# Opacity Enhancement of the On-Road Remote Sensor for HC, CO and NO

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## Introduction

Some components of diesel soot have recently been declared by the California Air Resources Board to be human carcinogens. The USEPA has tightened the National Air Quality Criteria for Airborne Fine Particulate Matter. There is an interesting dilemma in mobile source inventories resulting from the Northern Front Range Air Quality Study (Lawson and Smith 1998, Cadle et al, 1999a and b). and similar studies in the Los Angeles (LA) Basin (Rogge et al , 1993). Using chemical mass balance (CMB) analyses, including in Denver $^{14}C$ "age" determinations, the relative contribution of diesel and gasoline powered primary $PM_{2.5}$ emissions is about 25%:75%, respectively. The Denver inventory prepared by CDPHE for the State Implementation Plan using MOBILE modeling, and inventories derived in LA using CMB both arrive at about the opposite conclusion, namely 75% diesel, 25% gasoline. This disagreement is so important that an atmospheric chemist from Battelle was retained carefully to review both studies (Kelly et al, 2000). He reported that both studies appeared to have been done correctly and no fatal flaw in either was apparent to him. However, the difference remains and is very important to future decision making for particle pollutant reduction.

For these reasons, improved on-road measurements of particulate emissions from motor vehicles are an important goal. In the past we have used IR extinction at 3.9μm to measure opacity (Stedman et al, 1997). This wavelength suffers from the problem that particles have no strong spectral features, and are not strong scatterers in the IR wavelengths. To improve the system a visible laser (He/Ne) was added to improve the capability to directly measure opacity. The advantage of a visible laser wavelength is the approximately $1/\lambda$ increased scattering coefficient (Hidy, 1984). Also added was an opacity determination from the available 240nm UV data (Popp et al, 1999). Software modifications were needed as well as hardware because prior software intentionally rejected the most opaque plumes as having too much opacity for the reference correction to be considered valid. The proposed system also has the laser beam going twice across the road, doubling the optical path, and thus doubling the absorption over the IR and UV beams.

The ability to accurately monitor HDD emissions for NO has recently been demonstrated (Morris et al, 1999, and Morris' M.Sc. thesis from the University of Denver in 1999). One of the more important results of this work, the correlation between average opacity and average CO emissions, is shown as Figure 1. The apparent altitude effect on on-road HD diesel NO emissions was presented at the 10th CRC On-road Emissions Workshop by Gary Bishop, (2000) and was published by Bishop et al, (2001) after the reviewers had recovered from their reaction to our ugly truck picture.

Fine particle emissions from gasoline and diesel vehicles are a very complex issue. For diesel vehicles, the emissions are a strong function of load (Heywood, 1988) while for gasoline powered vehicles the situation is more complex yet. A recent Colorado roadside study identified only 0.16% of on-road gasoline powered vehicles as visible smoke emitters (Barrett and MacRae, 1997), while a more comprehensive CE-CERT study (Norbeck et al. 1996) showed high smoke

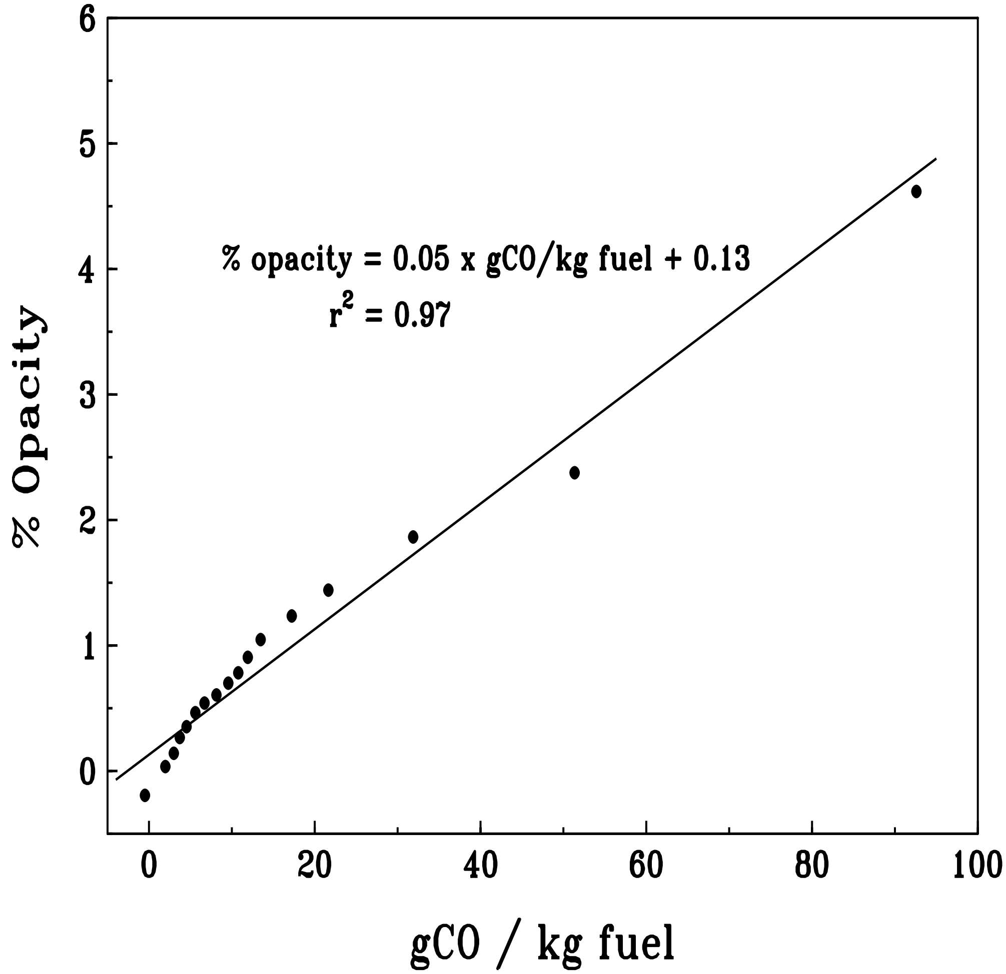


**Figure 1**. A correlation of the average % opacity and g CO/kg fuel of heavy-duty diesel trucks binned by opacity in the Austin-San Marcos Texas area. $r^2 = 0.97$.

