# Updates to

# Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming

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## Introduction

This document describes three updates that affect two figures (Figures 1 and 14) in the above-entitled paper (hereafter J2002): (1) An analysis suggests that the overall lifetime range of $CO_2$ should be 30-95 years instead of 50-200 years, (2) the $CO_2$ emission rate is no longer assumed to be in equilibrium with its atmospheric mixing ratio, and (3) for the diesel versus gasoline comparison, a comparison for a range of scenarios (15%-30% better mileage for diesel), rather than for one scenario (30% better mileage for diesel), is performed, based on data for high-mileage vehicles available in the U.S. The updates do not change the three main conclusions in J2002, namely,

(1) "any emission reduction of fossil-fuel particulate BC plus associated OM may slow global warming more than may any emission reduction of $CO_2$ or $CH_4$ for a specific period,"
(2) diesel cars emitting continuously under the most recent U.S. and E.U. particulate standards (0.08 g/mi; 0.05 g/km) may warm climate per distance driven over the next 100+ years more than equivalent gasoline cars,"
(3) Toughening vehicle particulate emission standards by a factor of 8 (0.01 g/mi; 0.006 g/km) does not change this conclusion, although it shortens the period over which diesel cars warm to 13-54 years,"

except that, for conclusion (1), the period in Figure 1 of J2002 during which eliminating all f.f. BC+OM has an advantage over all anthropogenic $CO_2$ decreases from about 25-

100 years to about 11-13 years and (3) the period in Figure 14 of J2002 during which gasoline vehicles may have an advantage broadens from 13-54 years to 10-100 years.

Early versions of the updated figures were first presented at several conferences and seminars [e.g., *Jacobson*, 2002, 2003a-f]. *Bond* [2003] subsequently but independently discussed parts of the second update. The purpose of this document is to describe the updates.

**1. Lifetime of $CO_2$**

In J2002, it was assumed that the lifetime of $CO_2$ was between 50 and 200 years. This range is commonly used in the literature. As shown here, though, the upper lifetime does not appear to be physical, even within the range of reasonable uncertainty, and the lower lifetime appears to be too high to explain the rate of change of the observed mixing ratio of $CO_2$.

The lifetime of $CO_2$ can be estimated from data with just a few parameters: (a) the current anthropogenic portion of the mixing ratio of $CO_2$, (b) the current rate of change of the mixing ratio of $CO_2$, and (c), the current anthropogenic emission rate of $CO_2$. Specifically, the rate of change of the mixing ratio ($\chi$, ppmv) of a well-mixed gas whose only source is emission is

$$\frac{d\chi(t)}{dt} = E - \frac{\chi(t)}{\tau} \qquad (1)$$

where $E$ is the emission rate (ppmv/yr) and $\tau$ is the overall *e*-folding lifetime (years) of the gas. Rearranging Equation 1 gives the lifetime as

$$\tau = \frac{\chi(t)}{E - \frac{d\chi(t)}{dt}} \qquad (2)$$

[e.g., *Gaffin et al.*, 1995]. Here, it is assumed that $\chi(t)$ is the anthropogenic mixing ratio of $CO_2$ (the difference between the current mixing ratio and that during preindustrial times) and $E$ is the anthropogenic emission rate. This requires the assumption that the preindustrial mixing ratio [$\chi_p(t)$=275 ppmv in 1750] of $CO_2$ is in equilibrium with its

natural emission rate, $E_p$. In other words, $\chi_p(t) = \tau E_p$, which is obtained by setting the derivative in Equation 1 to zero.

In the year 2000 ($t$=0), the overall mixing ratio of $CO_2$ was approximately 370 ppmv (Figure 1), so the anthropogenic portion was about $\chi(0)$=95 ppmv (=370-275 ppmv). From 1995-2000, the rate of change of the mixing ratio was about $d\chi(0)/dt$=1.8 ppmv/year (data from Figure 1). The global fossil-fuel emission rate of $CO_2$ in 2000 (and from 1995-2000) was near 6600 Tg-$CO_2$-C/yr (data from Figure 2).

**Figure 1.** Yearly and seasonal fluctuations in carbon dioxide mixing ratio at Mauna Loa Observatory, Hawaii, since 1958. Data from *Keeling and Whorf* [2003].

