Atmos. Chem. Phys. Discuss., 9, C2842–C2848, 2009
www.atmos-chem-phys-discuss.net/9/C2842/2009/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of mobile emissions contributions to Mexico City's emissions inventory using on-road and cross-road emission measurements and ambient data" *by* M. Zavala et al.

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Received and published: 14 July 2009

The authors would like to thank the reviewer for the comments and suggestions on the manuscript. We describe below the modifications made to the manuscript.

The paper attempts a comparison of the official road transport inventory of Mexico City with the results on emission factors produced by on-road measurements conducted by a mobile lab. The emission factors produced in this way are compared with remote

sensing measurements and ambient pollutant ratios from the air quality monitoring network. This is a useful study in the process of validating and quantifying the uncertainty of urban inventories. I would therefore recommend publication of the paper. However, I have a few points that the authors should consider and resubmit a revised version, before a final approval.

Major Points

Point 1: There is not a detailed description of the total measurement sample used in this study. In p.6369, l.16, the authors refer to "28 gasoline vehicle fleet average sampling intervals. . .during the analysis of approximately 6.5 h of on-road data". They also suggest that "in the fleet average sampling periods hundreds to thousands of individual exhaust plumes are actually sampled". I don't find this enough information to judge the quality of the input data. I would therefore suggest that the authors provide more detailed information to respond to the following questions: What is the total duration of the 28 gasoline sampling intervals? How can one separate gasoline intervals only, out of a driving condition involving hundreds to thousands cars? What is the split of the intervals to SAG, TRA, CRU? A SAG condition is defined as a condition of 16 kmfor a period of 5 min or more. In such conditions, measurements are mostly affected by the plume of the cars in the immediate vicinity of the mobile lab (mostly the car directly in front). How many SAG intervals were measured to obtain a representative measurement sample? In general, I feel the authors need to better support the representativity of their sample.

[Response] We agree with the referee that it is important to better clarify the description of the sample used in the study. We have improved the description of the total measurement sample by stating the total duration of the sampling intervals. We have also included further description on the classification of the fleet averaging sampling intervals by the SAG, TRA, and CRU sampling modes, the number of individual

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sampling events, as well as the distinction between gasoline and diesel emissions. As described in the manuscript, during the analysis we purposely filter out individual detected plumes from certain vehicles (including smokers) to avoid sampling bias. As a result, the measurement technique in fleet averaging mode is performed by the mobile laboratory when emissions from the surrounding vehicles are obtained continuously for a relatively long period of time (about 5 mins or more). During the analysis the exact number of vehicles from which their emissions are obtained in fleet averaging mode in a given time is not known. However, given the high traffic activity in Mexico City, during the sampling of 6.5 hrs of on-road data a –rather conservative- lower limit estimate of the sample size is likely in the hundreds. Not knowing the exact number of vehicles sampled in a given time does not totally preclude the assessment of the representativity of the sample. Two other indirect ways of assessment include the intercomparison with other high sampling volume techniques and the observed dependency of the emission factors by driving mode. In the manuscript we treat in some detail both of these issues by 1) comparing our results with on-road emissions measurements obtained preciously in Mexico City using the same technique and also with remote sensing measurements in Mexico City and U.S. cities, and 2) describing the variability of the on-road measured emission ratios and observing that their averages and standard deviations were sensitive to different driving modes under real-world driving conditions.

Point 2: A discussion on the effect of ambient conditions might be useful to include in the paper for two reasons. First, as Mexico City is situated at high altitude, the authors should discuss whether effects of the altitude on emissions are expected. In particular when comparing results with US cities (e.g. p.6372, l.15-20). In addition, information on the period and weather conditions between the 2003 and 2006 studies should be given, in particular following the discussion in Table 2.

[Response] The reviewer correctly points out that in this study the effects of altitude

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would only be relevant for the comparison of our measurements with results obtained at different ambient pressures in other cities. Unfortunately, there is very little information reported in the literature on the effects of altitude on the emissions from 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 unburnt components of exhaust gases increase. Although gasoline emissions in Mexico City could, in principle, tend to be higher than in U.S. cities 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. For this reason, the observed (significant) differences between the Mexico City and the U.S. fleets are more likely to result from differences in fleet age and to the fraction of vehicles with emission control technologies. We have also included information on the prevailing ambient conditions in Mexico City during the measurement periods. On average, temperature and relative humidity during the monthly measurements campaigns in March 2006 and April 2003 did not differ significantly (19C, 45

Point 3: I feel that the conclusions in p.6371, l.13 and p.6372, l.13 on the effect of calculation year to VOC species emissions contradict each other. Have the VOC species emission decreased or increased in the period 2000 to 2006?

