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Constraining a Historical Black Carbon Emission Inventory of the United States for 1960-2000

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Key Points:

1. Systematic evaluation of long-term United States black carbon observations identifies a small number of poorly estimated emission sources.

2. Updated black carbon emission is higher than the previous estimate by 80% for 1960-1980, showing a decreasing trend as found in observation.

3. Emission factors for pre-regulation vehicles, off-road engines and residential heating stoves in 1980 and before should be increased.

1

Abstract

We present an observationally-constrained United States black carbon emission inventory with explicit representation of activity and technology between 1960 and 2000. We compare measured coefficient of haze data in California and New Jersey between 1965 and 2000 with predicted concentration trends and attribute discrepancies between observations and predicted concentrations among several sources based on seasonal and weekly patterns in observations. Emission factors for sources with distinct fuel trends are then estimated by comparing fuel and concentration trends and further substantiated by in-depth examination of emission measurements. We recommend (1) increasing emission factors for pre-regulation vehicles by 80%-250%; (2) increasing emission factors for residential heating stoves and boilers by 70% to 200% for 1980s and before; (3) explicitly representing naturally aspired off-road engines for 1980s and before; (4) explicitly representing certified wood stoves after 1985. We also evaluate other possible sources for discrepancy between model and measurement, including bias in modeled meteorology, sub-grid spatial heterogeneity of concentrations, and inconsistencies in reported fuel consumption. The updated United States emissions are higher than the *a priori* estimate by 80% between 1960 and 1980, totaling 690 Gg/yr in 1960 and 620 Gg/yr in 1970 (excluding open burning). The revised inventory shows a strongly decreasing trend that was present in the observations but missing in the *a priori* inventory.

1.Introduction

Aerosols play an important role in altering the radiation balance of the Earth, and this effect is critical in explaining the historical pattern of temperature change (Santer et al., 1995) as well as in predicting future climate. The climate effect of aerosols mainly depends on atmospheric concentrations, optical properties, and interactions with clouds. Most aerosols have a net cooling effect by scattering light. However, some aerosols can also absorb sunlight, heat the atmosphere and oppose the cooling effect of the scattering aerosols. The aerosol warming effect has been recognized since the 1960s (Charlson & Pilat, 1969) and has been mainly attributed to black

2

carbon (BC) (Rosen et al., 1978). Later studies demonstrated the important influence of BC on regional and global climate (J. Hansen et al., 2000; J. Hansen & Nazarenko, 2004; Jacobson, 2001, 2002; Menon et al., 2002; Ramanathan & Carmichael, 2008). BC warms the atmosphere by absorbing visible light (Bergstrom et al., 2002; Bond et al., 2011; J. Hansen & Sato, 2001; Kirchstetter et al., 2004; Schulz et al., 2006; Smith et al., 2000), influences the formation and properties of water and ice clouds (Chung & Seinfeld, 2005; J. Hansen et al., 2005; Johnson et al., 2004; Riemer et al., 2009), and its deposition on snow reduces the planetary albedo and accelerates glacial melting (Chỳlek et al., 1983; Flanner et al., 2007).

BC is a type of carbonaceous material produced by incomplete combustion of carbon-based fuel. Major sources include emissions from fossil fuel for transportation, biofuel for industrial and residential use, and open burning of biomass (Allen et al., 1999; Bond et al., 2007; Carcaillet et al., 2002; Cochrane, 2003; Cooke & Wilson, 1996; Dickerson et al., 2002; Penner et al., 1993; Seiler & Crutzen, 1980). It is mainly removed from the atmosphere by wet deposition after several days (Jacobson, 2004; Koch, Menon, et al., 2009). Because of its short atmospheric lifetime, mitigation of BC can yield climate benefits that occur immediately, complementary to benefits of greenhouse-gas reductions that are observed over long periods (J. Hansen et al., 2000; Jacobson, 2004). Because BC contributes to anthropogenic climate forcing, accurate long-term historical emission inventories are important to discern the signal of anthropogenic influence against the natural variability of climate. BC is also a good tracer of incomplete combustion due to its chemical stability and short lifetime. In contrast, other incomplete combustion products (carbon monoxide, primary organic aerosol and volatile organic compounds) either have non-combustion sources or are transformed into other species. Therefore, the observation of historical BC changes may also provide an indication of changes in other emitted combustion products.

