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Impact of California's air pollution laws on black carbon and their implications for direct radiative forcing

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ABSTRACT

We examine the temporal and the spatial trends in the concentrations of black carbon (BC) – recorded by the IMPROVE monitoring network for the past 20 years – in California. Annual average BC concentrations in California have decreased by about 50% from 0.46 μg m^{-3} in 1989 to 0.24 μ gm^{-3} in 2008 compared to the corresponding reductions in diesel BC emissions (also about 50%) from a peak of 0.013 Tg Yr^{-1} in 1990 to 0.006 Tg Yr^{-1} by 2008. We attribute the observed negative trends to the reduction in vehicular emissions due to stringent statewide regulations. Our conclusion that the reduction in diesel emissions is a primary cause of the observed BC reduction is also substantiated by a significant decrease in the ratio of BC to non-BC aerosols. The absorption efficiency of aerosols at visible wavelengths – determined from the observed scattering coefficient and the observed BC – also decreased by about 50% leading to a model-inferred negative direct radiative forcing (a cooling effect) of −1.4 W m^{-2} (±60%) over California.

1. Introduction

Black carbon (BC) is a substantial component of carbonaceous aerosols produced from fossil fuel combustion and biomass burning (Andreae and Merlet, 2001) and is the principal absorber of visible solar radiation in the atmosphere. The absorption leads to a decrease of solar irradiance at the surface, but the net effect on the surface–atmosphere column, referred to as direct radiative forcing, is positive and results in a warming of the climate (Bond, 2007; Jacobson, 2005). Recent studies (Jacobson, 2010) indicate that heating of the atmosphere by BC particles could be a major contributor to global warming with an estimated radiative forcing of 0.55 W m^{-2} to 0.9 W m^{-2}, but some model estimates are as low as 0.2 W m^{-2} (Forster et al., 2007). The main importance of BC is that given its short atmospheric lifetime of one to two weeks (Koch et al., 2010; Ogren and Charlson, 1983) it offers a unique possibility for mitigating climate change (Jacobson, 2010). Current model estimates of radiative forcing (Chen et al., 2010a; Forster et al., 2007; Jacobson, 2010; Ramanathan and Carmichael, 2008) as well as a recent observational study (Ramana et al., 2010), indicate that mitigation of fossil fuel BC is a viable control strategy for climate warming. In particular, BC emitted by diesel combustion emits less organic aerosols (compared with biomass burning) which scatter significantly more solar radiation than BC and can lead to cooling. Hence it is fundamentally important to understand the role of diesel emissions on the direct radiative forcing of BC.

Given the large uncertainty in model estimates of BC emissions from various sources, their atmospheric concentrations and their optical properties (Forster et al., 2007) it would be highly advantageous to have empirical data to validate model predictions. Empirical measurements of atmospheric BC over a few decades time scale and few hundred kilometers spatial scale in a region where BC emissions from diesel fuels were reduced significantly would prove highly advantageous for validating the model predictions. In this study, we present such a unique data set that is ideally suited for the purpose of understanding the role of diesel fuel emissions in BC concentrations and its direct forcing. This data set was generated as a consequence of California's aggressive laws to reduce particulate emissions. Such laws led to the introduction of Diesel Particulate Filters (DPFs) in non-road machines since 1980, and in automobiles since 1996. The first statewide regulation on diesel engine emissions was introduced in 1987 with the first California Heavy Truck rule capping particulate emissions at 0.60 g BHP^{-1} h^{-1}, and progressively tighter standards have been subsequently implemented. While no jurisdiction has made DPFs mandatory, the increasingly stringent emissions regulations mean that eventually all on-road diesel engines will be fitted with them – for instance, the American 2007 heavy truck engine

