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Characterization of On-Road Vehicle NO Emissions by a TILDAS Remote Sensor

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ABSTRACT

A tunable infrared laser differential absorption spectrometer (TILDAS) was used to remotely sense the nitric oxide (NO) emissions from 1,473 on-road vehicles. The real-world measurement precision of this instrument in the limit of low NO concentration is 5 ppm of the vehicle exhaust, which corresponds to a 3σ detection limit of 15 ppm. Our analysis of the distribution of negative concentration measurements produced during this experiment supports this claim, showing that the instrumental noise for this set of measurements was at most 8 ppm in the limit of low NO concentration. The high sensitivity of this instrument allowed us to measure the NO emissions of even the cleanest vehicles. The measured vehicle fleet NO emissions closely fit a gamma distribution with 10% of the fleet contributing about 50% of the total fleet emissions. Newer vehicles had lower NO emissions than older ones, but high NO emitters were found in every vehicle age cohort. On a vehicle-by-vehicle basis, NO emissions correlated very weakly with vehicle velocity, acceleration, power per unit mass, carbon monoxide (CO) emissions, and hydrocarbon (HC) emissions. High NO emitting vehicles could not be identified by remote sensing of CO or HC emissions and vice versa. When we compared the NO emissions for 117 vehicles measured more than one time, about half of the high NO emitters were found to be very consistent, while the other half varied significantly.

IMPLICATIONS

On-road motor vehicles are the largest source of NO_x in the United States. NO_x interacts with hydrocarbons in the presence of sunlight to form tropospheric ozone, one of our most serious and persistent air quality problems. This manuscript describes the application of a highly sensitive technique for measuring the NO_x emissions of on-road vehicles while they are in use on the roadway. Using this technique, it is now possible to precisely measure the instantaneous NO emissions of even the cleanest vehicles. This allows the NO_x emissions of individual vehicles to be evaluated efficiently and unobtrusively. This method can also be used to characterize the distribution of NO_x emissions within a vehicle fleet and to monitor the effectiveness of automobile inspection and maintenance programs.

INTRODUCTION

Nitric oxide (NO) and nitrogen dioxide (NO_2), collectively referred to as NO_x, are important air pollutants. NO_x interacts with hydrocarbons in the presence of sunlight to form tropospheric ozone,[1] one of our most serious air quality problems. NO_x can also form nitric and nitrous acids that contribute to acid deposition.[1] Other undesirable effects include visibility degradation (through particulate nitrates), lung damage (by NO_2), and plant damage.[1]

On-road motor vehicles are the largest source of NO_x in the United States, contributing 35% of total NO_x emissions.[2] With high traffic density in the Los Angeles area, this figure goes up to 57%.[3] Despite the dominance of motor vehicles among NO_x sources, considerable uncertainty persists in the understanding of this emissions source,[3] and better quantification and control of on-road NO_x emissions are needed.

Remote-sensing instruments monitor individual vehicle pollutant emissions by referencing a measurement of the pollutant's concentration to the carbon dioxide (CO_2) concentration in the vehicle exhaust. These measurements are made simultaneously and immediately behind the moving vehicle and, thus, have the potential to provide the best possible information on actual in-use vehicle emission profiles. Remote sensing can also be used as a way to identify high emitters to be directed to inspection and maintenance (I/M) facilities for testing, or to identify low emitters for exemption from I/M testing. These actions can lead to more acceptable I/M programs and more cost-effective improvements in air quality.[4-5]

More than 95% of the NO_x released by light-duty gasoline vehicles is released in the form of NO.[6] Remote sensing of NO from automobiles has been traditionally accomplished by nondispersive infrared absorption[7] and ultraviolet absorption methods.[8] Unfortunately, neither of these methods has been shown to be sensitive enough in the field to measure the full range of vehicle emissions. Only the highest emitters can be reliably identified with these instruments, due to their low sensitivity. However, an improved dispersive UV instrument with a noise level of 20 ppm for NO under parking lot conditions has been presented.[9]

Tunable infrared laser differential absorption spectroscopy (TILDAS) can measure the NO emissions of even the cleanest on-road vehicles, as it is at least 50 times more sensitive than the two demonstrated nondispersive measurement techniques and 4 times more sensitive than the dispersive UV technique.[10-11] This high sensitivity will likely become increasingly important as automobile emissions standards become stricter. Because this method has high spectral resolution, it is inherently interference free. Finally, the long path-length capability of the TILDAS technique makes it particularly well suited for on-road vehicle emissions measurements.