Several historical BC emission inventories have been developed (Bond et al., 2007; Cofala et al., 2007; Klimont et al., 2016; Novakov et al., 2003). It is estimated that BC was primarily emitted from North America and Europe in the early 20th century and then greatly reduced (Bond et al., 2007; Novakov et al., 2003). However, discrepancies have been found between

3

ambient observations and emission estimates. Novakov and Hansen (2004) found different rates of reduction between atmospheric BC concentrations and estimates of BC emissions between 1962 and 2000 in Great Britain. Kirchstetter et al. (2017) found three to five fold reduction in BC concentrations in seven states reduced in the US from 1965 to 1980, while the fuel use and bottom-up emissions by Bond et al. (2007) increased during the same period. In this work, we quantitatively examine this apparent contradiction.

The aim of this work is to improve the accuracy of the BC historical emission inventory in the United States (US) from 1960 to 2000, when concentration measurements are available in the US. Other studies have given top-down constraints of total BC emissions with observations over much shorter time scales (Dubovik et al., 2008; Q. Wang et al., 2014), sometimes focusing on particular sectors (Hakami et al., 2005; Hu et al., 2009; Palmer et al., 2003; Park et al., 2003; Y. Wang et al., 2011). In this work, we explicitly represent historical activities that have taken place throughout history, including changes in combustion technology, and identify those requiring updates by comparison with decadal observations. In so doing, we have had to develop comparison techniques that have not appeared in previous literature, and have explored a variety of methods to distinguish source categories responsible for observed discrepancies and to update emission representations with literature support. We then present a new US BC emission inventory for 1960-2000, which we believe to be the best estimate of emissions given all possible constraints.

2.Methodology

2.1 Overview

An overview of our constraining procedure is shown in Figure 1. We started with the *a priori* US BC emission inventory for 1960 to 2000, which is described in Section 2.2. We connect emissions with ambient air concentrations using source-receptor relationships derived from a model (Section 2.3). Since the model may not reproduce the meteorology well, these transport relationships were adjusted with meteorological measurements. Historical concentrations were

4

predicted by applying the transport relationship to *a priori* historical emissions. In Section 2.4, we describe coefficient of haze (COH) measurements in California and New Jersey that are available for 1963 to 2000 and the BC concentrations that can be inferred from these data. While other states also have historical COH measurements, their records are less than 15 years in duration. Finally, we use a ratio called the "discrepancy factor" (DF, Section 2.5) to quantify the difference between observed and predicted BC concentrations. We describe how we identify fuels, technologies and sectors needing adjustments by examining trends in DF. This process is indicated by the red arrows in Figure 1.
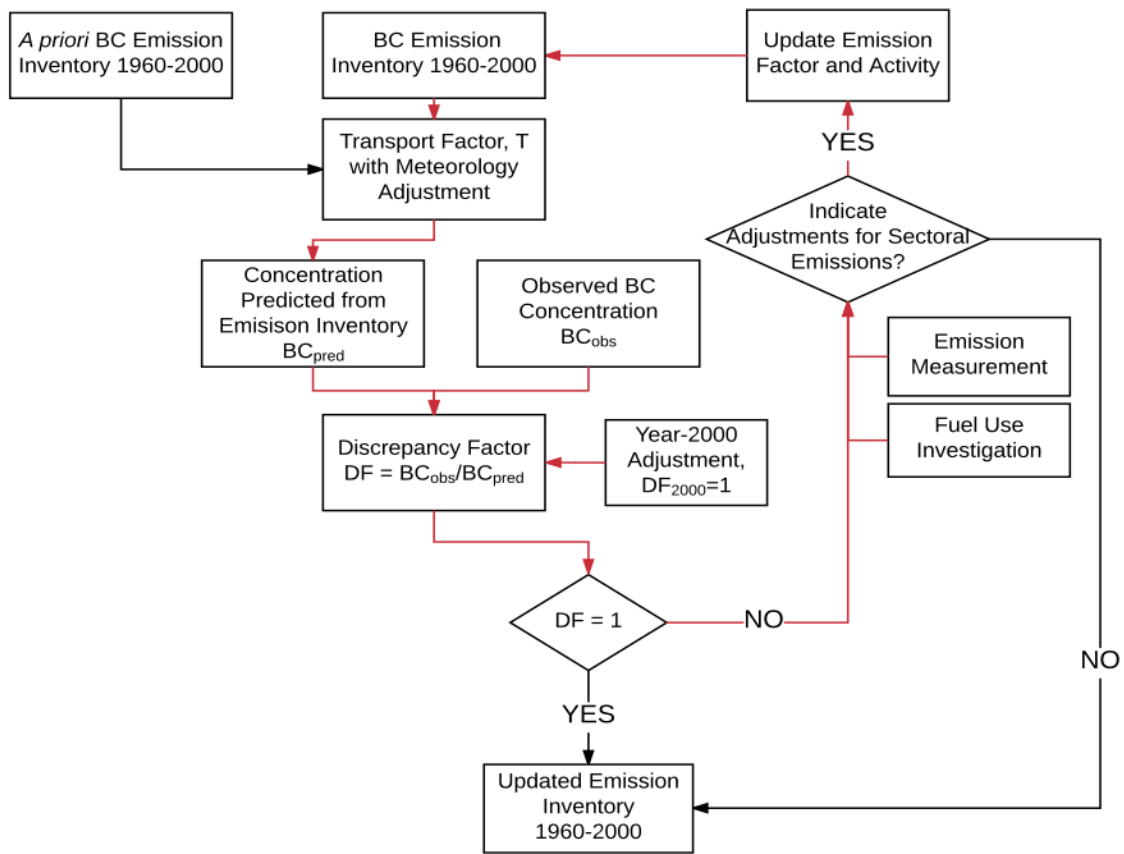