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emissions regulations cannot be met without filters. The "Risk Reduction Plan to Reduce Particulate Matter Emissions from Diesel-Fueled Engines and Vehicles" adopted by the California Air Resource Board (CARB) established further goals to reduce diesel emissions in California by 75% in 2010 and 85% in 2020. To support the diesel risk reduction plan CARB has introduced regulations including (but not limited to) the control equipment verification program (2007), public transit bus rules (2000), transport refrigeration unit regulation (2003), and proposed future idling regulations, details of which may be obtained under the California Code of Regulations Section 13 (CCR, 2010). These regulations have a mandatory character (in California) but allow a flexibility of approach which can be met by a variety of approaches including retrofitting engines with emission controls systems (such as DPFs and oxidation catalysts), replacement of existing engines with newer technology engines, and restrictions placed on the operation of existing equipment. Significantly, the reduction – 0.007 Tg Yr^{-1} in diesel emissions and 0.008 Tg Yr^{-1} in total BC emissions (CARB, 2008) – between 1990 and 2008 – is comparable, indicating the total decrease in BC is almost entirely attributable to diesel emission standards.

In addition to this opportune experiment in diesel emission reductions, statewide measurements are also available for elemental carbon (EC), organic carbon (OC), sulfate, and nitrate in the fine aerosol (<2.5 μm) phase from 22 Interagency Monitoring of Protected Visual Environments (IMPROVE) sites in California from 1988–2008 to establish spatially resolved trends of BC and non-BC aerosols California and their radiative properties. Furthermore, aerosol scattering coefficients at the surface and column measurements of the aerosol single scattering albedo were also available. These unique data sets enable us to ask the following questions: What was the impact of diesel emission control on BC concentrations in the atmosphere? And how did it alter the radiative properties of aerosols? And lastly, what was the impact on the direct radiative forcing of the atmosphere?

2. BC measurements and diesel emissions

Elemental carbon (EC) measured using thermal optical reflectance techniques as defined by the IMPROVE protocol (Chow et al., 2001, 2004) is used as a surrogate for BC. Since the optically-based definition of BC includes EC and a small fraction of organic carbon (OC), this corresponds to an atmospheric lower bound. The IMPROVE protocol has been applied consistently on the measurements in this study, making the observed trends reliable. The IMPROVE protocol has been shown to compare well with other evolved gas and optical measurements, although different ambient compositions (with different contributions of diesel and wood burning contributions, as well as interference from dust) could result in greater discrepancies as discussed by (Chow et al. 2005) in a recent review. For example, adsorption of volatile species and dust minerals associated can lead to discrepancies in the corrections for charring that have been shown to cause both positive and negative biases in EC measurements (Countess, 1990; Schmid et al., 2001; Turpin et al., 1990; Watson et al., 2009).

Studies in regions dominated by fossil fuel combustion (Allen et al., 1999; Liousse et al., 1993) have found strong correlations between EGA EC and aethelometer BC, indicating that EC from diesel emissions are likely to be well represented by the IMPROVE measurements (Turpin et al., 2000). To investigate correspondence between trends in measured EC and diesel fuel emissions in California, we incorporate two independent emission inventories in this study. An estimate of fossil fuel emissions – attributed to diesel and gasoline – is available from (Ito and Penner, 2005), and the California Air Resource Board (CARB, 2008) maintains an almanac of total particulate emissions and source related emission factors that can be used to estimate diesel fuel BC in California.

Fig. 1(a) illustrates the annual averages for BC in California for the twenty year period between 1989 and 2008. The annual mean concentrations show a steady decline within this time period (from 0.46 μg m^{-3} in 1988 to 0.24 μg m^{-3} in 2008), with annual variability expressed as a standard deviation (1σ, adjusted for multiple sites) approximately 40% of the mean value. The major portion of the decline in BC concentration appears to occur between 1989 and 1998 (from 0.46 to 0.26 μg m^{-3}) with a slower decrease between 1999 and 2008. The spikes in BC concentration in 1999 and 2008 correspond to major California forest fire years. The rate of decline in BC concentration agrees well with the decline in both total fossil fuel emissions (Ito and Penner, 2005) and diesel vehicle emissions (CARB, 2008) which show emissions peaked between 1985 and 1990 and declined thereafter. This correspondence in trend between reduced diesel emissions and BC concentrations is consistent with, and reinforces the conclusions of Kirchstetter et al. (2008) who report a reduced emission factor for diesel fuels between 1960 and 2000.

