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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Abstract

This document describes two updates and a correction that affect two figures (Figures 1 and 14) in *Jacobson* [2002] (hereafter J2002). The modifications have no effect on the numerical simulations in the paper, only on the post-simulation analysis. The changes include the following (1) The overall lifetime of CO_2 is updated to range from 30-95 yr instead of 50-200 yr, (2) the assumption that the anthropogenic emission rate of CO_2 is in equilibrium with its atmospheric mixing ratio is corrected, and (3) data for high-mileage vehicles available in the U.S. are used to update the range of mileage differences (15%-30% better for diesel) in comparison with one difference previously (30% better mileage for diesel). The modifications do not change the 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," and (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. 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 as follows: for conclusion (1), the period in Figure 1 of J2002 during which eliminating all fossil-fuel black carbon plus organic matter (f.f. BC+OM) has an

advantage over all anthropogenic CO_2 decreases from 25-100 yr to about 11-13 yr and for conclusion (2) the period in Figure 14 of J2002 during which gasoline vehicles may have an advantage broadens from 13 to 54 yr to 10 to >100 yr. Based on the revised analysis, the ratio of the 100-yr climate response per unit emission of f.f. BC+OM relative to that of CO_2 is estimated to be about 90-190.

1. Lifetime of CO_2

In J2002, it was assumed that the atmospheric lifetime of CO_2 against all loss processes combined was between 50 and 200 yr. This range is commonly used in the literature. However, 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 data-constrained overall lifetime of CO_2 can be estimated as follows. First, the rate of change of the mixing ratio (χ, 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 τ 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. These assumptions require the further

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 [*Keeling and Whorf*, 2003], 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/yr [*Keeling and Whorf*, 2003]. The global fossil-fuel emission rate of CO_2 in 2000 (and from 1995-2000) was near 6600 Tg-CO_2-C/yr [*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*, 2004a]. Thus, the total global anthropogenic emission of CO_2 in 2000 may have ranged from 8100-9300 Tg-CO_2-C/yr. With 1.095602×10^{44} air molecules in the global atmosphere (column abundance of air of 2.14797×10^{25} molec. cm^{-2} and an area of the earth of 5.10064×10^{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 the numbers above into Equation 2 gives an estimated data-constrained lifetime of CO_2 for the year 2000 of 39-45 yr.

Figure 1 shows the data-constrained lifetime of CO_2 for 1960-2000, calculated using the methodology described. The lifetime ranged from 20-100 yr, with an average between 30.6-43 yr. *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 yr. In no case in Figure 1 did the data-constrained lifetime approach 200 yr. Based on Figure 1 and uncertainties associated with it, it is assumed here that the lifetime of CO_2 ranges from 30-95 yr although a more likely upper limit may be 50 or 60 yr.

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

The second update relates to the two CO_2 curves in Figure 1 of J2002. The curves show the time-dependent temperature difference resulting when anthropogenic CO_2 is not versus is emitted, each at a different assumed overall lifetime of CO_2 (50 or 200 yr). The curves were obtained by running global climate response calculations at current and pre-industrial mixing ratios of CO_2, then scaling the resulting equilibrium temperature difference over time proportionally to the CO_2 mixing ratio, which was assumed to be in equilibrium with its emission rate. Whereas the equilibrium assumption holds under the current CO_2 emission rate if CO_2's lifetime were shorter (e.g. ~25 yr or less) than it currently is or if CO_2's anthropogenic emission rate were lower than it currently is, this assumption is not valid under the current data-constrained lifetime or anthropogenic emission rate of CO_2. Here, this assumption is corrected.

Integrating Equation 1 gives the analytical solution to the change in CO_2 mixing ratio over time as

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

Figure 2 here 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 yr and 22.32 yr 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 is constant 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 τ in Equation 3. It can also be derived by setting $d\chi(t)/dt = 0$ in Equation 1.

The difference in the time-dependent mixing ratio when anthropogenic CO_2 emission is absent versus present is

$$\Delta\chi(t) = \left[\chi(t)\right]_{\text{noemis}} - \left[\chi(t)\right]_{\text{w/emis}} = -\tau E\left(1 - e^{-t/\tau}\right) \tag{4}$$

where

$$\left[\chi(t)\right]_{\text{w/emis}} = \chi(0)e^{-t/\tau} + \tau E\left(1 - e^{-t/\tau}\right) \qquad \left[\chi(t)\right]_{\text{noemis}} = \chi(0)e^{-t/\tau} \tag{5}$$

are obtained from Equation 3 when $E \neq 0$ and $E=0$, respectively.