Figure 1. Process schematic for constraining BC emission inventory with concentration observations. Red arrows describe the iteration process of updating emission according to

indicated constraints. A key indicator is the discrepancy factor (DF), or the ratio between observed and predicted concentration. An overview of the method is given in Section 2.1, with supporting details in Sections 2.2 to 2.5.

2.2 *A Priori* Emission Inventory

The *a priori* US BC emission inventory for 1965 to 2000 is based on the one developed by Bond et al. (2007) with updates as described by Lamarque et al. (2010). The inventory is technology-based; that is, its underlying assumption is that emission per quantity of fuel (the "emission factor (EF) ") depends solely on the technology used to combust that fuel. Aggregate EFs are not an inherent property of the year or region, but rather depend on the technology mix in different times and places. The evolution of emissions is represented as a change in both fuel quantities and technology composition. This principle allows an EF for a specific combination of fuel and technology to be applied outside of the time and place where it was measured.

The global emission inventory is calculated every five years with fossil fuel use data from United Nations Statistics Division (1995) through 1980, data from International Energy Agency (IEA) (2004a, 2004b) after 1980, and biofuel data from Fernandes et al. (2007). Unlike meteorological factors, fuel consumption and emissions have little interannual variability, so that a single year's emission estimate can represent adjacent years.

The emission inventory for the US used in this work has two major updates. First, fuel consumption data are taken from the State Energy Data System of the US Energy Information Administration (US EIA) (2010), the US national agency responsible for collecting energy statistics. US EIA data are disaggregated by state, whereas IEA data are given by country. The second difference is the use of the SPEW-Trend vehicle fleet model (Yan et al., 2011). SPEW-Trend is a dynamic model of vehicle population linked to emission characteristics. It estimates the number of vehicles built according to different standards that remain on the road to emit in each year by tracking technology transitions for each vehicle group, including new vehicle introduction, emission degradation, retirement, and the transition to high-emitting conditions. Emissions do not cease when emission standards come into force, but rather when old vehicles

6

retire from the road. In Bond et al. (2007), vehicle emissions were calculated using an assumed time lag after introduction of standards, while SPEW-Trend gives a more realistic estimate of emission trends by using actual growth and retirement rates.

