

Interactive comment on “Comparison of emission ratios from on-road sources using a mobilelaboratory under various driving and operational sampling modes” by et al.

et al.

Received and published: 15 October 2008

The authors would like to thank the referee for the comments, which helped to improve the manuscript. We have made changes in the manuscript according to the suggestions regarding the analysis of the data as follows:

[Anonymous Referee 2] Zavala and coauthors present emission ratios of various gaseous and particulate pollutants relative to exhaust CO₂ concentrations for low-duty gasoline vehicles (LDGVs) and heavy-duty diesel trucks (HDDTs) in Mexicali, Mexico. The emission ratios were calculated from highly time-resolved measurements of pollutant concentrations using a mobile laboratory both in stationary sampling mode probing bypassing vehicles and from mobile sampling of individual vehicle emission

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plumes and fleet-averaged emissions.

While fleet-averaging measurements provide information on emission ratios for different driving conditions, probing of individual vehicles with the other two sampling modes provides information on different vehicle types. In addition to these data emission ratios are compared to values obtained in an earlier measurement campaign in Mexico City and to a limited number of measurements collected in Austin, Texas.

The strongest point in these measurements is the use of a large variety of highly time resolved measurement techniques for parallel determination of emission ratios for a broad variety of relevant pollutants. This - together with comparisons of emission ratios for different vehicle types and driving conditions - has the potential to extract detailed information not only on average emission characteristics, but also on the variability of these parameters and on factors affecting these values.

Unfortunately this information is not provided in the paper and likely cannot be extracted from the dataset due to the very limited statistics of probed vehicles and the large variability in the individual emission rates. Within some of the categories (e.g. driving conditions, vehicle types) presented, not much more than a handful of measurements were performed, resulting in limited information about typical emission behavior within this category. As a consequence, these variability-dominated results do not show clear differences between different driving conditions or measurement locations such that an in-depth analysis of the variability of emission ratios and their causes cannot be performed.

[Response] The paper addresses the variability of the on-road measured emission ratios by different driving modes under real-world driving conditions. We show that different driving conditions such as cruising, stop-and-go, traffic, and idling modes have distinctively sampled emission ratios for gasoline vehicle fleets (see, for example, Table 1 and Figure 5) during the fleet-average sampling mode. The reviewer is right in pointing out that, due to the nature of this type of experiments, relatively few sampling

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periods were obtained during the intensive field campaign. However, we show that during the mobile laboratory fleet-average sampling mode measurements the plume-by-plume analysis technique used allows capturing information on thousands of individual exhaust plumes. When the analysis is done according to the criteria described in the paper, information on fleet average characteristics can be obtained. In the cases of much smaller sampling size when dedicated 8211;individual- chase experiments are performed, these are clearly noted as such and are less likely to represent fleet average conditions. We show that these individual measurements usually present higher variability than with fleet-average sampling modes. As such, these individual measurements of given vehicles are useful for understanding the emission characteristics of selected types of vehicle (e.g. Figures 1 and 2) but not for fleet average conditions.

Another weak point in the manuscript is that the literature on emission factors or emission ratios is largely ignored. Besides the results of a former measurement by the same group in Mexico City no other measurements were presented and no comparison to other values is made. This leaves the reader without information where to locate the presented values within other measurements.

[Response] We have corrected this by including a number of comparisons of our results with other studies. We have further transformed the units of the reported emission ratios from ppb/ppb-CO₂ to grams of pollutant by kilogram of fuel to allow an easier comparison with other studies. In particular, we show that CO and NO emission factors measured in Mexicali (60-147 g/kg and 2.7-13.7 g/kg, respectively) are within the ranges of measurements of the late 1990s in U.S. cities but smaller than those measured in Mexico City and Monterrey in the early 1990s.

Finally another major point of criticism is the broad absence of a self-critical assessment of the values presented in the manuscript. For example no discussion of a potential bias of measured emission ratios towards those of "dirty" vehicles in the measurement modes where clear plume signatures are needed to identify an emission plume is made. No discussion of uncertainties in the extracted emission ratios due to

co-measurement of other emissions during the plume measurements is presented.

