

Large reductions in urban black carbon concentrations in the United States between 1965 and 2000



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HIGHLIGHTS

- Mined retired coefficient of haze (COH) data to estimate black carbon (BC) trends.
- Determined COH-BC relationship with two reinstated COH monitors and aethalometers.
- BC concentrations decreased markedly between 1965 and 2000 across the U.S.
- BC decreased from 13 to 2 $\mu\text{g m}^{-3}$ in New Jersey and 4 to 1 $\mu\text{g m}^{-3}$ in California.
- Declining BC concentrations contrast increasing energy use and CO₂ emissions.

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ABSTRACT

Long-term pollutant concentration trends can be useful for evaluating air quality effects of emission controls and historical transitions in energy sources. We employed archival records of coefficient of haze (COH), a now-retired measure of light-absorbing particulate matter, to re-construct historical black carbon (BC) concentrations at urban locations in the United States (U.S.). The following relationship between COH and BC was determined by reinstating into service COH monitors beside aethalometers for two years in Vallejo and one year in San Jose, California: $\text{BC} (\mu\text{g m}^{-3}) = 6.7\text{COH} + 0.1$, $R^2 = 0.9$. Estimated BC concentrations in ten states stretching from the East to West Coast decreased markedly between 1965 and 1980: 5-fold in Illinois, Ohio, and Virginia, 4-fold in Missouri, and 2.5-fold in Pennsylvania. Over the period from the mid-1960s to the early 2000s, annual average BC concentrations in New Jersey and California decreased from 13 to 2 $\mu\text{g m}^{-3}$ and 4 to 1 $\mu\text{g m}^{-3}$, respectively, despite concurrent increases in fossil fuel consumption from 1.6 to 2.1 EJ (EJ = 10^{18} J) in New Jersey and 4.2 to 6.4 EJ in California. New Jersey's greater reliance on BC-producing heavy fuel oils and coal in the 1960s and early 1970s and subsequent transition to cleaner fuels explains why the decrease was larger in New Jersey than California. Patterns in seasonal and weekly BC concentrations and energy consumption trends together indicate that reducing wintertime emissions – namely substituting natural gas and electricity for heavy fuel oil in the residential sector – and decreasing emissions from diesel vehicles contributed to lower ambient BC concentrations. Over the period of study, declining concentrations of BC, a potent and short-lived climate warming pollutant, contrast increasing fossil fuel carbon dioxide (CO₂) emissions in the U.S. Declining BC emissions may have had the benefit of mitigating some atmospheric warming driven by increased CO₂ emissions with complementary health benefits.

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1. Introduction

1.1. Background

Black carbon (BC) is an air pollutant produced during fossil and biomass fuel combustion. It is a major component of diesel engine exhaust particulate matter (PM), which is classified as a toxic air contaminant and linked to significant health impacts including cancer (IARC, 2012). It also constitutes a significant portion of the PM emitted during biomass fuel combustion, a leading factor in premature mortality worldwide (Lim et al., 2012). In addition to adversely impacting public health, BC is a strong absorber of sunlight and contributes to climate change, alters regional precipitation patterns, and enhances ice melting in the Arctic and Himalayas (Hansen and Nazarenko, 2004; Menon et al., 2002). Recent assessments have found that the positive radiative forcing of BC emissions is comparable to or larger than that of other pollutants except carbon dioxide (CO₂) (Bond et al., 2013). Since BC has a short atmospheric lifetime, controlling PM sources with high BC mass fraction has been proposed as a means of combating near-term global warming in the face of increasing CO₂ emissions, with the co-benefit of improving air quality (Grieshop et al., 2009; Jacobson, 2002).

This study presents a historical record of ambient BC concentrations in the United States (U.S.) between the years 1965 and 2000. Such records are useful for illustrating the air quality benefits associated with emission controls and transitions to cleaner energy sources. For example, a retrospective examination of ambient lead concentrations shows the efficacy of unleaded gasoline use (EPA, 2014; Needleman, 2000). Likewise, long-term carbon monoxide and sulfur dioxide trends demonstrate how automobile catalytic converters, reducing sulfur emissions from power plants, and reducing the sulfur content of fuels improved ambient air quality (EPA, 1998; Hand et al., 2012). Pollution trends can also support epidemiological studies, emission inventory development, and air quality modeling.