This paper presents the results of a study in which the NO emissions of more than 1,400 vehicles were measured using a TILDAS instrument developed by Aerodyne Research, Inc. The main objectives of this project were to demonstrate the ability to accurately measure NO tailpipe emissions of on-road vehicles and to characterize these emissions based on field data obtained from a fleet of vehicles in the Los Angeles area.

FIELD EXPERIMENT

The field experiment was performed in El Segundo, CA, from November 13 through 22, 1996. During this period the TILDAS instrument obtained NO/CO_2 emission ratio profiles for 1,473 passing vehicles. The spectral region for CO_2 was chosen to include nitrous oxide (N_2O) absorption features, so that N_2O emission data were obtained coincident with the NO emission data. The N_2O emission results have been reported elsewhere.[12]

To provide a context for the interpretation of vehicle NO emissions, the NO measurements were supplemented with simultaneous measurements of vehicle velocity, acceleration, and CO and hydrocarbon (HC) emissions. We used a video camcorder to record images of the vehicle license plate and a custom-built computer-based optical system to measure and record vehicle velocity and acceleration data. Vehicle make, model year, and identification number (VIN) were retrieved from the California Department of Motor Vehicles (DMV) from the recorded license plate data. CO and HC emissions data were provided by "Smog Dog" instrumentation through a collaboration with Hughes Santa Barbara Research Center (SBRC).[7]

Experimental Site Description

The measurements were performed at Hughes Way in El Segundo, an urban street with a 4% uphill grade and a central divider. Hughes Way is an access road to a large business facility. A traffic light was located about 100 m before the remote sensing site. Some of the vehicles measured had stopped at this traffic light, but others had not. El Segundo has a population of about 15,000 people, while about 150,000 people work in the city. Since most of the vehicles we measured were those of commuters coming to work from other cities, we presumed that they were fully warmed up.

We employed our two-laser TILDAS apparatus to simultaneously measure NO, N_2O, and CO_2 column densities with high detection sensitivity and high temporal resolution. The heart of the TILDAS instrument is a pair of infrared diode lasers, whose frequency of emission can be varied by changing the electrical current through each diode. The laser frequency was tuned across known absorption lines of NO, CO_2, and N_2O, and the strength of the absorption was related to the column densities of the species in the absorption path. The precision of the instrument for low NO emitters was estimated to be about 3 ppm of the vehicle exhaust for optimal overlap of the laser with a substantial plume. This translates to an ideal 3σ detection limit of 9 ppm. In practice, measurements were accepted even if the CO_2 column density was as small as 10% of the ideal case. On average, this increased the noise in the system by almost a factor of 2, from 3 ppm to 5 ppm. The TILDAS instrument used in this study is described in detail elsewhere.[10-11]

Assuming stoichiometric combustion of gasoline and a negligible amount of CO in the exhaust, one can determine the NO concentration in the vehicle exhaust from the NO and CO_2 column densities (in parts-per-million meter or ppm-m) as follows:

$$[NO] = \frac{NO\ (ppm - meter)}{CO_2\ (ppm - meter)}\ [CO_2]_s \qquad (1)$$

where $[CO_2]_s$ is the concentration of carbon dioxide produced in the stoichiometric combustion of gasoline. If the CO and VOC concentrations are also measured, they can be used to correct the NO measurement for carbon present in these incomplete combustion products.

Note that all of our emission data are reported as parts per million (ppm) of all the exhaust gases (including H_2O), and not as ppm of the dry exhaust. The NO values in ppm of the dry exhaust would be 14.3% larger than the values presented here for stoichiometric combustion.

The experimental setup is shown in Figure 1. The TILDAS instrument was inside a truck parked in the rightmost lane behind the traffic cones. The combined laser beams from the TILDAS instrument were sent to a turning mirror and then to a corner cube retroreflector. The total optical path length (to the retroreflector and back) of the TILDAS laser was 36 meters. Set up on each side of the TILDAS optical path were one of a pair of velocity and acceleration measurement instruments and one of a pair of Hughes "Smog Dog" remote sensors for CO and HC measurement.[7] Because of the range limitation of the Smog Dog instrument, the three-lane roadway was "coned down" to one lane of traffic.

