

# **Black Carbon and the Regional Climate of California**

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## LIST OF ACRONYMS

BC	Black Carbon
EC	Elemental Carbon
OC	Organic Carbon
BrC	Brown Carbon (Absorbing Fraction of OC)
ATOFMS	Aerosol Time of Flight Mass Spectrometry
GCM	General Circulation Model
AERONET	Aerosol Robotic Network
MISR	Multi-angle Imaging Spectro Radiometer
COH	Coefficient of Haze
AQMD	Air Quality Management District
AAE	Absorption Angstrom Exponent
SAE	Scattering Angstrom Exponent
EAE	Extinction Angstrom Exponent
SSA	Single Scattering Albedo
TOA	Top Of Atmosphere

## ABSTRACT

This report provides an assessment of the impact of black carbon on the regional radiative forcing and climate trends of California. The present regional integrated assessment is the first such attempt to estimate the radiative forcing of BC for one region (California in this case), both from a bottom-up approach (starting with emission inventory as input to aerosol-transport models) and a top-down approach (adopting satellite data in conjunction with ground based column averaged aerosol optical properties). This approach enabled us to uncover three unanticipated major findings: i) The first finding concerns the large decadal trends in BC concentrations largely in response to policies enacted to decrease PM emissions from diesel combustion. ii) The second is the discovery of the large effects of brown carbon (a form of organic carbon aerosols) on radiative forcing. iii) The third is the large discrepancy between the top-down and the bottom-up approach of estimating radiative forcing and ways to close the gap.

**Observed multi-decadal BC trends:** The trends in BC concentrations were determined from assimilation of mass-based measurements (from the IMPROVE network that typically samples remote areas), and from analysis of the large set of available COH (Coefficient of Haze) measurements (which typically sample urban areas). The more reliable IMPROVE data showed that the annual average BC concentrations in California have decreased by about 50% from  $0.46 \mu\text{g m}^{-3}$  in 1989 to  $0.24 \text{mgm}^{-3}$  in 2008. The COH data revealed that BC concentrations in California decreased markedly from about  $3.9 \mu\text{g m}^{-3}$  in 1966 to  $2.3 \mu\text{g m}^{-3}$  in 1980 to  $1.1 \mu\text{g m}^{-3}$  in 2000, agreeing with the trends reported from the IMPROVE data sets. Trends in other co-emitted aerosols such as OC, sulfates were much smaller or statistically insignificant. BC trends are related to an order of magnitude reduction in diesel PM emissions since the first smoke reduction standards were introduced in the 1970s. Other determining factors likely include BC emissions reduction from other sources in the transport sector, the cleanup of BC emissions in the industrial sector, and decreasing wood and waste burning since 1990. *Observations of large negative trends in BC and the lack of corresponding negative trends in co-emitted OC and sulfate aerosols gives compelling observational support to the conclusion of Jacobson (2010) and Bond et al (2013) that mitigation of diesel BC would mitigate global warming.*

**Importance of Brown Carbon for Solar absorption:** Analysis of the spectral dependence of solar absorption measured in-situ as well as over the column indicates an enhanced absorption at shorter (<500 nm) wavelengths that deviates from the expected behavior of BC. This enhanced absorption is attributed to organic, "brown", carbon. The solar absorption due to brown carbon is found to contribute as much as 40% of the BC forcing in the near-UV (<440 nm) wavelengths. Field measurements in several independent campaigns using aethalometers for absorption, and the ATOFMS for detailed chemical analysis find that brown carbon is primarily related to residential wood burning. However, a new class of particles related to secondary organics, i. e., aged large organic particles, is also found to contribute to solar absorption, thus raising the possibility that fossil fuels also contribute – via their contribution to secondary

organic particles – to brown carbon absorption. *We find that the direct warming effect of brown carbon, ignored in most models, offsets about 60% to 90% of the direct cooling effects of other organic carbon aerosols.*

**Direct Radiative Forcing Over California:** We determine the radiative forcing using a top-down approach developed by the PI's (VR) group that relies on NASA's ground based AERONET data and assimilated satellite measurements (MISR) of aerosol optical properties. This observationally constrained top-down (OC\_TD) estimate includes solar absorption by brown carbon while the emission inventory based bottom-up (EI\_BU) estimates included in this study do not account for brown carbon. The uncertainty in the OC\_TD forcing is about  $\pm 40\%$ . For the annual mean, the current top-of-atmosphere (TOA) forcing of BC+OC varies from about  $0.2 \text{ Wm}^{-2}$  over Northern California (NCA) to as large as  $1.9 \text{ Wm}^{-2}$  over Southern California (SCA). The implication is, in the 1980s when BC concentrations were higher by about 100%, the TOA forcing for BC+OC could have been as large as 0.4 to  $3.8 \text{ Wm}^{-2}$ . *Overall we conclude that the large negative trend in BC radiative forcing and the lack of corresponding negative trends in OC, confirms the assessment of some other studies (Jacobson, 2010 and Bond et al, 2013) that diesel related BC emission reduction would lead to global cooling.*

