

Decreases in elemental carbon and fine particle mass in the United States

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Abstract. Observations at national parks and other remote sites show that average elemental carbon and fine particle mass concentrations in the United States both decreased by over 25 % between 1990 and 2004. Percentage decreases in elemental carbon were much larger in winter than in summer. These data suggest that emissions controls have been effective in reducing particulate concentrations not only in polluted areas but also across the United States. Despite the reduction in elemental carbon, the simultaneous decrease in non-absorbing particles implies that the overall radiative forcing from these changes was toward warming. The use of a 2005 instead of 1990 as a baseline for climate-relevant emissions from the United States would imply a significantly lower baseline for aerosol emissions. The use of older data will overestimate the possibility for future reductions in warming due to black carbon controls.

1 Introduction

Fine particles in the atmosphere, or aerosol, are important climate forcing agents. Non-absorbing particles cool the Earth whereas sufficiently light-absorbing particles warm the Earth. Controls on black carbon (BC) have been proposed as part of a strategy to reduce abrupt climate change (Molina et al., 2009). Trends in BC therefore have implications for

policy, showing both the effectiveness of past control strategies and the possibilities for future reductions. We use data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network of aerosol samplers (Malm et al., 1994, 2004) to examine trends in the United States between 1990 and 2004. These data indicate decreases in both fine particle mass and elemental carbon (EC) over wide areas of the United States, in places by more than 50 %. Trends in EC in the US at single sites and/or shorter periods (Junker et al., 2004; Husain et al., 2008; Kirchstetter et al., 2008) as well as trends in fine mass in Canada (Hidy et al., 2010) support these decreases.

As motivation for national trends, Fig. 1 shows EC and BC measurements from northern California, chosen because independent data sets are available at Trinidad Head (Fiebig and Ogren, 2006) and the San Francisco Bay area (Kirchstetter et al., 2008). The trend in EC measurements at Redwood National Park is similar to that for the San Francisco Bay area except that absolute concentrations are a factor of 10 lower at Redwood National Park. A site at Point Reyes (not shown) shows a similar time series with absolute concentrations intermediate between San Francisco and Redwood NP. Aerosol light absorption, measured using a Particle Soot Absorption Photometer, also decreased at the NOAA monitoring site at Trinidad Head, California.

Despite a lack of strong local sources, concentrations at the Northern California sites respond primarily to US emissions. Even a coastal site such as Trinidad Head has significant contributions from California sources during offshore flow and periods of recirculation (Goldstein et al., 2004).



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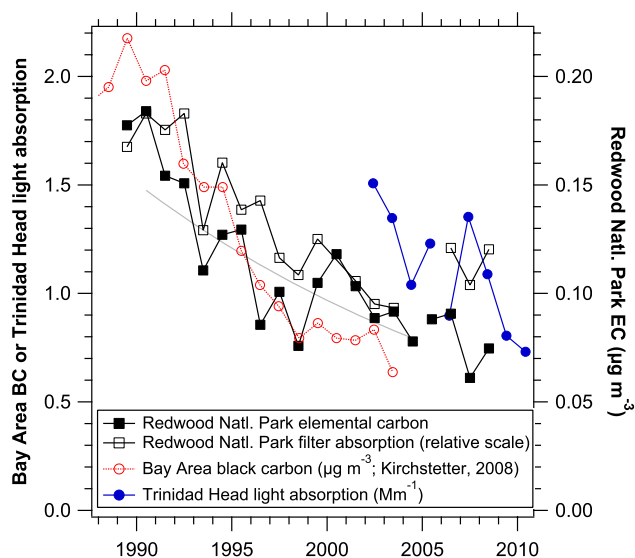


Fig. 1. Elemental carbon (EC) and light absorption trends for sites in northern California. The thin gray line is the fit for Redwood National Park (to the solid black squares). Redwood National Park data in 2004 and 2005 are not connected because of the change in instrumentation at the start of 2005. There was also an instrumentation change at Trinidad Head in late 2005.