Vehicle Velocity and Acceleration Measurement

A low-cost, laser diode-based-instrument was developed by EMDOT Corporation to provide an accurate means of measuring vehicle velocity and acceleration that would be compatible with the remote-sensing field environment. This instrument timed with microsecond accuracy the interruption of two light beams directed perpendicularly across the vehicle path from two roadside sources onto two photo detectors on the opposite side of the road. The two stations were separated by about 10 m. The two-station setup provided the information needed to calculate the velocity and a constant rate of acceleration for the vehicle between the two stations.

The average velocity of the vehicles with valid NO emission measurements was 13.9 m/sec, with a standard deviation of 2.6 m/sec. The average acceleration was 0.59 m/sec^2, with a standard deviation of 0.49 m/sec^2. Two-thirds of this average was due to the 4% road grade, which contributes 0.4 m/sec^2, since the forces due to acceleration and grade are indistinguishable.

The precision of these measurements was estimated from the calculated length of those vehicles that were measured more than once. The precision of the velocity and acceleration data were estimated to be 0.033 m/sec and 0.05 m/sec^2, respectively. This represents a fractional accuracy of 0.2% of the range of velocities and 1.3% of the range of accelerations observed in this study.

CO and HC Remote Sensing

As mentioned above, during our two days of measurements at El Segundo we set up our instruments side-by-side with two nondispersive infrared (NDIR) Smog Dog remote sensors from Hughes SBRC.[7] Each of the Hughes remote sensors measured CO and HC emissions simultaneously with our NO emission measurement. The two Hughes instruments were located 0.5 m before and after the TILDAS laser path, as indicated in Figure 1.

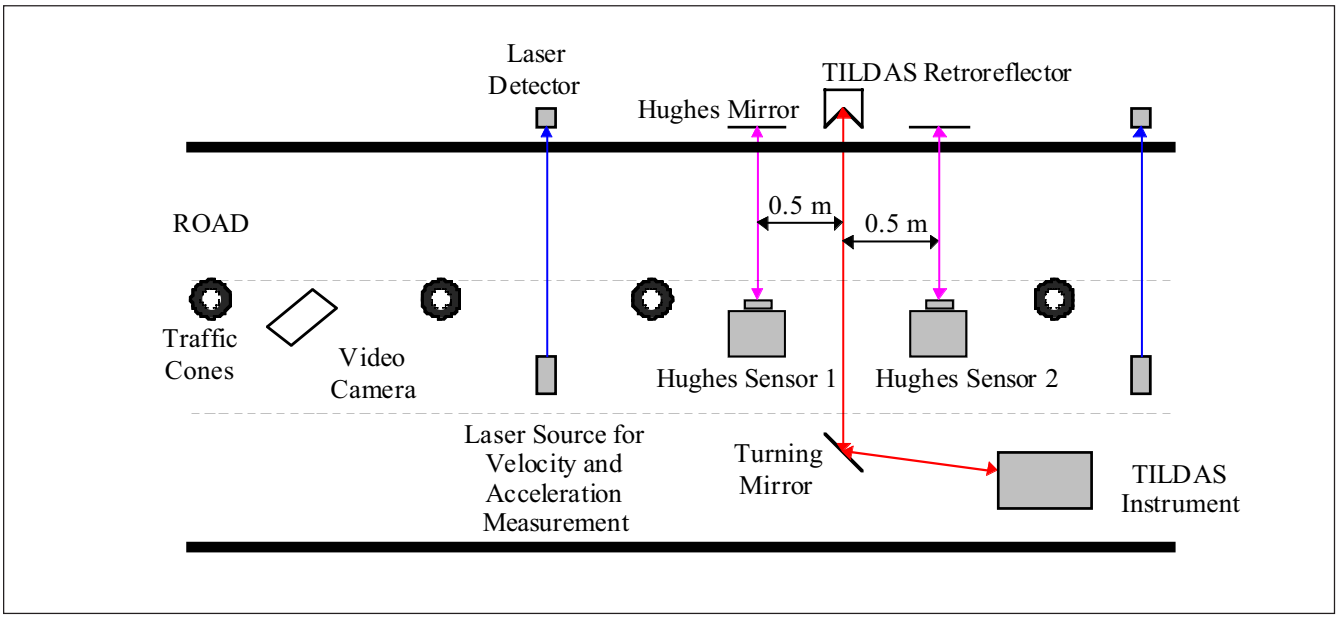

Figure 1. Field instrumentation arrangement.