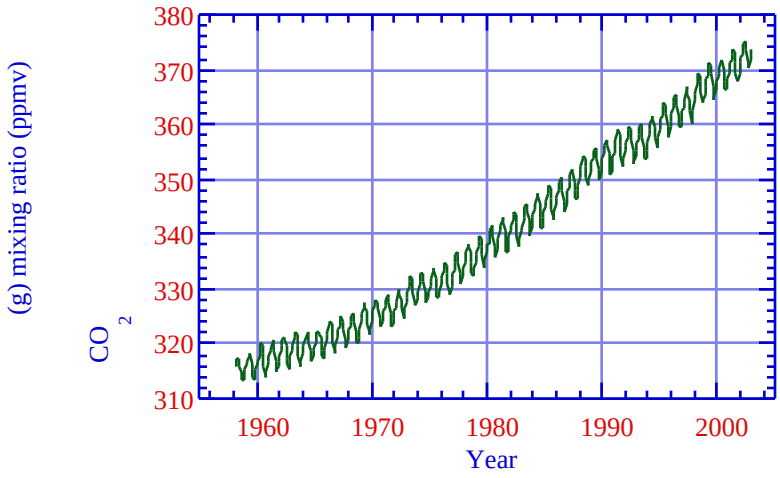
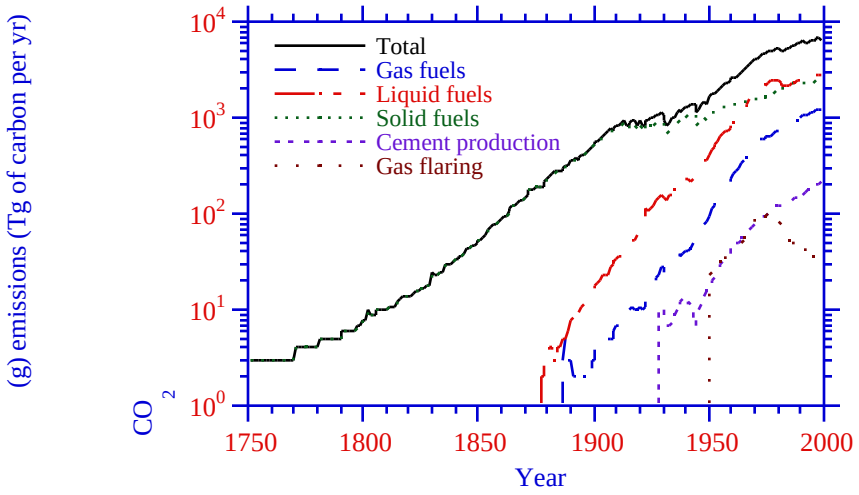



**Figure 2.** Global anthropogenic emission rate of carbon dioxide from 1750-2000 from *Marland et al.* [2003].

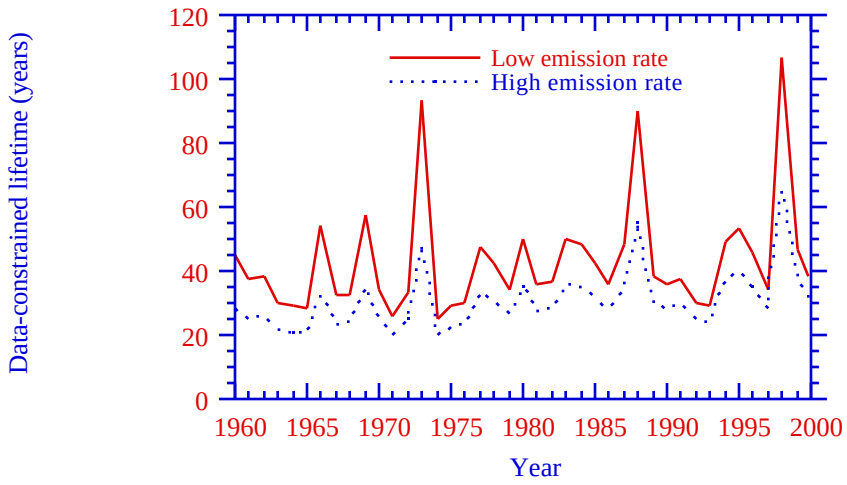



An estimated range of the anthropogenic portion of the outdoor biomass-burning emission rate is 1500-2700 Tg-$CO_2$-C/yr [*Jacobson*, 2003g]. With these numbers, total global anthropogenic emissions of $CO_2$ in 2000 may range from 8100-9300 Tg-$CO_2$-C/yr. With $1.095602 \times 10^{44}$ air molecules in the global atmosphere (column abundance of air of $2.14797 \times 10^{25}$ molec. $cm^{-2}$ and an area of the earth of $5.10064 \times 10^{18}$ $cm^2$), this translates to a globally-averaged emission rate of $E$=3.7074-4.2566 ppmv/yr (2184.82 Tg-$CO_2$-C/yr = 1 ppmv/yr). Substituting these numbers into Equation 2 gives an estimated data-constrained lifetime of $CO_2$ for the year 2000 of 39-45 years.

Figure 3 shows the data-constrained lifetime for the years 1960-2000, calculated using the methodology described above and data from Figures 1 and 2. The figure shows that the data-constrained lifetime ranged from 20-100 years, with an average value between 30.6 and 43 years. *Gaffin et al*. [1995] performed a similar calculation with slightly different assumptions (preindustrial mixing ratio of 280 instead of 275 ppmv, a single biomass-burning emission rate, and for the years 1959-1989) and found a mean lifetime on the order of 30 years. In no case in Figure 3 did the data-constrained lifetime approach 200 years. Based on Figure 3 and uncertainties associated with it, it is assumed

here that the lifetime of $CO_2$ ranges from 30-95 years although a more likely upper limit may be 50 or 60 years.