Whereas the abovementioned criteria air pollutants have been monitored for many decades in the U.S., similar long records of BC concentrations do not exist. Historical BC concentrations must be estimated from other PM air pollution measurements. Murphy et al. (2011) examined elemental carbon (EC) concentration trends measured at remote locations across the U.S. by the Interagency Monitoring of Protected Visual Environments (IMPROVE) network of aerosol samplers. Annual and wintertime average EC decreased by 30% and 50%, respectively, between 1990 and 2004. Bahadur et al. (2011) interpreted California IMPROVE EC as BC and indicated a 50% decrease over the 20-year period beginning 1998.

In this study, BC concentrations are estimated using archived records of coefficient of haze (COH). Whereas IMPROVE samplers are remotely located mostly in national parks, COH monitors were primarily operated in urban regulatory monitoring networks. The COH record begins in the early 1960s, which affords an opportunity to look back several decades, as well as over different time scales, to provide some insight about dominant BC sources and other factors affecting concentration trends. This study extends an earlier analysis of BC concentration trends in the San Francisco Bay Area (Kirchstetter et al., 2008) to other urban air basins in California and nine additional states stretching from the East Coast to the West Coast of the U.S.

1.2. Coefficient of haze

COH was an early air pollution particulate matter measurement method in the U.S. (Hemeon et al., 1953). Air quality agencies in many states deployed COH monitors in urban locations beginning

in the mid-1960s. COH has been related to contemporary measures of atmospheric PM, including total suspended particles and coarse and fine particle mass concentrations, but the technique for measuring COH closely resembles that now widely used for measuring BC rather than PM, which is sensitive to light-absorbing material and insensitive to nonabsorbing light-scattering material (Wolff et al., 1983). As newer, more specific methods for measuring PM were adopted by regulatory agencies, COH instruments were retired. For example, California operated COH monitors at more than 50 locations in 1980, but that number had dropped to 15 by 2003. Only one monitor remained in use in California by 2005.

The COH instrument, like the aethalometer used for measuring BC (Hansen et al., 1984), is a filter-based aerosol absorption photometer. In both instruments (shown in Fig. S1 of the Supporting Information), air is continuously drawn through a white filter and the intensity of light transmitted through the filter is periodically measured. Transmission decreases as light-absorbing particles are collected on the filter. COH and BC are analogously calculated with Equations 1 and 2, respectively:

$$\text{COH}(1000\text{ft}^{-1}) = \log(1/T) \frac{A_{\text{COH}}}{Q_{\text{COH}} \Delta t} \quad (1)$$

$$\text{BC}(\mu\text{g m}^{-3}) = \frac{1}{\sigma} \ln(1/T) \frac{A_{\text{aeth}}}{Q_{\text{aeth}} \Delta t} \quad (2)$$

T is transmission calculated from successive light intensity measurements over the period Δt (s), A is the area of the filter used to collect particles (m²), and Q is the volumetric air sampling rate (m³ s⁻¹). In Equation (2), σ is an attenuation coefficient (m² g⁻¹) relating particle light attenuation, $\ln(1/T)$, to BC mass. The aethalometer reports BC in units of mass concentration. COH is defined as the aerosol amount that produces an optical density, $\log(1/T)$, of 0.01 and concentrations are reported in terms of COH per 1000 linear feet (305 m) of sampled air.

The COH monitor uses a broadly emitting incandescent lamp to illuminate the particle sample and a photocell to measure the intensity of transmitted light, whereas the contemporary aethalometer uses a discrete light-emitting diode focused at 880 nm and a photodiode sensor. Light scattering within the aethalometer's quartz fiber filter (Pallflex type Q250F) amplifies its response to light-absorbing particles and diminishes its response to light-scattering particles (Hansen et al., 1984; Arnott et al., 2005). Similarly, Hemeon et al. (1953) reported that the COH filter paper (Whatman No. 4) amplified the instrument's response to smoke particles.