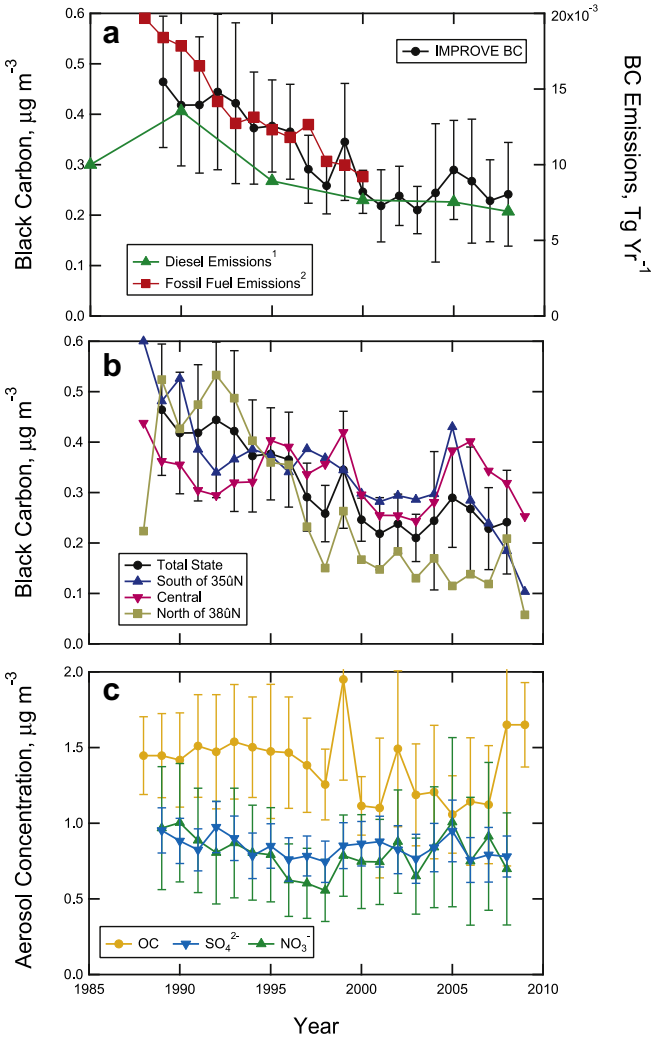

Fig. 1. (a) Annual means of measured Black Carbon (left axis) and BC fossil fuel emissions (right axis) in California from 1985 to 2008. Error bars correspond to standard deviation between measurements at each station. Dashed lines indicate a linear fit. Aerosol measurements from the IMPROVE network, emission inventories from (1) CARB, (2) (Ito and Penner, 2005) (b) Annual means of BC measured in Southern (South of 35 N), Northern (North of 38 N), and Central California (c) Annual means of measured Sulfate, Nitrate, and OC from IMPROVE network.

Fig. 1(b) compares the statewide annual mean BC with the mean calculated in three geographic zones – South California (South of 35 N) which contains the Los Angeles-Long Beach-Santa Ana metropolitan areas, Central California (between 35 and 38 N) which contains the San Joaquin Bay Area and most of the central valley, and North California (North of 38 N). While the background BC concentrations (as measured by the IMPROVE network) are higher in the more populous Southern and Central zones, a similar decline is observed in all three regions indicating that the observed decline in BC is uniform statewide, and not dominated by a few stations.

The measured sulfate, nitrate, and OC (Fig. 1(c)) show no statistically significant trend in concentration between 1989 and 2008, with the approximately 10% change in annual mean values being significantly smaller than the standard deviation in annual measurements. These contrasting trends suggest that the reduction in diesel fuel aerosol emissions is likely to be the dominant contributing factor to the reduction in BC concentration – particularly in the time frame following the introduction of Diesel Particulate Filters in California. These results further reinforce our conclusion that the IMPROVE data constitute an ideal set of ambient measurements for evaluating the climate impact of mitigating BC via diesel emission control.