Without sorting by a tracer or wind speed and direction, 24-hour samples, like IMPROVE, will sample significant amounts of local pollution from land breezes at these sites (Parrish et al., 2009).

2 Sampling methods

The IMPROVE network uses 24-hour filters that are collected every third day and analyzed at a central laboratory. Strengths of the IMPROVE data are the length of the record, the number of sites, and consistent techniques. All IMPROVE elemental and organic carbon analyses on quartz-fiber filters are performed at the same laboratory with the same protocols, the same suite of thermal/optical instruments, and consistent calibrations and quality assurance (Chow et al., 1993). The thermal/optical instruments were replaced in January 2005 so we focus on prior trends without the complication of this analytical change, which was flagged at: http://vista.cira.colostate.edu/improve/Data/QA_QC/Advisory/da0016/da0016_TOR2005.pdf. The IMPROVE network uses a parallel module with a Teflon-membrane filter to measure the filter blackness and fine particle mass ($PM_{2.5}$; particles with aerodynamic diameters less than $2.5\ \mu\text{m}$). Except as noted, the IMPROVE sites are located in national parks, monuments, or similar areas and represent regional background conditions. The IMPROVE sampling protocol is described in de-

tail at: http://vista.cira.colostate.edu/improve/Publications/SOPs/UCDavis_SOPs/IMPROVE_SOPs.htm.

It is customary to distinguish between BC and EC even though they are closely related in the atmosphere. BC is derived from light absorption measurements, often using the attenuation of light reflected from or transmitted through a filter. EC is determined by physical analyses such as heating of the sample in oxidizing and non-oxidizing atmospheres, sometimes with an optical correction for charring of some organic carbon to EC during analysis. “Soot” is a specific form of elemental carbon produced by combustion. The quantitative relationship between BC and EC depends on the size of the particles, their morphology, and coating by non-absorbing species (Watson et al., 2005; Bond and Bergstrom, 2006). There are large differences between BC or EC measured by different methods (Watson et al., 2005).

3 Trend analysis methods

Trends were computed starting in 1990 because there were insufficient sites in earlier years. The thermal/optical carbon analyzers were changed at the end of 2004 and it will require additional work to understand a trend computed across a change in instrumentation. Preparatory testing (Chow et al., 2007) showed that organic and elemental carbon values would remain consistent. Average EC during 2004 was lower at most sites than either the 2003 and 2005 annual averages, which may have been caused by instrument deterioration before replacement. As a result, we have chosen to end the trend computations in early 2004. An integral number of years (1 March 1990 to 29 February 2004) was chosen to minimize dependence of the trends on the seasonal cycle. The sampling frequency was changed in 2000 from Wednesday and Saturday to every third day based on the US EPA calendar for air pollutant sampling. This should have little effect on trends because Wednesday and Saturday are almost precisely at the mean of the weekly cycle in EC (Murphy et al., 2008).

For each IMPROVE site, individual samples were averaged to quarterly values (e.g. December through February). At least 10 valid samples were required in each quarter or else the entire quarter was treated as a missing value. Elemental carbon (EC) values were capped at $5\ \mu\text{g m}^{-3}$ to minimize the influence of a few of the most intense smoke plumes. This criterion affected less than 0.03 % of the data. For the trends from 1 March 1990 through 29 February 2004, sites were chosen that were operational before January 1995 and had at least 35 quarterly values (of 56 possible). Most missing quarterly values were because a given site did not become operational until after March 1990. Fifty sites had sufficient data to compute trends. A list of these sites is given in supplemental material along with their quarterly averaged data.

Trends were computed with simultaneous removal of the seasonal cycle. Removing the seasonal cycle is necessary because the seasonal cycle can bias the trends at those sites where the data start and stop in different seasons. Trends of annual averages were computed both for the linear model $A_0t + A_1\delta_{DJF} + A_2\delta_{MAM} + A_3\delta_{JJA} + A_4\delta_{SON} + \varepsilon$ and the exponential model $(A_1\delta_{DJF} + A_2\delta_{MAM} + A_3\delta_{JJA} + A_4\delta_{SON})\exp(A_0t) + \varepsilon$, where A_n are fitted coefficients and δ_{DJF} is 1 for data in December to February and 0 otherwise, etc. These two models have the same number of fit parameters and gave extremely similar trends. The exponential model is used in the figures because it had much smaller residuals at a few sites, generally those with the largest trends. There were no sites where it performed significantly worse than the linear model. Separate trends for each season naturally do not require removing the seasonal cycle.