emissions from different vehicles under different load conditions. In all cases high emissions were from a very small fraction of the fleet. The fact that a very few vehicles dominate the particle emissions from gasoline powered vehicles is a strong justification for an improved vehicle particle emissions detector, because remote sensing can observe very large fleets of vehicles in a short time (10,000 vehicles per day is not unusual for automobiles, Stedman et al, 1997).

Chen et al (1996) have shown that remote sensing of particle emissions from diesel vehicles with visible light opacity meters is entirely possible and furthermore state that:

"..the relative extinction caused by soot and $CO_2$ did give a reliable indication of the specific soot emission index for a wide range of operation conditions"

Their system measured $CO_2$ and soot only and did not have NO, HC or speed/acceleration capability.

There is no real certainty as to what should be measured in the case of particle emissions. The observed health effects may be correlated to at least six parameters including mass, mass in a given size range, particle surface area, number, composition and/or morphology. Any single measured parameter can not be expected to correlate with a multidimensional set of parameters as complex as the above. For instance; health effects may be related to particle mass which in turn is proportional to $r^3$. However surface area, proportional to $r^2$ may be the main factor. It may even be that some health effects outcomes are not caused by the same particle components as others.

Government standards are related to particle mass $\sim r^3$. This arises partly because mass is relatively easy to measure and calibrate. Visibility is related to particle area $\sim r^2$, through the processes of scattering and absorption. Climate effects (by cloud nucleation) are related to particle number $\sim r^1$

In Colorado where there is a State visibility standard, visibility reduction is also a reason to monitor PM emissions from vehicles. In this case the opacity measurements described herein may (or may not, I do not believe we know at this stage in the science) be more closely correlated to the desired indicator than would be mass or number measurements. In all cases there will be a rough correlation, in that larger particle numbers are related to larger surface area and larger mass, BUT all correlations are rough. For instance, McCormick et al, 2001 studied a sample of HD Diesel trucks on a chassis dynamometer. The particle mass emissions were more closely correlated to the CO emissions (easily measurable by means of remote sensing) than to the results of the standard J1667 opacity I/M test.

Kittleson, (1999) suggested that in view of the non-linear coagulation/condensation of nanoparticles, diesel particle emissions sampled by means of a dilution tunnel would not be the same as those measured on-road, or measured in a wind tunnel, wherein realistic dilution rates can be observed. Consistent with this suggestion, Maricq et al (2002) have shown in a series of wind tunnel experiments that the nanoparticles from gasoline fueled engines peak in their number distribution around 50nm while those from diesels peak under 100nm. This is also consistent with results from the Caldecott Tunnel by Allen et al (2001) who show the mass distributions peaking at 150 nm.

Moosmuller et al (2001) have studied diesel emissions with fast-response particle mass and optical instrumentation. The exhaust was coupled to a dilution tunnel, so the size distributions may not be as realistic as would be obtained with a wind tunnel. From their results and references therein, we chose an approximate coefficient for diesel particle scattering plus absorption of $5m^2/gm$ at 780nm which we further decomposed into $3m^2/gm$ of scattering and $2m^2/gm$ of absorption. If one further assumes that the absorption is wavelength independent, which will under predict in the UV because of polynuclear aromatic compounds, and that the scattering component varies inversely with wavelength to the 1.4 power, then effective extinction cross sections for the three wavelengths used can be generated (Hidy, 1984). These become 2.3, 6.0 and 18 $m^2/gm$ for the three wavelengths we use of 3900, 633 and 240nm. The final analysis of our data uses these coefficients. These are certainly an approximation because Kittleson (1999) has shown that the EC (black) percentage of the total aerosol mass can vary as widely as 5% to 95% depending upon wear, engine type and operating conditions. Clark et al (2002) have recently reported that the total mass from a given vehicle and engine is also very dependent upon the same variables. Yanowitz et al (2002) further implicate the rate of change of load as an important variable. The preliminary studies reported herein were attempting to achieve a moderate level of control over these variables by driving the high smoke emitting vehicles at constant speed up the highway entrance ramp.

It would appear on the surface to be straightforward to shoot a beam of He/Ne laser (633nm) radiation across a road and measure its intensity. In our minds, the major challenge was to interface the laser to the current FEAT UV and IR system (fully described elsewhere). The measurement is hard to make in large part because the expected loss of transmission signal, either by absorption or scattering, is small. Suppose we allow an acceptable plume maximum $CO_2$ amount to be five data points above 0.2% $CO_2$ (assuming an 8cm pathlength). This is the criterion we use for valid gas measurements with FEAT. This represents exhaust diluted by a factor of about 100. Suppose the actual opacity at the tailpipe is 3%, then the IR reference deflection which we try to correlate with the $CO_2$ increase is only 3 parts in 10,000. It is asking a lot from any measurement system to be noise free to 3/10,000 at 100 hz data rate! This problem can be partially alleviated by requiring valid data to be accompanied by a larger minimum $CO_2$ plume; however this criterion requires a very good loaded mode site and is a trade-off between data invalidity due to inadequate plume size and data invalidity because of too much noise for small plumes. Plume strength can be further exacerbated in light-duty diesel exhaust which contains a large excess of air.