2. Methods

2.1. Sampling

To support constructing a historical record of ambient BC, COH monitors and aethalometers were collocated and operated at two locations. Two COH monitors were restored to working condition with original lamps, photocells, and filter paper. Each unit was upgraded with a mass flow controller (see Fig. S1) and connected to a computer for data acquisition and tape advance control. Prior to field deployment, both COH monitors were evaluated in the laboratory using soot generated with a methane-air flame (Kirchstetter and Novakov, 2007). Measured COH concentrations from the two monitors were in good agreement: linearly correlated with a slope of 1.03, zero intercept, and correlation coefficient $R^2 = 0.96$ (Fig. S2). These tests established that time-resolved COH concentrations, like time-resolved BC concentrations measured using the aethalometer, are affected by a "filter loading" artifact (Weingartner et al., 2003;

Arnott et al., 2005; Kirchstetter and Novakov, 2007; Coen et al., 2010), wherein COH concentrations erroneously decrease as the instrument's particle collection filter becomes increasingly loaded with BC (Fig. S3). These tests also established that averaging COH concentrations over one or more sampling cycles largely eliminates the influence of this artifact (Fig. S3). Therefore, in this study, we examine trends in concentrations averaged over days, months, and years rather than hours.

After laboratory evaluation, the COH instruments were collocated with aethalometers in the air-monitoring network of the Bay Area Air Quality Management District (BAAQMD) where COH was historically measured in Vallejo and San Jose. Vallejo's population is 120,000 (in year 2010) and the monitoring station sits in a mixed commercial and residential neighborhood one mile east of downtown and 0.5 miles west of a major freeway. San Jose is the largest city in the San Francisco Bay Area with a population of 950,000 (in year 2010). The air monitoring station is located in a commercial and residential part of downtown San Jose encircled by major freeways. The aethalometers and COH monitors were operated for 14 months (March 2010 through April 2011) in San Jose and 25 months (March 2010 through March 2012) in Vallejo. One aethalometer had recently been serviced and was active in the monitoring network. The other had been inactive and was serviced by the manufacturer at the start of this study.

Consistent with historical practice, the COH monitors in this study were installed with 0.25 inch Teflon sample tubing, without an inlet cyclone, and with an airflow rate of 6.25 L min^{-1} . Consistent with contemporary sampling practice, each aethalometer sampled ambient air through a $2.5 \mu\text{m}$ diameter cut-point inlet cyclone (BGI, model SCC1.829) and electrically conductive tubing (0.375 inch aluminum in San Jose and 0.625 inch stainless steel in Vallejo). The aethalometers were operated at an airflow rate of 5 L min^{-1} .

The COH monitors were initially programmed to advance their filter tapes once every 2 h, again consistent with historical practice. However, current ambient BC concentrations were too low to provide sufficient signal after only 2 h of sampling. To increase the amount of PM collected on the COH filter and thereby increase the measurement sensitivity, the instruments were reprogrammed for tape advances once every 6 h. With each advance, a 6-h average COH concentration was recorded. In effect, the 6-h sampling interval provided the same filter loading that a 2-h interval would have accumulated when COH concentrations were three times higher than they are now. The aethalometer was programmed to advance its filter when measured attenuation reached the factory default value of 75, which occurred about four times every five days.

2.2. Data sources

The California Air Resources Board (CARB) and the New Jersey Department of Environmental Protection (NJDEP) provided COH data collected in California and New Jersey from the early 1960s to 2000. COH monitors were located predominantly in urban areas and the location and number of active monitors varied over time (Fig. S4). The Environmental Protection Agency's (EPA) Air Quality System (EPA, 2012) supplied COH data for several other states. The COH records of some states were not preserved as well as those kept by California and New Jersey. As such, the data obtained from the EPA were comparatively limited both temporally and spatially. The following presentation includes the data-rich states of California and New Jersey and eight other states with COH records covering at least ten years: Colorado, Illinois, New York, Missouri, Ohio, Pennsylvania, Virginia, and Washington.

Since BC is a primary air pollutant, urban emissions and local concentrations are tightly linked (Apte et al., 2012). Therefore, long-

term trends in annual average BC concentrations and weekly cycles in daily average BC concentrations are discussed in relation to energy consumption and CO_2 emissions trends as well as temporal patterns in emission sources. Energy consumption and CO_2 emissions data are from the U.S. Energy Information Administration (EIA, 2012; EIA, 2016).

Meteorological variability over the course of a year moderates air pollution levels. Thus, seasonal trends in BC concentrations are discussed in relation to the local meteorology, namely wind speed (u) and mixing height (H), which govern pollutant dispersion. Meteorological data were acquired with minimal processing as described by Apte et al. (2012) for the 2007–2009 period from NASA's Modern-Era Retrospective Analysis for Research and Applications (MERRA) (Rienecker et al., 2011). We assume the meteorological trends are the same for the whole period of COH record. MERRA uses the Goddard Earth Observing System atmospheric model to assimilate meteorological observations into a global data set with a spatial and temporal resolution of 0.5° longitude \times 0.76° latitude \times 1 h. At mid-latitudes in the U.S., this corresponds to a spatial resolution of approximately $50 \times 75 \text{ km}$. Data for cities was extracted by matching latitude and longitude coordinates to the nearest MERRA grid point. The harmonic means of u , H , and uH were calculated. The product uH is the normalized dilution rate ($\text{m}^2 \text{ s}^{-1}$) and provides a measure of atmospheric dilution for an urban area (Apte et al., 2012; Marshall et al., 2005).