[Response] We have added a discussion on this in the methodology section. As explained below (see our response to the specific reviewer comment below on possible sampling bias towards dirty plumes) in the chasing mode, usually larger 8211;easier to follow- vehicles are sampled, but no attempt was made to chase exclusively dirtier (black) plumes. Further, individual exhaust plumes are all identified and detected by their above background CO₂ signatures, which is nearly the same in both clean and dirty plumes as characterized by their PM and/or trace gaseous pollutant content. This fact ensures that there is no instrumental detection discrimination against 8220;clean8221; plumes from combustion powered vehicles. A more detailed description of the analysis methodology of the on-road data, including sampling of other non-road emission sources as well as self-sampling, is given in our previous work (Zavala et al., 2006) but we have now included some of this discussion in the text.

In Figure 7 emission ratios for NO_y are presented for Austin, Mexico City and Mexicali. Here the statement is made that the Austin ratios are "significantly" (p8077, L17) lower than the other ones while the error bars in Figure 7 (1 sigma) show that the values agree within their uncertainties.

[Response] We have corrected this statement in the text (see our response to this specific reviewer comment below).

In Figure 2 "linear" (log) relationships are found in data points that appear more like a point cloud with some trends.

[Response] We have included additional discussion of the data scatter in the figure. Note, however, that the purpose of Figure 2 is to show a comparison (in terms of positive versus negative correlations) of observed emission ratios of CO, NO, aromatic VOCs and fine particle number density from individual gasoline and diesel vehicle plumes sampled in roadside stationary mode, and that observation is still valid. The figure also shows the co-emission nature and variability of various pollutants for a given

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vehicle type (gasoline versus diesel).

Here and at several other places in the manuscript results are presented without critical assessment and leave the impression that not much information on emission ratios and their dependence on external factors can be extracted from the data set. At the end the reader wonders what the implication of the results presented here are. I suggest that the implications of the results as well as the uncertainties within the measurements and calculations are presented more clearly.

[Response] We have addressed the reviewer comments on self-critical assessments (see our responses below). This paper addresses the variability of the on-road measured emission ratios for different driving modes under real-world driving conditions. We show that different driving conditions for the gasoline vehicle fleet produce distinctively different sampled emission ratios for the fleet-average sampling mode. We make clear in the paper that stationary and individual 8211;dedicated- chasing experiments are useful for understanding the emission characteristics of various types of vehicles but not likely for fleet average conditions. We have added a more thorough discussion on the possible external factors affecting the results, including the ambient conditions and the possible effects of altitude on the comparison of the results between Mexicali and Mexico City. Finally, we compare our results with other studies and find that fuel use normalized emissions in Mexicali are much higher than in Mexico City and U.S. cities, possibly due to the large differences in the fleet age.

Detailed minor comments: A large fraction of the abstract is spent on the MCMA campaign and the methods developed there. This would fit better in the introduction. Also the duplicate use of the whole name of the campaign (Border Ozone Reduction and Air Quality Improvement Program for the Mexicali-Imperial Valley in 2005) should be avoided in the abstract.

[Response] We decided to include the reference to the MCMA campaign directly in the Abstract and in the Introduction because the paper has been submitted to the ACPD

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Special Issue on the MCMA campaign. As such, we show the connections of this work and the papers in the ACPD Special Issue. The duplicate use of the BORAQIP campaign has been corrected in the Abstract.

In the introduction statements are made about the variability of emission factors due to driving conditions or vehicle parameters. Here it would be helpful to provide an overview over this variability due to these factors as presented in the literature.

[Response] Thanks for this excellent suggestion we have included in the introduction key references that address the effect of vehicle speed and/or driving conditions (in the form of idling, acceleration, stop and go, etc.) on pollutant emissions in both laboratory and real-world conditions. The inclusion of these references strengthens the point in the paper that multiple factors directly affect the vehicle speed and engine load and therefore the emission rates from mobile sources under real world driving conditions.

Later in the introduction a statement about the difficulty of inter-comparisons of mobile measurements due to a variety of differences in the measurement process is made. Does this imply that the results obtained in such measurements are more a question of the measurement setup than of the actual emission? Here it would be helpful if it is made clearer what kind of solid information (which is not dependent on the measurement setup) can be extracted from such measurements.