**Figure 3.** Data-constrained overall lifetime of $CO_2$ versus time calculated from Equation 2 using yearly ambient $CO_2$ mixing ratio data from *Keeling and Whorf* [2003], yearly fossil-fuel $CO_2$ emission data from *Marland et al.* [2003] and biomass-burning emission rates ranging from 1500-2700 Tg-$CO_2$-C/yr. The low and high emission rate curves in the figure represent the sum of the yearly fossil-fuel emission rate plus the fixed low or high biomass-burning emission rate. The 40-year (1960-2000) low- and high-emission rate mean data-constrained lifetimes are 43.0 and 30.6 years, respectively.




## 2. $CO_2$ emissions were no longer assumed to be in equilibrium

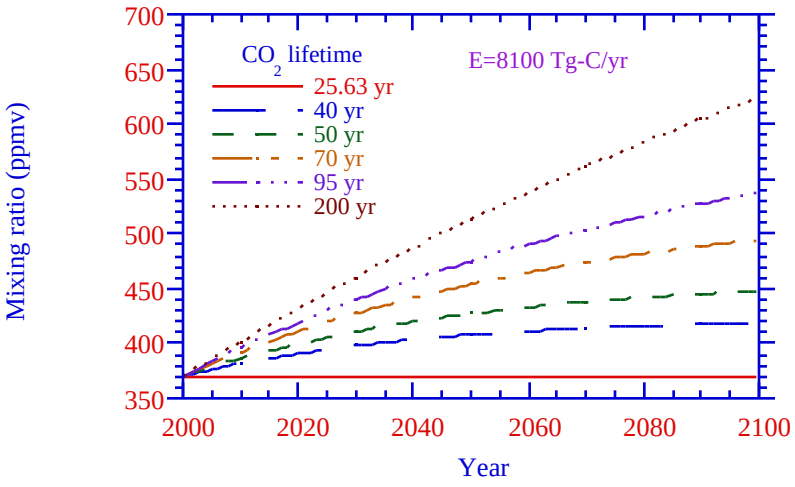
The second update here relates to the two $CO_2$ curves in Figure 1 of J2002. The curves represent the time-dependent difference in temperature resulting when anthropogenic $CO_2$ is not versus is emitted, each at a different assumed lifetime of $CO_2$ (50 or 200 years). The two curves were obtained by running global climate response calculations at current and pre-industrial mixing ratios of $CO_2$, then scaling the temperature difference over time proportionally to the $CO_2$ mixing ratio, where the $CO_2$ mixing ratio change with time was determined as a function of the $CO_2$ lifetime.

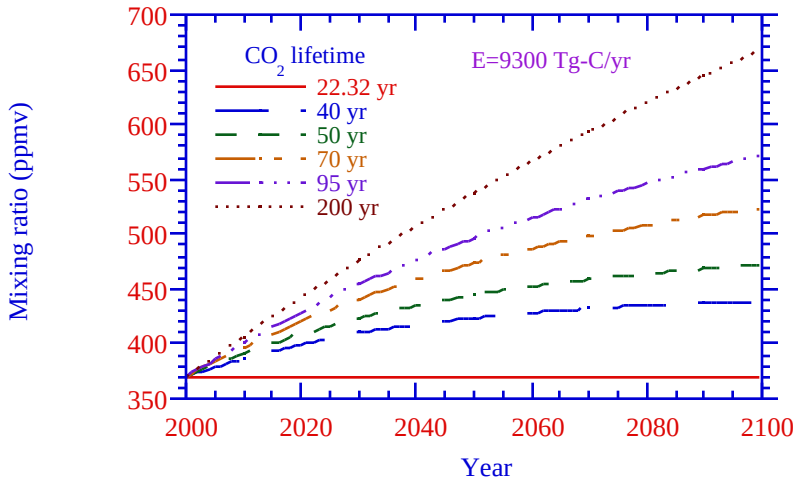
Mathematically, the analytical solution to the change in $CO_2$ mixing ratio with time can be obtained by integrating Equation 1 as

$$\chi(t) = \chi(0)e^{-t/\tau} + \tau E\left(1 - e^{-t/\tau}\right) \quad (3)$$

Figure 4 shows the time-dependent mixing ratio of $CO_2$ as a function of $CO_2$ lifetime for two respective emission rates from Equation 3. In each case, an "equilibrium lifetime" exists (25.63 and 22.32 y for the low and high emission rates, respectively), which is the lifetime at which the mixing ratio of $CO_2$ is always in equilibrium with a given emission rate (in other words, $CO_2$'s mixing ratio does not change over time when the emission rate is constant). This equilibrium lifetime is $\tau = \chi(0)/E$, derived by setting $\chi(t) = \chi(0)$ and solving for $\tau$ in Equation 3. It can also be derived by setting $d\chi(t)/dt = 0$ in Equation 1.