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

$$\Delta\chi(t) = -\chi(0)\left(1 - e^{-t/\tau}\right) \tag{6}$$

which was the mixing-ratio expression used to generate the CO_2 temperature-difference curves in Figure 1 of J2002.

The equilibrium assumption is always correct when either (a) CO_2's lifetime equals its equilibrium lifetime ($\tau=\tau_{eq} = \chi(0)/E$, where E is the actual emission rate) for any time t, (b) CO_2's emission rate is constant for a sufficiently long period ($t \gg \tau$ in Equation 4), or (c) CO_2's emission rate equals its equilibrium emission rate ($E=E_{eq} = \chi(0)/\tau$, where τ is the actual lifetime).

For example, when CO_2's actual emission rate is 9300 Tg-C/yr, Figure 2b shows that the equilibrium assumption is correct (a) for any t when CO_2's actual lifetime equals its equilibrium lifetime, τ_{eq}=22.3 yr or (b) for all lifetimes when $t \gg \tau$. Alternatively, the equilibrium assumption is correct (c) at an actual CO_2 lifetime of 31 yr (Figure 1, lower

curve) if CO_2's emission rate decreases to the equilibrium emission rate of E_{eq}=6695 Tg-CO_2-C/yr.

Figure 2, however, shows that under the current estimated range of CO_2 emission (8100-9300 Tg-C/yr) and under the current estimated range of CO_2 lifetime (30-95 y, from Figure 1), the mixing ratio of CO_2 is not in equilibrium with its emission rate. As such, the CO_2 mixing ratio will increase with time at a constant emission rate. For example, for average estimated CO_2 lifetimes of 31 yr and 43 yr from Figure 1 and a current emission rate of about 9300 and 8100 Tg-C/yr resulting in those respective lifetimes, the anthropogenic CO_2 mixing ratio will increase from 95 ppmv to 132 and 159 ppmv, respectively over the next 100 y. Similarly, for every 1000 Tg-C/yr increase in the emission rate, the mixing ratio should increase by another 14-20 ppmv.

To revise Figure 1 of J2002 with the information above, it is necessary to recalculate the estimated temperature change over time due to the time-dependent mixing ratio change from Equation 4. Climate-response simulations from J2002 showed that the temperature change per unit mixing ratio of CO_2 differed upon a decrease (eliminating all anthropogenic emission) of CO_2 versus an increase (doubling) of CO_2. Eliminating the anthropogenic mixing ratio of CO_2 ($\Delta\chi_{eq,dec}$=-95 ppmv) resulted in an equilibrium temperature decrease of $\Delta T_{eq,dec}$=–0.9 K whereas doubling CO_2 ($\Delta\chi_{eq,inc}$=370 ppmv) resulted in an equilibrium temperature increase of $\Delta T_{eq,inc}$=3.2 K. The reason for the different climate response per unit mixing ratio is that the response is a function of the mixing ratio itself and the feedbacks associated with it.

The time-dependent temperature change accounting for the different climate responses upon a decrease or increase in mixing ratio is

$$\Delta T(t) = \left\{\left[\chi(t)\right]_{\text{noemis}} - \chi(0)\right\}\frac{\Delta T_{eq,dec}}{\Delta\chi_{eq,dec}} + \left\{\chi(0) - \left[\chi(t)\right]_{\text{w/emis}}\right\}\frac{\Delta T_{eq,inc}}{\Delta\chi_{eq,inc}} \qquad (7)$$

$$= \chi(0)\left(e^{-t/\tau} - 1\right)\frac{\Delta T_{eq,dec}}{\Delta\chi_{eq,dec}} + \left(\chi(0) - \tau E\right)\left(1 - e^{-t/\tau}\right)\frac{\Delta T_{eq,inc}}{\Delta\chi_{eq,inc}}$$

where the second expression was obtained by substituting Equation 5 into the first. This equation differs from that used in J2002 only in that J2002 assumed $\tau E = \chi(0)$, resulting in $\Delta T(t) = \chi(0)\left(e^{-t/\tau} - 1\right)\Delta T_{eq,dec}/\Delta\chi_{eq,dec}$.

Figure 3 shows modified time-dependent temperature-change curves when Equation 7 is used and when the lifetime of CO_2 ranges from 30-95 yr instead of 50-200 y. A similar curve, but based on a new set of simulations accounting for the effects of soot on snow albedo, is given in *Jacobson* [2004b].

After the modification, Figure 3 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 y) than does Figure 1 of J2002 (25-100 y). 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 3 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).

