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Rapid Identification of High Particle Number Emitting On-Road Vehicles and Its Application to a Large Fleet of Diesel Buses

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Pollutant concentrations measured in the exhaust plume of a vehicle may be related to the pollutant emission factor using the CO_2 concentration as a measure of the dilution factor. We have used this method for the rapid identification of high particle number (PN) emitting on-road vehicles. The method was validated for PN using a medium-duty vehicle and successfully applied to measurements of PN emissions from a large fleet of on-road diesel buses. The ratio of PN concentration to CO_2 concentration, Z, in the exhaust plume was estimated for individual buses. On the average, a bus emitted about 1.5×10^9 particles per mg of CO_2 emitted. A histogram of the number of buses as a function of Z showed, for the first time, that the PN emissions from diesel buses followed a gamma distribution, with most of the values within a narrow range and a few buses exhibiting relatively large values. It was estimated that roughly 10% and 50% of the PN emissions came from just 2% and 25% of the buses, respectively. A regression analysis showed that there was a positive correlation between Z and age of buses, with the slope of the best line being significantly different from zero. The mean Z value for the pre-Euro buses was significantly greater than each of the values for the Euro I and II buses.

Introduction

Over 80% of the particle number (PN) in urban ambient air occurs in the ultrafine size range (smaller than 100 nm) (*1*). Roadside measurements have shown that particles smaller than 10 nm account for more than 40% of the total PN concentration and that most of them originate from motor vehicle emissions (*2*). These particles contribute very little to the total mass; however, they have attracted increasing attention as they potentially pose a significant risk to human health (*3*). Because of their small size, ultrafine particles are able to pass through the upper respiratory tract, penetrating deep into the alveolar region in the lungs, aggravating conditions such as asthma and bronchitis, and they have even been linked to tissue damage and premature death (*4*, *5*). However, there are no current regulatory standards for motor vehicle PN emission factors. Present air quality and vehicle emission regulatory standards for particle matter are based on mass. A major reason for this deficiency is the difficulty in measuring PN emissions. A further complication arises due to the wide variations in PN emissions observed between vehicles operating under very similar conditions (*6*). The observed differences are in large measure due to the unpredictably high proportion of semivolatile nanoparticles (particles smaller than 50 nm) that are present in the exhaust gases (*7*, *8*). Nanoparticles are formed through a secondary particle production process and their concentrations are very sensitive to dilution conditions and exhaust cooling rates.

Thus, there is an urgent need to develop a more robust method for assessing PN concentrations in vehicle emissions. Furthermore, if such a testing method were capable of assessing the emissions from vehicles on-road, without removing them from service, its application would have a greater practical significance. Studies of emissions from individual vehicles in on-road fleets are important for several reasons, including inspection and maintenance procedures, policy decision making on the acquisition of new vehicles, and the introduction of new emission control strategies with a focus on the overall improvement of urban air quality. There are several methods for estimating PN emission factors from motor vehicles. However, all of them have major shortcomings in application to rapid testing of a large number of individual vehicles. Roadside measurements near busy motorways and traffic tunnels yield useful information but they are generally average estimates based on a number of assumptions on vehicle types, fuel, and operating conditions. Dynamometer measurements, although more controlled and accurate, are not appropriate due to the involved time and costs. An alternative method that has been used recently is remote sensing of vehicle emissions using IR and UV lasers. A narrow laser beam was directed across a single lane roadway to intercept the exhaust plumes of moving vehicles with the receiver positioned on the opposite side of the road (*9*). The method has been applied to measurements of carbon monoxide, carbon dioxide, nitric oxide, and gaseous hydrocarbons emissions. It has also been modified to quantify the particulate matter in vehicle exhaust by using the backscatter and transmission of an ultraviolet laser beam (*10*). However, this method depends upon a prior laboratory calibration as well as knowledge of the physical, chemical, and optical characteristics of the exhaust particles, giving rise to many uncertainties.

As such, remote sensing techniques have not been very effective in monitoring PN and mass emissions from vehicles travelling on the road, mainly because it requires the extraction of a portion of the exhaust plume for analysis by measuring instruments. This leads to an additional problem which is the uncertainty involved with dilution by ambient air. Depending on various parameters such as the wind speed and direction, vehicle speed, and turbulence, the extent of dilution at the point of measurement may vary widely.