**Figure 4.** Time-dependent mixing ratio of $CO_2$ versus year as a function of $CO_2$ lifetime for two constant emission rates. From Equation 3 using 2184.82 Tg-$CO_2$-C/yr = 1 ppmv/yr and $\chi(0)$=95 ppmv.






The difference in the time-dependent mixing ratio from Equation 3 when $CO_2$ emissions are absent ($E$=0) versus present is

$$\Delta\chi(t) = [\chi(t)]_{\text{noemis}} - [\chi(t)]_{\text{w/emis}} \tag{4}$$

$$= \left[\chi(0)e^{-t/\tau}\right] - \left[\chi(0)e^{-t/\tau} + \tau E\left(1 - e^{-t/\tau}\right)\right]$$

$$= -\tau E\left(1 - e^{-t/\tau}\right)$$

In J2002, it was assumed that when $CO_2$ was emitted, its emission was in equilibrium with its ambient mixing ratio ($\tau = \chi(0)/E$). Substituting $\tau E = \chi(0)$ into Equation 4 gives

$$\Delta\chi(t) = -\chi(0)\left(1 - e^{-t/\tau}\right) \tag{5}$$

which was the mixing-ratio expression used to generate the $CO_2$ temperature-difference curves in Figure 1 of J2002. The equilibrium assumption $E = \chi(0)/\tau$ is always correct if either one of two conditions holds: (1) if $CO_2$'s emission rate is constant for a sufficiently long period ($t$»$\tau$ in Equation 4) or (2) for any period if $CO_2$'s lifetime equals its

equilibrium lifetime. When the emission rate is 9300 Tg-C/yr, for example, Figure 4 shows that the equilibrium assumption is correct for all lifetimes of $CO_2$ when $t \gg \tau$ or for any $t$ when $\tau$=22.3 years, $CO_2$'s equilibrium lifetime. Figure 3 (lower curve), though, shows that the average data-constrained lifetime of $CO_2$ is closer to 31 years when the emission rate (in the last years) is 9300 Tg-C/yr, suggesting that the lifetime of $CO_2$ is greater than its equilibrium lifetime (22.3 years).

$CO_2$'s mixing ratio can be in equilibrium with its emission rate today if its emission rate is held constant for next several decades. For example, given an average estimated $CO_2$ lifetime of 31 and 43 years from Figure 3 and current emissions of about 9300 and 8100 Tg-C/yr resulting in those lifetimes, the equilibrium mixing ratio of $CO_2$ in the two respective cases is 132 and 159 ppmv. At those mixing ratios, the current lifetime becomes the equilibrium lifetime. This result has physical significance. It suggests that, if its emission rate is held constant for the next several decades, $CO_2$'s mixing ratio should increase by 37 to 64 ppmv (132-95 and 159-95 ppmv). A similar calculation also shows that, for every 1000 Tg-C/yr increase in the emission rate, the mixing ratio should increase by another 14-20 ppmv. The last 1000 Tg-C/yr increase in the emission rate took about 14 years (1986-2000).

Another way for $CO_2$ emission to be in equilibrium with its mixing ratio is if the emission rate decreases. For example, at a 31-year lifetime, $CO_2$'s emission rate is in equilibrium with the mixing ratio if the emission rate decreases to 6695 Tg-$CO_2$-C/yr. In sum, Equation 5 is exactly correct under some conditions, but under current conditions, it is not.

**Figure 5.** Corrected Figure 1 of J2002. The figure shows the comparative cooling of global climate due to eliminating all anthropogenic emissions of f.f. BC+OM, $CH_4$ (with a 10-year *e*-folding lifetime) and $CO_2$ (with 30-, 50-, and 95-year lifetimes). It is obtained by subtracting Equation 7 from Equation 6 (or Figure 7 from Figure 6).

