The effect on photochemical smog of converting the U.S. fleet of gasoline vehicles to modern diesel vehicles

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[1] With the increased use of particle traps and nitrogen oxide (NO_x) control devices to reduce air pollution, ''modern'' diesel vehicles are being encouraged over gasoline vehicles globally as a central method of slowing global warming. Data to date, though, suggest that the NO_2:NO ratio from modern diesel may exceed that of gasoline, and it is difficult to reduce diesel NO_x below gasoline NO_x without increasing particle emissions. Here, it is calculated that, unless the diesel NO_2:NO ratio and total NO_x are reduced to those of gasoline, modern diesel, which should have lower hydrocarbon (HC) and carbon monoxide (CO) emissions than gasoline, may still enhance photochemical smog at the surface and aloft, on average, over the U.S. relative to gasoline. The reason is that vehicle-produced smog in the U.S. depends more on NO_x and the NO_2:NO ratio than on HCs or CO. It is also found that vehicle NO_x controls may be more effective than NO_2:NO ratio controls at reducing ozone. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0317 Atmospheric Composition and Structure: Chemical kinetic and photochemical properties; 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry. **Citation:** Jacobson, M. Z., J. H. Seinfeld, G. R. Carmichael, and D. G. Streets (2004), The effect on photochemical smog of converting the U.S. fleet of gasoline vehicles to modern diesel vehicles, *Geophys. Res. Lett.*, *31*, L02116, doi:10.1029/2003GL018448.

1. Introduction

[2] Many countries are developing strategies to slow global warming. One strategy already being implemented in much of the European Union and considered in California is to encourage diesel over gasoline vehicles because diesel vehicles are thought to emit less CO_2 (see auxiliary material[1]) and ''modern'' diesel vehicles (those with particle traps and, potentially, NO_x control devices) are cleaner than diesel vehicles of the past. Despite this widespread belief, no study has examined the potential effects on air pollution of replacing gasoline with modern diesel vehicles. Given that changes in air pollution can affect health, such a study is warranted.

[3] Table 1 compares estimated emissions of several pollutants from modern gasoline and diesel passenger vehicles, where the diesel contains a particle trap but no NO_x device. In the absence of a trap (not shown), diesel CO and hydrocarbon (HC) emissions are similar to those from gasoline. A trap added to a diesel not only collects then burns off particles (during regeneration); it also catalyzes the oxidation of CO to CO_2 and HCs to CO_2 and H_2O.

[4] Diesel vehicles, with or without a trap and without a NO_x device, emit 4–30 times more NO_x than do gasoline vehicles (Table 1). Under California Low Emission Vehicle II emission standards (LEV II), NO_x emissions from passenger cars and light-duty vehicles must be <50 mg/mi (31 mg/km) and particle emissions must be <10 mg/mi (6.2 mg/km) by 2004. These are the toughest proposed standards worldwide. Currently, many new gasoline vehicles meet both standards and others are close (e.g., Table 1). Table 1 suggests that, with the addition of a particle trap, diesel may meet the particle standard but not the NO_x standard. As such, technologies to reduce diesel NO_x (and gasoline NO_x further) are being improved. A tradeoff, though, exists between NO_x and particle reductions in diesel [*Johnson*, 2001] such that although diesel NO_x emissions will decrease, they may or may not drop to those of gasoline because of the need to ensure maximum particle controls. Here, it is assumed that all diesel vehicles will have a NO_x device, reducing diesel NO_x emission to or nearly to that of gasoline.

[5] Another issue with modern diesel is the NO_2:NO ratio. In diesel vehicles without a trap and gasoline vehicles, the ratio is about 10:90 [*Ayala et al.*, 2002; *Ullman et al.*, 2002]. In Europe, many gasoline passenger vehicles emit about 4% NO_2 whereas light-duty trucks may emit about 18% NO_2 [*Soltic and Weilenmann*, 2002] again suggesting that a reasonable fleet-average of 10:90. In heavy-duty diesel vehicles with a particle trap, NO must be converted to NO_2, which oxidizes soot in the trap. The resulting NO_x exhaust contains 30–70% NO_2 [*Ayala et al.*, 2002; *Ullman et al.*, 2002; *Guo et al.*, 2003]. Some passenger vehicles with a trap use a catalyst, such as cerium, instead of NO_2, for oxidizing soot. In the exhaust of such vehicles, though, NO_2 is still 13–39% of NO_x [*PSA Peugeot-Citroen, pers. comm.*, 2002]. In sum, diesel vehicles with a trap and without an NO_x device emit more NO_x and a higher ratio of NO_2:NO than do gasoline vehicles.