Table 1 summarizes annual means and rate of change in BC concentration, BC/sulfate ratio, and BC/non-BC aerosol ratio. A faster drop in BC concentration is observed for the winter and spring months (Nov–Apr), compared to the summer and fall months (May–Oct) which correspond to the wildfire season in California. This seasonal variation is consistent with a greater reduction in anthropogenic emissions compared to forest fire emissions and trans-Pacific transport. The ratios also show a steady reduction from 1989 to 2008 for all seasons, indicating that the decline in BC has outpaced the decline in total aerosol concentration. The BC/sulfate ratio decreased at approximately double the rate (−6% compared to −3%) of BC/non-BC aerosol ratio, further supporting our conclusion that reduction in emissions from low sulfate fuels (such as diesel) and the proliferation of cleaner emission vehicles are primarily responsible for the reduction in BC aerosols.

3. Spatial distribution of BC

The twenty year average BC concentration measured at the IMPROVE sites is illustrated in Fig. 2(a). A low background concentration between 0.1 and 0.2 μg m^{-3} is observed over the majority of the state, with significantly higher concentrations (up to 0.7 μg m^{-3}) in the central valley and the south coast air basin, indicating that anthropogenic emissions are the principal source of BC in the state. Fig. 2(b) and (c) illustrate the rate of change in BC concentration and BC/non-BC aerosol (represented here by sulfate, nitrate, and OC) respectively, which are negative over the entire state indicating that anthropogenic aerosols generated by BC emissions have been declining in California between 1989 and 2008.