[Response] The noted statement in the introduction makes the point that cross-validation and inter-comparisons of different measurement techniques are difficult to make for a number of reasons. It does not imply that the comparisons between measurement techniques can not (or should not) be made but that a number of considerations have to be accounted for during the comparisons of the observed emissions from various studies. The typical example of this idea is when comparing dynamometer studies with on-road measurements (e.g. Sjödin and Lennerb, 1995; Walsh et al., 1996), but it is also valid for cross-comparisons among on-road measurements techniques. For example sampling times may vary from less than a second for remote

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sensing up to several hours for tunnel and mobile laboratories, all in which different air-to-fuel ratio transient conditions of the engines may be captured, as well as possible differences in cold versus hot exhaust emissions may be present in the observations. Perhaps a good example of considerations needed to take into account for inter-comparisons of results using the same measurement technique is the large effect of road-grade in tunnel studies on measured emissions (Kean et al., 2003).

Still, as the reviewer rightly points out, solid data can be obtained from any of these techniques when properly implemented. Inter-comparisons are indeed useful with appropriate consideration of the different conditions. We have expanded this idea in the revised version of the paper and included some references as examples.

In the introduction the three measurement modes are described three times, in the abstract they are described another time and in the methodology as well as in the results sections they are again described in detail. I suggest reducing this redundant information to a minimum.

[Response] We agree that it was redundant to describe the measurement modes in various sections of the paper. We have removed such redundancy by presenting the description of the measurement modes only in the Methodology section of the paper.

P8064, L8-11: The measurements with same techniques provide information on actual differences in fleet emission characteristics of Mexico City and Mexicali. Can you discuss the influence of environmental differences in the two measurement locations like ambient pressure, RH or temperature?

[Response] We have included information on the prevailing ambient conditions in Mexico City and Mexicali during the measurement period. On average, Mexico City presented higher relative humidity and lower temperature (45%, 19 C) than Mexicali (28%, 24 C). Higher humidity generally results in lower NO_x emissions whereas higher temperatures would increase emissions. However, such differences in ambient conditions are not large enough to explain the observed differences: the difference between the

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NO_x-correction factors obtained after applying the standard equations from the EPA-CFR Title 40 is only 9%. The main difference between the two cities is the ambient pressure (76 kPa for Mexico City versus 100 kPa for Mexicali). Unfortunately, there is very little information reported in the literature of the effects of altitude on gasoline fueled vehicles to make a quantitative statement. At high altitude, the air-fuel ratio supplied to the engine may be reduced because air density is reduced. With richer fuel-air mixtures the combustible un-burnt components of exhaust gases increase.

Although gasoline emissions in Mexico City could, in principle, tend to be higher than in Mexicali because of the higher altitude, vehicles are generally provided with a mechanism for compensating for the effect of altitude on air density, minimizing this effect. Manufacturers conduct certification testing in the laboratory (for example by restricting the flow of air to the engine intake and equalizing intake and exhaust pressures) to comply with on-road standards for regulated emissions. With respect to the case of the effects of altitude on the emissions from HDT, Bishop et al., (2001) also report an increase in emissions with altitude. Note our results show lower NO_y emissions in Mexico City than in Mexicali despite the altitude effect (as the altitude effect would tend in fact to put the measurements in the upper limit during the comparison), implying that actually other effects (e.g. fleet type and driving mode) are larger.

P8064, L11-12: How does this point measurement (several hours on a single day 8211; or a few days?) presented in this manuscript provide information on the evolution of the characteristics of the emissions of the vehicle fleets?

[Response] It is true that a single fleet-average measurement does not provide information on the evolution of the emission characteristics of the fleet. The statement has been removed. Originally, this statement was motivated by the idea that future measurements in the same city would be valuable to understand the fast-evolving characteristics of the vehicle fleet. This has been accomplished in Mexico City (in 2002, 2003 and 2006) but not in Mexicali.

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P8064, L12-15: What is the point of comparing the emission ratios of Mexicali with those of Austin, Texas? This seems somewhat arbitrary.

[Response] Dedicated chase experiments with the same set up for the different instruments were conducted in Austin, TX as the ARI mobile lab was heading back to its headquarters after the MCMA-2003 campaign. The motivation was that with the implementation of NAFTA there is an increasing likelihood that Mexican trucks will be allowed to transport goods north of the border rather than transferring their cargo to US trucks at the border. This change could have an impact on Texas air quality if the Mexican and US truck have significantly different nitrogen oxide emissions. Comparative measurements of on-road heavy-duty diesel NOy emissions for US and Mexican vehicles allow this possibility to be evaluated.