RESULTS AND DISCUSSION

NO Emissions Distribution

NO emissions measurements with adequate plume-laser overlap were taken from 1528 vehicles at the Hughes Way site. Post analysis of these data sets yielded 1,473 valid data points. Only 3.5% of the initially accepted measurements were discarded as invalid, typically due to suspected interference from the exhaust of a previous vehicle. The mean NO emission was 321 ppm with a standard deviation of 525 ppm. The median value was 90 ppm and the maximum was 3,753 ppm.

The distribution of the NO emissions is presented in Figure 2. It is very skewed, with 10.5% of the vehicles, those emitting more than 938 ppm, responsible for 50% of the NO emissions. This is similar to the distribution found previously for CO and HC emissions[13] and also for NO emissions using a less sensitive remote sensor.[8] Remote sensing studies have typically found automobile CO and HC emissions to follow a gamma statistical distribution function.[13] A previous study also concluded that this is true for NO emissions.[8] In order to assess this point, Figure 2 also shows a gamma distribution of the same mean and standard deviation as the experimental distribution. The agreement between the two distributions is very good. A gamma distribution signifies that "dirty" vehicles are different from "clean" vehicles, with a few "broken" vehicles contributing disproportionately to the total emissions. This is distinct from a normal statistical distribution, in which the emissions of individual vehicles would cluster about the mean, and the contribution of dirty vehicles to the total would be smaller. The gamma distribution suggests the feasibility of reducing vehicle emissions by intervening with a selective I/M program.

The high sensitivity of the TILDAS instrument is clearly demonstrated by the distribution of negative concentration readings produced by the instrument. Since negative emissions are physically impossible, their magnitude provides an estimate of the instrument's precision. An instrument with a precision of 300 ppm will produce many negative readings with magnitudes of order 300 ppm. In contrast, the data obtained with the TILDAS instrument show a distribution of negative concentration measurements with a standard deviation of only ~8 ppm. Figure 3 shows a detailed histogram of the lowest concentration bin in Figure 2 spanning the NO concentration range from –50 to 50 ppm. We have modeled our data as the numerical convolution of the measured gamma distribution with a normal distribution having a standard deviation of 8 ppm. The real distribution is well approximated by this convolved distribution, from which we infer that the precision of the measurements obtained during this study was about 8 ppm of NO in the vehicle exhaust. The expected instrumental precision[11] was 5 ppm. The slight difference between these two noise estimates may be due, at least in part, to variations in the NO background due to previous vehicles.

We observed that the NO emissions of most of the vehicles were low relative to the average NO emissions permitted under existing NO standards. For example, 51% of the vehicles emitted less than 85 ppm. If a vehicle produced an average of 85 ppm over the certification test driving cycle, it would satisfy the California ultra low emission vehicle (ULEV) standard at 50,000 miles (assuming a fuel economy of 30 mpg). This result may be due to the fact that most vehicles were measured in a fully warmed-up, mild-power condition.

A decile plot of the NO emissions distribution is presented in Figure 4. The first decile represents the fraction of the total NO produced by the 10% dirtiest vehicles, the second decile by the next 10% dirtiest vehicles, and so on. It is easier to appreciate in this graph how clean most

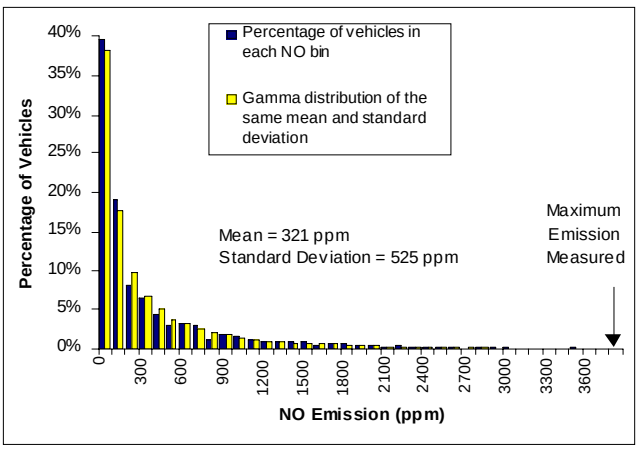

Figure 2. Experimental NO emissions distribution and the gamma distribution of the same mean and standard deviation.