The national average trends shown in the figures were computed from the percentage trends at various sites weighted by the absolute concentration at each site. If all sites had complete data records this would be to first order the same as the trend in the national average concentration. The extra step of computing separate trends for each site was necessary to minimize the impact of relatively clean or dirty sites entering the network in different years.

Trends in filter absorption (IMPROVE fabs) or “blackness” of the Teflon filters were computed after subtracting time-dependent zero offsets. These offsets are due to a 1994 change in the optical measurement as well as variations between different batches of filters. The correction is based on using the cleanest ambient filters as field blanks. In practice there are only a few very clean ambient filters and it is better to extrapolate back to zero concentration after excluding the dirtiest filters to avoid non-linear absorption. The offsets were computed 8 times per year from a multiple regression of filter absorption against EC and iron as a surrogate for light-absorbing mineral dust. These regressions used filters from the entire IMPROVE network, not just the sites that met our criteria for long-term trends. Similar offsets can be derived for a variety of techniques for estimating the field blank. The offset corrections reduce the computed trends in filter blackness over this period. No adjustment is made for the removal, in different years at different sites, of masks formerly used to reduce the effective collection area of the Teflon filters. Adjusting for the effects of unmasking would increase computed trends in filter blackness, as described at: http://vista.cira.colostate.edu/improve/Data/QA_QC/Advisory/da0028/da0028_DA_mask_Fabs.pdf.

4 Trend results

Figure 2 shows the percentage trends in EC and PM_{2.5} mass for the 50 IMPROVE sites with sufficient data to compute a trend before 2005. There are decreases across the continental

US and at Denali National Park in Alaska. Only a handful of sites exhibit increases. We do not investigate the causes of the increases at individual sites. Some of the increases may be due to wildfire influence, as discussed in Sect. 5.1.

Figures 3 and 4 show the trends by season in EC and PM_{2.5} mass. The most important feature evident by season is the increase in EC and to a lesser extent PM_{2.5} in summer in the mountain west. Some individual sites are noisy for single seasons.

Figure 5 shows national average trends by season. The largest seasonal difference is that decreases in EC were larger in winter than summer. Trends in filter blackness (not shown), measured independently on different filters in the same samplers, showed the same pattern as EC. For example, the average 14-year trend in EC concentrations was -49% for December through February and -15% for June through August (Fig. 5). The corresponding average changes in filter light absorption were about -53% and -27% . Washington DC, the only urban site with a long enough data record, was excluded from Fig. 5 because it is clearly different than the background stations. Most obviously, the average EC value was larger. Including Washington DC would have slightly increased the magnitude of the national average trends for EC and PM_{2.5} mass.

Some IMPROVE sites have nephelometers or transmissometers in addition to filter samplers. The limited data show decreases in light scattering or extinction at most sites, as expected from the decreases in fine particle mass. However, the fine particle mass is a more reliable long-term measurement (<http://vista.cira.colostate.edu/improve/Publications/GrayLit/TransDataUseWarning/TransDataUseWarning.htm>).

5 Discussion

5.1 Emission history

The decreasing concentrations are almost certainly due to decreases in emissions from the United States. There is no reason to believe that removal processes changed by so much over this period. Long-range transport from Asian sources would cause an increase, not a decrease, during the period 1990–2005 (Bond et al., 2004). The importance of United States emissions is supported by consistently negative trends at eastern sites least susceptible to transport from outside the US (Fig. 2). From 1990 to 2004, EC decreased by almost 50% for the entire winter and spring (December through May) period (Figs. 3, 5). This would reduce the melting of snow cover induced by deposition of BC. Indeed, when trajectories came from North America, BC concentrations in the Arctic decreased significantly between 1990 and 1998 (Fig. 9b in Sharma et al., 2004).