**Regional Climate Effects:** The regional climate changes associated with the observed reduction in BC concentrations were estimated with a regional climate model. *The declining trends in BC (by a factor of five to approximate the 1960s) cause a reduction in the TOA direct radiative forcing everywhere and the cooling effect ranges from  $-0.5$  to  $-3.5 \text{ Wm}^{-2}$ , consistent with the OC\_TD estimates, with larger effects during summer than winter.* In response to this change, the lower atmosphere is found to cool everywhere, accompanied by a smaller warming near the surface, which is not statistically significant in any season. Overall, the local climate effects are small.

#### **Primary Finding: Mitigation of Global Warming**

*Reductions in emissions of BC mostly from diesel engines, since the 1980s have contributed significantly to mitigation of global warming, equivalent to mitigation of 21 million metric tons of  $\text{CO}_2$  emissions annually. This climate benefit may date back to at least the 1960s, and is currently ongoing.*

Because of the long lifetime of  $\text{CO}_2$  (one century or longer) compared with the much shorter lifetime of BC (about one week), mitigation of  $\text{CO}_2$  is critical for limiting long term (>50 years) global warming, while mitigation of BC is critical for limiting near-term (<50 years) warming.

## EXECUTIVE SUMMARY

### OVERALL FINDING

BC emission reductions since the 1980s, attributed in large part to diesel engine emissions mitigation, are equivalent to *reducing CO<sub>2</sub> emissions by 21 million metric tons annually. This is approximately equal to 5 % of the total direct CO<sub>2</sub> annual emissions of 393 million metric tons.*

As on-road diesel is very low in sulfur in developed regions and lowering elsewhere, and since compared to other major BC sources, diesel PM has more BC and less OC, it follows that controlling diesel BC would have a cooling effect. The control of BC from diesel therefore is an effective means of mitigating near-term global climate change. This conclusion is consistent with findings of recent studies [Jacobson, 2010; *Bond et al.*, 2013], which conclude that reduction of BC from diesel sources would lead to global cooling. However, we would like to caution that, without simultaneous reduction of CO<sub>2</sub> emissions, it will not be possible to limit future warming to below 2°C as required by the Copenhagen accord.

### Background

Black Carbon (BC): Soot contains black carbon and organic carbon aerosols, which absorb and scatter solar radiation and thus impacts the climate system from local to regional and global scales. The component of soot that absorbs solar radiation is usually referred to as black carbon (BC) or elemental carbon (EC). The two terms, BC and EC, are used interchangeably and we follow the same practice in this report. BC or EC is simply a functional definition that depends on the measurement technique. In principle, the relatively strong light absorption properties of BC can be used to infer BC from an optical measurement and knowledge of the mass specific absorption of BC. In thermal methods, the filter media used for sample collection is heated and the thermally evolved carbon in the specified temperature plateau and analysis atmosphere defines the concentration of EC and organic carbon (OC).

Brown Carbon (BrC): Organic carbon is normally assumed to be a pure scattering aerosol. However, recent experimental studies have demonstrated that a fraction of OC also absorbs sunlight with their absorption increasing dramatically towards shorter wavelengths (<500 nm wavelengths). The absorbing part of OC referred to as brown carbon. In this report, the term OC includes BrC.

Radiative Forcing: The net effect of absorption and scattering of solar radiation by, BC and OC, is to alter the solar radiation absorbed by the surface and the atmosphere, which is the fundamental driver of the global climate system. The

change in the radiative heating of the climate system is referred to as the ‘Radiative Forcing’ [see the primer on radiative forcing later in the summary]. The change in the forcing due to scattering and absorption of solar radiation is referred to as ‘Direct Forcing’. But the addition of BC and OC aerosols as well as the alteration of solar heating by BC and OC alters the cloud fraction and cloud properties, which in turn alters the radiative forcing since clouds are the largest modulators of solar radiation. These cloud induced radiative forcings are referred to as indirect and semi-direct forcings [see the primer]. These changes to the solar radiative forcing [units of  $W m^{-2}$ ] are the primary metric used to assess the importance of BC and OC on climate.