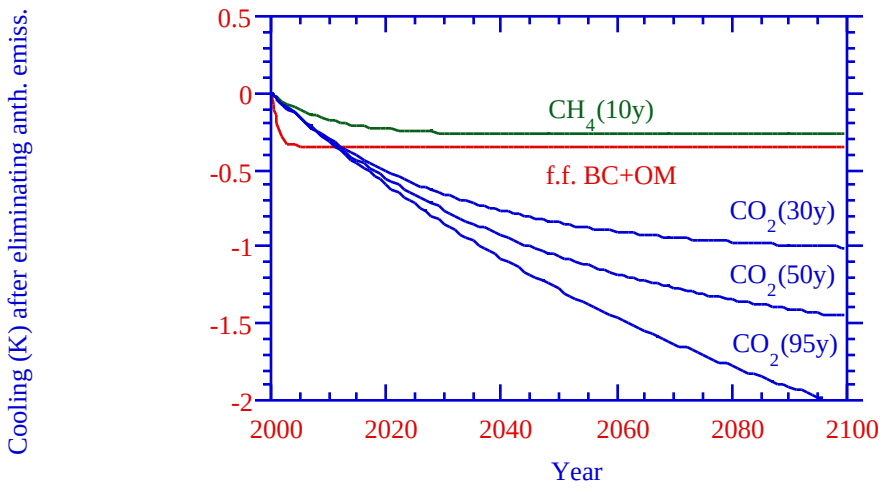



Figure 5 shows the modified temperature-change curves when Equation 4 instead of Equation 5 is used to model the $CO_2$ mixing ratio change with time and when the lifetime range of $CO_2$ is 30-95 years instead of 50-200 years. To generate the temperature difference curve in Figure 5, it is necessary to consider how temperature changes with an increase in $CO_2$ versus with a decrease in $CO_2$. J2002 calculated that removing all anthropogenic $CO_2$ ($\Delta\chi_{eq,dec}$=95 ppmv) might decrease temperature by $\Delta T_{eq,dec}$=–0.9 K, whereas doubling total $CO_2$ 370 to 740 ppmv ($\Delta\chi_{eq,inc}$=370 ppmv) might increase temperature by $\Delta T_{eq,inc}$=3.2 K. The difference in temperature change per unit mixing ratio arises because the climate response per unit mixing ratio is a function of the mixing ratio, itself, and the feedbacks associated with it. In sum, the temperature change curves in Figure 5 are calculated from the difference between curves in Figures 6 and 7. Figure 6 represents the temperature change due to the decrease in mixing ratio associated with Equation 4 (the difference between the initial mixing ratio and that in the absence of emission, which is the same as the mixing ratio difference arising when the emission rate is in equilibrium with the mixing ratio). This temperature change is

$$\Delta T_{dec}(t) = \left\{ \chi(0) - [\chi(t)]_{\text{noemis}} \right\} \frac{\Delta T_{eq,dec}}{\Delta \chi_{eq,dec}} = \chi(0)\left(1 - e^{-t/\tau}\right) \frac{\Delta T_{eq,dec}}{\Delta \chi_{eq,dec}} \qquad (6)$$

Figure 7 represents the temperature change due to the increases in mixing ratio associated with Equation 4 (the difference between the mixing ratio at a constant emission rate and the initial mixing ratio). This temperature change is

$$\Delta T_{dec}(t) = \left\{ [\chi(t)]_{\text{w/emis}} - \chi(0) \right\} \frac{\Delta T_{eq,inc}}{\Delta \chi_{eq,inc}} = \left(\tau E - \chi(0)\right)\left(1 - e^{-t/\tau}\right) \frac{\Delta T_{eq,inc}}{\Delta \chi_{eq,inc}} \qquad (7)$$

Subtracting Equation 7 from Equation 6 (Figure 7 from Figure 6) gives the curves in Figure 5.

**Figure 6.** Temperature change (from Equation 6) due to eliminating anthropogenic $CO_2$ emission when, in the presence of emission, the emission rate is in equilibrium with the ambient mixing ratio.

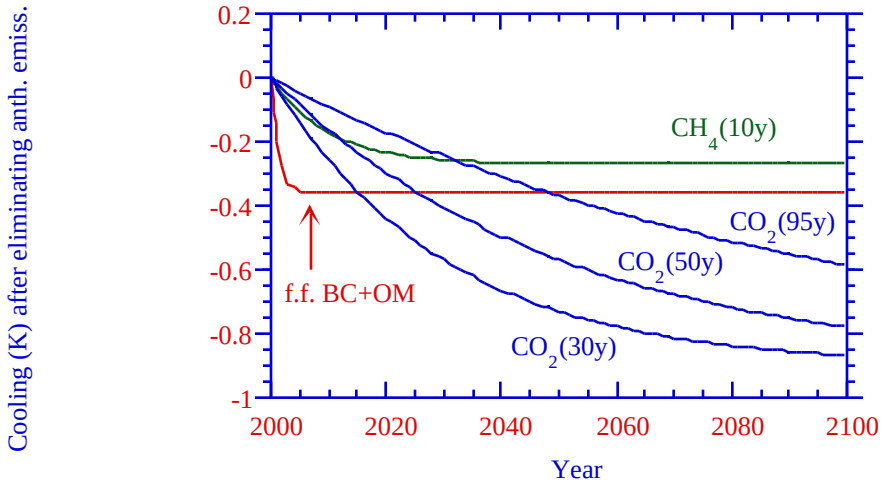



**Figure 7.** Temperature change (from Equation 7) due to the increase in $CO_2$ mixing ratio at a constant emission rate, which occurs because the $CO_2$ emission rate is not in equilibrium with its ambient mixing ratio, except at a lifetime of below 30 years.