3. Results and discussion

3.1. Relationship between COH and BC

Fig. 1 shows the relationship between daily averaged concentrations of BC and COH at the Vallejo and San Jose sites. Lowest concentrations were measured in May, June, and July and the highest in December and January. A linear regression of the data from both locations yields the equation that we use in this study to calculate BC concentrations:

$$\text{BC}(\mu\text{g m}^{-3}) = 6.7\text{COH} + 0.1 \quad (3)$$

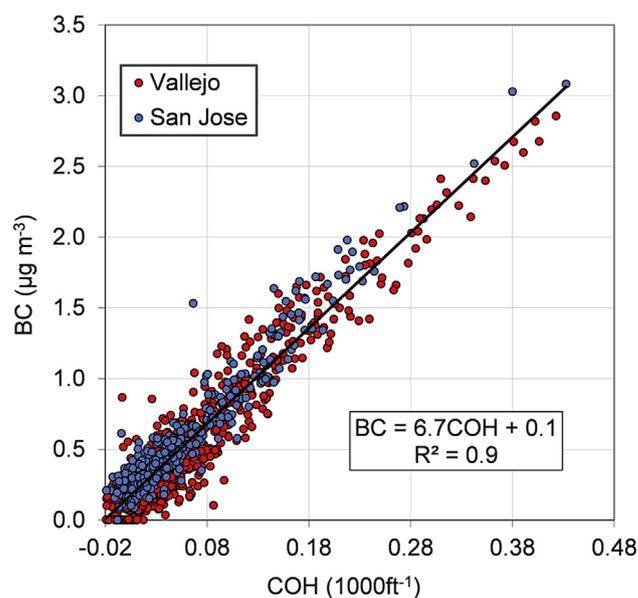


Fig. 1. Scatter plot of daily averaged BC and COH concentrations measured in San Jose (March 2010 through April 2011) and Vallejo (March 2010 through March 2012). The linear regression applies to the entire data set.

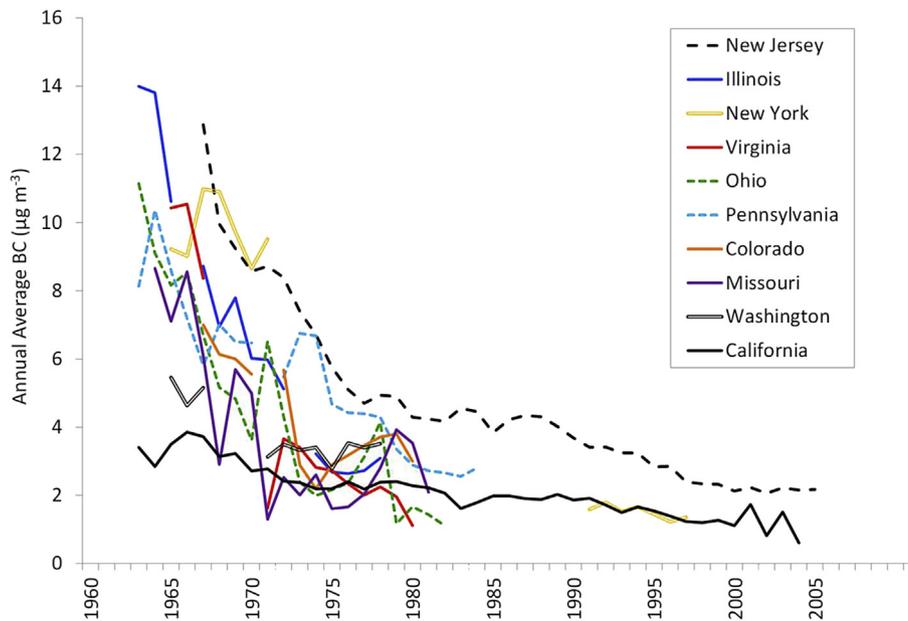


Fig. 2. Annual average urban BC concentrations in ten states. California's BC concentration trend includes the densely populated South Coast air basin only from 1969 to 1976 because that region's COH data were unavailable outside of this period. BC concentrations in the South Coast air basin were higher than in other California air basins (Fig. S6), but increased statewide average BC concentrations only by a small amount.