P8064, L21ff: Information on the time resolution and detection limits of the instruments would be valuable.

[Response] Although the details of the characteristics and set up for the different instruments (including time resolution and detection limits) was given in our previous study (Zavala et al, 2006), we have included a brief statement summarizing instrumental characteristics.

P8065, L11: 98 valid mobile emission periods seem to be a very limited number, especially when these periods are further divided into different driving, vehicle and measurement mode categories.

[Response] Note that although the number of mobile emission measurement periods may seem small, the plume by plume analysis technique used with the mobile laboratory allows capturing information on thousands of individual plumes. When the analysis is done according to the criteria described in the paper, information on fleet average characteristics can be obtained. In the cases when dedicated 8211;individual- chase experiments are performed, these are noted as such and are less likely to represent fleet average conditions.

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Methodology section: Only very limited information is given on the measurement locations (highways, city streets, etc.) and their environments (free field, city, etc.). Also no information on the weather conditions is provided. In addition I am wondering how other sources of various pollutants are considered in the calculation of emission ratios. It is explained that the excess CO₂ (CO₂ above background) was used to calculate the ratios and likely this was also done for the other pollutants. However, how are variations in the background or advection of pollutants from other sources considered? This could be important especially for "clean" plumes.

[Response] We have added a sentence indicating the type of measurement locations and their environment. With respect to the analysis of the on-road data and the discrimination against possible sources of pollutants others than the mobile sources, including self-sampling, we have also added a sentence briefly describing it, but more detailed information is given in our previous work (Zavala et al., 2006).

P8066, L19: What was the distance of the mobile laboratory to the sources?

[Response] Our experience has shown that the distance of the mobile laboratory to the sources should not be too large during normal city driving conditions. Distances smaller of about 3-5 meters are good enough to allow the plumes to reach the sampling port before excessive dilution. During highway chase experiments, the distance increases some more, not only for sampling purposes but also for safety.

P8067, L18-20: Since individual plumes have to be detected in the stationary sampling mode (and in the chasing mode, see P8069, L8-11) does this generate a systematic bias towards dirty (easy-to-detect) plumes?

[Response] As noted, the high sensitivity of the instrumentation on board the mobile laboratory allows for the sampling of relatively clean plumes, not only the dirty ones. The CO₂ enhancement is not really affected by whether the plume is dirty or not, and so if the CO₂ hits signals (plumes) guide the choice of plumes, then there should not be a bias towards clean/dirty plumes. Nevertheless, as explained in the paper (P8067,

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L5-18), when sampling in stationary mode there is a characteristic time window of only a few seconds before emissions are diluted in the background air. The appropriate location of the sampling port is very important and we have accounted for it. As shown in Figure 1, the traffic density of the selected road allowed the unequivocal distinction of emission signatures for individual passing vehicles when a relatively large time elapsed between them. In the chasing mode, usually larger 8211;easier to follow- vehicles are sampled, but no attempt was made to chase exclusively dirtier (black) plumes.

P8068, L3-8: This explanation is hard to understand. It should be clearly stated that for the inorganic compounds measured with the AMS no correlation to the CO₂ concentration is seen, because these particulate compounds are not from the vehicle.

[Response] The idea of the sentence in the paper has been simplified as suggested.

P8069, L23-27: Could you provide clearer information on how the data were separated into the various driving conditions? Is "vehicle speed" the average speed over a sampling interval of >5 min or are the given speed ranges the ranges between the highest and smallest vehicle speed within this interval? In line 26 the CRU speed has to read "56 km/hr".

[Response] They are the speed ranges between the highest and lowest vehicle speeds in the 5- min interval. This has been clarified in the text.

P8070, L25-28: In the fleet average sampling mode the emissions are dominated by gasoline vehicles. Can you give an estimate of the fraction of HDDTs in these data sets? Would it be possible to split these data into data sets with high and low HDDT contribution?

[Response] As part of the analysis to obtain fleet average estimates, the periods when there is influence from surrounding heavy-duty trucks (from real-time notes taken during the measurement and from reviewing the video images later on) are marked and not included in the gasoline emission estimations. As such, the reported data set should

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include minimum or low contribution from HDDT sources.