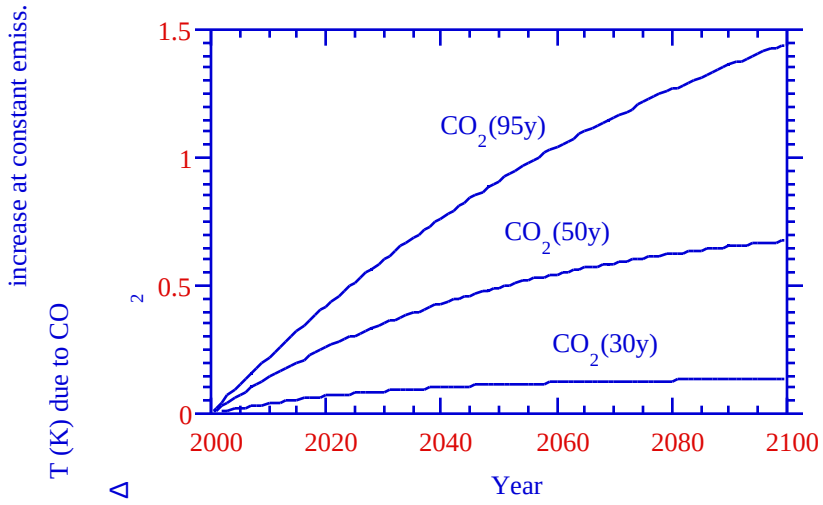



Figure 5 represents a more realistic time series than Figure 1 of J2002. Even after the modification, though Figure 5 still shows that controlling all f.f. BC+OM has an advantage over controlling all anthropogenic $CO_2$, but for a shorter period (about 11-13 years) than does Figure 1 of J2002 (25-100 years). Thus, the conclusion in J2002 that controlling f.f. BC+OM may be the most effective method of slowing global warming for a specific period still holds, but for a shorter period than originally estimated.

**3. Comparison of diesel versus gasoline**

Third, the comparison of diesel versus gasoline, embodied in Figure 14 of J2002, was updated to account for (1) the revision to Figure 1 of J2002, as shown in Figure 5 here and (2) a range of mileage differences of diesel versus gasoline rather than one difference. In addition, a lower estimate of the density of diesel (840 g/L) than the 856 g/L used in J2002, was assumed (a modification that benefits diesel).

In J2002, it was assumed that diesel vehicles obtained 30% better mileage than equivalent gasoline vehicles. This assumption, though, does not apply to the highest-mileage vehicles in the U.S. nor does is necessary apply to vehicles compared based on

their costs to society. Whereas, consumer decisions are based on cost of a product, policy decisions are based on costs to society, which include externality costs related to air pollution. The highest-mileage gasoline vehicles available in the U.S. not only emit comparable $CO_2$ as do the best diesel vehicles, but they also emit lower levels of particulate matter and oxides of nitrogen. The addition of particle traps and $NO_x$ control devices may or may not reduce the air pollution damage due to diesel, depending on the level of reduction (Jacobson et al., 2003h), but the point is moot since U.S. vehicles do not presently include such devices. In addition, the addition of such devices decreases the fuel efficiency of diesel vehicles. Here, new curves are derived that consider mileage differences of such high-mileage cars as well as differences of low-mileage cars.

Table 1 shows the highest-mileage diesel and gasoline vehicle available in the U.S. The table shows that the highest-mileage diesel vehicle obtains only 14% better mileage than the highest-mileage gasoline vehicle (45.5 versus 40 mpg). This translates into only slightly greater $CO_2$ emissions for the gasoline vehicle (Table 1). The addition of a particle trap to the diesel increases its fuel use by 3.5-8.5% [*Salvat et al.*, 2000, *Ullman et al.*, 2002; *Durbin and Norbeck,* 2002]. Assuming a 5% increase, the same diesel now emits more $CO_2$ per unit distance than does the gasoline (Table 1). In all cases, gasoline-electric hybrid vehicles available in the U.S. emit less $CO_2$ than diesel with or without a trap.

Table 1. Highest-mileage passenger vehicles in the U.S. in 2003, ranked by their $CO_2$ emissions (with and without a particle trap in the case of diesel).

| Vehicle | Energy source | Avg. mpg | $CO_2$ (g-C/km) | $CO_2$ (g-C/km) w/trap |
|---|---|---|---|---|

| | | | | |
|---|---|---|---|---|
| Honda Insight (M) | Gas/electric | 64.5 | 23.0 | |
| Honda Insight (A) | Gas/electric | 56.5 | 26.2 | |
| Toyota Prius (A) | Gas/electric | 48.5 | 30.6 | |
| Honda Civic (M) | Gas/electric | 48.5 | 30.6 | |
| Honda Civic (M) | Gas | 40 | 37.1 | |
| Toyota Echo (M) | Gas | 39 | 38.0 | |
| VW Golf, Jetta (M) | Diesel | 45.5 | 37.8 | 39.7 |
| VW Golf, Jetta (A) | Diesel | 39.5 | 43.5 | 45.7 |

(A) denotes automatic transmission; (M) denotes manual transmission. The table assumes a gasoline and diesel density of 737 g/L and 840 g/L, respectively, a gasoline and diesel carbon content of 85.5% and 87.0%, respectively, and an increase in fuel use with a trap+filter of 5% (see text). Source of fuel economy: *DOE* [2003].