We also added seasonality to emissions in the residential sector by assuming that activities and emissions are proportional to heating degree days (HDD). We assume that all other emissions are constant throughout the year. HDD is the difference between the desired temperature in a building and the outdoor temperature, and should therefore be proportional to heating energy (Kaynakli, 2008; Quayle & Diaz, 1980; Stohl et al., 2013). Equation (1) gives the definition:

$$HDD = \sum_{d=1}^{n} \max(T_{set} - T_d, 0) \qquad (1)$$

where *HDD* stands for heating degree-days, T_d is daily mean outdoor temperature for day *d*, T_{set} is the temperature of the indoor environment, and *n* is the number of days for each month. We calculated the average monthly HDD for each decade. We use HDD data from the National Climate Data Center of National Oceanic and Atmospheric Administration (www.cpc.ncep.noaa.gov/products/analysis_monitoring/cdus/degree_days/, 2013) to calculate a seasonality factor for each month (i), S_i, or the monthly HDD normalized by its annual average, as shown in Equation (2):

$$S_i = \frac{HDD_i}{\overline{HDD}} \qquad (2)$$

We calculated the average monthly *Si* over each decade. *Si* shows small inter-decadal change with a coefficient of variation of 7% and 8% for New Jersey and California, respectively. Seasonal emissions for each month are calculated by multiplying the average residential emissions by the seasonality factor. Seasonality factors for California and New Jersey are shown in Table S1 (Supporting Information).

Vehicle emissions may also have seasonality. This is discussed later in Section 4.2.1.

7

2.3 Atmospheric Transport

For comparison with measured concentrations, we predict concentration at each receptor based on emissions and atmospheric transport. Our primary analysis relies on trends rather than concentration comparisons (Section 3.2, SI Section Text S1), so the transport plays only a small role in determination of missing sources.

2.3.1 Representation of atmospheric transport: Transport factor

We formulate the relationship between emissions and ambient air concentrations as a *transport factor*. Each transport factor represents the sensitivity of concentration in the receptor region to the emission in the source region. The transport factor T transfers the emissions E (Gg/mo), into concentrations C ($\mu g/m^3$).

$$C(m, y, x) = T(m, x) \cdot E(m, y, x) \qquad (3)$$

where m and y indicate that the transport factor depends on month and year, respectively, and x represents dependence on location. Separate transport factors were developed for each month and for the two states, as described below. With the transport factor, we predict concentrations from emissions without performing a simulation for each year. The concept of the transport factor is same as the Jacobian matrix in inverse modeling (Brasseur & Jacob, 2013; Kopacz et al., 2009; Rodgers, 2000; Y. Wang et al., 2004). In this study, as in inverse modeling studies, we assume the transport relationship is linear and independent of the magnitude of emission.

The transport factors were numerically constructed from a simulation over the period of 2000-2006, conducted with the NCAR Community Atmospheric Model (CAM) version 4.0 (Neale et al., 2010) running in 1.9°×2.5° resolution with specified dynamics, forced with transient sea surface temperatures, and using the emission inventory reported by Bond et al. (2007). We modified the bulk aerosol module to tag BC emissions from different source regions and simulate multiple BC-like aerosol tracers as described in Bond et al. (2011).

We developed two transport factors with California and New Jersey as receptors, respectively (see Table S2 in Supporting Information), using the average concentration over

8

multiple grid boxes within each state. The contribution from regions outside the US is small, as is the influence of wildfire (see Text S2 and Table S3 in Supporting Information).

2.3.2 Errors in model meteorology

Errors in simulating meteorology could cause discrepancies between predicted and observed concentrations, which should not be attributed to inaccuracy of the emission inventory. We examined the fidelity of the planetary boundary layer height (PBLH) and the wind speed because both affect atmospheric concentrations. Modeled values of these two parameters for the period 2000-2006 were compared with measurements acquired from Modern-Era Retrospective Analysis for Research and Applications, National Aeronautics and Space Administration 2 (MERRA-2) (2014). Wind speed is averaged within the mixing height or planetary boundary layer. We also evaluated whether BC concentrations were, in fact, affected by PBLH and wind-speed in the CAM simulation results.

2.3.3 Long-term trends in atmospheric transport

One concern about using the transport factor to predict historical concentrations might be the fluctuation of meteorology during the period of interest. We examined the variation of PBLH for 1979-2005 in California, which is the longest historical observation available in the MERRA-2 database for the time period of this work. We also examine the precipitation observations from 1965 to 2000 for California and New Jersey (NOAA National Centers for Environmental information, 2018). In addition, we calculated variations of deposition within a global climate model simulation over the period of 1850-2005, conducted with CAM3. The simulation had prescribed interannually changing sea surface temperatures over this period, but constant BC emissions, to isolate the variation due to transport alone. Changes in horizontal transport and wet removal would cause differences in BC deposition, so we used fluctuations in BC deposition to estimate possible long-term trends in transport.