Dynamometer studies have shown that PN emissions from vehicles of a given type operating under identical running conditions may vary by over an order of magnitude (*6*). However, the CO_2 emission factor is related to the fuel nature by stoichiometry. Therefore, it has been shown in a series of dynamometer studies that, at a given speed and load, the CO_2 emission factor of a vehicle is relatively stable (*6*, *11*). The amount of CO_2 emitted is directly related to the fuel burnt. The combustion of a liter of diesel fuel will yield about 2.5 kg of CO_2. The concentration of CO_2 at the vehicle tailpipe will depend on the exhaust air flow rate. Fuel consumption rates and exhaust air flows both increase with engine speed and load.

Consequently, it has been shown that, in estimating the emission factor of a vehicle, CO_2 may be used as a measure

of the dilution factor of the exhaust gases as it mixes with ambient air (*11*−*14*). The method has been applied to the measurement of soot emissions from passing vehicles (*13*, *14*) and shown to be applicable to PN emissions (*12*).

The main objectives of the present study were to (a) validate the above method of using CO_2 as a measure of the dilution factor of the exhaust gases as it mixes with ambient air in the monitoring of PN emissions from vehicles on the road, and (b) to demonstrate that the method has a particular application in identifying high PN emitting buses in a large fleet in the shortest possible time at minimal cost. The latter objective has an important significance because of its potential use in minimizing environmental pollution by identifying and possibly removing high PN emitting vehicles from the road for repair and correction.

Materials and Methods

Technique. The method uses CO_2 as a measure of the dilution factor of the exhaust plume to identify high polluting vehicles as they travel on the road. In this study, it was used to monitor PN emissions. A small sample of the exhaust plume was extracted at a point close to the edge of the road. PN and CO_2 concentrations were measured with fast-response instrumentation and recorded in real time. The concentrations contributed by each vehicle were corrected for background fluctuations by calculating the difference between the peak concentration soon after the passage of the vehicle and the background concentration just before the vehicle passed. In the analysis, the ratio of these two step heights, that is PN concentration to the CO_2 concentration contributed by the vehicle, was calculated and denoted *Z*. This ratio was not expected to vary with dilution as the exhaust plume mixed with ambient air and, so, the value of *Z* measured at any point within the plume may be equated to the value at the tailpipe. Thus, the value of *Z* measured in the exhaust plume of a vehicle provided a method for the identification of high PN emitting vehicles. It must be stressed that, using fast response monitors, the method would be equally applicable to emission parameters other than PN, particularly if they show large variability between vehicles.

Instrumentation. The PN concentration was measured with a TSI 3025A condensation particle counter (CPC). This is a fast-response continuous flow instrument that can detect airborne particles down to a size of 3 nm in total concentrations up to 10^5 cm^{-3}. The response time was 1.0 s for 95% response to concentration step change when sampling at a flow rate of 1.5 L min^{-1}. Particle count data were stored at 1.0 s intervals on an interfaced laptop computer. As the PN concentrations on and near busy roads often exceeded the maximum detectable level of the instrument (10^5 cm^{-3}), a HEPA filter was attached to a T-junction in the tube feeding the inlet of the CPC to dilute the sample below the maximum detectable limit. The flow rate through the filter was monitored with a TSI flowmeter and controlled with a pinch valve to provide a steady dilution factor of 10, that is, every unit volume of exhaust sample air was diluted with nine units of clean filtered air. The CPC readings (particles cm^{-3}) were then multiplied by 10 to obtain the actual sample PN concentrations. The CO_2 concentrations were measured with a Sable CA-10A analyzer. This instrument uses a dual wavelength infrared beam and has a response time of 1.0 s at a sampling rate of 1 L min^{-1}. Data were stored at 1.0 s intervals in real time. Wind speed and direction were also monitored in real time using an automatic portable weather station.