The high correlation coefficient ($R^2 = 0.9$) indicates that COH is a good proxy for BC. The linear relationship suggests that the COH-BC relationship is not seasonally dependent. Moreover, since the BC fraction of ambient PM varies throughout the year as a result of meteorological influences on secondary PM, the relationship between COH and BC does not appear to be highly sensitive to ambient PM composition. The intercept of the linear equation is small compared to past BC concentrations (see Fig. 2), which means that historical BC and COH are approximately proportional. Analysis of monthly averaged concentrations of BC and COH yields essentially the same result (Fig. S5), indicating that the same relationship can effectively be used to convert daily and monthly averaged values of COH to BC.

Others have previously examined the relationship between COH and EC. While EC and BC are not the same, they are often well correlated (Venkatachari et al., 2006), such that examining the relationship between EC and COH may be instructive. The BAAQMD derived EC ($\mu\text{g m}^{-3}$) = 7.5COH from measurements during the period 1991–2004 in San Francisco and San Jose, California (BAAQMD, 2007). A decade prior in the summer of 1981 in Detroit, Michigan, Wolff et al. (1983) measured EC ($\mu\text{g m}^{-3}$) = $8.9\text{COH} - 0.63$ with $R^2 = 0.9$ for EC concentrations up to $10 \mu\text{g m}^{-3}$.

Together, the data collected in this work (Fig. 1) and the findings of the studies cited above demonstrate that COH correlates with light-absorbing PM, indicate that the relationship does not change dramatically with location or time, and thus support using COH to estimate past BC concentrations. While uncertainty remains in the use of a constant COH-BC relationship at all sites, over all years, and at concentrations exceeding those in Fig. 1 and the above-cited studies, it facilitates a reasonable analysis of the COH database.

3.2. Black carbon concentration trends

Fig. 2 shows the trends in annual average urban BC concentrations for the ten states considered in this study, in each case computed as the arithmetic mean of BC concentrations at all monitoring sites in the state. At the start of this record, BC concentrations were highest in New Jersey, New York, and Illinois and

lowest in California. Over the 15 year period from the mid-1960s to 1980, BC concentrations decreased by 5-fold in Illinois, Ohio, and Virginia, 4-fold in Missouri, and 2.5-fold in Pennsylvania. From the mid-1960s to 2000, annual average BC concentrations in the urban regions of New Jersey and California decreased from 13 to $2 \mu\text{g m}^{-3}$ and 4 to $1 \mu\text{g m}^{-3}$, respectively. Fig. S6 shows BC concentrations separately for different air-basins in California and New Jersey.

New Jersey's greater reliance on BC-producing fuels in the 1960s is most likely why the BC concentration reduction was more dramatic in New Jersey than California. Clean burning natural gas was the single largest energy provider in California from 1960 to 2000, whereas energy production in New Jersey relied more on residual and distillate fuel oils than natural gas until 1974 (Fig. S7) and coal was consumed in significant amounts by New Jersey's industrial sector up until about 1970 (Fig. S8).

Fig. 3 and Fig. S9 show that BC concentrations decreased much more in winter than summer in many East Coast states. The transition to lower wintertime BC concentrations was especially quick for Pennsylvania and Illinois, where the seasonal BC cycle essentially disappeared (i.e., the curve flattened out) between the 1960s and 1970s. Pennsylvania's wintertime BC concentrations decreased from about 10 to $5 \mu\text{g m}^{-3}$, whereas its summertime concentrations did not decrease much at all. An annual cycle is evident in New York's data in the 1960s, but much less so in the 1990s. Similarly, the seasonality evident in New Jersey's BC concentrations in the 1970s steadily diminished to the point where concentrations were relatively uniform throughout the year in the 1990s and 2000s.

Seasonal variations in BC concentrations reflect the combined influence of emissions and meteorology. Whereas the latter probably has not changed when averaged over many years, a transition to cleaner fuels used for wintertime heating has occurred. For example, beginning in the early 1970s, BC-producing distillate fuel oil in New Jersey's residential sector (EDF, 2009) was substituted with cleaner burning natural gas and electricity (Fig. S10). Therefore, reduced wintertime emissions associated with heating likely caused the seasonality in BC concentrations to disappear and drove the major reductions in annual BC concentrations early in the record (Fig. 2), as previously discussed.