Motivation [BC is the second Largest Contributor to Global Warming]: Black carbon (BC), the main light-absorbing component of soot is the principal absorber of visible solar radiation in the atmosphere. *Jacobson, [2002]* and *Ramanathan and Carmichael [2008]* concluded that BC is the second largest contributor to global warming, next to  $CO_2$ . For present day BC, due to both natural and anthropogenic sources, Ramanathan and Carmichael estimated a direct forcing of  $0.9 W m^{-2}$  [0.45 to  $1.35 W m^{-2}$ ] compared to  $1.6 W m^{-2}$  for  $CO_2$  [*Forster et al., 2007*] - this conclusion was debated because most estimates of forcing from models were a factor of two to three lower. A major landmark study of the BC forcing problem was published this year [*Bond et al., 2013*] by a group of researchers that included many modeling groups, and their estimate of  $0.88 W m^{-2}$  for the BC direct forcing (due to all BC sources) is nearly identical to Ramanathan and Carmichael’s estimate of  $0.9 W m^{-2}$ . Because of its short life times of few weeks, BC is concentrated close to the sources and hence a regional evaluation is critical for a better understanding of the global effects. Furthermore, California has witnessed major decreases in its BC concentrations and we need to understand the impact of these regional trends in climate.

### **The unique integrated approach**

We have developed a balanced approach between observations, data analyses, and modeling studies, allowing us to uniquely constrain the estimates provided in this study using measurements conducted by ground based network, aircraft and satellite instruments. The study consisted of four primary components (i) analysis of available measurements, and documentation of multi-decadal BC trends constrained by field measurements, (ii) estimation of the direct aerosol forcing due to black and brown carbon using integrated observations as well as models, (iii) source apportionment based upon chemical mixing state, and (iv) estimating the climate impact of BC emissions under various mitigation scenarios. The full climate impact of BC on the regional climate of California is evaluated by using regional climate models in a series of numerical experiments with varying BC emissions to determine changes in the surface temperature and hydrology. The regional climate models used to evaluate the climatic impacts of BC are also used in step (ii) to estimate the BC forcing, so their uncertainty can be assessed by comparing their estimated BC forcing with the observation based estimates. Statewide temporal and spatial resolved BC concentrations have been derived

by analyzing the coefficient of haze [COH] (directly correlated to the BC) recorded at 100 locations throughout California (data available from CARB). A key component of this work involves determining the actual single particle mixing state of soot particles in California. These measurements allow us to proceed without making major assumptions regarding the size, mixing state, and optical properties of ambient soot particles that have resulted in tremendously large uncertainties in prior studies. The details of our unique approach are given in the Methods section at the end of this section.

## **Principal Findings**

### **I. BC concentration results**

We examine the temporal and the spatial trends in the concentrations of black carbon (BC) using filter based mass measurements recorded by the IMPROVE monitoring network, and optical measurements reconstructed from the available Coefficient of Haze (COH) records in California.

- 1. Annual average BC concentrations measured at California IMPROVE sites have decreased by about 50% from  $0.46 \mu\text{g m}^{-3}$  in 1989 to  $0.24 \mu\text{g m}^{-3}$  in 2008 compared to a corresponding reductions in diesel BC emissions (also about 50%) from a peak of  $0.013 \text{ Tg Yr}^{-1}$  in 1990 to  $0.006 \text{ Tg Yr}^{-1}$  by 2008 (Figure 2(a)).*
- 2. A larger set of COH measurements is also used to determine BC concentrations and reveals that these trends are uniform across the state and persistent in several major air basins (Figure 2(b)).*

The consistency between the IMPROVE and COH trends is important since the COH data are largely from urban sites whereas IMPROVE is from remote sites.

- 3. A corresponding trend in co-pollutants such as nitrates, sulfates, and organic carbon is not observed (Figure 2(a)).*

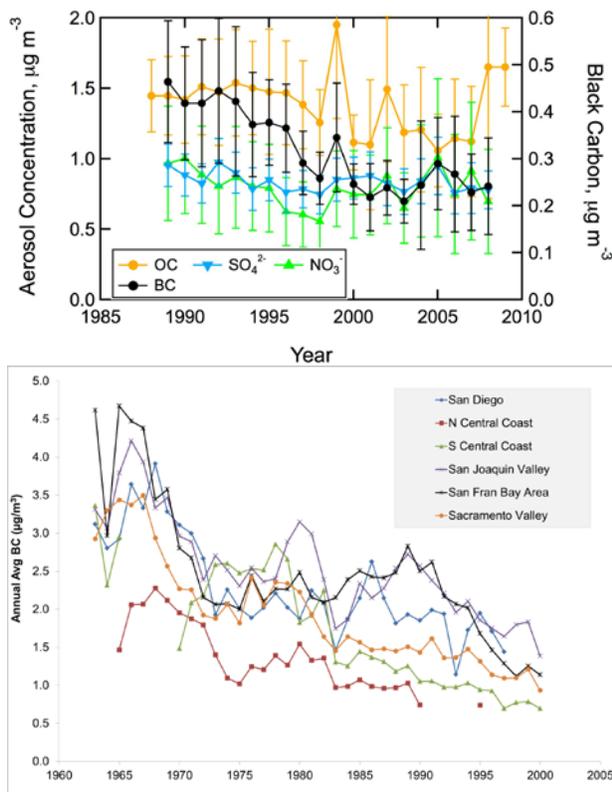
This finding is crucial, since the co-pollutants are largely cooling aerosols, through their direct and indirect effects on clouds. Since they are not showing negative trends, it implies that the decrease in BC will most likely lead to a global cooling effect from California's BC reductions.