Validation of Method. The method was first validated using a Toyota Landcruiser 4.1 L passenger vehicle operating on 300 ppm sulfur diesel fuel. The tests were conducted on a straight, level track with a steady wind of about 3 m s^{-1} normal to the road. The instruments were placed at a distance of about 2 m from the road on the downwind side. The vehicle was repeatedly driven past in one direction at a fixed cruising speed of 40 km h^{-1}. The PN concentration (particles cm^{-3}) and CO_2 concentration (ppm by volume) were monitored in real time. For each traverse, each of these two parameters showed data spikes, from which the corresponding ratio, *Z*, was calculated in units of millions of particles per milligram of CO_2, assuming that 1% of CO_2 by volume equates to 17.7 g of CO_2 in 1 m^3 of air at the ambient temperature of 30 °C. The CO_2 concentration in the exhaust of the vehicle was measured directly with a CODA engine gas analyzer, which is a generic, portable, PC-based engine analysis system which includes a 5-gas exhaust analyzer and Windows based software for data logging and sensor testing of engine management systems. The corresponding value of *Z* was used to estimate the PN concentration at the tailpipe of the vehicle. This value was compared with the emission factor of the vehicle estimated under similar running conditions, using a bag-capture method (*15*).

TABLE 1. . Composition of the BCC Diesel Bus Fleet at the Time of the Study

bus type				
chassis	body	no. in service	in-service date	emission standard
MAN SL200	PMC	5	1982	pre-Euro
MAN SL200	Denning	70	1982−86	pre-Euro
Volvo B10M	Comeng	95	1987−89	pre-Euro
Volvo B10M	Volgren Articulated	7	1988	pre-Euro
Volvo B10M	QBB	60	1989−90	pre-Euro
Volvo B10M	Austral Articulated	6	1991	Euro I
Volvo B10M	Austral	133	1991−94	Euro I
Volvo B10M	Austral/ Denning	39	1994−95	Euro I
Volvo B10L	Austral/ Pacific	52	1997−2000	Euro II
Volvo B10L	Volgren	6	2000	Euro II
Volvo B6	Volgren	2	2000	Euro II
Merc Benz	Volgren	40	2003−04	Euro II

Application of Method. After validation, the method was applied to the detection of high PN emitting buses in the Brisbane City Council (BCC) fleet of diesel buses. Monitoring was carried out on 7 days between August and December 2005 at the Mater Hill Busway Station, yielding approximately 30 h of data. This station is located on the South East Busway, Brisbane, which was opened in September 2000 as the first of a series of busway networks to be developed in South East Queensland. A BCC transport bus passes the station per direction approximately every 50 s during the day. The BCC bus fleet consists of around 750 buses of which roughly 500 operate on diesel. In 2004, the entire diesel fleet was converted from low sulfur diesel (500 ppm) to ultralow sulfur diesel (50 ppm). Approximately 190 of these diesel buses operate through the Mater Hill Station each day, with each bus doing roughly 4−6 trips in each direction. Table 1 shows the composition of the BCC diesel bus fleet according to bus type, their corresponding number in service, and engine emission standard.

The instruments were placed on a trolley close to the curb of the departure end of the outbound platform. The busway at this point consisted of two lanes in each direction. The air samples were extracted from a point 0.6 m above the ground at the curb. Most buses passed within a distance of about 1.0 m from the sampling point. The inbound lanes, on the far side of the bus station, were at least 5 m away from the outbound platform, and separated by a central barrier. The passage times and registration numbers of buses were available on video footage from the permanent overhead

cameras on the platform. Specifications of individual buses, such as model, fuel type, and age, were obtained via the recorded registration numbers. Most of the buses stopped at the station and the small number that did not were required to slow down when passing. As the monitoring was carried out at the departure end of the platform, all buses were steadily accelerating past the sampling point. Thus, all the buses passed the monitoring point under the same approximate load. Following the arguments presented earlier, it was reasonable to assume that all buses had roughly the same CO_2 emission factor, and that the particle emission factor of a bus was directly proportional to the value of the ratio Z.

Results and Discussion

Validation Experiments. Figure 1 shows typical data spikes obtained over a 10-min period for (a) PN and (b) CO_2 concentrations, as the test vehicle was driven past the monitoring station. Each spike corresponds to a traverse of the vehicle. In each diagram, the spike close to time 11:37:00 was from a passenger car that passed by during the test period. The mean value of the Z ratio for the test vehicle was 1315 million mg^{-1} with a standard deviation of 280. The single value for the passenger car was 637 million mg^{-1}, which was clearly outside the range of values obtained with the test vehicle. The CO_2 concentration in the exhaust of the vehicle, measured directly with the engine gas analyzer was found to be 34 000 ppm by volume at a steady speed of 40 km h^{-1}. Using $Z = 1315$ million mg^{-1} gives a PN emission concentration of 5.2×10^7 cm^{-3}. The PN emission factor for this vehicle, measured using a bag-capture method, was found to be 1.5×10^{14} km^{-1} at 40 km h^{-1} (*13*). From this value and the measured exhaust air flow rate, the exhaust PN concentration was calculated and found to be 3.0×10^7 cm^{-3}. The reasonable agreement between these two values, together with the observation that the Z ratio, though highly variable between vehicles, was relatively stable for individual vehicles, provided confidence that the proposed method offered a viable technique for the identification of high PN emitting vehicles passing on a road.