[Response] We have clarified these statements in the manuscript by indicating that the comparison between 2000 and 2006 refers to the VOCs remote sensing measurements of Schifter et al (2009) whereas the comparison between the 2003 and 2006 refers to the selected VOC measurements in this study. As described, in our study we can only compare the individual VOCs measured in both campaigns which account only for a relative small fraction of the total VOC mass.

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Point 4: In general the VOC estimate is prone to a number of uncertainties. The authors correctly recognize that remote-sensing VOC measurements only approximate total NMVOCs (p.6374, l.8). Therefore use of the approximate VOCratio on the CO measurement, in order to calculate VOC and compare with the emission inventory, can only provide an approximate result. Just because of its uncertainty, this is not enough evidence to come up to the conclusion that the MCMA underestimates emissions by 1.4-1.7 times (p.6374, l.23). In addition, no big difference was found for the VOC species actually measured (p.6374, l.25). Therefore, it could be that the inferred VOC calculation overestimates emissions and not vice versa. In order to provide a more fair judgment to the difference, I would propose the following two revisions:

1. The blue bar of HC in Fig. 3 should be formatted differently (e.g. blue hashed) and a clarification should be added in the caption to make clear that this is an inferred and not a measured value. 2. The inventory should not be judged as providing an underestimation. Instead, the authors should talk about a difference between the inventory and the inferred value and explain the reasons (as they do) of the difference in a non-biased manner. This also includes the discussion in the abstract.

[Response] We agree with the reviewer on this suggestion. We have explicitly indicated now that the comparison is only an approximation due to potentially significant uncertainty in using the measured VOCratio for estimating VOCs. We have also included the reviewer's suggestions in Fig. 3.

Point 5: The authors suggest that the inventory significantly underpredicts PM emissions from gasoline cars (p.6375, l.7). As gasoline cars are not prime emitters of exhaust PM, the authors should provide more information on the exhaust vs non-exhaust (tire and brake wear) contribution to total PM. Does the inventory include

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non-exhaust sources? If yes, what is their share in PM1? What is the (estimated) share of nonexhaust sources in their measurements?

[Response] We have clarified this in the text by indicating that the PM from the inventory represents only the estimated PM2.5 emissions from gasoline exhaust. The inventory estimates mobile emissions of PM10 and PM2.5 from various combustion and non-combustion sources. In our comparison, we have only included the exhaust emissions estimates of PM from gasoline sources.

Minor Points 1. What is NOy? (p.6378, l.16).

[Response] NOy is the total concentration of reactive oxidized nitrogen-containing compounds (NOx +HNO3 + PAN, etc.). This has been clarified in the text too.

2. Where does the value 54.1 molC/lt come from? (p.6370, l.4). Is there any reference? This may significantly affect the gfuel emission factors produced.

[Response] This is the result of assumption of the fuel's carbon mass fraction content and density. Both of these values are well known for Mexico City through measurements (e.g. Schifter et al, 2005) and their variability is relatively small in part because there is only one fuel provider company in Mexico.

3. The abstract discusses about NO emissions (p.6364, l.10) while the main body discusses about NOx. What is the CLD analyzer tuned to?

[Response] Although most NOx is emitted in the form of NO (especially from gasoline vehicles), the reviewer is right in pointing out that we should be more specific. We have

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removed this inconsistency by clearly specifying when needed that NO emissions were obtained.

4. The VOC "underprediction" is quoted in the range of 1.4-1.9 in the abstract (p.6364, l.17) compared to 1.4-1.7 in the main body of the text (p.6373, l.26).

[Response] Thanks for catching the typo.

5. The discussion in the abstract on diesel vehicles effects on NOx does not make sense (p.6364, l.28). NOx is found not to have changed but this suggests that the contribution of diesel vehicles has increased.

[Response] We have clarified the discussion in the abstract regarding our finding of increased contribution of diesel vehicles in overall NOx levels. As explained in the manuscript, in addition to the relatively recent introduction of NOx emission standards for gasoline vehicles, pieces of evidence for this effect includes large increases in the dieselfuels per year combined with the much larger NOx emission factors of diesel powered vehicles compared to gasoline-powered vehicles. However, as correctly pointed by the reviewer 2, other factors also may affect the observed NOx variability including proliferation of air conditioning and different removal efficiency of catalytic converters.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 6363, 2009.