Major sources of elemental carbon in the United States are internal combustion engines (especially diesels), wild and

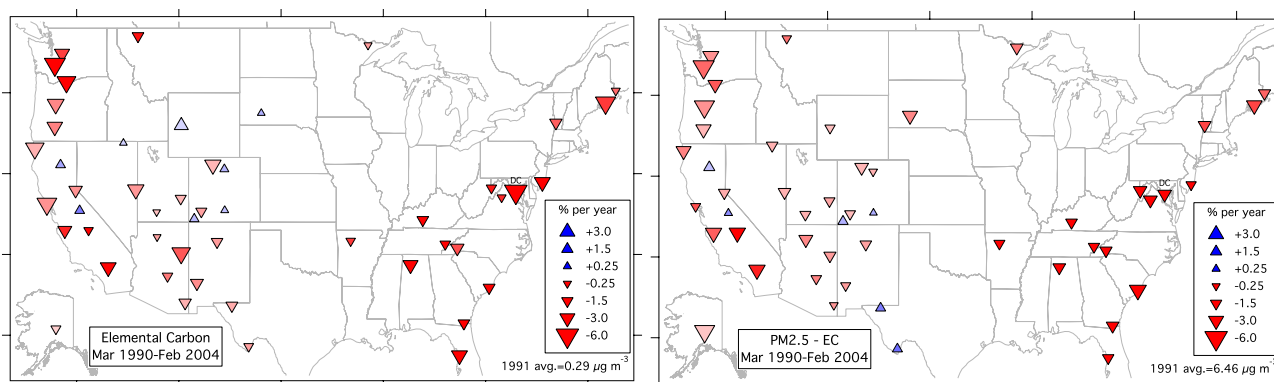


Fig. 2. Trends in EC and PM_{2.5} (fine particle) mass other than EC at all IMPROVE network stations with sufficient data between 1 March 1990, and 29 February 2004. Marker size indicates the magnitude of the trend. Triangle direction and blue or red color corresponds to the sign of the trend. Color saturation is proportional to the average concentration in 1991 with full saturation at twice the national median. The only urban site in Washington DC is marked. Averages in the bottom right corner exclude Washington DC.