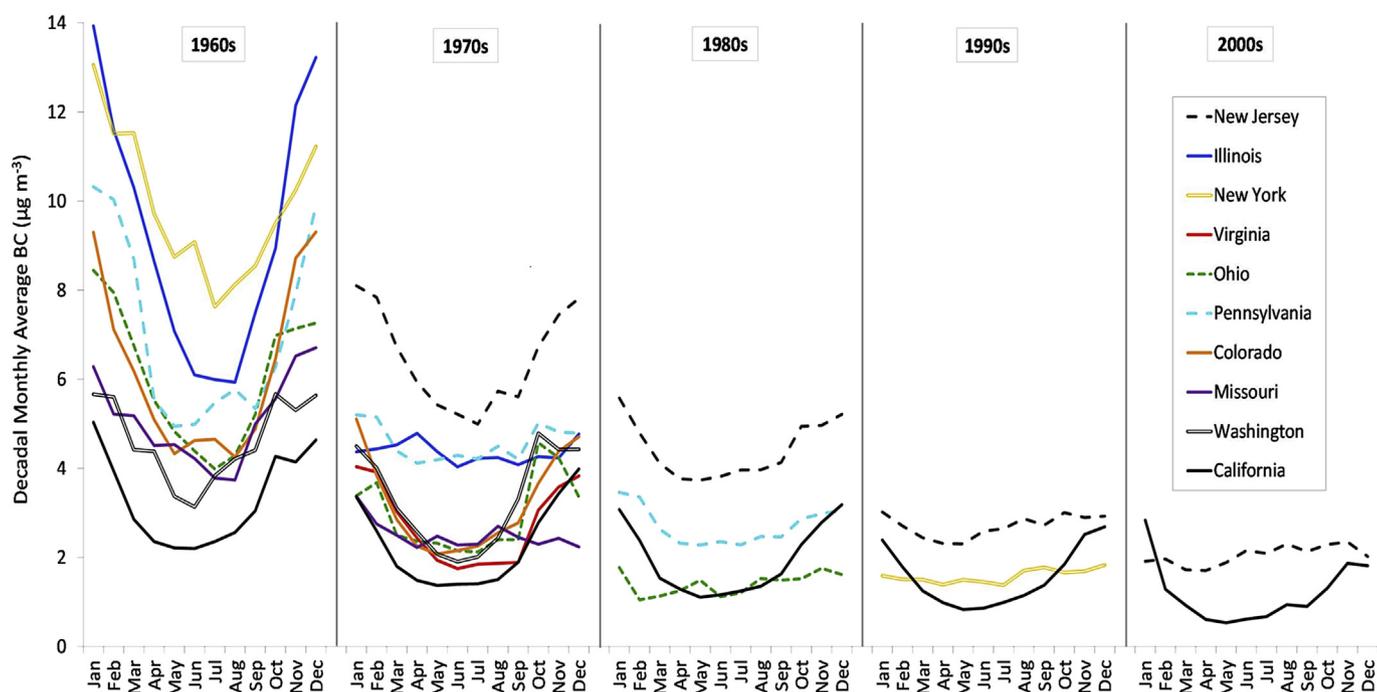


Fig. 3. Monthly average BC concentrations by decade in ten states.

In contrast, California's seasonal cycle persisted throughout the entire period of record: BC concentrations are consistently 2 to 3 times higher in the winter than in the summer (Fig. 3 and Fig. S9). This seasonality appears to be driven by meteorology rather than emissions. Whereas California's residential energy sector relied on cleaner energy sources over the entire study period (Fig. S10), atmospheric dispersion of pollutants in California is 2–3 times stronger in summer than in winter (Figs. S11 and S12; Glen et al., 1996). Thus, BC concentrations in urban California are naturally much higher in winter. The seasonal variation in atmospheric dilution in New Jersey and other eastern cities is weaker and opposite that of California, consistent with the absence of a strong seasonal cycle in BC after the wintertime heating emissions were largely reduced. The study by Glen et al. (1996) similarly attributed wintertime maxima and summertime minima in CO concentrations in San Francisco, California to lower wind speeds and mixing heights in the winter, and a weak or nonexistent seasonality in concentrations across several East Coast cities to the lack of a seasonal cycle in pollutant dispersion (Fig. S13).

