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 $\mu g\ m^{-3}$ in New Jersey and 4 to 1 $\mu g\ m^{-3}$ in California.
- Declining BC concentrations contrast increasing energy use and CO_2 emissions.

ARTICLE INFO

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: BC ($\mu g\ m^{-3}$) = 6.7COH + 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 $\mu g\ m^{-3}$ and 4 to 1 $\mu g\ m^{-3}$, 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 (CO_2) emissions in the U.S. Declining BC emissions may have had the benefit of mitigating some atmospheric warming driven by increased CO_2 emissions with complementary health benefits.

Published by Elsevier Ltd.

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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 (CO_2) (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 CO_2 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:

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

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

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 (m^2), and Q is the volumetric air sampling rate ($m^3 \, s^{-1}$). In Equation (2), σ is an attenuation coefficient ($m^2 \, g^{-1}$) 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;