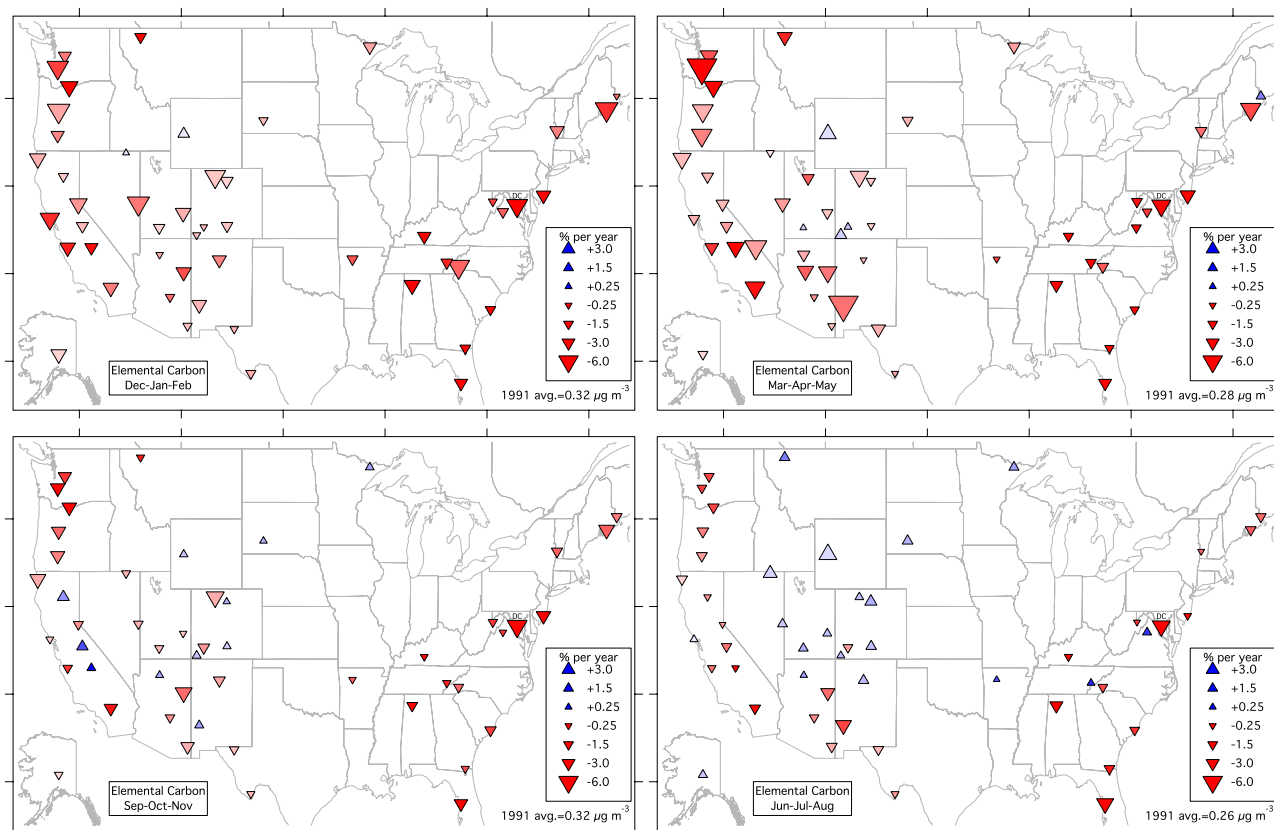


Fig. 3. Quarterly exponential trends in elemental carbon at IMPROVE network sites. As in Fig. 2, trends are for all stations with sufficient data between 1 March 1990, and 29 February 2004. Marker size indicates the magnitude of the trend. Triangle direction and blue or red color corresponds to the sign of the trend. Color saturation is proportional to the average concentration in 1991 with full saturation at twice the national median for each quarter. The only urban site in Washington DC is marked. Averages in the bottom right corners exclude Washington DC.

controlled burning of vegetation, and in places, wood stoves and other biofuels (Bond et al., 2004). Summer increases in EC were evident in the mountain western US (Fig. 3).

This is consistent with large decreases in controlled anthropogenic sources coupled with an increase in summer wildfires (Fig. 6).