Results section: A large amount of information about emission ratios is given in this section and the reader might wonder what the relevance or implication of the individual bits of information is. This would be easier to digest (and avoid repetitions of results) if this section would be merged with the discussion section.

[Response] We prefer to differentiate the Results and the Discussion sections because we want to present the results clearly distinguished by driving mode and then wrap them up in the Discussion section.

P8073, L17ff: The data presented in Figure 5 shows typically lower emission ratios determined with the fleet-averaged measurement mode compared to those modes where individual emission plumes have to be identified. This could be an indication that the latter measurements are biased towards "dirty" vehicles. This potential bias as well as the observed differences in the emission ratios should be discussed.

[Response] As explained above, during the chasing mode no attempt was made to chase exclusively dirtier (black) plumes and individual plumes are detected by their similar excess CO₂ content, not their PM or trace gas components. There is not much control over sampling vehicles with dirtier/cleaner plumes in stationary mode. These measurements can be used to deduce specific emission characteristics of gasoline/diesel vehicles. However, we mention in the paper that the sampling in these measurement modes could hardly be representative (particularly for the gasoline fleet) if the sampling size is small. In the paper we show that the results show higher variability in the sampling of individual plumes and chased vehicles than for the fleet average mode. We suggest that this could be explained by the 8220;micro8221; approach of these measurement techniques where a large number of factors (emission control system, vehicle age, maintenance state, fuel type, etc.) play a major role in determining the emissions from an individual vehicle. In the fleet average sampling mode, all these factors are smoothed by averaging (equally weighting) the measured emissions plumes.

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Nevertheless, an important observation is that despite the variation observed in the fleet average sampling mode, the averages and their standard deviations indicate that the sampling size was large enough to be sensitive to driving mode.

P8074, L9ff: This sentence is a repetition of a sentence a few lines before.

[Response] Thanks, this has been corrected.

P8075, L3ff: Table 2: What is the point of listing emissions for San Diego or Mexico City if these results are not mentioned or discussed in the text? How are the emissions calculated for those cities where the numbers were taken from the literature and how are they comparable to the values calculated from the measurements presented here? There is a detailed comparison of Mexicali and Calexico emissions for NO_x and CO for which the concentrations turn out to reflect nothing else than the size of the vehicle fleet (at least within their uncertainties). So what is the purpose of this comparison?

[Response] Note that, at least for Mexicali, there have not been any previously reported observation-based estimations of mobile emissions. As the measurements from the mobile laboratory can readily provide such estimations, we feel it is a good idea to present it even if it turns out to be a reflection of the size of the vehicle fleets. We agree to remove the estimates from San Diego since they add little value.

P8077, L5-6: In Figure 6 not the NO_y emissions as a function of driving speed are shown, but the NO_y emissions are presented as a bar chart in order of increasing NO_y emission ratio. I suggest generating the plot that is described in the text: NO_y emission ratio plotted versus driving speed.

[Response] We have modified the description to the figure to indicate the NO_y measurements ranked by the magnitude of the speed.

P8077, L7: What means that the trucks "were identified by their license plates"?

[Response] The license plates of the sampled trucks were recorded (which allows motor vehicle registration records to be used to determine vehicle model and age). This

has been indicated in the text.

P8077, L15-19: Here it is claimed that the Mexicali and MCMA NO_y emission ratios were "significantly" higher than those in Austin. In Figure 7 one can clearly see that within the uncertainty of the measurements the emission ratios agree with each other!

[Response] We have corrected this statement in the text. However, we still note that the differences in the averages of the measurements clearly indicating that these measurements represent emissions from a limited number of vehicles and it is possible that the sample size is not sufficient to produce accurate fleet average HDDT emission ratios.

Table 1: In this table everywhere 3 digits are presented for the emission ratios while in many cases the uncertainty of the values exceeds tens (up to 732;80) of percent. For values with such a degree of uncertainty it does not make sense to present numbers with so many digits.

[Response] This has been corrected. We have also changed the units of the results in table 1 to grams of pollutant per kilogram of fuel for an easier comparison with other studies.

Table 2: Since in the text it was shown that the emissions of Mexicali and Calexico mainly reflect the vehicle fleet size of the two cities I recommend adding information on vehicle fleet size, population and area of all four cities to the table.

[Response] This information has been included in the table.

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ACPD

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