In addition to the seasonal cycle, all states exhibit a weekly pattern of lower BC concentrations on weekends compared to weekdays and lowest BC concentrations on Sundays (Fig. 4). For example, BC concentrations in the 1970s in Missouri and Colorado were approximately 1.5 times higher during the week than they were on Sunday. This weekly cycle strengthened in most cases as baseline BC concentrations decreased over time. The persistent weekly pattern points to a source of BC emissions with the same weekly pattern that has been persistently dominant throughout the study period. Emission inventories include diesel trucks as a major BC source (e.g., Bond et al., 2004) and, as previously reported, diesel truck activity is appreciably lower on weekends. For example, on-road diesel truck volume on Sundays is 25% of the weekday average (Dreher and Harley, 1998) and weekend off-road diesel vehicle activity is less than 10% of weekday activity (Coe et al., 2003).

A number of studies have attributed declining ambient BC (or

EC) concentrations to diesel truck emission regulations (Christoforou et al., 2000; Murphy et al., 2011; Bahadur et al., 2011), notably smoke emission limits in 1970 and PM emission standards beginning in 1988 (Dieselnet, 2016). According to Sawyer and Johnson (1995) and Lloyd and Cackette (2001), PM emission factors from new heavy-duty highway diesel engines decreased by a factor of 10 from 1970 to 1990. Changes in in-use emission rates are more pertinent than new truck emission standards when relating to ambient concentration trends. The longest span of measurement of in-use heavy-duty diesel truck fleet PM emissions at one location is 1975–1999 in Pennsylvania's Tuscarora Tunnel. Over that 24-year period, the fleet average diesel PM emission factor decreased by a factor of 6 (Gertler et al., 2002). This is double the factor of 3 increase in taxable (i.e., highway) diesel fuel consumption in California and triple the factor of 2 increase in New Jersey's distillate consumption (not exclusively used by highway trucks) between 1975 and 2000 (Fig. S14). More recently, between 1997 and 2006, the average BC emission factor for in-use heavy-duty diesel trucks in California's Caldecott Tunnel decreased by 61% (Ban-Weiss et al., 2008), about twice as much as consumption of California taxable diesel increased (33%) over the same period (Fig. S14). These data suggest that the decrease in on-road diesel truck BC emission rates outpaced increased fuel consumption and thus contributed to the trend of decreasing BC concentrations shown in Fig. 2.

The recent study of McDonald et al. (2015) highlights the importance of off-road engines when assessing long-term ambient BC trends. Off-road diesel engines were not subjected to EPA emission standards until 1996 and have higher emission rates than highway diesel vehicles (Dieselnet, 2016). Data available for California indicate that off-road diesel fuel consumption declined by 40% from 1970 to 2000 (Fig. S14). Thus, BC emissions from off-road diesel engines likely also contributed to the ambient BC concentration decline over the period of record in California, due to decreasing consumption rather than reduced emission factors.