- 4. As no similar trends are observed in other chemical source tracers (such as K, for biomass burning), we therefore attribute the observed BC trends primarily to the emission reduction from transport-related PM emissions, primarily from diesel.*

A detailed analysis of technology based emission inventories and fuel use in California indicates that although the total consumption in diesel fuel has

increased, the emissions of BC from diesel fuel combustion have decreased significantly. The reduction in emissions is due to a number of factors, including the introduction of low sulfur fuel, tighter emission standards, cleaner burning engines, and other improvements in technology mandated by statewide regulations. California therefore appears as a success story in mitigating the anthropogenic impact on climate. While, decreasing trends in BC have been noted in other states (of US), we are unable to comment on their climate mitigation efficacy since data on other co-pollutants have not been analyzed in published literature.

5. Our conclusion that the reduction in diesel emissions is the primary cause of the observed BC reduction is also substantiated by a significant decrease in the ratio of BC to non-BC aerosols.



**Figure 0.1** (a) Statewide trends in concentrations of common aerosol species from the IMPROVE network, and (b) historical BC concentrations resolved by air basin. A clear reduction in BC is observed.

## II. Role of brown carbon absorption

We have developed an observationally based analytical method for rigorously partitioning measured aerosol absorption optical depths (AAOD) and single scattering albedo (SSA) among EC and BrC using multi wavelength measurements of total (EC, OC and Dust) absorption.

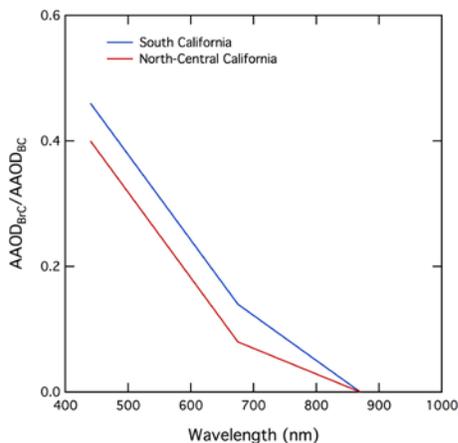
1. We have quantified column-integrated absorption in multiple wavelengths by brown carbon from observations for the first time.

The new method we developed separates dust, BC and BrC absorption from spectral solar observations. It can now be applied to other regions of the world.

2. Organic carbon (OC) is shown to absorb strongly at visible to UV wavelengths, an effect typically not represented in climate models. The BrC absorption at 440 nm is about 40% of the EC, while at 675 nm it is less than 10% of EC (Figure 2).

We find an enhanced absorption due to BrC in the summer months, and in South California (related to forest fires and secondary OC). The contribution of BrC to the total aerosol absorption is largest over northern and central California.

3. BrC emissions are likely both from biomass burning (forest fires and residential wood burning) as previously thought, and also from large aged particles indicating that secondary organics may also be absorbing.



**Figure 0.2** Wavelength dependent relative Absorption Aerosol Optical Depths (AAODs) for BC and BrC determined from ground-based estimates of the SSA and AOD in California.

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### III. Source Attribution and Speciation

1. Major differences exist in the PM sources in northern and southern California. In-situ measurements of optical properties and chemical mixing state reveal fossil fuel sources contribute the most strongly in southern California, whereas biomass burning and biogenic sources dominate in northern California. This has been shown in other recent studies as well (Cahill et al. 2012).

2. Using optical properties for estimating aerosol speciation using satellites or networks such as AERONET has the potential to provide extensive input into global climate and air pollution studies. This study demonstrates that the interpretation of the AERONET results is strengthened by complementary measurements of aerosol sources.

Speciation of dust, BC and BrC, are well classified by optical data, but the separation between fossil fuel and biomass burning sources has limitations because of their overlapping optical properties.

3. Despite these limitations, detailed comparison reveals the significance of aerosol absorption due to secondary organic aerosol (OC) which is currently underestimated in climate models, in addition to black carbon. Brown carbon and secondary sources impact many absorption events, indicating these processes must be given consideration when developing future climate mitigation policies.