[6] Here, the possible effects of replacing gasoline with diesel vehicles are examined when NO_x emissions are 0, 25,

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[1]Auxiliary material is available at ftp://ftp.agu.org/apend/gl/2003GL018448.

Table 1. Emissions from a diesel vehicle with a particle trap and without an NO_x device compared with those from a gasoline vehicle

	CO (g/km)	HCs (g/km)	NO_x (g/km)	% NO_x as NO_2	PM (mg/km)
Diesel, w/trap	0.10[a]	0.030[a]	0.34[a], 0.3–0.4[h]	13–39[b] 30–70[c,d,e]	1.5–15[a,g,h,i,j]
Gasoline	0.26[a]	0.059[a]	0.042[a], 0.01–0.1[h]	10[c,d,e] 4.3–17.6[f]	0.05–2[i,k]

This study assumes an NO_x device will be added to diesel, reducing NO_x emissions to 0–100% higher than those of gasoline.
[a]*Ahlvik* [2002] citing Peugot 307 HDi FAP diesel and Peugot 307 1.6 110 hp gasoline certification data.
[b]PSA Peugeot-Citroen, *pers. comm.* [2002].
[c]*Ayala et al.* [2002].
[d]*Guo et al.* [2003].
[e]*Ullman et al.* [2002].
[f]*Soltic and Weilenmann* [2002] (4.3% for passenger vehicles; 17.6% for light-duty trucks).
[g]*Salvat et al.* [2000].
[h]*Durbin and Norbeck* [2002].
[i]*ACEA* [2002].
[j]*Jeuland et al.* [2002].
[k]*Maricq et al.* [1999].

50, and 100% higher for diesel than for gasoline and NO_2:NO ratios are 10:90, 20:80, and 30:70 for diesel and 10:90 for gasoline. In all cases, modern diesel is assumed to emit 50% less HCs and CO and twice as much primary particle mass (Table 1). Since HC and CO emissions from diesel are assumed to be less than those of gasoline, it is assumed that diesel will pollute less than gasoline if its NO_x emission falls below that of gasoline.

[7] This study was carried out with GATOR-GCMOM, a nested global-to-urban pollution/weather/climate model [*Jacobson*, 1997a, 1997b, 2001a, 2001b, 2001c, 2001d, 2002]. It treated time-dependent gas, aerosol, radiative, dynamical, cloud, land, and ocean processes over two layers of nesting, the globe and the U.S. The U.S. resolution was 0.5°S-N × 0.75°W-E (about 55 km S-N × 68 km W-E), so cities were not resolved. The relatively coarse inner grid allows the whole U.S. to be treated but increases uncertainty, some of which is addressed in a previous study with nested grids [*Jacobson*, 2001b, Table 2].

[8] Gridded, hourly anthropogenic emissions of speciated total organic gas (TOG), NO_x, SO_x, CO, NH_3, $PM_{2.5}$, and PM_{10} for the U.S. and parts of Canada/Mexico were obtained from the 1999 U.S. National Emission Inventory [*USEPA*, 2003]. The inventory accounts for over 370,000 stack and fugitive sources, 250,000 area sources, and 1700 source classification code (SCC) categories of on- and nonroad mobile sources (including 837 categories of gasoline vehicles: motorcycles, cars, trucks, recreational/construction/farm/industrial vehicles, etc.). Yearly U.S. emissions of NO_x, CO, and TOG from gasoline-only mobile onroad plus nonroad sources in the inventory were 4.06×10^6, 7.92×10^7, and 1.09×10^7 metric tonnes/yr, respectively, representing 19.2%, 73.0%, and 37.0% of all U.S. anthropogenic emissions of these pollutants in the inventory. Other mobile-sources (mostly diesel) accounted for another 35.0% of NO_x but lower levels of other pollutants. The model also included biogenic emission of isoprene, monoterpenes, VOCs, and NO. The auxiliary material describes processes in the model and its initialization and compares model results with measurements.