2.4 BC Concentration Measurements

BC ambient air concentration data are estimated from COH data acquired from California Air Resources Board (www.arb.ca.gov/aqd/aqdcd/aqdcddld.htm, 2014), New Jersey Department

9

of Environmental Protection (http://www.njaqinow.net/, 2014), and US Environmental Protection Agency (US EPA)'s Air Quality System (www3.epa.gov/ttn/airs/airsaqs/, 2015). COH was one of the first measures of PM air pollution, introduced in the early 1950s (Hemeon et al., 1953). The measurement of COH is similar to that of BC (A. Hansen et al., 1984) because both are based on the quantification of light transmission through a filter upon which particles are collected. Therefore, BC concentrations can be estimated from records of COH concentration (Allen et al., 1999; Kirchstetter et al., 2008). Kirchstetter et al. (2017) conducted side by side COH and BC measurements at two sites (Vallejo and San Jose, California) for two years. They found a seasonally independent relationship of BC (μg m^{-3}) = 6.7COH + 0.1, with R^2 = 0.9. Wolff et al. (1983) measured EC (mg m^{-3}) = 8.9COH - 0.63 with R^2 = 0.9 for EC concentrations up to 10 mg m^{-3} in Detroit, Michigan, 1981. These studies suggest the dependence of light-absorbing PM on COH is relatively constant with location and time. In this study, the relationship given by Kirchstetter et al. (2017) is used to translate COH to BC. The COH data are available at two-hour resolution for California and New Jersey during 1963-2011 and 1967-2005, respectively. Nearly all of the COH sites were located in urban areas. At its peak in the 1980s, the California COH sampling network included more than 50 sites. The number of COH monitoring locations decreased to 30 around 2000. New Jersey contained 25 sites, 12 of which were closed after 1985. For a consistent trend, we use only observations from sites with records longer than 15 years.

Figure 2 shows averaged concentrations in California and New Jersey. To evaluate seasonality, concentrations in each month were averaged over a five-year period to decrease the influence of natural meteorological fluctuations (e.g. "January 1970" in the figure representing January concentration for 1968 to 1972). The California BC decreasing trend from 1990 to 2000 is similar to that interpreted from Interagency Monitoring of Protected Visual Environments (IMPROVE) by Bahadur et al. (2011), with a correlation coefficient of 0.87. During 2000 to 2008, COH-BC shows a reduction of 49% compared with previous decade, while IMPROVE-BC decreases by 35%. The predicted BC concentrations in Figure 2 will be discussed later. COH

10

records from other states could not be used, as their sites were not continuously operated for more than fifteen years (except for one site in Pennsylvania which has record for twenty years (US EPA, 2018)).

2.5 Analysis of Discrepancy between Observation and Prediction

2.5.1 Raw Discrepancy Factors

With the *a priori* inventory as described in Section 2.2 and the transport factor adjusted to measured meteorology as described in Section 2.3 and later in Section 3.1.2, we predict BC concentrations and compare them with observed BC concentrations (Section 2.4). The transport factor for each month is applied to emissions representing each month in a five-year period, and the observations are also averaged for each month in the same five-year period. The influence of wildfire is small but is removed from the analysis, as described in Text S2 of Supporting Information.

We refer to the ratio of observed BC concentrations to BC concentrations predicted from the *a priori* emissions as a raw *discrepancy factor* (DF). The general definition of a discrepancy factor is:

$$DF_{raw} = BC_{obs} / BC_{pred} \tag{4}$$

A raw DF of unity indicates perfect agreement between observations and BC concentrations predicted from the *a priori* emissions, and a raw DF greater than one indicates that observed concentrations are greater than predicted BC concentrations.

Throughout this work, the discrepancy factor is derived for separate months and for different types of emissions. The seasonality of BC concentrations is used to distinguish baseline and seasonal discrepancy. Separate DFs are developed for emissions that we term "baseline" and "seasonal" for each five-year period. We assume that emissions have a baseline component that is constant throughout the year, and a seasonal component that varies by month. We also assume seasonality in the emissions is caused by varying heating use, as described in Section 2.1, so that only baseline emissions occur in summer. Therefore, concentrations in summer (June, July, and August) are used to evaluate baseline emissions, and concentrations averaged over the rest of the

11

year are used to evaluate seasonal emissions. Calculations of DFs are given in Text S1 of the Supporting Information. We exclude international shipping emissions from the *a priori* emissions when calculating DFs, assuming that they do not contribute to the regional, land-based BC observations.