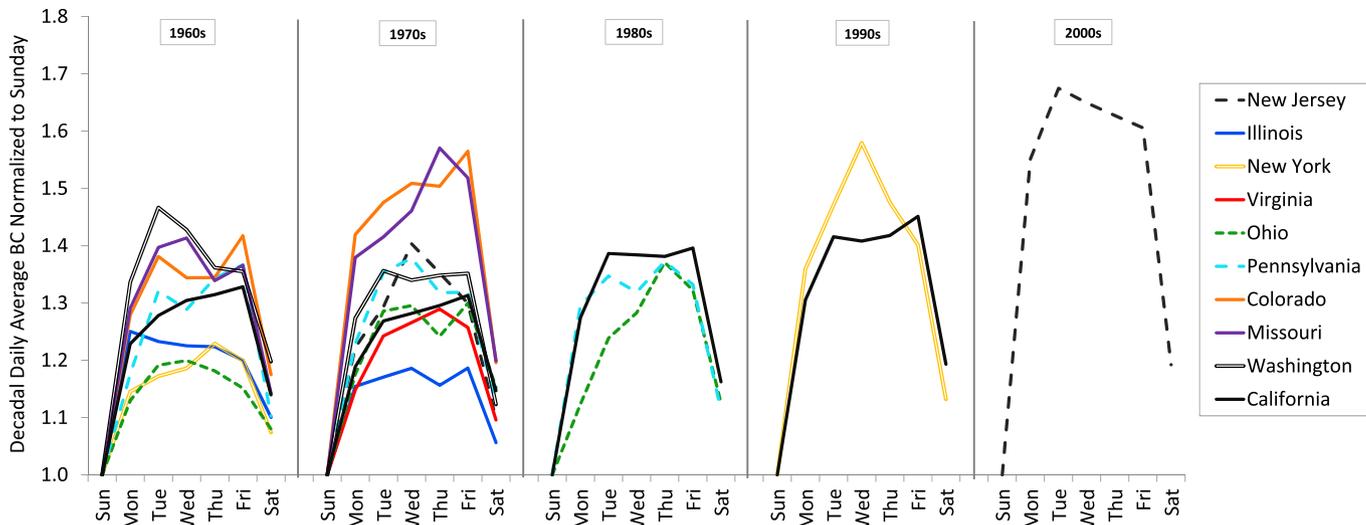


Fig. 4. Weekly cycle of BC concentration by decade. The vertical axis reports the ratio of average BC concentrations each day of the week to the average Sunday concentration.

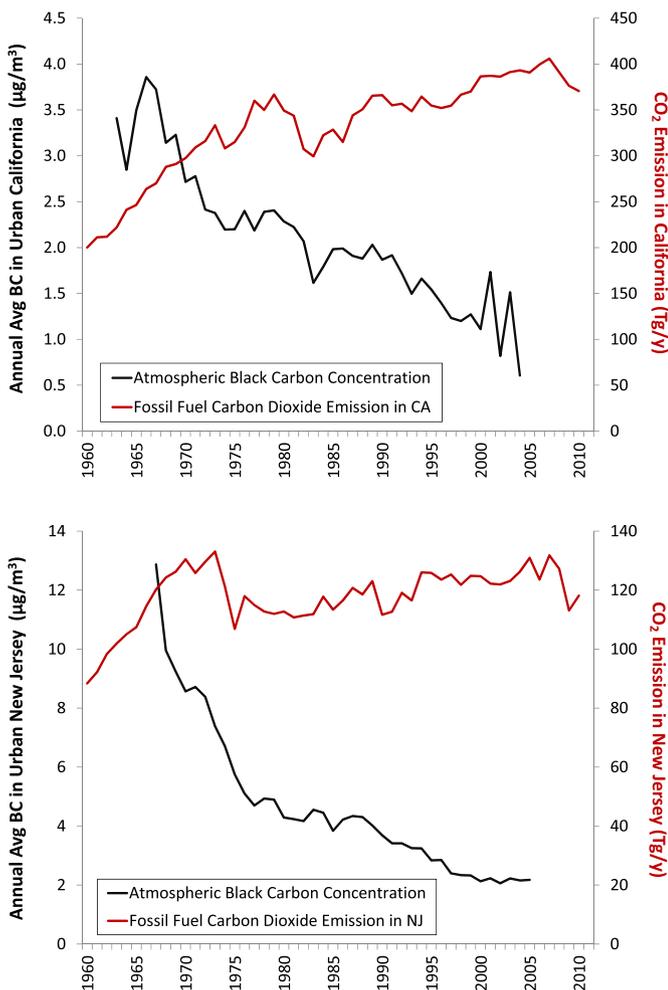


Fig. 5. Major reductions in BC concentrations over time contrast with increases in CO₂ emissions in both California (top) and New Jersey (bottom).

3.3. Relationship between BC and CO₂

Over the period from the mid-1960s to the early 2000s, when annual average BC concentrations in New Jersey and California decreased from 13 to 2 $\mu\text{g m}^{-3}$ and 4 to 1 $\mu\text{g m}^{-3}$, respectively, fossil fuel consumption increased from 4.2 to 6.4 EJ in California and 1.6 to 2.1 EJ in New Jersey (Fig. S15). Fig. 5 shows the rise in energy consumption trends in terms of fossil fuel greenhouse gas CO₂ emissions, which strongly contrast with declining BC concentrations. Since BC is a potent but short-lived climate warming pollutant, reducing BC emissions may have had the benefit of mitigating some atmospheric warming driven by increased CO₂ emissions. For example, Bahadur et al. (2011) reported a negative radiative forcing (i.e., climate cooling) over California due to a reduction in BC concentrations. Given that BC is a major component of diesel PM, which is a toxic air pollutant and a human carcinogen, these reductions may have had complementary health benefits (Anenberg et al., 2012).

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.atmosenv.2016.11.001>.

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