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Memorandum

Date: January 30, 2008
To: Chung Liu, SCAQMD
Cc: Henry Hogo, SCAQMD, Jon Leonard, TIAX Irvine

From: Michael D. Jackson
Loc: Cupertino Office
Phone: 408 517-1560

Subject: Mates III PM Results

Summary

I investigated possible reasons for understanding the dichotomy of MATES III results that simultaneously show measured reductions in elemental carbon and inventory estimates that showed increases in PM emissions. I found that one possible explanation is the introduction of newer technologies in both the light- and heavy-duty vehicle segments. Interestingly, the ratio of elemental carbon to particulate emissions is changing as a function of vehicle fleet: the ratio is increasing for light-duty vehicles and decreasing for heavy-duty vehicles. Based on simplifying assumptions, I conclude it is possible to have a 23% decrease in elemental carbon emissions and a 10% increase in PM emissions. The use of elemental carbon to determine PM emissions is quite complex. In my view, the current write up in the public draft version of MATES III[1] provides a better methodology for estimating PM emissions from elemental carbon measurements than that previously used in MATES II.

Introduction

In December 2007, I evaluated possible reasons for the measured reduction in elemental carbon in the recent MATES III results while inventory estimates showed increases in the total PM emissions. I looked into two possible mechanisms that could explain the PM increase with the measured EC reductions. One question I had was the possible effect of light-duty vehicles compared to heavy-duty vehicles on the emissions inventory. Although HDVs dominate the PM inventory, it could be possible that PM increases in the LDV segment could offset PM decreases in the heavy-duty sector. My second question involved the accuracy of the EC to diesel PM correlation that was used in the MATES II study.[2] These two issues are discussed further below.

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[1] South Coast Air Quality Management District, "MATES-III," Draft Report, Multiple Air Toxics Exposure Study in the South Coast Air Basin, January 2008
[2] Elemental carbon (EC) was used to estimate diesel PM using a conversion factor in of 1.04 in MATES II.

Effect of Light-Duty Vehicle Fleet

To get an indication of the contribution of LD and HD combustion PM, TIAX staff ran EMFAC20007 for the South Coast in 1999 and 2006. Table 1 shows the EMFAC2007 LD and HD PM results.

Table 1. EMFAC2007 South Coast Inventory Estimates for Particulate Emissions

Inventory Element	*1999*	*2006*	*Change*
LDV PM (tpd)	2.77	4.15	50%
HDV PM (tpd)	13.51	13.75	2%
total PM (tpd)	16.28	17.9	10%
VMT (thousand miles)	325,735	392,218	20%
Vehicles	8,941,640	10,513,500	18%

My speculation that the LDV segment could have increased overall PM emissions is borne out by these data. In fact, LDV PM emissions increased considerably more than HDV emissions; this had the effect of increasing total PM instead of reducing total PM (as might be inferred from the EC reductions in the MATES III data). Intuitively, one could expect that PM emissions would be reduced rather than increased with the introduction of PZEV technologies in the LDV segment. Any reduction associated with phasing in of better emissions controls must be offset by increases in number of vehicles and vehicle miles travel (VMT), or better accounting of high-emitting vehicles.

EMFAC2007 inventory estimates suggest a 10% growth in total PM emissions from 1999 to 2006. The ambient measurements of elemental carbon suggest an overall basin wide reduction of about 38%. With the PM to EC ratio used in MATES II of 1.04 one would expect a comparable reduction in PM. Possible explanations are discussed further below.

Changes in Vehicle Technology

One possible explanation of decreased EC but increased PM may be the differences in vehicle/engine technologies. Perhaps newer engine technologies affect the composition of PM. As particulate emission standards where implemented in heavy duty engines, the composition of PM changed from so called “wet” to “dry” engines. The older engines had more soluble organic matter (so called soluble organic fraction--SOF) compared to the newer “drier” engines. EGR technology also increases the insoluble component of diesel particulate emissions. Thus, it is possible in the case of HDV emissions that there was a shift in PM composition, which resulted in low SOF components and high insoluble components. This raises a key question of how this affects the EC/PM ratio.