Busway Study: Application of the Methodology. No discernible PN and CO_2 emission signals were recorded from buses in the inbound lanes behind the separating barrier, except for a few spurious readings that were neglected in the analysis. Both instruments recorded distinct signal spikes as each bus passed the sampling point on the outbound near lane, especially when the wind was blowing from the busway to the instruments. Smaller, less distinct signals were sometimes obtained when the wind was in the opposite direction and kept the exhaust plume away from the instruments. However, the extent to which the exhaust plume from a passing bus washed over the sampling point was not important as the validation experiments showed that the Z ratio was independent of dilution. In spite of this, there were several difficulties in acquiring consistent data from every outbound bus that passed. A small number of buses (approximately 1 in 10) passed on the outbound far lane, particularly when overtaking a stationery bus at the platform. Only a small fraction of these buses yielded measurable signals, with hardly any when the wind was blowing away from the instruments toward the busway. When a bus followed too close behind another, it was often difficult to discriminate between the two data spikes. If the time taken for the prevalent wind to dissipate the emissions from one bus before the following bus passed was too short, the reading from the second bus was affected by emissions from the first bus and had to be discarded. Owing to these reasons, useful data were obtained from around 30% of the buses that passed. However, as each bus made several traverses during the period of measurement, it was possible to obtain at least one reliable data set from every bus. Of just over 1000 bus passages through the outbound busway of the station during the period of observation, 317 passes yielded sufficiently clear and reliable data spikes as required for the analysis.

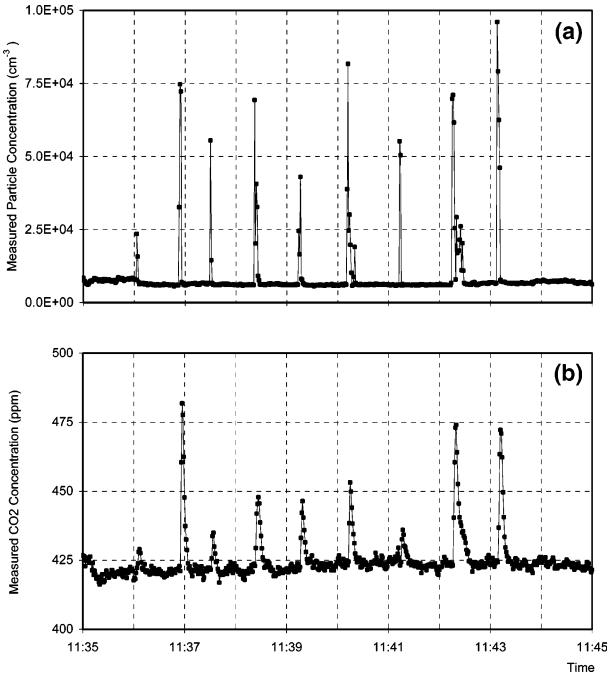

FIGURE 1. (a) PN and (b) CO_2 concentrations measured at a point 2 m downwind of the road along which the test vehicle was driven at a steady speed of 40 km h^{-1}. Each data spike is from a single traverse of the vehicle. The spike at 11:37:00 is from a passenger car that passed by during the study.

Passage times of buses were required in great accuracy for the purpose of differentiating between the buses. However, very often, buses clumped together and passed the sampling point in rapid succession, sometimes in both outbound lanes. Due to the unfavorable angles of the overhead video cameras, these were not very useful in determining the precise time when the tailpipe of each bus passed the sampling point, especially when a bus overtook another bus at that point. Therefore, passage times and registration numbers were also recorded manually using a stop watch to an accuracy of 1 s. This information, although tedious to obtain, proved to be much more useful in the analysis when differentiating between buses.

There are several ways of improving the rate of detection of bus emissions. Placing the measuring instruments downwind of the road is an obvious choice. Controlling the flow of buses with traffic-control devices, so that they do not pass too close to each other and are allowed to travel past the monitoring station in a consistent driving pattern, is highly recommended. Such options are not always convenient without disrupting normal traffic flow, but may be feasible in specific areas such as in or near bus depots.