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### III.IV. Radiative forcing results: Present day values

#### Radiative Forcing: A Primer

*Direct Radiative Forcing:* BC and OC increase the amount of solar radiation absorbed by the atmosphere, by intercepting the direct incoming solar radiation and also by intercepting the solar radiation reflected by the surface, the atmosphere and the clouds. The interception of solar radiation also leads to a decrease in solar radiation absorbed at the surface (dimming). The alteration of the solar absorption of the surface and the atmosphere by BC and OC is referred to as *direct radiative forcing*. The net effect is the sum of the atmospheric heating and the surface dimming and is referred to as the top-of-the atmosphere (TOA) forcing. The TOA forcing is the most relevant quantity for assessing the significance of California's BC policy actions for mitigating global warming. However, for assessing the impacts on California's regional climate, we have to examine the impact of EC and OC on atmospheric solar heating (atmospheric forcing) and surface solar dimming (surface forcing).

*Indirect and semi-direct Radiative Forcing:* BC and OC also influence cloud formation. They nucleate cloud drops that increase the number of cloud drops and also increase cloud fraction, both of which enhance the cloudy-sky albedo and lead to cooling. This is the indirect effect of BC and OC. On the other hand, the solar heating of the atmosphere by BC and BrC leads to burn-off of clouds. In addition, inclusion of BC in cloud drops also enhances solar absorption by the drops. This cloud-inclusion effect also enhances cloud burn-off. The burn-off in turn decreases the cloudy-sky albedo and thus provides an offset to the cooling effect of the indirect effect.

The annual average direct radiative forcing associated with BC+OC from the observationally constrained Top-Down (OC\_TD) approach, initialized with two assimilated data sets is summarized in Table 1. The total aerosol optical depth (AOD) is retrieved from the MISR satellite whereas the single scattering albedo (SSA) is based upon observations from the AERONET network, and from the GOCART climate model. The two different values of SSA allow us to estimate the upper and lower limits for the BC and BrC (details in Chapter 9). However, our baseline estimate is that given by the version of OC\_TD with SSA from AERONET. Table 1 also shows simulated forcing by the emission-inventory based bottom up (EI\_BU) WRF-Chem models. PNNL and LBL refer to the two modeling groups participating in our study.

1. *Direct Radiative Forcing by BC+OC is positive everywhere in California. It ranges from  $0.2 \text{ Wm}^{-2}$  in northern California to  $0.88 \text{ Wm}^{-2}$  over S. California.*

- 2. The annual average BC+OC direct forcing for California estimated using the WRF-Chem regional climate model (with the published emission inventory) is  $0.16 \text{ W m}^{-2}$ , compared to  $0.51 \text{ W m}^{-2}$  ( $\pm 40\%$ ) from the observationally constrained estimate. Thus, even regional models with region specific emissions, severely underestimate the BC+OC warming effects by a factor of three.*

The WRF-Chem model also underestimates surface BC concentrations by a factor of two. We show the forcing with BC emissions increased by a factor of 2 (4<sup>th</sup> column) that reduce this discrepancy, but the model forcing of  $0.29 \text{ W m}^{-2}$  is still much lower than the OC\_TD approach. Some of the remaining discrepancy can be further explained due to the neglect of BrC absorption in the regional models.

- 3. The underestimation (by a factor of three) is consistent with Bond et al. [2013]'s conclusion that models underestimate absorption by BC by a factor of three on a global mean basis.*
- 4. The BrC absorption has a major impact on the direct forcing due to OC aerosols. Without BrC absorption, models estimate direct forcing due to OC to be between  $-0.30$  and  $-0.15 \text{ W m}^{-2}$  depending on the location and region, whereas inclusion of BrC brings the net cooling down to between  $0.00$  and  $-0.10 \text{ W m}^{-2}$ . Thus models that neglect BrC severely overestimate the OC cooling effects.*

Table 2 presents a comparison in the atmospheric heating rates from the observationally constrained calculations, and the WRF-Chem regional model operating in two independent configurations. All three calculations produce similar trends in the magnitude of the forcing. This agreement obtained from three independent calculations indicates the robustness and significance of the results.

- 5. The atmospheric heating, or energy added to the atmosphere, is a factor of four to eight larger than the TOA forcing (Table 2).*

Carbonaceous aerosols are found to be warming in all regions and seasons with a magnitude ranging between  $0.5$  and  $3.5 \text{ W m}^{-2}$ . The greatest warming is observed in Southern California (which typically has a higher anthropogenic aerosol concentration and lower cloud cover), and also in the summer (possibly due to a higher net solar flux).