2.5.2 Normalized discrepancy factors

Sub-grid spatial heterogeneity is a concern when predicted concentrations are compared with observations for short-lived species. Point observations cannot represent grid-averaged concentrations predicted by models, especially for urban areas where concentration gradients are steep. R. Wang et al. (2014) showed that more detailed emissions and higher resolution models performed better in reproducing observations. Most of the historical COH measurements are from urban regions.

We acknowledge the discrepancy caused by comparing point observations with grid-box averages and provide a brief evaluation of the potential magnitude. Using WRF-Chem simulations (Grell et al., 2005) of BC concentrations over Los Angeles, California, with a resolution of 4km×4km, we imitated a lowered model resolution by aggregating the concentrations in small grid boxes into larger grid boxes. This examination shows that, in urban areas and depending on the location of the observation, the concentration in a large grid box can underestimate a point observation by a factor of two (see Figure S3). This discrepancy would differ for each paired observation and grid box, so this simulation cannot be used for the entire analysis; it indicates only the potential magnitude of discrepancy. However, if the discrepancy attributed to spatial heterogeneity is relatively constant throughout the historical record, then observed trends may reflect emission trends even if the absolute magnitude cannot be reproduced.

To adjust for discrepancies due to spatial heterogeneity alone, we produce normalized DFs by dividing the raw DF by the DF from the year-2000 period. The principle is that the governing factors affecting emissions, such as fuel use and emission rates, are better known recently than historically, so that the latest emissions (1998-2002 average) for both states are approximately correct and discrepancies are largely attributable to spatial heterogeneity. This calculation is

12

detailed in Text S1 of Supporting Information and only normalized DFs are presented in this paper. Supporting the notion that recent emissions are reasonably well known, an inverse model by Hu et al. (2009) estimated the US total emission to be 400 Gg in 2004, which is similar to our *a priori* inventory. Koch, Schulz, et al. (2009) found the median BC concentrations predicted by the AeroCom suite of global models to be 20% higher than the observations of BC surface concentrations from IMPROVE in US for year 2000. An estimate by Park et al. (2003) is 70% higher than our *a priori* emission. If this value, rather than the others, is correct, then it would affect the total magnitude of emissions estimated here.

2.5.3 Modified emission estimates

Normalized DFs indicate the required multiplier to the *a priori* emission inventory to predict BC concentrations that match the observations. Multiplying the baseline and seasonal emissions in the *a priori* emission inventory with the normalized DFs yields a quantity that we refer to as *scaled emissions*. Definitions of the terms used in this work are summarized in Table 1.

Although the scaled emissions give good agreement between predicted BC concentrations and observations, this top-down constraint does not identify the emission sources that are poorly estimated. Emission sources that are in error could comprise only a fraction of the total emissions. Applying these normalized DFs in other locations and times would not guarantee improvement in a different emission inventory. Therefore, we search for the sources responsible for the disagreement by investigating specific combinations of fuel and technology that could be responsible for the changes in normalized DF. Each fuel-technology group is subject to nationwide standards and produced by common manufacturing, and thus has a distinct EF that changes over the entire US at approximately the same time. Emissions based on this physical reasoning are called *updated emissions*.

13

Table 1. Definition of Terms in This Work

Data	Name in this work		
	Emission	**Concentration** [1]	**Emission Factor**
Observation from COH. No adjustments.	N/A	Observed	N/A
A priori	*A priori*	Predicted from *a priori* emission	Original
Multiply all *a priori* emissions by discrepancy factor	Scaled	Predicted from scaled emission	N/A
Alter specific fuel and technology representation and recalculate emissions	Updated	Predicted from updated emission	Updated

[1]See section 2.5.5 for process of predicting concentrations from emissions.

