000. Environmental science & technology, 34(7), 1132-1142.
- [5]. A. Seaton and D. Godden et al. 1995. The lancet, 345(8943), 176-178.
- [6]. S. Vedal. (1997). Journal of the Air & Waste Management Association, 47(5), 551-581.
- [7]. C. Pope and S. Ziebland et al. 2000. BMJ: British Medical Journal, 320(7227), 114.
- [8]. J. Ferin and G. Oberdorster et al. 1992. American Journal of Respiratory Cell and Molecular Biology, 6 (5), 535-542.
- [9]. K. Donaldson and X. Y. Li et al. 1998. Journal of Aerosol Science, 29(5-6), 553-560.
- [10]. F. Raes and R. Van Dingenen et al. 2000. Atmospheric environment, 34(25), 4215-4240.
- [11]. M. Tiwari and S. K. Sahu et al. 2013. Microchemical Journal, 106, 79-86.