2. Simulations and Results

[9] A baseline and six sensitivity simulations, in which all SCC categories of gasoline vehicles were switched to diesel (as described in Figure 1), were run for August and February, 1999. Figure 1 shows the August difference in the frequency distribution of hourly near-surface O_3 over the U.S. for the six cases. The switch to diesel increased O_3 > 60 ppbv in all six cases. The number of increases in case 2 (50% NO_x increase) was between those of cases 1 (25% NO_x increase) and 3 (100% NO_x increase). Changes in NO_2:NO at the same NO_x level (cases 2, 4, 5) had less effect on O_3 than did changes in total NO_x. For case 3, diesel increased O_3 > 120 ppbv by 51% and >80 ppbv by 17%.

[10] In case 6, ozone increased when NO_2:NO increased from 10:90 to 20:80 with no change in NO_x. This occurred

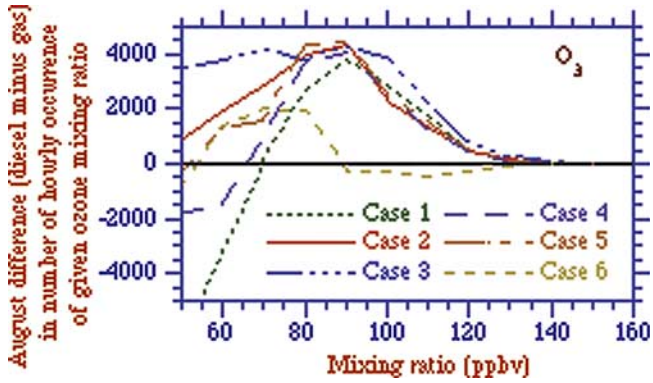

Figure 1. Difference (diesel minus gasoline) in the number of occurrences of different ozone mixing ratios (in 10 ppbv increments), summed over every hour of August and over every near-surface regional-domain (U.S.) grid cell, for six cases. Case 1: diesel:gasoline NO_x = 1.25 and diesel NO_2:NO = 20:80; Case 2: diesel:gasoline NO_x = 1.5 and diesel NO_2:NO = 20:80; Case 3: diesel:gasoline NO_x = 2.0 and diesel NO_2:NO = 20:80; Case 4: diesel:gasoline NO_x = 1.5 and diesel NO_2:NO = 10:90; Case 5: diesel:gasoline NO_x = 1.5 and diesel NO_2:NO = 30:70; Case 6: diesel:gasoline NO_x = 1.0 and diesel NO_2:NO = 20:80. In all cases, gasoline NO_2:NO = 10:90, diesel CO and primary HC emissions equaled half those of gasoline (Table 1), and diesel primary particle emissions were twice those of gasoline (a low estimate from Table 1). The summed number of occurrences of all ozone mixing ratios (down to 0–10 ppbv) was equal in all cases.

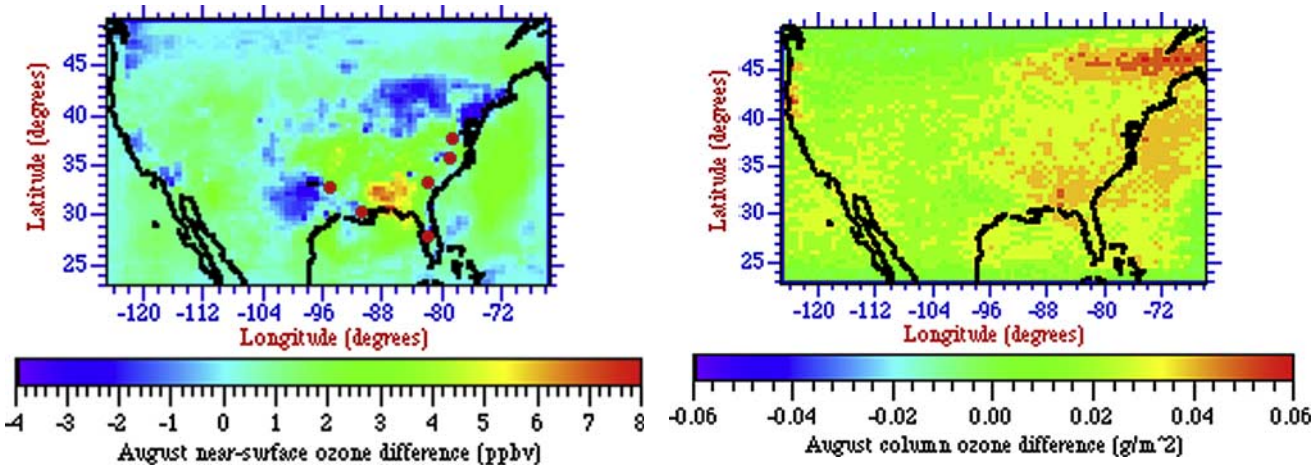

Figure 2. Modeled August difference (averaged over every hour in August) in (a) near surface O_3 and (b) column O_3 from case 2 of Figure 1. Red dots are locations for comparisons with data in the Supplementary Information.