I did a very brief look at several data sets to see if any conclusions could be drawn regarding changes to the EC/PM ratio. The first data set that I looked at was contained in recent published work by Robert et. al.[3] Figure 1 (taken from their Figure 1) shows fine and ultrafine PM mass emission rate for various model year diesel engines. Also shown is the mass of elemental carbon and organic matter. Of interest here are the changes in elemental carbon (EC) and organic matter (OM) as engine technologies change. Key captions on the figure correspond to the following engines:

- HDDV-6 1985 Caterpillar 3406
- HDDV-5 1991 Caterpillar 3406B
- HDDV-3 1997 Cummins N-14 460E+
- HDDV-2 1998 Detroit Diesel Corp (DDC) Series 60

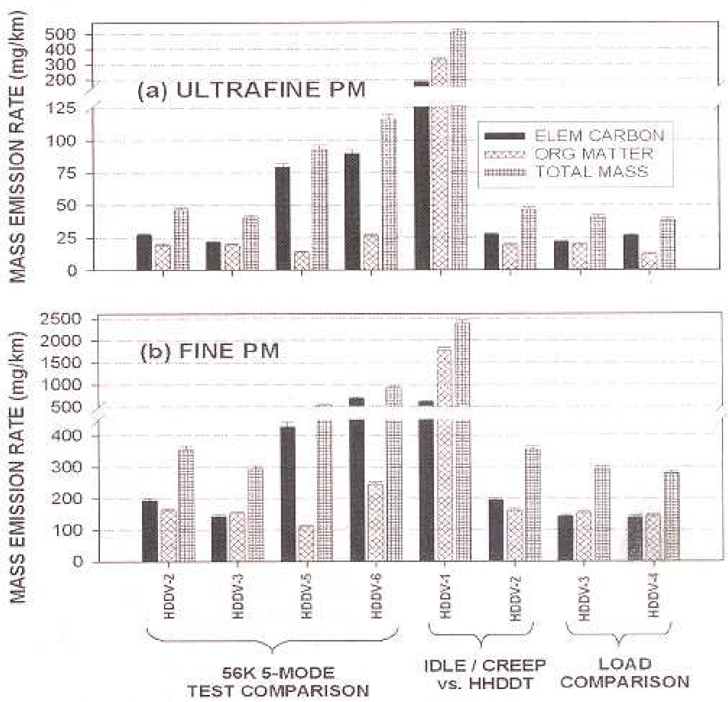

Figure 1. (a) HDDV ultrafine and (b) fine PM emissions under 56,000-lb (56k) and 66,000-lb (66k) inertial loads.

As shown in this figure, newer engines have lower PM emissions as expected. But also shown is a dramatic difference in EC and OM composition. In the older engine technologies (HDDV-5 and -6), EC varies from 0.8 to 0.7 of total PM. For the newer

[3] Robert, Michael A, Michael J. Kleeman, and Christopher A. Jakober, "Size and Composition Distributions of Particulate Matter Emissions: Part 2—Heavy-Duty Diesel Vehicles", Journal of the Air and Waste Management Association, Volume 57, pg 1429, December 2007.

engines (HDDV-3 and -2), EC composition of total PM varies from 0.4 to 0.5--an obvious decrease in EC composition compared to the older engines. This is also shown graphically in Figure 2 (taken from their Figure 2). MATES II assumed that EC to Diesel PM was 0.67 (or DPM=1.49EC) which is closer to the older engines than the newer engines. The newer engines suggest DPM=1.8 to 2.4 times EC.