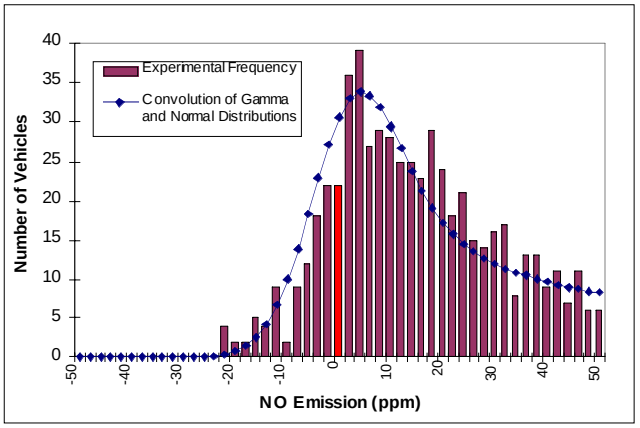

Figure 3. Comparison of the frequency of NO emission readings from –50 to +50 ppm; and the convolution of a normal distribution of standard deviation 8 ppm to the experimentally determined gamma distribution.

of the vehicles were; that is, the cleanest 50% of the vehicles were responsible for only 4.2% of the total NO emissions.

Correlation between NO Emissions and CO/HC Emissions

The NO, CO, and HC emissions of 822 vehicles were measured simultaneously. There was little correlation between NO and CO emissions or between NO and HC emissions on a vehicle-by-vehicle basis. A regression analysis returned very small correlation coefficients in both cases (R^2=0.001 for NO to CO; 0.007 for NO to HC). This means that one cannot predict the NO remote sensing emission level of a vehicle from the CO or the HC remote sensing emissions of the same vehicle, and vice-versa.

Figure 5 shows the NO and CO binned by rank-ordered CO emission vehicle population deciles. For example, the first point represents the average NO emissions of the 10% of the vehicles with the lowest CO emissions, plotted against the average CO emissions of those vehicles. The next point contains the average CO and NO for the next 10% cleaner vehicles for CO, and so on. For the 80% of the vehicles with the lowest CO emissions, there was a weak positive correlation between the binned emissions of both pollutants. This is the opposite of what was found in the previous study.[8] For the 20% of vehicles with the highest CO emissions, CO and NO appear to be weakly anticorrelated.

A plausible explanation for these trends is that cleaner vehicles have a totally or partially functioning emissions control system; and as the performance of the system degrades, it does so for both CO and NO, and we would expect them to be positively correlated. The dirtiest vehicles tended to have a completely inoperative control system. On these vehicles we see the well-known inverse relationship between untreated engine NO and CO emissions as a function of combustion stoichiometry.[6]

Figure 6 presents the NO versus HC emissions, binned by HC emissions rank-ordered vehicle population deciles. The trends are similar to those of the NO versus CO graph, that is, increasing NO with increasing HC for the vehicles with lower HC, and decreasing NO with increasing HC for the 20% or so vehicles with the highest HC emissions. These trends can be qualitatively explained by the same mechanism described above.

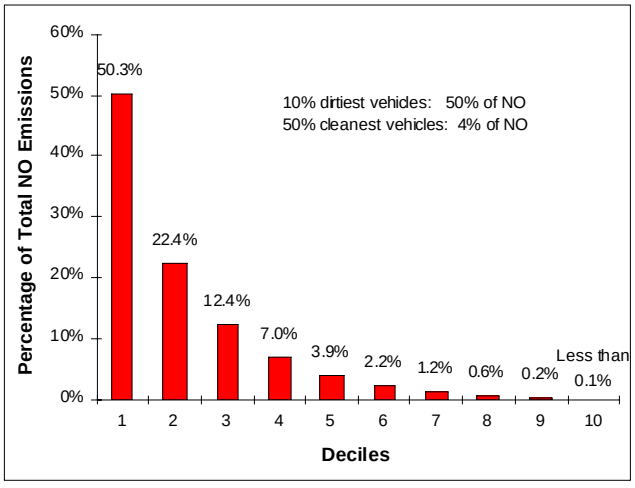

Figure 4. Decile plot of NO emissions in El Segundo.