The IR opacity system has been previously shown to have the capability to statistically distinguish fleet average on-road opacity by model year from gasoline and diesel vehicles (Stedman et al, 1997). ESP (formerly RSTi) in Tucson purchased a light-duty diesel vehicle whose opacity could be varied by means of driver aggressiveness between about 3% when unloaded and 24% when loaded. The remote sensed IR opacity readings on that car at 3% are noisy enough that a point reading 7% opacity and a point reading -7% both appear in 25 runs. When tested at about 24% opacity, the readings from the remote sensor are about +/- 3% opacity averaging 24%. As previously discussed, the signal to noise in the IR opacity channel was correlated with plume strength and for that reason a minimum $CO_2$ value of 0.8% (in a 8cm pathlength) was added to the software validity criteria. One observed sensing limitation is an apparent inability at this wavelength (3.9µ) to detect oil smoke from gasoline vehicles.

An additional data processing complication with the IR opacity channel involved very opaque plumes. The design role of the reference detector in the FEAT system is to allow the instrument to distinguish between gas absorptions and physical blockages, such as vehicle body parts.

However, the reference channel is also capable of detecting road dirt, water or snow spray, and smoke. Therefore, limits were placed in the software which would reject data for gas analysis which contained reference deflections of more than ±2.5%. In some of the first testing of the IR opacity channel in Hong Kong in 1993, we learned that very opaque plumes from a diesel taxi led the software to invalidate all of the data points because the reference deflection criterion had been violated. This eliminated many of the vehicles that one would set out to identify as black smokers. This problem cannot be completely avoided since an optical transmission technique will always have problems with plumes that are completely opaque. The current work has helped us to improve in this area with an understanding that we can allow up to a 20% deflection in the reference signal without adversely affecting the gaseous measurements. We made this discovery after the CRC shootout and corrected it before our own parking lot studies.

Instrumental

Low cost He/Ne lasers have an output which looks stable to the eye but in actuality is quite variable in intensity and quite variable in the angle at which it exits at the laser head. This situation was much improved upon the arrival of a stable Melles Griot 5mw He/Ne laser.

Interfacing this laser to the FEAT was attempted in two ways. In the initial system the laser was interfaced to a single 100 micron diameter optical fiber (Oz Optics interface). This fiber made up of one arm of a “Y”. The 1m long fiber terminated (at the base of the “Y”) in a ferrule, placed as close as possible to the spinning mirror in the FEAT detector system without interfering with the outside IR beam (CO). The light from the fiber sprayed off the spinning mirror and a segment of that band of light illuminated the main mirror, from which it was transmitted across the road to the source and returned via the retro reflector. The returned light passed in reverse through the optics and some actually returned to the laser up the original fiber. However, the retro reflector also laterally displaced the return radiation, and this displaced radiation entered a set of six fibers which surrounded the main fiber in its ferrule at the base of the “Y” and which terminated at an SMA (shaped memory alloy) connector in front of a narrow band filter and silicon photo diode.

This system provided a measurable signal from across the road and also a reference signal for the outgoing laser light as a flash of light at a different phase angle from the returned (across road) radiation. The two signals are detected as a series of intensity peaks at the spinning frequency of the mirror, namely 2400 Hz. Because of the phase angle difference, the reference and opacity signals could be separated with a synchronous demodulator for averaging and further processing.

Figure 2 shows the major system components on the source side of the road. Two different optical configurations were tested (components are labeled A and B in the figure, elements common to both are not labeled) for collimating the laser light into the current IR and UV FEAT source and returning the laser light to the detector. The first arrangement (labeled A) used an IR/visible beamsplitter close to the IR source which transmitted the IR radiation and reflected the