Busway Study: Identification of High-Emitting Buses. Ambient PN concentrations on the busway platform during the period of observation ranged from 5×10^3 to 4×10^4 cm^{-3}, rising as high as 1×10^6 cm^{-3} for brief periods following passage of high-emitting buses. The corresponding concentrations of CO_2 ranged from 480 to 520 ppm by volume and peaked at nearly 1500 ppm due to some buses.

Figure 2 shows a typical set of usable data spikes due to the passage of three buses. The Z values for these three buses were 696, 1021, and 508 million mg^{-1} from left to right, respectively. Thus, they ranged over a factor of 2. For a given bus, the heights of spikes were determined by the wind direction while the widths of the spikes were chiefly

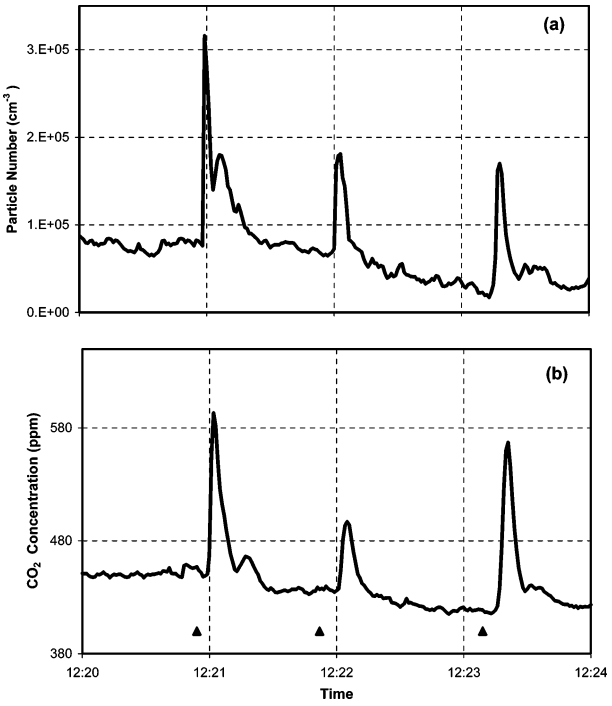

FIGURE 2. A typical set of (a) PN and (b) CO_2 concentration data spikes from three successive buses. The triangles show the passage times of the buses. The *Z* values for the buses were 696, 1021, and 508 million mg^{-1}, respectively, from left to right.

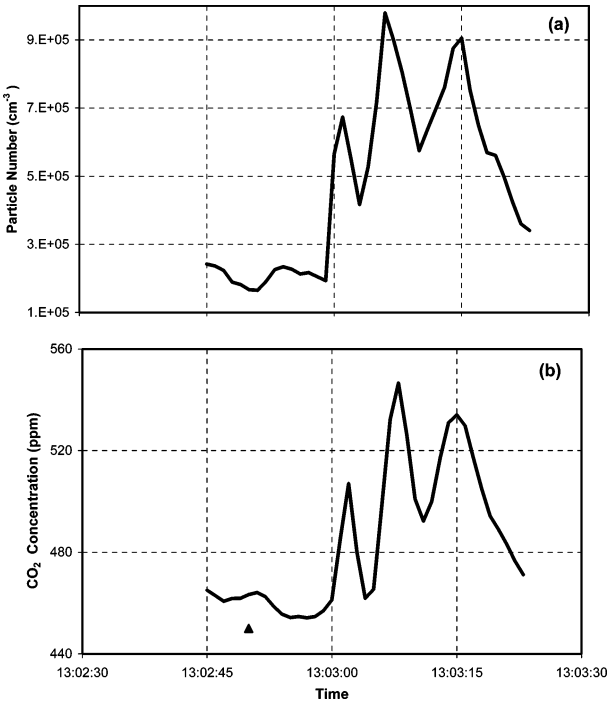

FIGURE 3. A typical example of multiple data spikes from the same bus. The triangle indicates the passage time of the bus. The *Z* values for the three spikes from left to right were 5216, 4828, and 5093 million mg^{-1}, respectively.