Effect of Velocity and Specific Power on NO Emissions

We measured velocity and acceleration together with NO emissions for 867 vehicles. There was little correlation between NO emissions and vehicle velocity (R^2=0.004). There was not a clear trend of NO emission with speed for the binned population deciles data either. This result may be expected since the load on the engine coming from rolling resistance and aero-

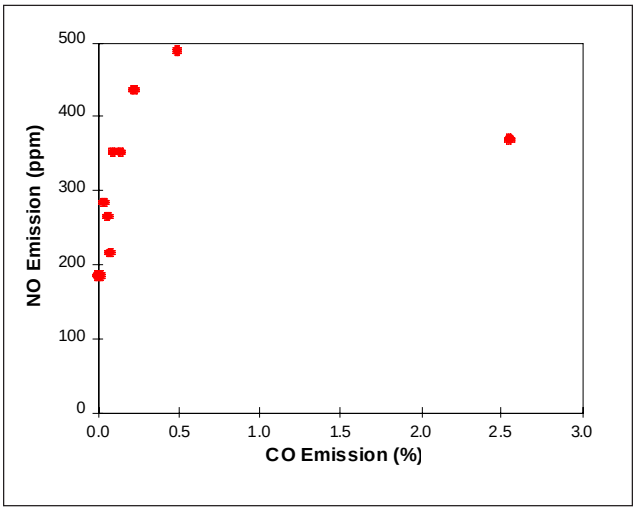

Figure 5. Average NO vs. CO emissions for vehicle population deciles.

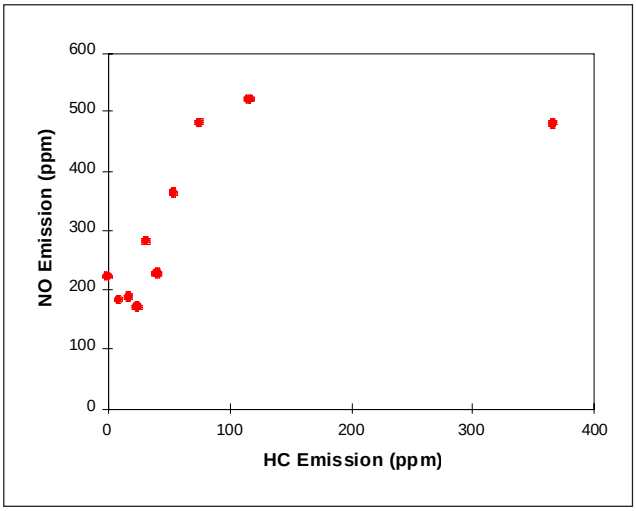

Figure 6. Average NO vs. HC emissions for vehicle population deciles.

dynamic drag, which are functions of velocity, was only a small fraction of the total load for most of the vehicles whose emissions we measured.

We present here the effect of another parameter, vehicle-specific power, on NO emissions. It was chosen instead of vehicle acceleration because it is a more accurate measure of engine load. Specific power is defined here as the product of velocity and the sum of acceleration and roadway grade.

$$\text{Specific Power} = \text{Velocity} * (\text{Acceleration} + \text{Grade} * g) \quad (2)$$

where *Grade* = vertical rise/slope length, and g = acceleration of gravity.

Specific power expresses the power per unit mass of the vehicle required to overcome acceleration and grade forces on the vehicle. Since maximum engine power generally scales with vehicle weight, specific power is also a surrogate of the load on the engine.

The average specific power calculated for the vehicles in this experiment was 8.4 ± 7.4 kW/metric ton. The range of values calculated was -6.5 to 59.5 kW/metric ton.

As with other parameters, the correlation between NO emissions and specific power was very small on a vehicle-by-vehicle basis (R^2 = 0.023). Figure 7 shows the average NO emissions of each vehicle population decile rank-ordered by specific power. There was a strong correlation between NO emissions and specific power for the lower nine deciles, which we attribute to the expected increase of NO emissions with engine load.[6] The 10th decile seems to show the reduction of NO emissions due to the power enrichment of some vehicles that were operating at the highest powers.

Effect of Model Year on Emissions

The correlation between NO emissions and model year (or vehicle age) is small on a vehicle-by-vehicle basis (R^2 = 0.14). Looking at any single model year, one can see a very wide variation in emissions values, typically spanning almost three orders of magnitude. High NO emitters were found in every vehicle model year.