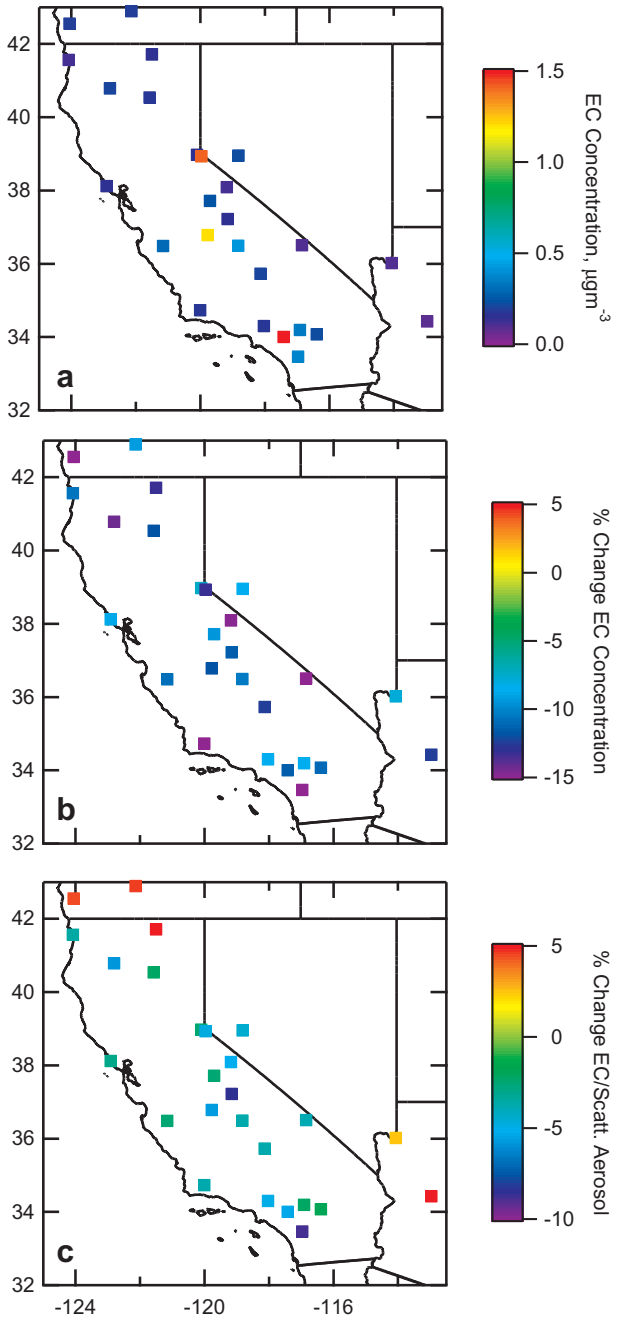

Fig. 2. (a) 20-year average BC Concentration from IMPROVE network in California expressed in μg m^{-3} (b) Average rate of change of BC Concentration, % (c) Average rate of change of BC/non-BC aerosol ratio. Locations of the measuring stations are indicated.

Table 1
Annual mean and standard deviations of measured Black Carbon concentration and BC/non-BC Aerosol ratios in California. Rates of change are calculated based on a linear fit for annual mean values between 1989 and 2008.

	1989	2008	Annual	Average rate of change, %			
				Spring (Feb–Apr)	Summer (May–Jul)	Fall (Aug–Oct)	Winter (Nov–Jan)
BC concentration, μg m^{-3}	0.46 ± 0.13	0.24 ± 0.10	−4.8 ± 0.5	−5.9 ± 0.2	−3.6 ± 0.2	−4.3 ± 0.5	−6.6 ± 0.4
BC/SO_4^{2-} ratio	0.39 ± 0.11	0.36 ± 0.12	−6.1 ± 0.4	−7.8 ± 0.5	−4.6 ± 0.3	−4.6 ± 0.6	−7.6 ± 0.8
BC/(SO_4^{2-} + NO_3^- + OC) ratio	0.14 ± 0.01	0.07 ± 0.01	−3.0 ± 0.3	−3.3 ± 0.3	−3.5 ± 0.2	−3.5 ± 0.4	−3.1 ± 0.2

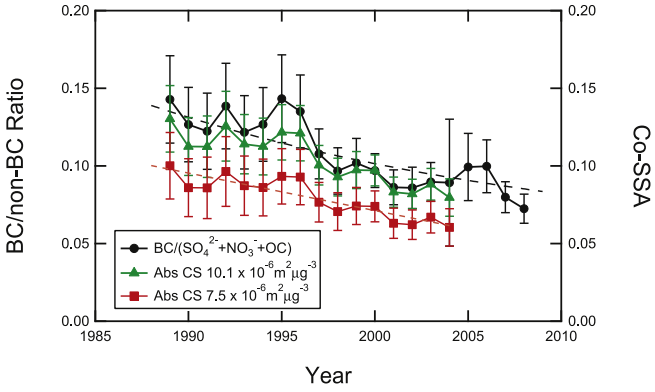

Fig. 3. Time series of measured BC/non-BC Aerosol ratio from the IMPROVE network in California (left axis) and derived co-Single Scattering Albedo for visible light (right axis). Co-SSA is calculated using 10.1×10^{-6} m^2 μg^{-1} (Horvath, 1993) and 7.5×10^{-6} m^2 μg^{-1} (Bond and Bergstrom, 2006) for BC absorption cross section.

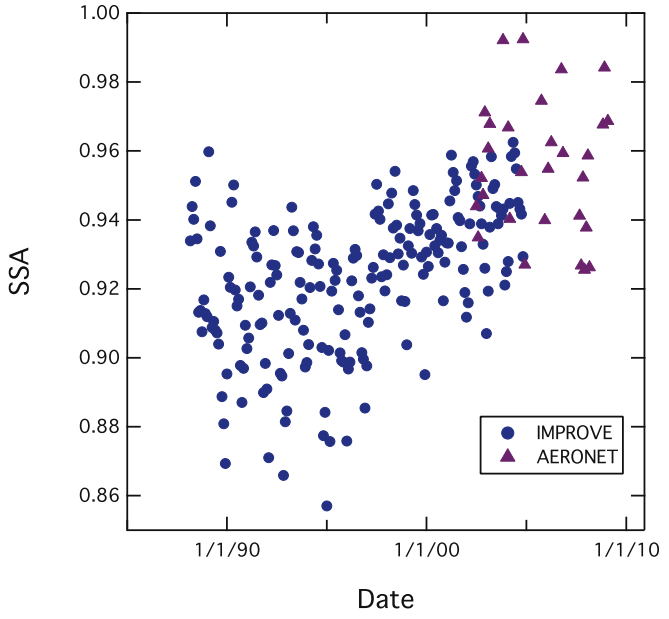

Fig. 4. Monthly averaged SSA calculated from IMPROVE measurements and retrieved from the AERONET network in California from 1988–2010.