determined by the prevalent wind speed. The time resolution of the data is 1.0 s for both parameters. At times, more than one spike was observed from a single bus. Figure 3 shows such an example. These were due to multiple gusts of wind that moved the exhaust plume around. As expected, in contrast to spikes from different buses, these multiple spikes from the same bus generally yielded relatively similar *Z* values. For example, in this instance, *Z* values for the three spikes

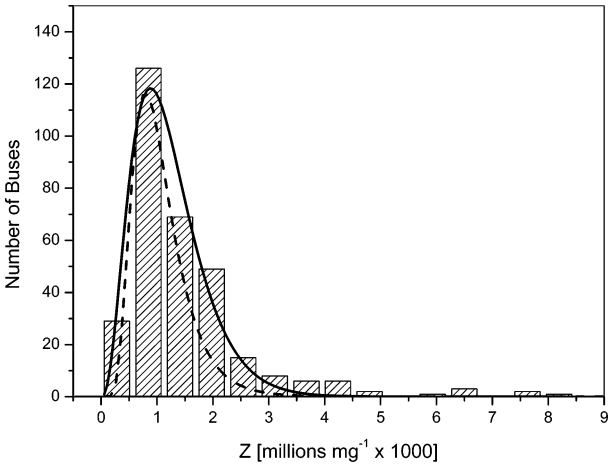

FIGURE 4. Histogram of the number of buses as a function of *Z*. The distribution is skewed and follows a gamma function, with the best fit curve ($R^2 = 0.97$) shown by the thick line. The broken line shows the lognormal curve fit ($R^2 = 0.89$). Note the small number of high emitters at the right.

from left to right were 5216, 4828, and 5093 million mg^{-1}, respectively. Coincidentally, this was one of the highest emitting buses.

Figure 4 shows the distribution of the number of buses as a function of *Z*. The median value of *Z* was 1150 million mg^{-1} with a standard error of 67. The mean value of *Z* was 1508 million mg^{-1}. Most of the buses showed *Z* ratios within a narrow range with a few buses exhibiting relatively large values. A statistical curve-fit showed that the distribution was not normal, being skewed with a long tail at the higher end of *Z*. It can be shown that the distribution follows a gamma function with a regression coefficient (R^2) of 0.97. This may be compared with the corresponding regression coefficient of 0.89 for a lognormal distribution. Previous studies have shown that emissions of carbon monoxide and hydrocarbons from motor vehicles are well represented by gamma distributions (*16*). Here, for the first time, using a large fleet of diesel buses, it is shown that PN emissions follow the same trend.

Since the *Z* value of a bus is a measure of its emission factor, using a cumulative function, it was estimated that roughly 10% of the PN emissions came from just 7 or 2% of the buses while roughly 50% of the emissions came from 79 or 25% of the buses. Bishop and Stedman (*9*) found a similar trend in the carbon monoxide and hydrocarbon emissions from passenger cars in California, and noted that one-half of the pollution was produced from less than 10% of the vehicles. The larger fraction of high-emitters is not unexpected as a significant number of passenger cars have badly tuned engines when compared to the regularly serviced buses in a well-maintained transport fleet. Bishop and Stedman also showed that, although most of the high emitters were older vehicles, this was not always true. Service history was found to be equally, if not more, important than age, with many new cars being identified as high emitters.

The last two columns in Table 1 show the date of service and the Euro emission standards of the bus engines in the BCC fleet. Figure 5 shows the median *Z* values of the buses as a function of age. Despite the large scatter, a regression analysis showed that there was a positive correlation between *Z* and age, with the slope of the best line being significantly different from zero at the 95% confidence level. Next, the buses were classified into four age groups, 5−9, 10−14, 15−19, and 20−24 years, and the median *Z* value for each group was calculated. The result is shown in Figure 6. Again, the *Z* value shows an increasing trend from the newest group of buses to the older groups. The error bars indicate the

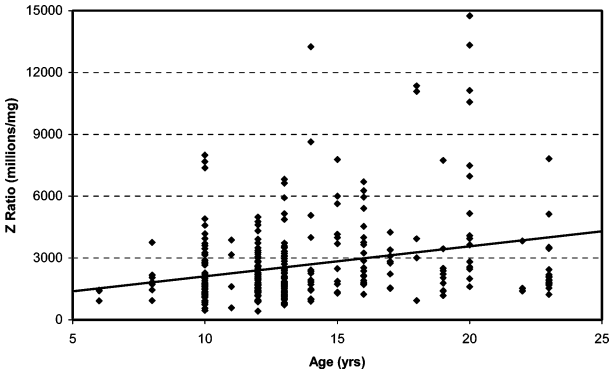

FIGURE 5. *Z* as a function of the age of the buses.