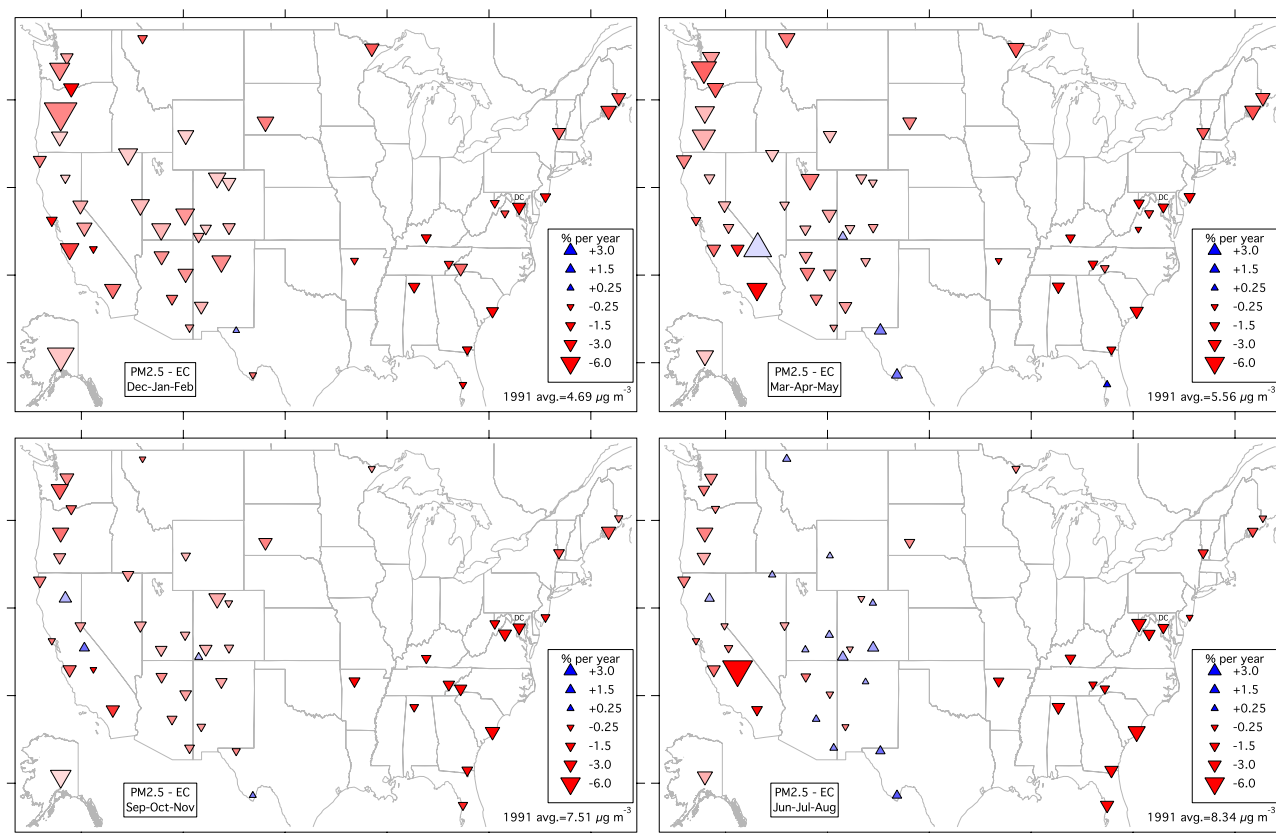


Fig. 4. Quarterly trends in fine mass other than elemental carbon at IMPROVE network sites. Symbols are as in Figs. 2 and 3.

A general smoke standard for heavy-duty highway diesel vehicles was introduced in 1970. Particulate standards for diesels (Yanowitz et al., 2000; Lloyd et al., 2001) were introduced at 0.6 g/bhp-hr in 1988 and reduced to 0.25 in 1991, 0.1 in 1994, and 0.01 in 2007. Due to the slow turnover of the truck fleet, a continuing response would be expected from earlier regulations. On-road measurements show that the emission factor of particulate matter from diesel trucks fell by 4 to 5 % per year between 1988 and 1996 (Yanowitz et al., 2000) and 1997 to 2006 (Ban-Weiss et al., 2008). Total emissions would have decreased less because emissions factors are multiplied by increasing fuel use (Dallmann and Harley, 2010). A strong Sunday-Monday minimum in EC implies that even the remote IMPROVE sites are significantly affected by diesel trucks and off-road equipment which are used less on Saturdays and Sundays (Murphy et al., 2008).

Particulate emission standards for wood stoves were introduced in 1988 (US EPA). As with diesel trucks, slow turnover would mean that these standards had a gradually increasing impact. The large decreases in EC in winter are consistent with a contribution from controls on wood stoves.

Starting in 1999, PM_{2.5} concentrations have been monitored at a large network of sites by the US Environmental Protection Agency. Most of these sites are urban, so they

represent different conditions than the IMPROVE sites used here. Analyzing these sites is beyond the scope of this paper. Nevertheless, recent decreases in PM_{2.5} concentrations at the EPA sites (<http://www.epa.gov/airtrends/pm.html>) are broadly consistent with the decreasing trends in PM_{2.5} at the IMPROVE sites.