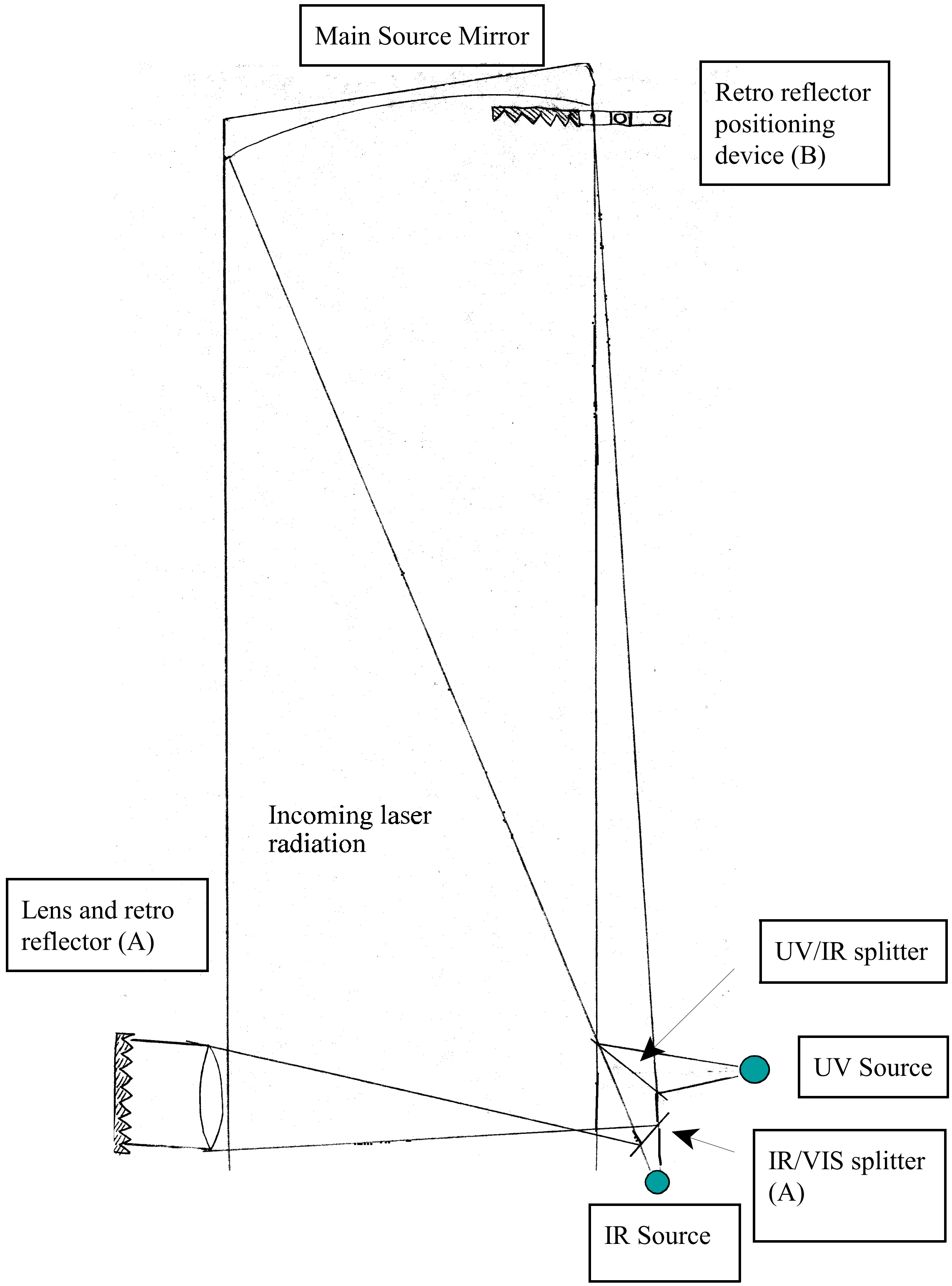


**Figure 2.** Schematic of two source (final design used configuration B) configurations tried to collimate the laser light with the current IR/UV sources. Common elements are not lettered.

laser light. This reflected light was collimated with a glass lens then retro reflected with a single, high precision corner cube. This arrangement had several drawbacks, the first was that the UV/IR beamsplitter had to be traversed twice, resulting in light loss each time. The second drawback was that the IR/visible beamsplitter was very close to the hot IR source (1400C) and tended to suffer thermal stress when the source was illuminated. Overall S/N was not as good as the second and simpler arrangement (labeled B) which simply retro reflected the light using a small strip of plastic corner cubes positioned in front of the main source mirror. This improved signal strength of the returned laser light and eliminated several costly optical elements.

The final configuration of the detector system (shown in Figure 3) has the laser directly illuminating the spinning mirror through a gap in the IR pickup mirrors. This produces a plane of laser light which illuminates the IR detectors and the focusing mirror located at the rear of the detector unit. A portion of this light strikes a reflective blade which illuminates the detector to provide a laser reference beam, electronically this signal is phase shifted from the signal beam allowing one detector to sample both beams. The important element in this design is a circular IR/Visible beam splitter (Infrared Optics, Inc) with a slit cut in the middle (nicknamed the "Pac-Man") to allow the plane of laser light out to the focusing mirror. This laser light is reflected across the road and is retro reflected back to the detector by the plastic strip of corner cubes. During the trip the plane of laser light grows in size. This laterally displaces the beam so that now much of the return beam does not pass through the slit in the beam splitter but is reflected by the Pac-Man to the interference filter and photo diode. Again, synchronous demodulation at 2400 Hz provides the intensity and reference signals which are averaged down to 100 Hz.

Software development has provided a system which reports CO, HC, NO, IR, Laser and UV opacity with error bars. Laboratory testing has shown that realistic $CO_2$ plumes can be measured, and the opacity readings reported for zero opacity plumes are within plus or minus a few percent.

Each of the new opacity channels approaches the detection problem similarly by attempting to quantify the loss of light transmission correlated to the $CO_2$ in the exhaust of passing vehicles.

Adapting the UV NO system to monitor opacity involved changing the software to allow for the collection by the monochromator of some additional intensity data which was previously not being collected. The monochromator detector is a linear silicon photo diode array with 128 detector elements. The majority of the diodes are not used since the NO measurement is optimized to be made in the middle of the array. We modified the software to collect the average signal intensities for the last 10 diodes for each of the 50 10ms data sampling scans. These intensities are then normalized for total intensity and correlated against the $CO_2$ absorptions collected by the IR system and converted into an opacity measurement.

The opacity readings that are reported are fundamentally ratios to $CO_2$. They have been scaled to 10% $CO_2$ in an 8 cm path length for comparison to an on-board opacity meter and have been expressed as a % thus $I/I_o \times 100$ scaled to 10% $CO_2$ in an 8cm path. Negative readings are a result of instrument noise or interferences which could not be accounted for properly. Note that this scaling to $CO_2$ provides a fuel-based ratio reading. For gasoline vehicles which run rich or