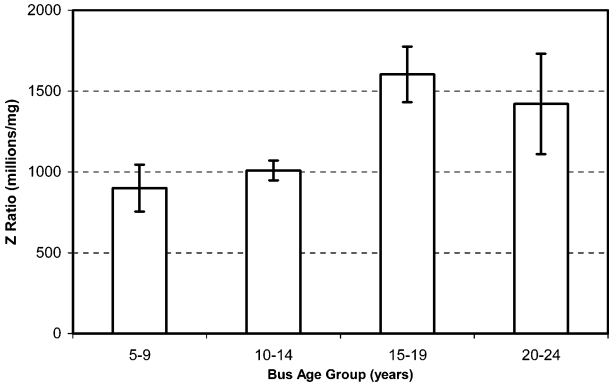

FIGURE 6. *Z* for the buses classified by age into four groups. The bars show the median values for each group with the error bars showing the respective standard errors. Statistically significant differences are observed between the first two groups and the last two groups.

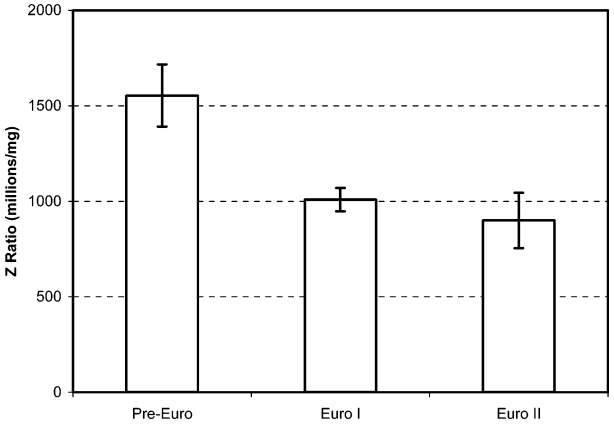

FIGURE 7. *Z* for the buses classified according to their emission standards into three groups, pre-Euro, Euro I, and Euro II, as shown in the last column in Table 1. The error bars show the respective standard errors. There is a statistically significant difference in the medians between the pre-Euro group and the other two groups.

respective standard errors. A *t*-test analysis showed that there was a statistically significant difference in *Z* between the two newest groups and the two oldest groups, with an increase of about 50%. Finally, the buses were classified according to their emission standards into three groups (pre-Euro, Euro I, and Euro II) as shown in the last column in Table 1 and the *Z* values for each group were calculated. The results are shown in Figure 7. The median *Z* value was greatest for the pre-Euro group (1554 million mg^{-1}) and lowest for the Euro II group (900 million mg^{-1}), with the error bars again indicating the standard errors. A *t*-test analysis showed that the value of *Z* for the pre-Euro group was significantly greater than the values for the other two groups at a confidence level of 95%.

Ristovski et al. (*6*) attempted to derive a correlation between the PN emission factor and age of vehicles in a sample of twelve diesel buses from this same fleet. However, they noted that the sample size was too small to show a statistically significant dependence. In the present study, using a large number of buses, it is shown for the first time that there is a significant increase of PN emission factor with age of buses. The *Z* ratio increased by roughly 75 million mg^{-1} per year of service and the dependence of *Z* on age was statistically significant at the 95% confidence level. There are several possible reasons for an increase of *Z* with age. Increasing wear and tear leads to deterioration in engine performance and a general reduction of efficiency. Moreover, recent years have seen the introduction of improved engine technologies to comply with more stringent emission standards. These factors have no doubt contributed to lower emissions.

It must, however, be stressed that, although most of the highest PN emitters were among the oldest buses, this was not always true. For example, Figure 5 shows three relatively new buses, all aged 10 years, within the top ten emitters detected. Bishop and Stedman (*9*) observed a similar trend among motor cars for gaseous pollutants. It is, thus, clear that other factors such as service history and proper engine tuning play an important role in determining PN emissions from buses. Although age is a quick and convenient measure of estimating emission factors of vehicles in a large fleet, it is not entirely satisfactory. The method is shown to be suitable for rapidly identifying and correcting high PN emitting vehicles in a large fleet.

Acknowledgments

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