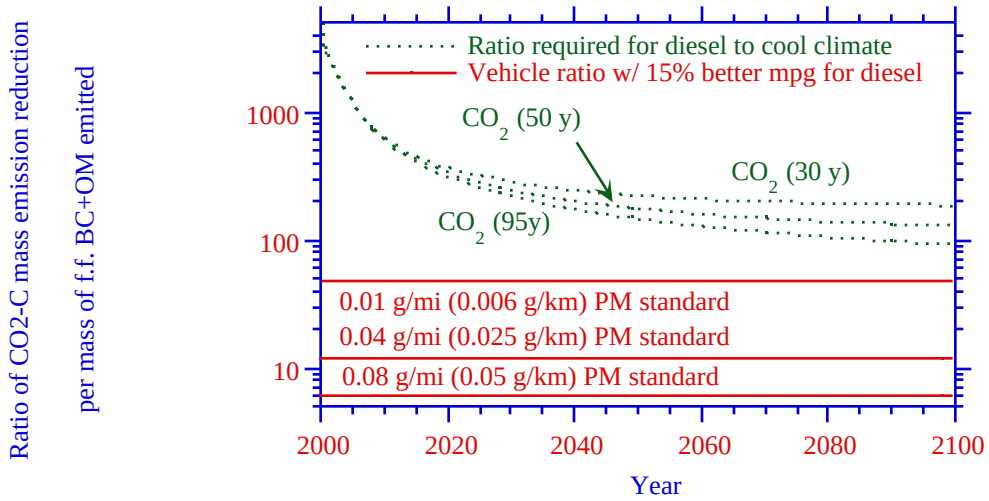
Here, the effect of diesel versus gasoline on climate is examined when a range of mileage differences between diesel and gasoline (15-30% better for diesel instead of just 30% better, assumed in Figure 14 of J2002) is considered. When the mileage of a diesel is <13% better than that of gasoline, gasoline always has a climate advantage, so no curves are shown for those cases. The updated result also accounts for the modified temperature-change curves in Figure 5, and a range of $CO_2$ lifetimes from 30-95 years.

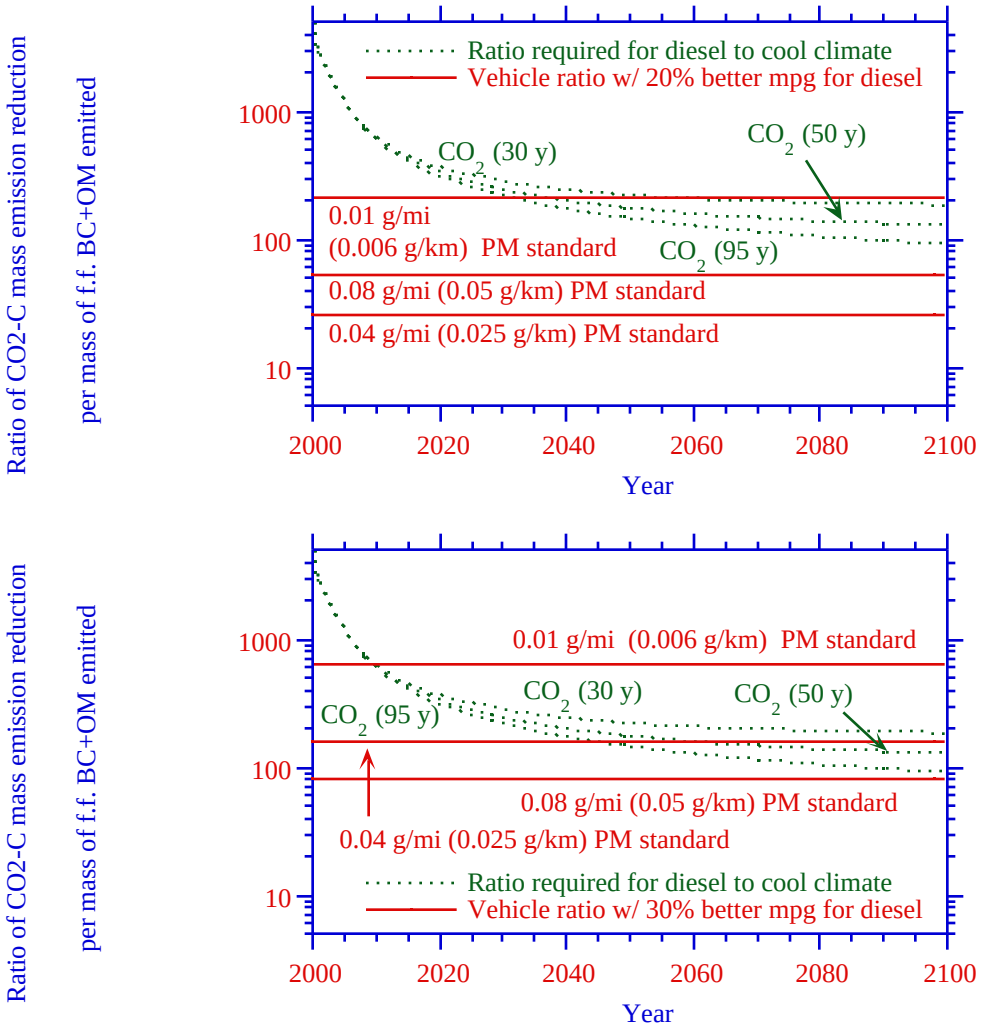
Figure 8 shows that, in all cases where diesel vehicles emit at a PM standard of 0.08 g/mi, diesel vehicles emitting continuously may warm climate more than gasoline vehicles for >100 years. In all cases where diesel mileage is 15% greater than that of gasoline, diesel also warms climate for more than 100 years. At 20% better mileage, diesel warms more than gasoline for >100 years when the PM emission rate is 0.04 g/mi or greater and for 30-50 years when the PM emission rate is 0.01 g/mi. At 30% better mileage, diesel warms for >100 years at 0.08 g/mi, 45-100 y at 0.04 g/mi, and 10 y at 0.01 g/mi.