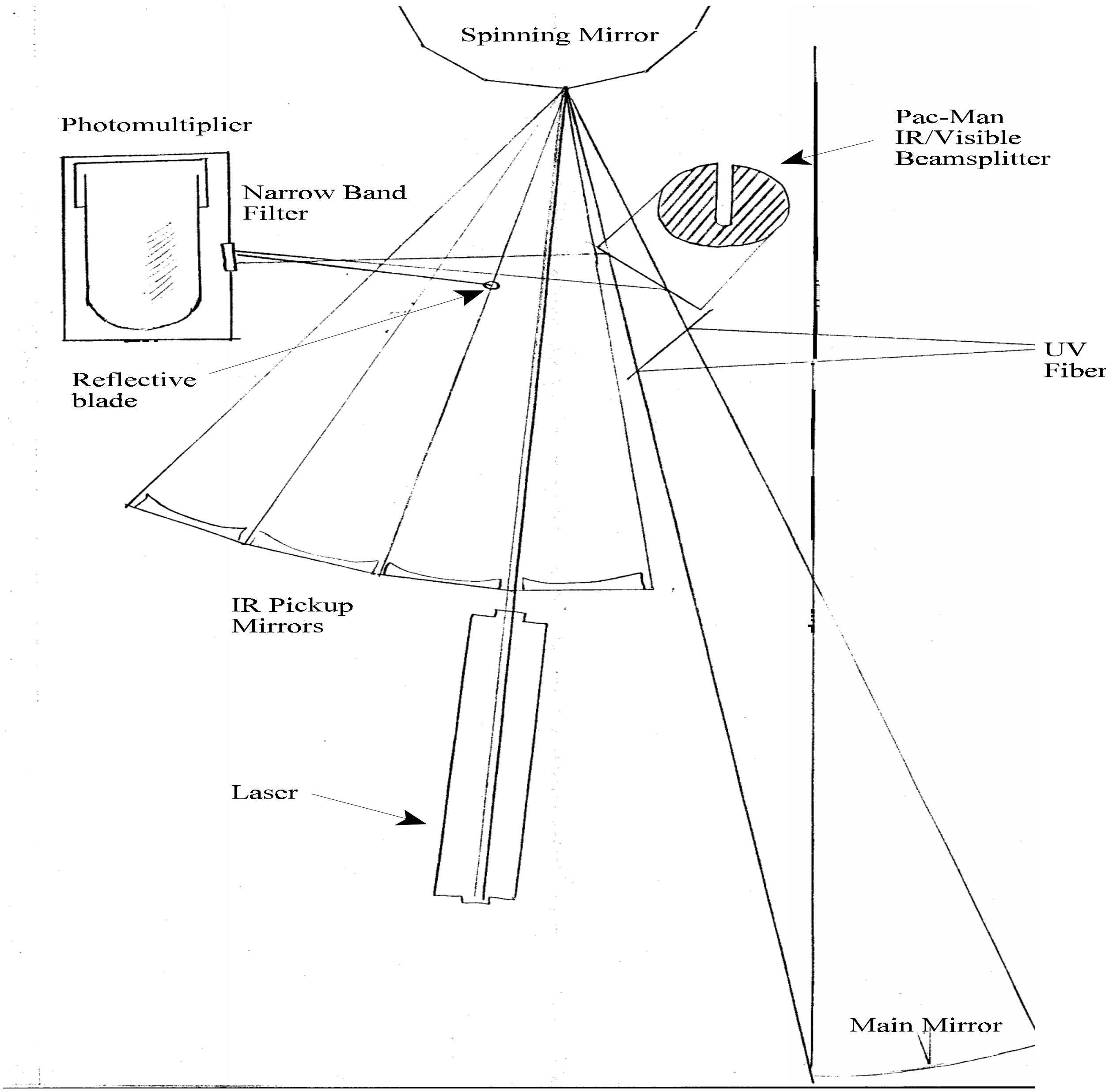


**Figure 3**. Detector system showing the laser beam striking the spinning mirror, exiting via the slit in the Pac-Man and returning to the Pac-Man, thence to be reflected to the PMT. The reference signal is provided by the small reflective blade.

stoichiometric, the scaling provides a reading which is directly related to a tailpipe probe reading. For diesel vehicles in which there is a large and variable added air component in the engine, remote sensing by definition "corrects" for excess oxygen not involved in combustion and thus might be expected to give a larger reading than observed by a tailpipe gas or opacity probe. Note also that simple linear scaling is reasonable for small opacities but quite incorrect for larger ones. If we measure 30% opacity for a 1% $CO_2$ plume then we would scale this to 300% for 10% $CO_2$ which we know is not cosmetically correct but does indicate that the vehicle is a bad smoker. To compare more accurately with a tailpipe opacity probe one should scale logarithmically, thus 30% opacity becomes 97% when scaled.

The raw data for opacity readings are the ratio of observed signal reduction [approximately -ln($I/I_o$)] to the observed increase in $CO_2$. The software used to report opacity herein has taken the wavelength differences into account only approximately. For the three wavelengths, 240, 638 and 3900nm, the reported opacity should be divided by 100, 500 and 1000, respectively. These raw ratios can be converted into gm particles/gm$CO_2$ using the estimated absorption and scattering coefficients described earlier. The unitless numerators are converted into gm/$m^2$ by division by 18, 6 and 2.3 $m^2$/gm respectively (the assumed $m^2$/gm scattering plus absorption coefficients in the UV, visible and IR respectively). The denominator must be multiplied by the HC calibration factor (in this case 1.6 to take into account the non-linear NDIR $CO_2$ absorption as measured by the morning gas cylinder calibration), and by a further 1.2, (the conversion factor from %$CO_2$ in 8cm to gm$CO_2$/$m^2$). From this calculation one obtains gm particles per gm $CO_2$. The only assumption is the scattering coefficients. To obtain particle emissions per kg of fuel one only has to multiply by the gm$CO_2$/kg of fuel which is obtained directly from the measured emission ratios thus:

$$\text{gmCO}_2\text{/kg of fuel} = 44/(0.014(1+Q+6*Q'))$$

where Q and Q' are the reported $CO/CO_2$ and $HC/CO_2$ ratios (for a detailed derivation see www.feat.biochem.du.edu, What's a FEAT?, "standard combustion equations").