	<u>Observationally Constrained Top-Down</u>		<u>Emission Inventory Based Bottom-Up</u>	
	<b>(OC TD)</b>		<b>(EI BU)</b>	
	SSA from AERONET	SSA from GOCART	WRF_PNNL (1*BC)	WRF_PNNL (2*BC)
<b>TOA forcing</b>				
<b>North</b>	0.20	0.28	0.09	0.16
<b>Central</b>	0.44	0.48	0.17	0.32
<b>South</b>	0.88	1.92	0.21	0.40
<b>State</b>	0.51	0.89	0.16	0.29
<b>Atmospheric heating, W m<sup>-2</sup></b>				
<b>North</b>	1.73	1.79	0.60	0.82
<b>Central</b>	2.40	2.78	0.86	1.25
<b>South</b>	2.44	4.05	0.96	1.41
<b>State</b>	2.19	2.87	0.81	1.16
<b>Surface forcing, W m<sup>-2</sup></b>				
<b>North</b>	-1.54	-1.47	-0.51	-0.65
<b>Central</b>	-1.94	-2.38	-0.68	-0.92
<b>South</b>	-1.62	-2.47	-0.75	-1.01
<b>State</b>	-1.70	-2.11	-0.65	-0.86

**Table 0.1** Annual average radiative forcing attributed to BC+OC at the top of the atmosphere, atmospheric heating, and at the surface determined using the observationally constrained MACR model and the WRF-Chem climate model simulations based on emission inventories.

#### **IV-V. Surface Brightening:**

*Radiative calculations also estimate that the 50% decrease in BC concentration from the 1980s to current levels should have led to an increase in the surface solar radiation (surface brightening) between 1.5-3.5 W.m<sup>-2</sup>.*

We looked for this brightening effect in a network of surface solar radiometers operated by California's CIMIS network since 1982. The inter-annual variability was much larger than the detected trend in solar flux measurements and when this is considered with the limited data duration of the measurements (about 25 years), we are led to the deduction that the detected trends during the 1980s to 1990s are not statistically significant to verify the predicted brightening trends due to decreasing BC concentrations.

#### **V.VI. Implication to mitigation of global warming**

1. *Our findings thus far suggest that policies enacted by California to reduce diesel emissions should have led to a substantive mitigation of global warming.*

This conclusion is derived from the following observations and model simulations:

2. *The observed decrease in surface-BC by a factor of 2 from 1989 to 2008 and by a factor of 3.5 from 1960s to 2008.*

3. *This decrease was not accompanied by a comparable decrease in OC or other cooling aerosols such as sulfates.*

4. *As a result the decrease of BC was accompanied by a decrease in ratio of BC to Non-BC aerosols, which implies the warming aerosols decreased more in relation to cooling aerosols.*

6. *The observed BC decreases were largely attributed to the decrease in diesel emissions of BC.*

7. *Targeted emission controls adopted by the State of California are shown to be effective in reducing BC concentrations, by a factor of at least 2 dating back to the 1980s, and by as much as a factor of 3.5 dating back to the 1960s. At the regional level this decrease is associated with a TOA cooling of between 0.5 and 1.5 W m<sup>-2</sup>.*

8. *The indirect and semi-direct forcing changes due to the observed decrease in BC is expected to be much smaller than the direct forcing changes given above, due to the following two reasons: The negative indirect forcing is large mainly for OC and we did not observed statistically significant changes in OC concentrations (as expected for diesel related BC changes); As estimated by UNEP(2011) and Bond et al (2013), the sum of the indirect effect and the semi-direct effect is nearly zero for BC.*

The above finding, while it is the most important scientific aspect of our results, need to be validated by doing similar analyses for other states of US which have experienced strong reductions in BC.

In order to estimate the global warming mitigation from this diesel related BC forcing decrease, we need to account for not only the direct radiative forcing due to BC but also the following: Brown Carbon absorption; indirect effects of BC and OC; cloud-inclusion effects of BC and BrC; semi-direct effects of BC and BrC. We are aware of only one such modeling study by Jacobson (2010) which has explicitly estimated the global warming potential of fossil fuel BC. Furthermore, Jacobson's direct forcing estimate is consistent with our OC\_TD estimates. As a result, we rely on Jacobson's 100 Year-GWP, which for fossil BC is 3000 (using his lower range).

9. *We estimate that reduction of BC emissions primarily from diesel engines from 1989 to 2008 has mitigated global warming equivalent to reducing annual CO<sub>2</sub> emissions by 21 million metric tons/year. This corresponds to about 5% of California's 2009 CO<sub>2</sub> direct emissions of 393 metric tons.*

#### **VI.VII. Validation and Improvement of regional climate model treatment of BC forcing**

Our goal is to use Emission Inventory based bottom-up (EI\_BU) regional models to understand the regional climate effects of BC mitigation measures. However, we must validate these models first. In this study, we use the WRF-Chem model in a variety of numerical experiments to estimate the seasonal scale changes in radiative forcing. Two configurations of the WRF-Chem model were used, the standard version 3.2.1, and one that includes more updated cloud microphysical and radiative transfer parameterizations [Zhao *et al.*, 2012].

1. *A comparison between the simulated BC concentration and measurements at over 30 sites in California revealed that WRF-Chem under-predicted BC by nearly a factor of two, which indicates that the emission inventory developed in the ARCTAS-CA campaign may be under representing BC.*

The climate impact of BC was evaluated by changing the emissions by factors of 10x, 5x, and 2x. The 2x case approximates the conditions for the present day emissions; the 5x and the 10x cases approximate the emissions during the 1980s and the 1960s respectively.