J2002 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. Table 1, for example, shows the highest-mileage diesel, gasoline, and gasoline-electric hybrid vehicle available in the U.S. in 2005. The table shows that the highest-mileage diesel vehicle obtains only 5% better mileage than does the highest-

mileage gasoline vehicle (42 mpg versus 40 mpg). This translates into greater CO_2 emissions for the highest-mileage diesel vehicle since diesel fuel has a greater density and carbon content than does gasoline (Table 1). The addition of a particle trap to a diesel vehicle 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, diesels with a trap emit even more CO_2 per unit distance than do the gasoline vehicles (Table 1). In all cases, gasoline-electric hybrid vehicles available in the U.S. emit less CO_2 than do diesel with or without a trap and gasoline vehicles.

Here, the effect of diesel versus gasoline on climate is reexamined when a range of mileage differences between diesel and gasoline (15-30% better for diesel instead of just 30% better, which was assumed in Figure 14 of J2002) is considered. When the mileage of a diesel is <13% better than that of gasoline (e.g., all cases in Table 1), gasoline vehicles are always found to have a climate advantage, so no curves are shown for those cases. The updated result also accounts for the modified temperature-change curves in Figure 3 and a CO_2 lifetime range of 30-95 y.

Figure 4a,b shows that, when diesel vehicles achieve 30% or 15% higher mileage than do gasoline vehicles, diesel vehicles emitting particles continuously at a PM standard of 0.08 g/mi may warm climate more than gasoline vehicles for >100 yr for all CO_2 lifetimes. When diesel achieves 15% higher, but not 30% higher, mileage than does gasoline, diesel vehicles emitting particles continuously at a tougher PM standard of 0.01 g/mi may also warm climate for more than 100 y.

J2002, calculated that, when diesel achieves 30% higher mileage than gasoline, diesel vehicles emitting 0.01 g/mi continuously for 100 yr may warm climate for 13-54 yr relative to gasoline vehicles. Based on the revised results in Figure 4b here, diesel may warm climate relative to gasoline for about 10 yr at 30% higher mileage. Because no

diesel vehicle available in the U.S. in 2005 emits less CO_2 than does the best gasoline vehicle available (Table 1), the 30% scenario is not applicable for the best available vehicles. As such, the upper end of the warming period due to diesel over gasoline must be >100 y.

Figure 4 (and Figure 14 of J2002) should be viewed cautiously, though, 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. Second, gasoline vehicles also emit particles (generally 0.00008-0.003 g/mi, or 0.05-2 mg/km). Although such emissions are generally lower than those of diesel vehicles with a trap, Figure 4 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 plus the standard.

Finally, the caption from Figure 4 suggests that the 100-yr climate-response per unit emission rate of f.f. BC+OM relative to that of CO_2, may range from about 90-190.

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 that they modify the period over which f.f. BC+OM has an advantage.

5. References

Department of Energy (DOE), Fuel Economy Ratings, 2005; see www.fueleconomy.gov.

Durbin, T., and J. M. Norbeck, Comparison of emissions for medium-duty diesel trucks operated on California in-use diesel, ARCO's EC-diesel, and ARCO EC-diesel with a

diesel particulate filter, Final Report to National Renewable Energy Laboratory Under Contract #ACL-1-30110-01 and the Ford Motor Company, July, 2002.

Gaffin, S.R., B.C. O'Neill, and M. Oppenheimer, Comment on "The lifetime of excess atmospheric carbon dioxide" by Berrien Moore III and B.H. Braswell, *Global Biogeochemical Cycles*, *9*, 167-169, 1995.

Jacobson, M.Z., Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming, *J. Geophys. Res., 107*, (D19), 4410, doi:10.1029/ 2001JD001376, 2002.

Jacobson, M. Z., The short-term cooling but long-term global warming due to biomass burning, *J. Clim.*, 17 (15), 2909-2926, 2004a.

Jacobson, M.Z., The climate response of fossil-fuel and biofuel soot, accounting for soot's feedback to snow and sea ice albedo and emissivity, *J. Geophys. Res.*, *109*, D21201, doi:10.1029/2004JD004945, 2004b.

Jacobson, M. Z., J. H. Seinfeld, G. R. Carmichael, and D. G. Streets, The effect on photochemical smog of converting the U.S. fleet of gasoline vehicles to modern diesel vehicles, submitted, 2003h.

Keeling, C. D. and T. P. Whorf, Atmospheric CO_2 concentrations (ppmv) derived from in situ air samples collected at Mauna Loa Observatory, Hawaii, http://cdiac.esd.ornl.gov/ftp/maunaloa-co2/maunaloa.co2, 2003.