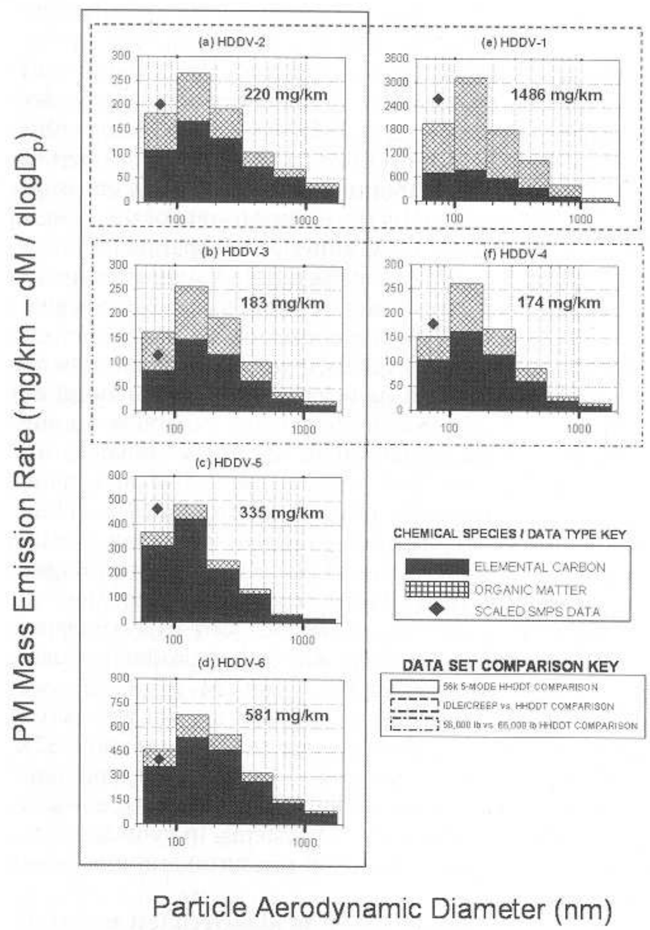

Figure 2. Size and composition distributions of PM emitted from HDDVs. Emissions profiles in the left column (a–d) were produced using the 5-mode HHDDT driving cycle with a simulated load of 56,000 lb. Emissions profiles in the right column used an idle-creep cycle (e) or 66,000-lb simulated load (f)

Where:

- (d) HDDV-6 1985 Caterpillar 3406
- (c) HDDV-5 1991 Caterpillar 3406B
- (b) HDDV-3 1997 Cummins N-14 460E+
- (a) HDDV-2 1998 Detroit Diesel Corp (DDC) Series 60

Similar results were obtained comparing mechanically injected to electronically injected diesel engines.[4] In this study, the authors found that elemental carbon and total carbon were reduced by 60% with the electronically controlled diesel engine. Organic carbon was also reduced but at a lower level. Using their data the mechanical engine had an elemental carbon to total carbon ratio (EC/TC) of 0.90 and the electronic engine had an EC/TC of 0.63. It is expected that this will again change with 2007 diesel technologies equipped with diesel particulate filters (DPFs). DPFs will substantially reduce EC and to a lesser extent OC. And, therefore, one can expect even lower future EC/TC ratios with the 2007 diesel technologies.

The next question was "How does light duty vehicles (LDV) change the possible correlation between PM and EC?" Again I did a very brief literature review and found several papers providing some insight into the EC/PM performance of LDVs as a function of vehicle technology, age, and level of emissions (gross polluter).

Schauer presents a good overall review of the use of EC as a marker for DPM.[5] He explains the different methodologies for measuring EC and provides estimates for various sources including fireplace combustion, agriculture burning, coal combustion, fuel oil combustion as well as spark ignited gasoline and compression ignited diesel technologies. Schauer shows diesel EC/TC varying from 0.50-0.80 but with not enough data to distinguish between factors such as driving cycle, engine type, engine age, and engine fuel, although he concludes these would be important factors.

Schauer also summarizes EC/TC for gasoline vehicles and found the ratio to vary from 0.14 for smoking vehicles to 0.27 to 0.38 for normal vehicles (non smoking). He points out that these results are consistent with the idea that smoking vehicles burn more lubricating oil which then adds more OC to TC. Schauer also correctly points out:

> *"..accurate estimates of the relative contributions of diesel vehicles and gasoline-powered motor vehicles will require an understanding of the distribution of vehicles, the average driving cycles for these vehicles, and the average EC content of the vehicle fleets' emissions under relevant driving conditions."*

[4] Bagley, Susan T., Winthrop F. Watts, Jr, Jason P. Johnson, David B. Kettelson, John H. Johnson, and James J. Schauer, "Impact of Low-Emissions Diesel Engines on Underground Mine Air Quality," National Institute for Occupational Safety and health Grant No. R01/CCR515831-01, May 2, 2002.
[5] Schauer, James J., "Evaluation of Elemental Carbon as a Marker for Diesel Particulate Matter," Journal of Exposure Analysis and Environmental Epidemiology, (2003) 13-443-435, June 2003.