The average NO emissions for vehicles grouped by model year shows an almost monotonic increase with vehicle age, as shown in Figure 8. Due to the small sample size, data on vehicles built prior to 1983 are not considered reliable for a single model year. However, the aggregated NO emission data for all vehicles prior to 1983 does show these vehicles to be higher NO emitters than subsequent models. Our data do not seem to conform to the detailed structure of the NO versus age curve described by Zhang et al.,[8] though a larger sample than that obtained in this study would be needed to ascertain this point.

One of the features of this study was the simultaneous recording of NO, CO, and HC emissions for a statistically significant and reasonably unbiased population. All emission levels increased with vehicle age. This relationship undoubtedly underlies the portion of positive correlation between NO/CO and NO/HC discussed above. In Figure 9, the total emission contributions, the product of the average model year emission level, and the number of vehicles of that model year are shown. The graph shows that for NO, CO, and HC the total contribution of each model year was remarkably similar. Vehicles about 10 years old produced the greatest proportion of the emissions for all three pollutants.

NO Emissions Variability

NO emissions variability for individual vehicles is an important issue since a high variability will make a single remote sensing measurement less meaningful. The data from the present study provide an opportunity to analyze

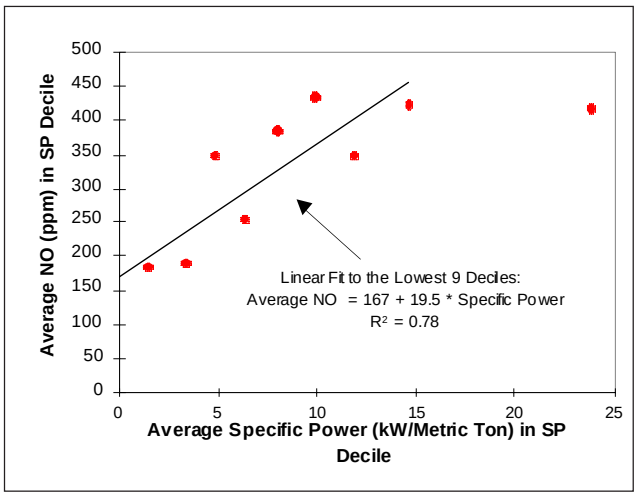

Figure 7. Average NO emission versus average specific power by population decile.

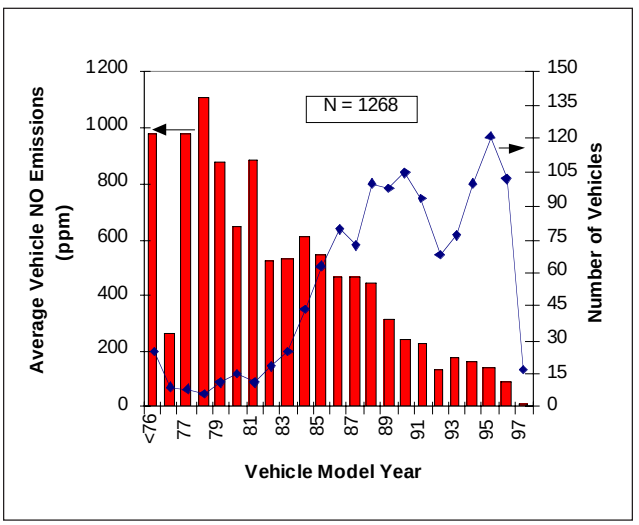

Figure 8. Average NO emissions versus vehicle model year.

this issue, as the NO emissions from 117 vehicles were measured more than once. These data are analyzed in Figure 10, which shows the first two measurements for each vehicle measured more than once plotted versus the average value of the measurements. Most of the vehicles were consistently low emitters. About half of the vehicles with high NO emissions had high variability, while the other half had markedly less variability. Thus, some of the high emitters were very consistent, while others ("flippers") were more variable. This is similar to what has been observed previously for CO and HC emissions.[14]

Practically all of the measured NO emission variability can be attributed to the vehicles and not to the TILDAS instrument, since the variability of most measurements was far larger than the TILDAS measurement precision.

Comparison with CO Emissions Variability

The variability of CO emissions was studied using the CO emissions data collected on the vehicles that were measured two or more times. It was found that CO relative variability, defined as the difference between the two measurements divided by their average value, was larger than the NO variability during these experiments. Given that CO remote sensing is generally accepted as a valid means of determining the CO emission level of a vehicle, this result implies that NO remote sensing, performed under appropriate conditions, should be accepted as well.