Experimental

Two days of side by side measurements were carried out with the Desert Research Institute in Denver during February 2001. Three light-duty diesel vehicles, a Ford Club van, an Isuzu pickup and a 2000 Ford F250 pickup (two separate EECC's, one normal and one programmed for high smoke were used, effectively making it two vehicles), were driven by CDPHE employees to intercompare the remote opacity measurements. On February 21, measurements were carried out in a large parking lot owned by DU and consisted of multiple passes of each vehicle under steady state cruise conditions at 10, 20, 30 and 40 mph. The two remote sensing systems were set up approximately 5 feet apart in a level portion of the parking lot and the measurements were made after the vehicles had approximately reached steady state operations. Concurrently, tailpipe opacity measurements were recorded by CDPHE for each vehicle by an on-board data acquisition system using a traditional tailpipe opacity meter. Figure 4 graphs the comparison between the Laser and UV opacity channels and the IR opacity channel.

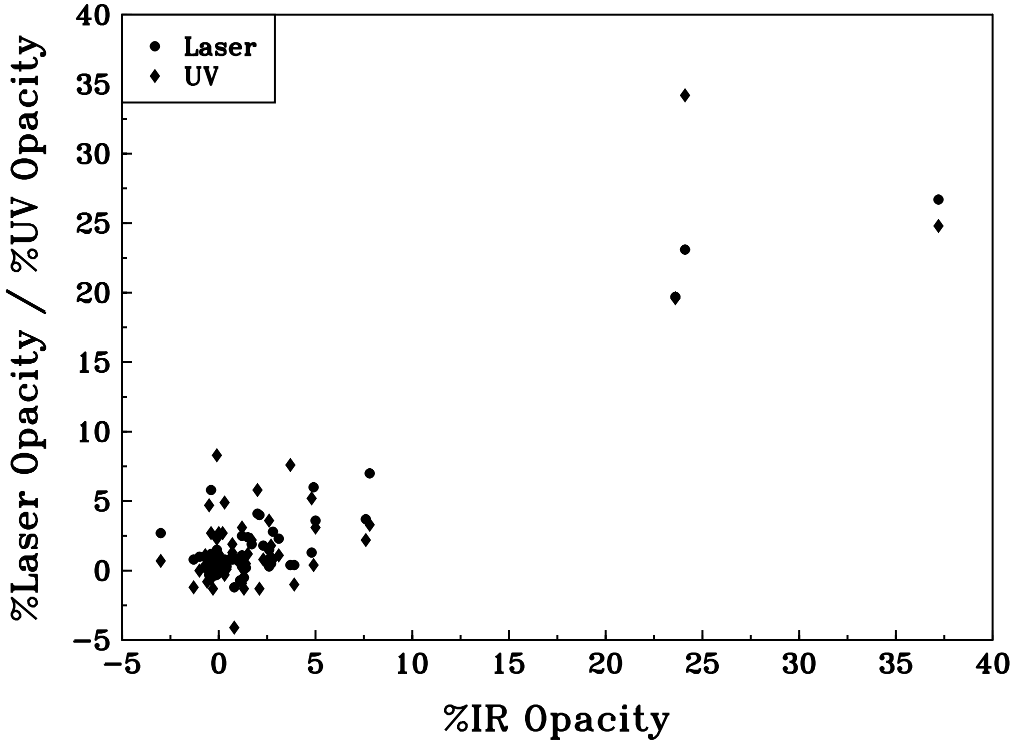


**Figure 4**. Correlation graph between the IR, UV and laser opacity measurements collected in the parking lot on February 21, 2001.

In general, visible opacity under steady-state driving conditions was rare from these vehicles. The Isuzu pickup and the Ford F250 with the high smoke EECC were capable of large amounts of smoke when accelerated hard. The only noticeable problem which we encountered during the first day of testing was that the exhaust from the Isuzu pickup was very difficult for our system to detect. For the majority of passes with the Isuzu pickup we were unable to make any exhaust measurements because the observed $CO_2$ plume sizes were too small. The Isuzu most likely proved so difficult because in addition to being a light-duty diesel (lots of excess air not involved in combustion) it only had a 4 cylinder engine producing low exhaust volumes. Maximum reported $CO_2$ concentrations, when valid, were a factor of 4 to 6 less than either of the other two vehicles.

For the second day of testing (February 22, 2001) the two remote sensors were set up three quarters of the way around a curved uphill on-ramp from northbound University Blvd. to northbound I-25. Traffic volumes are relatively low (300 - 500 light-duty vehicles / hour) and the tightness of the curve limits operating speeds. The DU FEAT attempted to collect data from 2300