5.2 Correlations with other species

EC accounts for about 5 % of the fine particle PM_{2.5} mass at non-urban IMPROVE sites. Therefore, the observed trend in mass must be caused by more abundant species. Most of the decrease in PM_{2.5} mass was due to organic carbon and sulfate. Silicon and other mineral dust elements in PM_{2.5} increased from 1990 to 2004.

Trends in EC at individual sites were uncorrelated with trends in sulfate. In contrast, trends in EC and organic carbon were correlated (Fig. 7). The total carbon, which can be measured more reliably than either component (Cavalli et al., 2010), decreased. Decreasing emissions from wood stoves would reduce both elemental and organic carbon. Decreases in EC in Japan were also correlated with organic carbon (Minoura et al., 2006). Recent work suggests that, because of semi-volatile compounds, the formation of

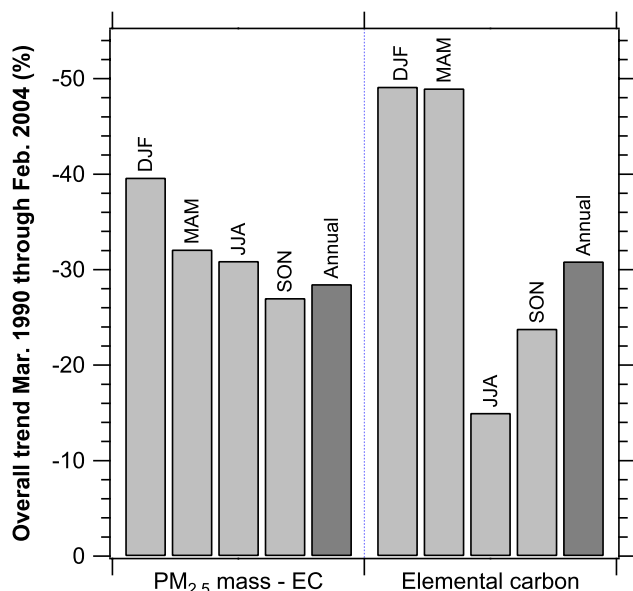


Fig. 5. Seasonal average national trends for the 14-year period 1 March 1990 to 29 February 2004 for elemental carbon and the balance of fine particle mass. (DJF: December, January, and February; MAM: March, April, and May; JJA: June, July, and August; and SON: September, October, and November).

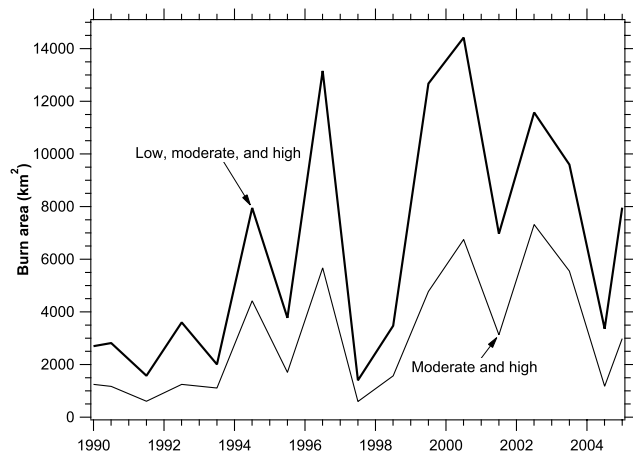


Fig. 6. Annual burn areas for the continental United States by fire severity from the Monitoring Trends in Burn Severity project of the US Forest Service and US Geological Survey (<http://mtbs.gov/data/search.html>).

secondary organic aerosol from diesel engines is much larger than previously thought (Weitkamp et al., 2007; Miracolo et al., 2010). If so, reductions in diesel emissions may be responsible for much of the trend in both EC and organic carbon mass.