Fujita et. al.[6] provides data suggesting average heavy-duty EC/TC on the order of 0.62 and average LDV (or SI gasoline) containing mostly OC with EC/TC of 0.17 for high emitters and 0.31 for low emitters. Robert et. al., in a companion paper to their HDV results, provide data on light-duty gasoline vehicles.[7] Their results also show an increasing EC/TC with newer LEVs compared to older (and higher mileage) technologies (three way catalysts, oxidation catalysts, and no catalysts). Smoker or high emitters had the lowest EC/TC. Their data suggest the following EC/TC values:

- LEV 0.8
- TWC 0.45
- Oxidation Cat 0.06
- Non Cat 0.33
- Smoker 0.02

It is not clear why the oxidation catalyst equipped cars had such a high content of organic matter compared to EC. The total PM emissions for oxidation catalyst equipped vehicles were higher than the non catalyst equipped vehicles.

Conclusions and Implications

What does all this mean? Table 2 shows a possible scenario were the total PM emissions have increased but the measured EC has decreased. The top part of table 2 shows the emissions inventory from EMFAC2007. This is the same summary data shown previously in Table 1. What has been added is a guess at the EC/PM for LDV and HDVs.

As summarized above one can make the case for EC/PM or EC/TC[8] for HDVs to move from higher EC/PM to lower EC/PM with newer vehicles (newer technology). For 1999 (the year MATES II data were taken) one could speculate EC/TC of 0.9 (i.e. greater than 0.8 for a 1985 MY HD diesel engine). MATES III data was collected in 2005-2006. In this period both fuel reformulations (ultra low sulfur diesel fuel) and newer engine technologies were being introduced into the Basin fleet. Using an EC/TC representing

[6] Fujita, Eric M., Barbara Zielinska, David E. Campbell, W. Patrick Arnott, John C. Sagebiel, Lynn Mazzoleni, Judith C. Chow, Peter Gabele, William Crews, Richard Snow, Nigel N. Clark, W. Scott Wayne, and Douglas R. Lawson, "Variations in Speciated Emissions from Spark-Ignition and Compression – Ignition Motor Vehicles in California's South Coast Air Basin," Journal of the Air & Waste Management Association, Volume 57, June 2007, page 705

[7] Robert, Michael A, Saskia VanBergen, Michael J. Kleeman, and Christopher A. Jakober, "Size and Composition Distributions of Particulate Matter Emissions: Part 1—Light-Duty Gasoline Vehicles", Journal of the Air and Waste Management Association, Volume 57, pg 1414, December 2007.

[8] For this analysis I am assuming total carbon and particulate manner are the same.

engine MYs 1997 and 1998 may be a reasonable, accurate estimate of the fleet average. These values were 0.4 to 0.5.

Table 2. Estimated Elemental Carbon Changes for the Combined Gasoline and Diesel Fleet in the South Coast Air Basin

Inventory Element	*1999*	*2006*	Change
LDV PM (tpd)	2.77	4.15	50%
HDV PM (tpd)	13.51	13.75	2%
Total PM (tpd)	16.28	17.9	10%
Assumed EC/PM for LDV and HDV Technologies			
LDV	0.4	0.6	
HDV	0.75	0.45	
Estimated Elemental Carbon Emissions			
LDV (tpd)	1.108	2.49	125%
HDV (tpd)	10.13	6.18	-39%
Total (tpd)	11.24	8.67	-23%

Similarly, the data for light-duty vehicles have shown that EC/PM ratio has increased with newer technology. In 1999 a reasonable guess at EC/PM might be 0.4 — representative of normal emitting vehicles in the fleet equipped mostly with three way catalyst technology. In 2005-2006 the fleet would have more LEV technologies and a guess of the ratio might be 0.6—half way between 0.8 seen for LEV technologies and normal three way catalyst vehicles.

Finally, the bottom part of Table 2 shows the calculated EC estimated from the inventory estimates and the assumed EC/PM ratios for LD and HDVs. As shown, it is possible to have an increase in total PM emissions of 10% but have a 23% decrease in total EC. MATES III results report a decrease in EC and the inventory show an increase in PM emissions. Concluding a reduction in EC corresponds to a decrease in PM emissions may not be supportable. As illustrated, it is much more difficult to estimate DPM from EC since vehicle technologies affecting EC emissions are changing and older, high-mileage vehicles are being phased out. EC/PM is different for LDV compared to HDV and the trends with improving technology are also different. EC/PM for newer LDV technology increases and EC/PM for newer HDV technology decreases. The Basin fleet mix in 1999 was different enough compared to 2005-06 to result in different overall EC/PM ratios and therefore result in different estimates of PM emissions.