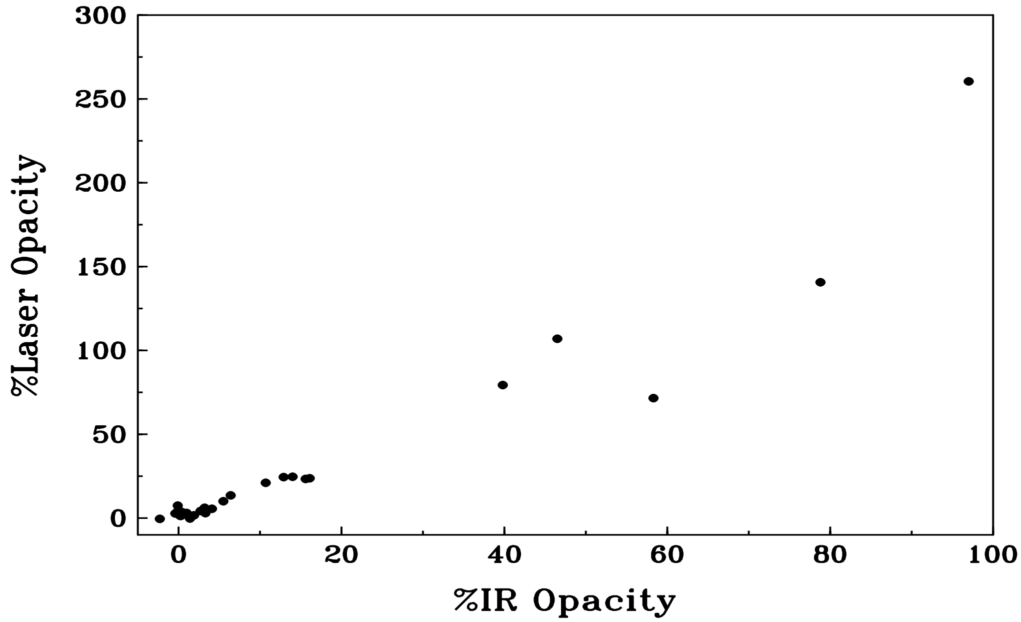


**Figure 5**. A correlation graph using data collected in our parking lot on March 7, 2001 with the Ford F250 truck. This data has been post processed using improved software which allowed the collection of more opaque plumes.

vehicles between 10:45 and 16:00. A total of 1686 measurements were collected with valid readings for all of the gaseous species. Because of the programming problems with our opacity rejection criteria (which rejected all of the elevated readings) we have chosen to plot all of the opacity data which accompanied the valid gas readings. Figure 5 shows all of the data (1686 measurements) from the laser and UV opacity channels graphed against the IR opacity channel and Figure 6 eliminates two extremely negative readings to enlarge the central group. All of the elevated points except one are from the light-duty diesel vehicles used in the shootout. The one exception is a gasoline powered early 90's Chrysler mini-van reported by the laser system only at 80% opacity. Since this is a gasoline powered vehicle we can only assume that it might be an oil smoker. We know from previous work that the IR opacity channel is not especially sensitive to oil smoke, however, the videotaped picture of this vehicle does not seem to show any visible oil smoke.

The same three light-duty diesel vehicles were measured under steady state conditions at 10, 20 and 30 mph (the 40 mph run was dropped because the tight curve of the ramp made this speed unsafe). In addition, several hard accel runs were made with the Ford and Isuzu pickup trucks. As during the previous day, exhaust from the Isuzu pickup was difficult to detect on most of its runs. In addition, software changes to deal with opaque plumes were not as successful as hoped (we believe we have improved in this area since the tests) and several of the acceleration runs were invalidated by a lack of data points meeting the reference variance criteria.

Separate from the E56 testing, we performed additional testing at DU with the Ford F250 and the

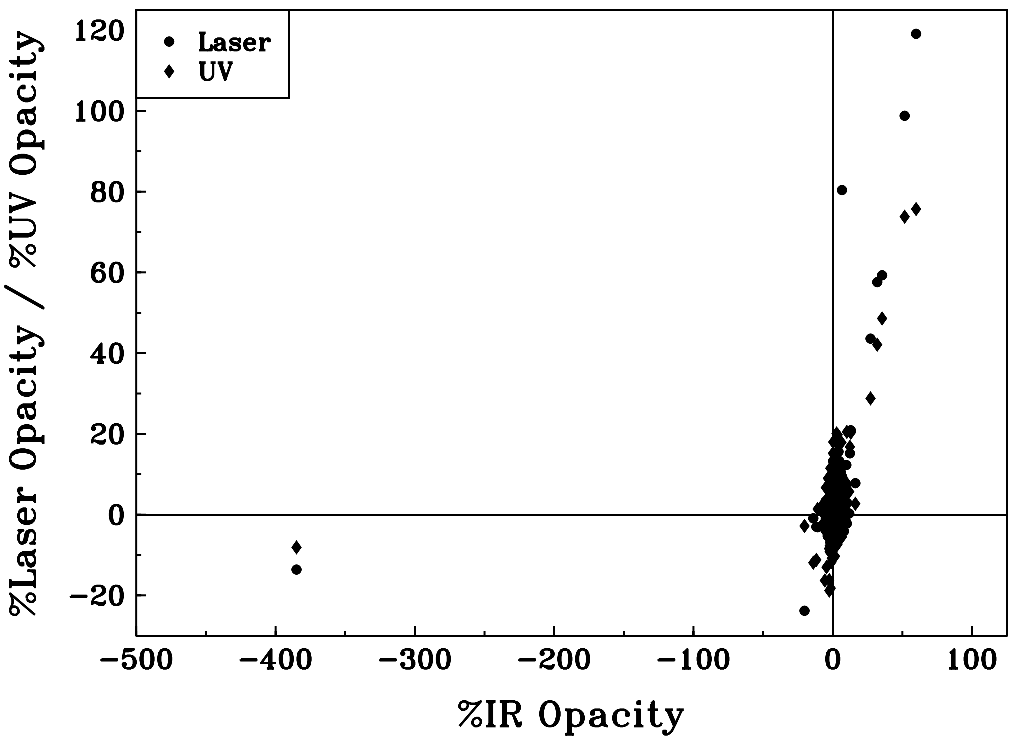


**Figure 6**. Correlation graph of 1686 on-road measurements collected on the University/I-25 on-ramp during the second day of testing (February 22, 2001).

high smoke EECC under hard acceleration conditions with revised software. This software allowed the IR reference channel to tolerate signal perturbations as large as ±20% (instead of the ±2.5% used in the original software) without invalidating that point. This greatly improved the acceptance rate from the opacity channels and Figure 7 shows the correlation obtained between the laser opacity channel and the UV opacity channel. We also used an on-board opacity meter; however, when testing under hard acceleration it is not trivial to ascertain the exact time when a highly variable opacity reading should be correlated to a remote sensing reading. Nevertheless, we were able to show that, of 26 passes, the six highest on-board opacity readings corresponded to the six highest remote sensing readings.