CONCLUSIONS

This study yielded a number of significant new findings with respect to the emissions of on-road vehicles and the utility of remote-sensing emissions measurements. The TILDAS instrument consistently and accurately measured the NO emissions of on-road vehicles. The NO emissions of low emitters were measured with a precision of approximately 5 ppm. In addition, the TILDAS instrument achieved high measurement accuracy with no need for on-site calibrations, while competing techniques require frequent field calibration to resist inherent drifts. Another major advantage of the TILDAS technique is that it is not as limited in range as the competing instruments. This eliminates the need for constricting the traffic to a single lane, which modifies driving behavior and vehicle emissions. For example, a path length of 36 m was used in this study, compared to 8 m for the Hughes NDIR instrument. A further extension of this path length can be achieved with only minor degradation of sensitivity.

The measured vehicle fleet NO emissions closely fit a gamma distribution with the 10% of the fleet having the highest emissions contributing about 50% of the total fleet emissions. The data clearly show that newer vehicles have lower NO emissions than older ones but also supports the conclusion that high NO emitters are found in every vehicle age cohort. Analysis of the NO data with respect to the other simultaneously recorded data for the subject vehicles shows that on a vehicle-by-vehicle basis NO emissions correlate only weakly with vehicle velocity, specific power, CO emissions, and HC emissions. When these same data are aggregated into rank-ordered population deciles, the positive correlation between decile-averaged NO emissions and the decile-averaged values of CO, HC, and specific power for most of the vehicles can be seen more clearly. The data show quite conclusively that high NO emitting vehicles cannot be identified by remote sensing of CO or HC emissions and vice versa.

When we compared the remotely sensed NO emissions measurements for 117 vehicles measured more than one time, most of them were consistently low emitters. About half of the high NO emitters were found to be consistently high, while the other half had significant variability.

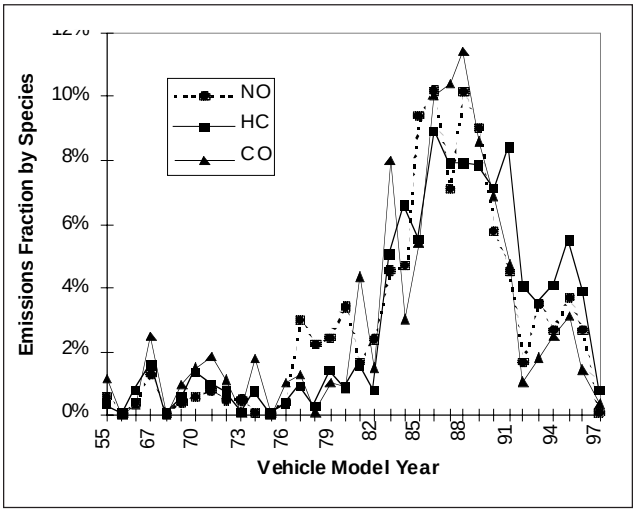

Figure 9. Total NO, HC, and CO emissions by model year.

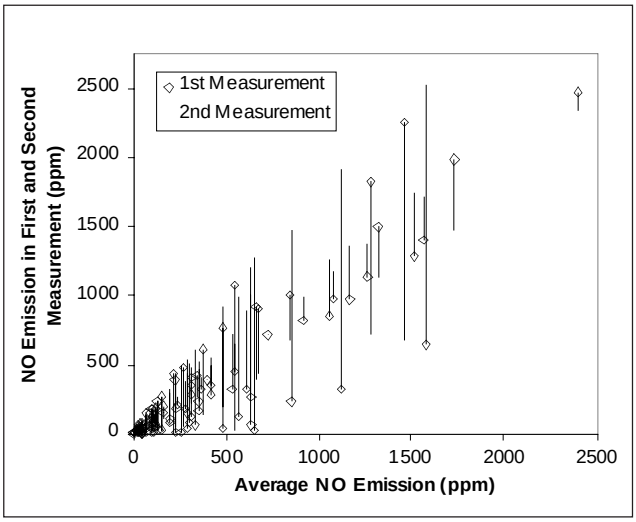

Figure 10. NO emissions in the first and second passes versus the average emissions for vehicles measured more than once.

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