because, when NO_x is emitted continuously as NO_2, O_3 forms almost directly by NO_2+UV light → NO+O followed by $O+O_2+M \rightarrow O_3+M$. When the same NO_x is emitted as NO, the NO must first react with O_3 or peroxy radicals to form NO_2 before creating new O_3, so this path destroys some of the O_3 that is created. There was little change in O_3 at high mixing ratios in case 6 possibly because additional O_3 formed by emitting NO_x as NO_2 at high O_3 was small relative to that formed by other means and/or because NO levels at high O_3 were high enough to titrate the additional O_3.

[11] Statistics for PAN, NO_2, HNO_3, particulate nitrate, and particulate BC also showed increases at all upper mixing ratios following the switch to diesel. Nitrate increases were 5–15 times greater than BC increases (which were small due to the small difference between diesel with a trap versus gasoline particulate emissions) and occurred primarily where HNO_3 increased (see auxiliary material). Primary particulate organic matter (OM) increased slightly, but secondary OM decreased, as expected. Statistics for CO and most primary HCs showed decreases at upper mixing ratios. Although overall mixing ratios of O_3 were lower in February than August, trends for February were similar to those for August. For example, for case 2 in February, diesel increased O_3 at all mixing ratios >45 ppbv.

[12] Figure 2a shows the August-average U.S. difference in O_3 following conversion to diesel in case 2. Ozone increased in more than 75% of the U.S. Most increases occurred in areas of high average O_3. The maximum increase (8 ppbv, in the Southeast) was twice the maximum decrease (4 ppbv, along the northeast corridor) and four times the decrease in Los Angeles (2 ppbv). Other O_3 decreases occurred in parts of the north-Midwest and Great Plains, where O_3 was relatively low. For these reasons, diesel increased the number of hours of high O_3 over the U.S. (Figure 1). At about 20% of Northeast locations where O_3 decreased overall, it increased in the afternoon, during its peak, but decreased more at night, when O_3 was low. In remaining decrease cases, O_3 decreased during day and night.

[13] Changes in O_3 in Figure 2a can be explained partly in terms of an ozone isopleth [e.g., *Seinfeld and Pandis*, 1998]. Increases in O_3 were caused by increases in NO_x in the presence of high total HCs, as expected from an isopleth. Although diesel reduced vehicle HCs, total HCs were high in the Southeast because biogenic HC emissions were high there (Figure 3). Decreases in O_3 were due to three factors: (1) in urban areas (e.g., Los Angeles, northeast corridor), increases in NO_x at high NO_x and at low biogenic HCs decreased O_3, (2) in areas of low biogenic HCs and moderate to high NO_x, reductions in vehicle HCs due to diesel decreased O_3 production, and (3) in areas of high nighttime NO_x, titration by excess NO_x decreased nighttime O_3. In nearly every area where O_3 decreased from diesel, biogenic HC emissions were low (compare Figures 2a and 3).

[14] Although near-surface O_3 decreased slightly in some places due to diesel, NO_2 increased in those locations (Supp. Info), and the number of increases in high-O_3 mixing ratios far outweighed the number of decreases (Figure 1). Ozone also increased in several grid cells containing nonattainment areas (e.g., Birmingham, Atlanta, Baton Rouge, St. Louis, Dallas-Fort Worth, Houston-Galveston-Brazoria) and areas close to nonattainment [*Chameides et al.*, 1997] (the model did not resolve the urban areas themselves).