Marland, G., T. A. Boden, and R. J. Andres, Global CO_2 emissions from fossil-fuel burning, cement manufacture, and gas flaring: 1751-2000. In Trends Online: A compendium of data on global change. Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tenn., USA, 2003.

Salvat, O., P. Marez, and G. Belot, Passenger car serial application of a particulate filter system on a common rail direct injection diesel engine, SAE 2000-01-0473, 2000.

Ullman, T. L., L. R. Smith, J. W. Anthony, Exhaust emissions from school buses in compressed natural gas, low emitting diesel, and conventional diesel engine configurations, Southwest Research Institute Report 08.05303, 2002.

Table 1. Highest-mileage passenger vehicles in the U.S. in 2005, 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	63.5	23.4	
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 (A)	Gas/electric	47.5	31.2	
Honda Civic (M)	Gas	40	37.1	
Toyota Echo (M)	Gas	38.5	38.5	
VW N. Beetle, Golf, Jetta (M)	Diesel	42	41.0	43.1
VW N. Beetle (A)	Diesel	39	44.1	46.3

(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* [2005].

Figure Captions

Figure 1. 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 [*Jacobson*, 2004a]. 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-yr (1960-2000) low- and high-emission rate mean data-constrained lifetimes are 43.0 and 30.6 y, respectively.

Figure 2. 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.

Figure 3. 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-yr *e*-folding lifetime) and CO_2 (with 30-, 50-, and 95-yr lifetimes). It is obtained from Equation 7.

Figure 4. Comparison of the modeled ratio of the CO_2-C emission reduction required per unit of f.f. BC+OM emitted for diesel vehicles to cool global climate with the actual ratio of CO_2-C emission reduction per unit mass f.f. BC+OM emission when diesel achieves (a) 15% and (b) 30% better mileage than gasoline and when diesel has different f.f. BC+OM emission rates. The modeled curves (dashed lines) were obtained by dividing the f.f. BC+OM-temperature curve in Figure 3 by each CO_2-temperature curve (30 y, 50

y, 95 y) then multiplying the result by the yearly emission rate of anthropogenic 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 show that a yearly 1 Tg/yr decrease in f.f. BC+OM emission cools climate by about 4200-4500 times more than does a 1 Tg/yr decrease in CO_2-C emissions during 1 y. After 100 yr of continuous 1 Tg/yr decreases in both, the resulting ratio of f.f. BC+OM to CO_2-C cooling is about 90-190:1 (this ratio is the 100-yr climate response of f.f. BC+OM per unit emission relative to that of CO_2). 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 during which diesel vehicles enhance global warming in comparison with gasoline vehicles under the given emission standard. For example, in the case of the 0.08 g/mi standard, diesel warms climate in comparison with gasoline for >100 yr for all CO_2 lifetimes and for both differences in diesel versus gasoline mileage.

Figure 1.

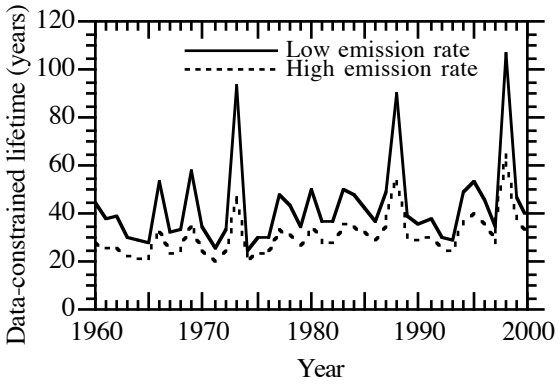


Figure 2.

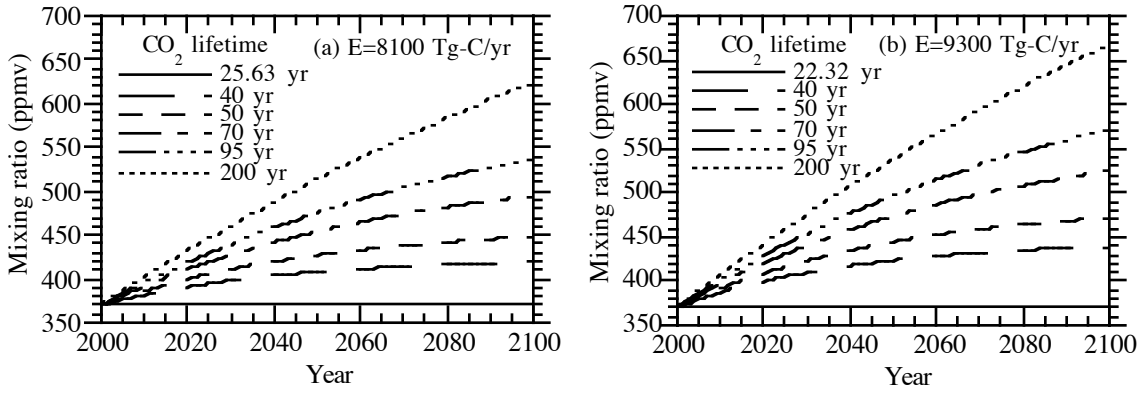


Figure 3.