4. Observed changes in aerosol radiative properties

Fig. 3 illustrates the approximately 50% reduction in ratio of BC (an absorbing aerosol) to the total sulfate, nitrate, and OC (primarily scattering aerosols), from 0.14 (1989) to 0.07 (2008). We can infer an aerosol absorption coefficient (k) from the BC concentrations using a conversion factor of 7.5×10^{-6} as recommended by Bond and Bergstrom (2006) or 10.1×10^{-6} m^2 μg^{-1} recommended under the IMPROVE protocol (Horvath, 1993). The k can then be combined with nephelometer measurements (Malm and Gebhart, 1996; Malm and Pitchford, 1997) of the aerosol scattering coefficient (s) from the IMPROVE network to obtain the single scattering albedo (SSA) defined as: $s/(s + k)$. A more useful quantity for tracking the forcing impact of absorbing aerosols is the co-single scattering albedo $A_e = (1 - SSA)$, which is the quantity shown in Fig. 3. A_e peaked at 0.13 in 1989 and decreased by about 40% to 0.08 in 2004 (the last year for which data were available). Values less than 0.05 will typically lead to net negative forcing while values greater than about 0.15 lead to net warming, and for values in between 0.05 and 0.15, the sign and magnitude will depend on the surface albedo, cloud fraction, vertical profile and latitude and season. The observed scattering coefficient changed very little with time therefore trends in A_e closely resemble the trend in BC. The negligible trend in s is consistent with corresponding small trends in SO_4 and NO_3 (which dominate scattering) shown in Fig. 1(c), thus demonstrating the over all consistency of the various data sets used in this study for establishing the impact of diesel emission reductions. In summary, the rate of decline in BC concentrations has outpaced the decline in non-BC aerosols (primarily sulfate), resulting in a reduced warming, or equivalently cooling, potential of aerosols in California.

5. Implications for direct radiative forcing

In order to use the surface data for exploring implications for radiative forcing, we need to examine the link between surface SSA and column SSA, which is discussed next. Fig. 4 illustrates the monthly average SSA over California between 1988 and 2010 from both the IMPROVE measurements at the surface and the AERONET network (Holben et al., 1998, 2001) data (available from 3 stations in California between 2005 and 2010) which estimates the column averaged SSA. Though the increase in calculated SSA is expected as a consequence of decreasing BC concentrations, the trend is consistent with the more recently available AERONET observations.

The impact on direct radiative forcing due to the changes in aerosol loadings is estimated with a Monte Carlo radiative transfer model (Kim and Ramanathan, 2008). The model adopts a satellite derived surface albedo of 0.2 and cloud fractions of 0.11, 0.1, 0.26, and 0.01 for low, middle, high and convective clouds over California. Aerosol vertical profiles below an altitude of 3000 m are constructed (Hadley et al., 2007) using IMPROVE annual mean data and assumed to be exponentially decreasing above 3000 m. Constructed vertical profiles for BC and non-BC aerosols in 2008 are illustrated in the Supplement. Sea salt and dust aerosol concentrations are from chemical model transport model simulations for 2001 (Feng and Ramanathan, 2010) and assumed to be super-micron. The sulfate, BC, organic matter ($1.4 \times$ OC), and nitrate are considered to be internally mixed with a modal radius of 0.05 μm. Literature values are used for the refractive indices of BC (Bond and Bergstrom, 2006) and non-BC aerosols (d'Almeida et al., 1991) in the visible band. The hygroscopic growth of sulfate and sea salt is calculated with a thermodynamic model (Jacobson, 1999).