Figure 8 shows that, under the assumptions of the figures, toughening particle standards by a factor of 8 (from 0.08 g/mi to 0.01 g/mi) still allows diesel to warm

climate for 10-100 years (when diesel mileage was 15-30% better than that of gasoline), a slightly broader range than found in J2002, calculated as 13-54 years when one mileage difference (30% better for diesel) was considered. For the 30% case, the new advantage of gasoline is 10 years at a 0.01 mg/mi standard instead of 13-54 years as in J2002. The benefit of gasoline, though, increases superlinearly with decreasing mileage benefit of diesel.

**Figure 8.** Comparison of the modeled ratio of the $CO_2$-C emission reduction required per unit of f.f. BC+OM emitted required for diesel vehicles to cool climate on the global average to the actual ratio obtained when a diesel versus a gasoline vehicle is used. The three subfigures represent cases when the diesel vehicle obtains (a) 15%, (b) 20%, and (c) 30% better mileage than the gasoline vehicle. The modeled curves (the same in all three subfigures) were obtained by dividing the f.f. BC+OM-temperature curve in Figure 5 by each $CO_2$-temperature curve (30 y, 50 y, 95 y) then multiplying the result by the yearly emission rate of $CO_2$ (8100 Tg-C/yr) and dividing by that of BC and associated OM from fossil fuels (5.1 Tg/yr BC+10.1 Tg/yr OM). The modeled curves shows that a yearly 1 Tg/yr decrease in f.f. BC+OM emissions will cool climate by about 4200-4500 times more than will a 1 Tg/yr decrease in $CO_2$-C emissions during 1 year. After 100 years of continuous 1 Tg/yr decreases in both, the resulting ratio of f.f. BC+OM to $CO_2$-C cooling is 90-190:1. The three solid, straight lines in each figure represent the actual ratio of $CO_2$-C saved to f.f. BC+OM emitted for a modern diesel vehicle emitting 0.08, 0.04, and 0.01 g/mi BC+OM. The intersection of each straight line with each modeled curve indicates the period of time during which diesel vehicles enhance global warming in comparison with gasoline vehicles under the given emission standard. In the case of the 0.08 g/mi standard, the period of time is >100 years for all $CO_2$ lifetimes and differences in diesel versus gasoline mileage.






Finally, Figure 8 (and Figure 14 of J2002) should be viewed cautiously when considering the comparison at a 0.01 g/mi standard. First, regardless of whether gasoline or diesel cools at that level, the total mass of emission is small at that standard, so the magnitude of cooling or warming by either vehicle type at that level will be small (e.g., more than an order of magnitude smaller than any cooling or warming due to f.f. BC+OM from vehicles today). Second, gasoline vehicles also emit particles. Although such emissions are generally lower than those of diesel with a trap (see discussion above), Figure 8 can be applied correctly for the 0.01 g/mi standard only if it is assumed

that diesel PM emissions are equal to gasoline PM emissions (generally 0.05-2 mg/km) plus the standard.

## 4. Summary

Two figures in J2002 were updated. The updates do not change the main conclusions in J2002 regarding the relative benefit of f.f. BC+OM control versus $CO_2$ control and that of gasoline versus diesel, except to the extent that the period over which f.f. BC+OM has an advantage is changed, as described in the introduction. Because $CO_2$ mixing ratios are expected to increase for awhile, even at a constant emission rate, due to the fact that the lifetime of $CO_2$ exceeds its equilibrium lifetime (the lifetime at which the $CO_2$ emission rate is in equilibrium with its ambient mixing ratio), control of global warming requires the control of both $CO_2$ and f.f. BC+OM.

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