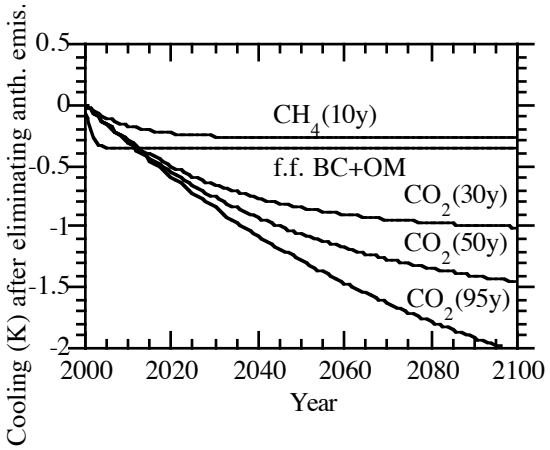
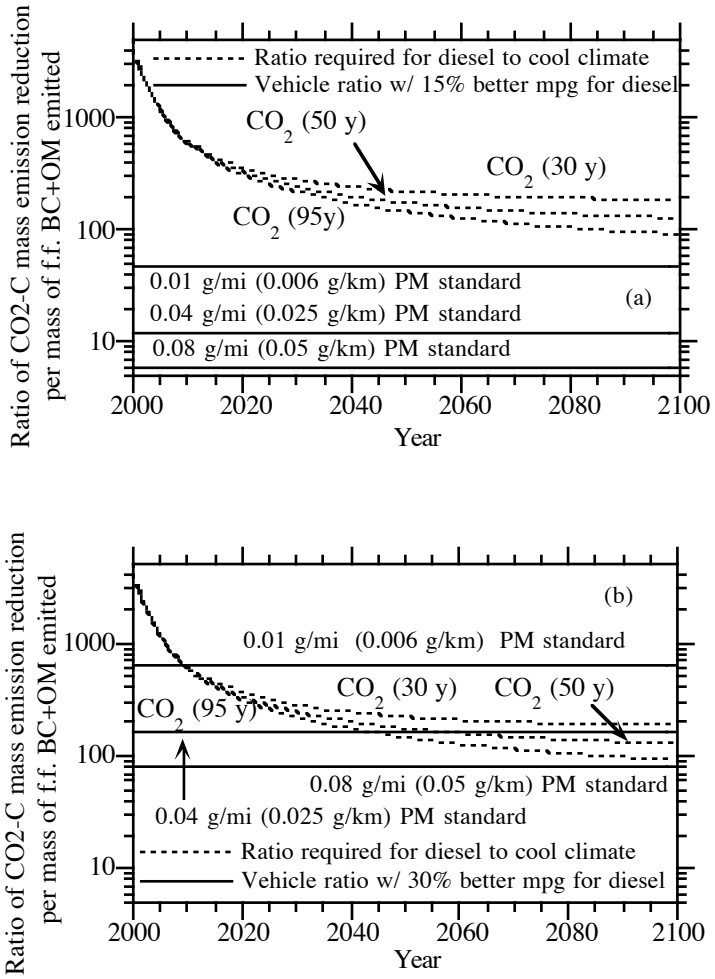


Figure 4.


Ratio of CO2-C mass emission reduction per mass of f.f. BC+OM emitted
Ratio required for diesel to cool climate
Vehicle ratio w/ 15% better mpg for diesel
CO2 (50 y)
CO2 (30 y)
CO2 (95y)
0.01 g/mi (0.006 g/km) PM standard
0.04 g/mi (0.025 g/km) PM standard
0.08 g/mi (0.05 g/km) PM standard
(a)
1000
100
10
2000
2020
2040
2060
2080
2100
Year
(b)
0.01 g/mi (0.006 g/km) PM standard
CO2 (30 y)
CO2 (50 y)
CO2 (95 y)
0.08 g/mi (0.05 g/km) PM standard
0.04 g/mi (0.025 g/km) PM standard
Ratio required for diesel to cool climate
Vehicle ratio w/ 30% better mpg for diesel