The calculated aerosol optical depth (AOD) decreased about 19% from 0.27 in 1989 to 0.22 in 2008. The modeled A_e decreased from 0.116 to 0.077, comparing well with the observed trend (Fig. 3). In the forcing estimates, we used 0.23 and 0.18 for AODs in 1989 and 2008 to match the regional mean MODIS/Terra AOD retrieval (Platnick et al., 2003) (0.18) in 2008 over California, but retained the AOD decrease based on the IMPROVE values.

The cooling effect of aerosols at the top-of-the Atmosphere (TOA) changed by -1.4 W m^{-2} between 1989 and 2008, with the absorption in the atmosphere decreasing by 6.5 W m^{-2} and incoming solar radiation reaching the surface increasing by 5.1 W m^{-2}. The uncertainty in the estimated changes in TOA forcing arises from the model-input values for AOD and the vertically integrated SSA. The estimated uncertainty in forcing is about 8% (surface), 14% (atmosphere) and 28% (TOA) due to the differences between the calculated and satellite-based AOD. With respect to SSA, the observationally based SSA (Fig. 4) is only for the near-surface air, which we assumed to hold for other levels in the atmosphere. We don't have observational data for the vertical variation in SSA which prevents from making an objective estimate for the uncertainty. Our guess is that it

should be at least ±50%. The estimated large cooling effect of the reduction in diesel related BC aerosols is consistent with recent model predictions (Jacobson, 2010).

The surface brightening of 5.1 W m^{-2} (±60%), caused by reduction in diesel based BC is about 3% of the net surface solar radiation (190 W m^{-2}), is large enough to be detectable. Such large increases in surface solar radiation as a result of aerosol (BC in our case) reductions is consistent with an observed clear-sky brightening of 5.0 W m^{-2} observed over the United States between 1997 and 2006 (Long et al., 2009). This does not necessarily imply that reduction in BC should have contributed to surface warming over California, since the decrease in atmospheric solar absorption (6.5 W m^{-2}) is much larger, leading to a TOA negative forcing and mitigation of human impact on global warming. The magnitude of the surface brightening and atmospheric decrease in solar heating are sufficiently large to have had an impact on regional heat and water budget. Black carbon reduction should also have led to alteration of clouds by deceasing cloud nuclei and cloud albedo (warming effect; see (Chen et al., 2010b; Jacobson, 2010)) and increased cloud fraction (cooling effect; see (Ackerman et al., 2000)). The net effect of BC interactions with clouds is estimated to be warming (Jacobson, 2010), but this requires further study and the California data reported here should offer an excellent opportunity to validate published hypotheses for BC-Cloud interactions.

The availability of aerosol phase measurements of BC from the IMPROVE monitoring network, in conjunction with California's air pollution laws for BC reduction, provide a comprehensive picture of atmospheric BC trends over California. The data indicates that the annual mean BC concentration has decreased by 50% from 0.46 μg m^{-3} in 1989 to 0.24 μg m^{-3} in 2008, and followed a parallel trend in the reduction of fossil fuel (primarily diesel) BC emissions (also about 50%). The correlation between BC decline and diesel emission decline is further supported by the relatively constant concentrations of sulfate, nitrate, and OC aerosols over the same time period. The use of multi-site aerosol phase measurements allows us to constrain changes in the SSA compared to retrievals from AERONET, and construct vertical concentration profiles within the boundary layer. These measurements provide an invaluable resource for determining the climate impact of BC, and in conjunction with emission inventories provide a direct link between regulatory control policies and the long-term impact of anthropogenic emissions. Our model calculation indicates that the decrease in BC in California has lead to a cooling of 1.4 W m^{-2} (±60%). The regulation of diesel fuel emissions in California therefore has proven to be a viable control strategy for climate change in addition to mitigating adverse human health effects.

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Appendix. Supplementary material

Supplementary material related to this article can be found online at doi:10.1016/j.atmosenv.2010.10.054.

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