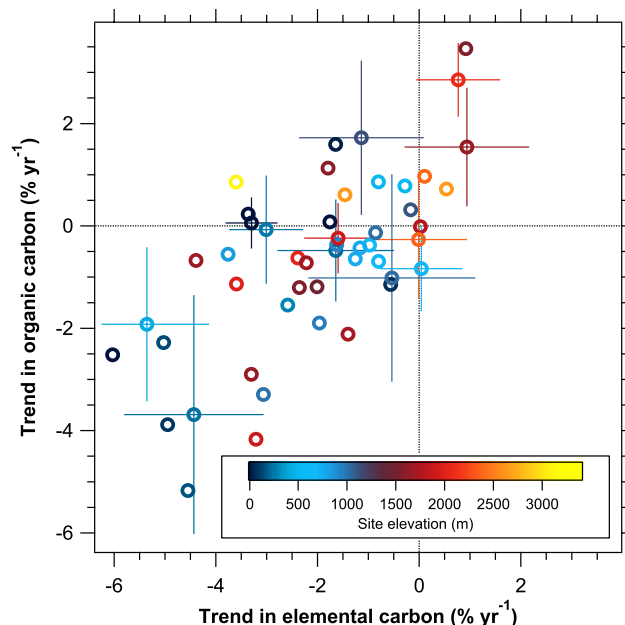


Fig. 7. The relationship between trends in organic and elemental carbon for each IMPROVE site shown in Fig. 2. Uncertainties (1σ) for the trend fits are shown for a subset of the sites.

5.3 Implications for radiative forcing

Black particles produce warming and non-absorbing particles cooling. Radiative forcing was calculated using the GISS GCM3 general circulation model with aerosol fields provided by the GEOS-Chem chemical transport model (Rind et al., 2007; Chen et al., 2010). These calculations are conducted with parallel radiative calculations within the model. The model is run with specified sea surface temperatures and sea ice coverage. Additional information about the GEOS-Chem and radiative forcing simulations can be found in Leibensperger et al. (2011). Radiative forcings for BC and the sum of sulfate, organics, and nitrate were computed for each month and aggregated to season and annual quantities. The BC forcing for each season was multiplied by the observed percentage trend in EC for that season, with a similar calculation for non-absorbing particles. The annual average change in forcing was computed from these seasonal changes.

Applying the observed percentage trends in concentration to US emissions in this pair of models, the decreases shown in Fig. 5 would produce a direct radiative forcing of -0.09 W m^{-2} because of decreased black carbon and 0.39 W m^{-2} because of decreased non-absorbing particles, for a net of 0.3 W m^{-2} over the contiguous US. The net global forcing was calculated as 0.014 W m^{-2} . GEOS-Chem underestimates absolute BC concentrations over the US but positive net forcing from the observed trends still holds if modeled BC over the US is doubled. The radiative forcings calculated here with the GISS GCM3 model do not include

aerosol indirect effects on clouds or the radiative forcing from reduced albedo of snow following deposition of black carbon (Flanner et al., 2007, 2009). These omissions likely do not change the qualitative conclusion that the net change in aerosol forcing has been towards warming.

There are several implications to these trends. Emissions standards and restrictions implemented during the 1990s on diesel engines and residential wood-burning stoves have apparently had a significant effect on BC concentrations across the United States. Further reductions can be expected from the stricter diesel standards implemented in 2007 and the complementary low-sulfur fuel introduced nationally in 2006. It is important to use up-to-date emissions inventories to evaluate both health effects and radiative forcing. In Copenhagen, the United States proposed reductions in climate-relevant emissions compared to emissions in 2005 (<http://www.state.gov/g/oes/rls/remarks/2009/121010.htm>), whereas the Kyoto Protocol specified a 1990 baseline. Proposed US legislation on greenhouse gases has variously used 1990 or 2005 as baselines (Energy Information Administration, 2009; Pew Center, 2008). For carbon dioxide, 2005 provides a higher baseline than 1990. This work shows that black carbon has the opposite behavior: using 2005 as a reference year would mean a significantly lower baseline for US BC emissions than a 1990 baseline. Decreased concentrations imply that the room for reducing warming by controlling BC may be less than expected if older inventories are used.

Supplementary material related to this article is available online at:

<http://www.atmos-chem-phys.net/11/4679/2011/acp-11-4679-2011-supplement.zip>.

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