2. *The simulated seasonal and regional variation in BC forcing is consistent with the patterns in OC\_TD and furthermore, in agreement with OC\_TD, the TOA forcing is positive everywhere. However, the direct forcing of (BC+OC) even with the 2x simulations is about 60% smaller than the OC\_TD values.*

Table 2 compares the atmospheric heating calculated using both the observationally constrained and climate model approaches. Two sets of observationally constrained calculations were performed – in the first, OC was

treated as a scattering species, and in the second, some of the OC was allowed to absorb light as well as the BC.

3. *The comparison indicates that about a third of this difference between the models and the observationally constrained approach is due to the fact that the models ignore BrC. With respect to the other 2/3, we must point out that the observational estimates have an uncertainty of ±40%.*

		Observationally Constrained BC Only	Observationally Constrained BC+OC	WRF-CHEM calculation using 2xBC emission: PNNL	WRF-CHEM calculation using 2xBC emission: LBL
Annual Average	North	1.36	1.54	0.82	0.99
	Central	2.10	2.26	1.25	1.31
	South	2.24	2.34	1.41	1.49
Spring Average	North	1.71	1.97	0.74	0.95
	Central	2.77	2.85	1.12	1.30
	South	2.41	2.43	1.37	1.55
Summer Average	North	1.95	2.21	1.35	1.85
	Central	3.17	3.49	2.04	2.14
	South	3.70	3.94	2.23	2.33
Fall Average	North	1.03	1.12	0.72	0.98
	Central	1.37	1.55	1.11	1.33
	South	1.95	2.09	1.25	1.48
Winter Average	North	0.75	0.85	0.45	0.45
	Central	1.08	1.15	0.70	0.64
	South	0.91	0.91	0.77	0.80

**Table 0.2** Comparison of Atmospheric Heating Rates in  $Wm^{-2}$  in California calculated using independent modeling studies. The observationally constrained heating rates have an uncertainty of about ±60% compared with the 40% uncertainty in the TOA forcing.

#### **VII.VIII. Regional climate impact results**

To determine the climate impacts of BC, we performed two simulations, each covering 5 years for the 2005 - 2009 meteorological conditions. In one simulation, we used 2xBC to represent the conditions of the 2000s, and in the other simulation, we used 10xBC to represent the conditions of the 1960s (with BC reduced by a factor of five to approximate the historical COH records). Following this decreasing trend, we expect the top of the atmosphere (TOA) to cool.

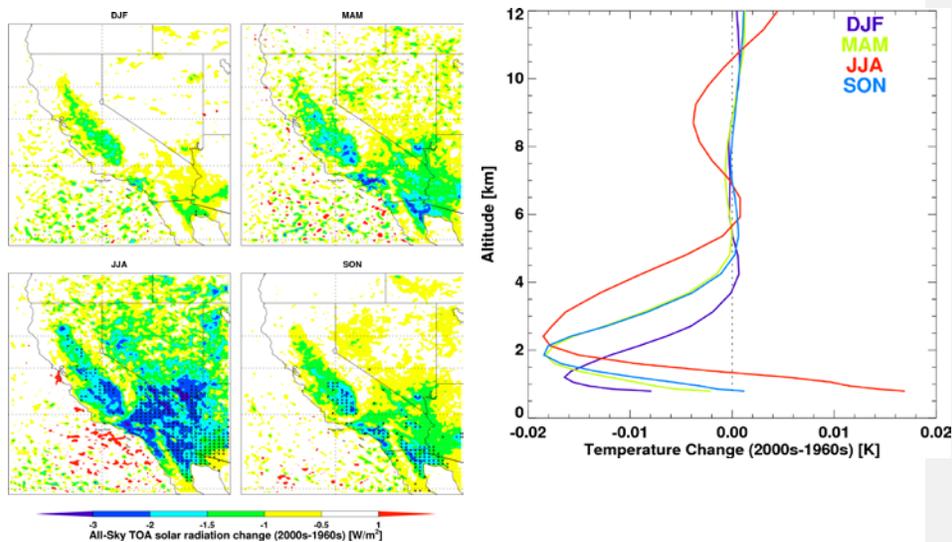
1. *Overall, comparing the 2000s to the 1960s we find that the TOA radiative forcing decreases by up to  $3 W m^{-2}$  (Figure 4) with larger and statistically*

significant changes at 90% confidence level in the summer and fall and over the Central Valley and southern CA.

The diabatic heating in the atmosphere is reduced by up to 0.06 K/day, and maximizes in the summer and at about 2 km above the surface. Reduction in BC leads to brightening at the surface as expected.