[15] Figure 2b shows that diesel increased ozone column abundance across the entire U.S. Excess column O_3 can be

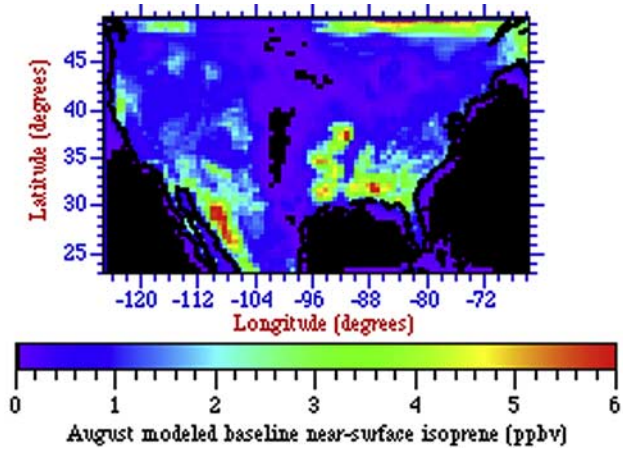

Figure 3. August baseline monthly-averaged modeled near-surface isoprene.

transported long distances and contributes to global warming. Excess O_3 aloft from diesel was either generated at the surface and convected aloft or generated from excess NO_x that escaped the boundary layer and reacted with background HCs and their products aloft. Winds aloft moved excess O_3 aloft faster than at the surface, increasing column O_3 over a wide area, including over areas where surface O_3 decreased.

[16] Diesel increased near-surface NO_2, PAN, and HNO_3 on average (auxiliary material)[1]. Since NO_2 and HNO_3 can only increase whereas O_3 can increase or decrease with an increase in NO_x, NO_2 and HNO_3 decreased in fewer areas than did O_3. Increases in HNO_3 are important because HNO_3 forms particles, which are known health hazards. Locations of PAN changes were similar to those of O_3 changes since PAN, like O_3, depends on HCs and NO_x. Mixing ratios of primary organics (e.g., primary aromatics, alkanes) decreased over the U.S. following the switch to diesel, as expected.

[17] The results here agree with those from several studies that have found that areas of high natural HC emission may be NO_x limited whereas urban areas (e.g., the northeast corridor, Los Angeles) may be HC limited [e.g., *Trainer et al.*, 1987; *Sillman et al.*, 1990; *NRC*, 1991; *Roselle et al.*, 1991; *McKeen et al.*, 1991; *Jacob et al.*, 1993; *Fiore et al.*, 1998; *Streets et al.*, 2001]. Here, it is similarly found that vehicle NO_x emission controls may be more effective than vehicle HC controls at reducing O_3 over most of the U.S., particularly in the Southeast, where biogenic emissions are high. An exception may be Los Angeles and along the Boston-Washington corridor, where natural HC emissions are low. This result is particularly consistent with that of *Jacob et al.* [1993] who modeled O_3 over the U.S. with resolution eight times coarser but found that a 50% decrease in NO_x decreased summer O_3 over the mid-southeastern U.S. by about 10–15 ppbv and slightly increased O_3 along the northeast coast, whereas a decrease in HCs of 50% decreased O_3 by only 3 ppbv.

[18] Previous studies have not examined the effect of NO_x versus HCs on column O_3 or pollutants aside from O_3 across the U.S or the effect of gasoline vehicle NO_x (as opposed to total NO_x) versus HC emission controls on near-surface O_3. The difference between gasoline vehicle emission and total emission is that the mass ratio of gasoline NO_x:HC in the U.S. is about 0.34:1 whereas that of total anthropogenic NO_x:HC is about 0.72:1. Finally, previous studies have not examined the effect of the NO_2:NO ratio on air pollution across the U.S. Here it was found that reducing the ratio may slightly reduce O_3, on average, but not so much as reducing NO_x.

3. Conclusion

[19] This study concludes that the replacement of gasoline with modern diesel vehicles in the U.S. may drive up photochemical smog, including total column ozone, near-surface ozone, and nitrogen-containing species, over the U.S. on average and in the Southeast in particular, unless NO_x emissions and the NO_2:NO ratio from diesel vehicles are reduced to or below those of gasoline vehicles. The study also finds that vehicle NO_x controls may be more effective than NO_2:NO ratio controls at reducing ozone. The results apply even though modern diesels emit less HCs and CO than gasoline vehicles because, on average, photochemical smog depends more on vehicle NO_x than hydrocarbon emissions. Thus, if gasoline vehicles meet the toughest planned NO_x regulations worldwide, and if diesel vehicles with a particle trap and NO_x device also meet such standards but emit NO_x at a slightly higher level, diesel vehicles may exacerbate smog relative to gasoline vehicles. To remedy this potential problem, technology is necessary to reduce NO_x and the NO_2:NO ratio of diesel to or below those of gasoline without increasing particulate matter emissions.

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