All data from the "shootout" studies in the parking lot (Feb. 20) and the on-road runs (Feb. 21) were sent to Dr. S. Cadle in March 2001, and will be posted on our web site www.feat.biochem.du.edu, together with this report upon approval by CRC.

The calculations described earlier were carried out for the five vehicles which DU FEAT software successfully measured as valid (despite their problems) on February 22, 2001. All five are vehicles intentionally smoking as a component of this study. The results are given below as Table 1.

We are very pleased by how closely these numbers correspond from left to right. The fact that reasonable assumptions from the literature give agreement within +/- 20% is very encouraging. A 17% lower IR coefficient and a 20% higher UV coefficient would give perfect agreement, but

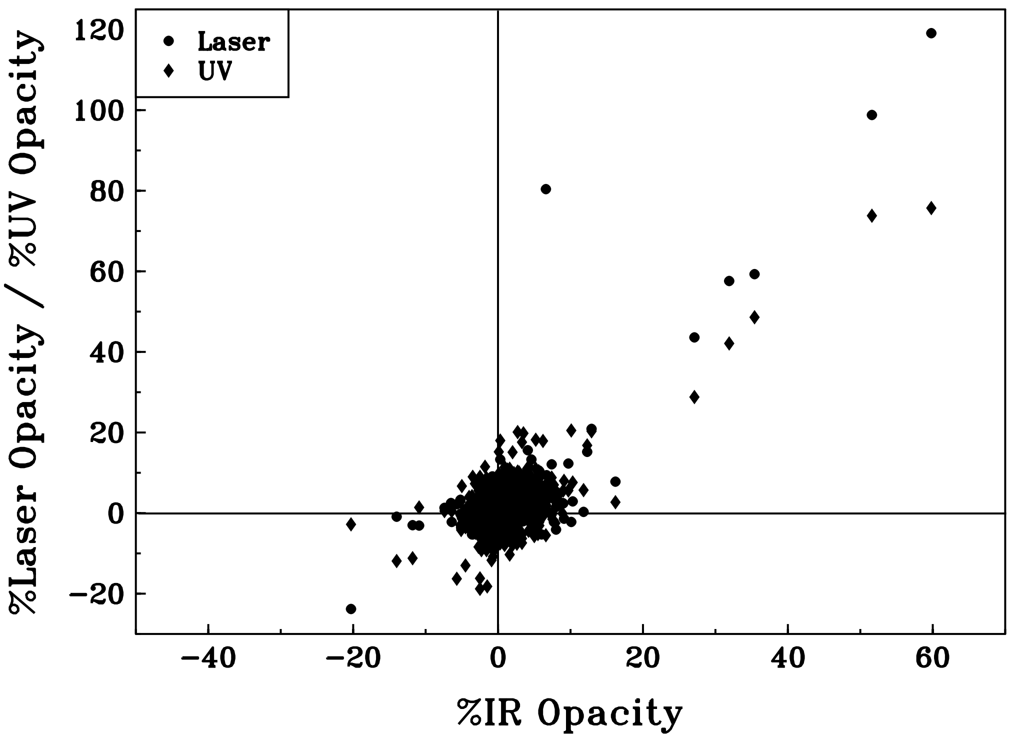


**Figure 7**. This is an enlargement of figure 4, eliminating several of the extreme values.

**Table 1**. Calculated particle mass emissions for the five high vehicles presented in order left to right as measured by IR, Visible and UV opacity techniques and using the assumed scattering and absorption coefficients.

| Vehicle | Time | IR gm/kg | Visible gm/kg | UV gm/kg |
|---|---|---|---|---|
| Isuzu pu (accel) | 16:01:32 | 33.1 | 50.6 | 53.6 |
| Isuzu pu (accel) | 15:57:13 | 29.1 | 42.8 | 53.2 |
| Isuzu pu (accel) | 15:53:34 | 21.9 | 28.1 | 38.4 |
| Isuzu pu (accel) | 15:55:13 | 19.6 | 27.1 | 33.1 |
| Isuzu pu (30 mph cruise) | 12:47:55 | 18.7 | 23.0 | 25.3 |

that sort of small adjustment is not justified by the accuracy with which these coefficients are otherwise known. The overall conclusion is that only vehicles which have been deliberately adjusted to smoke were measured as bad smokers in a realistic on-road situation, and reasonable assumptions about scattering and absorption coefficients give remarkably good agreement on estimated fuel based particle mass emissions.

## Conclusions

Unfortunately, software changes made to allow opacity measurement on very opaque plumes was incompletely removed for the shootout studies. Nevertheless, we were able to demonstrate real-time measurements of opacity at three wavelengths. We were also able to demonstrate that there were no false positive identifications in the sense that if our software did indicate high opacity on all three wavelengths, this only occurred for a vehicle which we had intentionally induced to smoke, in a sample totaling 1686 passing vehicles. After finally getting the software straightened out, the data shown in Figure 7 were obtained in our parking lot from the Ford F250 truck with 95% validity. We therefore believe that we now have available for on-road use, a real-time, three wavelength opacity measurement tool. If all three wavelengths agree on a gross smoker reading, there is a very high probability that the vehicle is indeed a gross smoker. If only the laser and UV channels provide gross smoking readings, the probability remains high. If only one channel reports a high reading, further action is not recommended. In principal, a three wavelength opacity system can provide information (in the form of the Angstrom alpha coefficient) bearing upon particle size distribution. We do not believe that our currently available S/N has reached that point. However, the level of agreement of particle mass emissions shown in Table 1 is quite encouraging.

## Acknowledgments

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