2. Corresponding to the enhanced surface solar radiation, skin temperature increases while the TOA radiative cooling leads to a cooling of atmospheric temperatures, but the average temperature changes are small ( $<0.1$  C) and are not statistically significant due to the large interannual variability.

Winds at 10m generally become weaker over CA but stronger off shore in southern CA. Finally, changes in surface hydrology are small because the BC effects are generally small during winter, and BC is mostly concentrated in the Central Valley and southern CA, so its effects are negligible in the Sierra Nevada, which is where hydrology is most critical.



**Figure 0.3** Changes in TOA solar radiation in the last 40 years attributed to BC decrease. Changes that are statistically significant at the 90% confidence level are stippled. The simulated atmospheric cooling and surface warming (only in summer) are shown on the right panel (not significant at the 90% level).

- 3. It appears that reduction of BC from the 1960s to 2000s produces a cooling of the lower atmosphere, accompanied by a statistically insignificant surface temperature changes.*

However, the larger scale and remote influence due to BC reduction over CA cannot be assessed in the regional modeling framework, which could also potentially affect the regional results over CA. In addition, the WRF-Chem model does not account for solar absorption by brown carbon. These uncertainties should be addressed in future studies.

### **Recommended future work**

A more comprehensive understanding of the climate impacts of California's BC (+OC) mitigation on global and regional climate (including impacts on California's snow packs) processes requires further work along the following lines:

#### **I. Global climate mitigation**

- 1. We need to estimate the magnitude and uncertainty associated with the regional indirect and semi-direct effects through the OC\_TD approach to better constrain the models.*

While the scope of this work was limited to determining the impact of BC, we discovered that BrC is also a significant absorber of radiation, particularly at short wavelengths and may serve as an extra warming agent due to its typically higher concentrations. The BrC is particularly important for California since our data reveals that open biomass burning is a large source for BrC. This opens up a whole new avenue of research as unlike BC, the sources, chemistry, and optical properties of BrC are less well understood. In particular we need to understand the following physic-chemical-optical properties of BrC:

2. What is the relative role of primary aerosols from biomass burning and diesel sources to BrC?
3. What is the relative role of secondary organics from biomass burning and diesel sources to BrC?
4. What are the intrinsic properties of BrC such as: size distribution, the single scattering albedo, and the angstrom exponent of absorption coefficient and scattering coefficient of BrC?
5. How does BrC within cloud drops and as interstitial aerosols in clouds enhance radiative forcing?

*We recommend extensive future work, including field studies, laboratory studies, and data assimilation and modeling studies to better understand BrC. Further, current climate models do not consider BrC absorption and treat OC as a cooling*

*agent – while there still exists an uncertainty in the final sign of the net OC forcing we assert that climate models are overestimating its cooling impact.*

In this report we presented several mitigation scenarios for BC that represent current and historical records of BC concentrations in California that are shown to be consistent with the control of diesel emissions. The mitigation scenarios considered in this study were constructed based on trends in California's aerosol concentrations. Although regional emissions are most important considering the short lifetime of BC, we did not consider variations in long-range transport (such as from Asia) in this study. Future studies should attempt to distinguish between transported and locally emitted BC.

## **II. Regional climate change**

*1. Continuation of EC/BC monitoring networks. The major findings of this study were made possible by the excellent networks of surface BC and column averaged aerosol optical properties. Unlike the long-lived greenhouse gases, documenting the impacts of mitigating short lived climate warmers such as BC requires continuation of such local and regional networks.*

In the course of this study, we also discovered large uncertainties in existing data sets and resources that are currently used by climate scientists, such as in the solar flux measurements (Chapter 9) and the BC emission inventory (Chapter 11).

*2. We recommend a more complete analysis of these resources and possible studies to implement better quality control, and to reduce the associated uncertainty.*

*3. Reducing uncertainty in regional model forcing: As documented here the OC\_TD and EI\_BU estimates of the forcing differ by a factor of almost two. Model treatment of EC and BrC emission inventory, optical effects, long range transport of BC from outside California, and vertical profiles of BC in models and simulation of clouds have to be validated first to bring the two approaches closer.*

For example, an earlier study by us [Hadley et al., 2007] revealed that about 75% of BC above 2 km over California during spring time is transported from Eastern Asia.

*4. A more detailed assessment of regional affects: The current study does not allow for the interaction with the larger scale circulation. Imbedding the WRF\_Chem in a global model is required to simulate the potential changes in large-scale circulation due to BC reductions in CA.*

*5. The model does not account for solar absorption of OC, so the climate influence may be underestimated. In addition, it has been shown in an earlier study [Hadley and Kirchstetter, 2012] that snowfall deposits about 90% of the BC emissions to the snow packs of California, contributing to their melting.*

6. Lastly, model biases and uncertainties in the model formulations and parameterizations, particularly related to clouds and aerosol-cloud interactions, should be addressed in the future to provide more robust simulations of BC climate effects.