Normalization to the year 2000 (Section 2.5.2) means that emissions estimated throughout the historical period are affected by inaccuracies in year-2000 emissions. If year-2000 *a priori* emissions are estimated low by 10%, then scaled emissions are also low by 10%. These inaccuracies, however, are much smaller than those caused by spatial heterogeneity, which was the reason for the normalization. Although the trend evaluations use long-term observations, the influence of each technology is dominant over 15-20 year sub-periods, not the entire period. A varying ratio of discrepancy from spatial heterogeneity due to urbanization or other factors would have limited influence on the inference.

2.5.4 Identifying Causes of Discrepancy Factors

To identify emission sources that are poorly estimated, we examine how baseline and seasonal normalized DFs vary between the two states and over time. First, we identify fuel-technology groups whose consumption and emission magnitudes could plausibly explain the observed decadal trends. Tools used to identify the causes of mismatches and suggested updates are regression, analysis of weekly cycles for vehicles, and literature reviews on EFs and accuracy of fuel reports.

14

When emissions from fuel-technology combinations have distinct trends, the temporal variation might be used to infer required adjustments using a linear regression of the scaled emissions on the *a priori* emissions:

$$y_i = \sum_j a_j \left(EF_j Fuel_{ij} \right) + Const \qquad (5)$$

where *i* indicates year; *j* indicates a target fuel and technology combination and includes a category of all other emissions; y_i is the scaled emissions calculated by multiplying the *a priori* emission by the normalized DF. EF_j is the original EF for the fuel-technology combination *j* in g/kg; $Fuel_{ij}$ is fuel use by technology *j*; and the regression coefficient a_j is a multiplicative adjustment for the *a priori* emissions from combination *j*. If all the adjustments were attributable to EF rather than to fuel, then the product a_jEF_j would suggest a single best-fit EF for the identified fuel-technology group. For marketed fuels, fuel quantities are usually well known.

We also use the weekly variation in BC observations for California to apportion discrepancies between heavy-duty diesel vehicles (HDDV) and other baseline emissions. HDDV emissions have a strong weekly pattern that is not expected for other vehicles or in other sectors (Dreher & Harley, 1998; McDonald et al., 2014). According to freeway motion sensor data, HDDV travel 25% as much on Sunday as they do during the week (California Department of Transportation, 2000) while this ratio is 90% for light-duty gasoline vehicles (LDGV). By assuming this ratio is constant during the study period, we manipulate the differences in COH observations between weekdays and weekends to apportion emission discrepancies to HDDV. Equations for this analysis are given in Text S3 of the Supporting Information. The weekly cycle analysis allocates the discrepancy between HDDV and non-HDDV baseline emissions, giving a different adjustment for each overall group in each year.

Finally, for all fuel-technology combinations that might require new EFs, we re-examined previous literature on emission measurements and assumptions made in the *a priori* inventory. Thorough reviews of EFs have already been published (e.g. Bond et al., 2004), and few additional data are available. Furthermore, there is a fundamental challenge in determining EFs

15

of early technology, which had poorer combustion and higher BC EFs. Improved combustion and emission standards were all driven simultaneously by air quality concerns. By the time the first emission measurements were reported, some technology improvements had probably already occurred, and less-polluting installations may have been selected for measurement. For that reason, the most polluting technologies may remain unrepresented in the emission literature.

2.5.5 Summary of connection between emissions and concentrations

In the search for discrepancies, it is sometimes more useful to compare concentrations, and sometimes better to compare emissions. We summarize here the connection between modeled emissions and modeled concentrations, as described in the preceding text. First, emissions are multiplied by a transport factor inferred from a model (Section 2.3.1), which is adjusted for the effect of inconsistencies in modeled PBLH (Section 2.3.2). The modeled concentrations are next multiplied by a normalization factor gleaned from a five-year period surrounding the year 2000, which accounts for spatial heterogeneity between the model gridbox and the point of observation (Section 2.5.2). Emissions can also be inferred from concentrations by reversing the process.

3. Results: Discrepancy Factors

In this section, we summarize discrepancies between observed BC concentration and BC predicted from the *a priori* emissions. We evaluate potential discrepancies caused by atmospheric transport (Section 3.1), describe normalization of the DFs to near-present day values in order to examine trends of discrepancy (Section 3.2), and present the final normalized DFs that will be used in Section 4 to constrain sectoral emissions.

3.1 Characteristics of Atmospheric Transport

3.1.1 Long-term trend of atmospheric transport

Trends in transport could influence the trend in predicted BC emissions. Transport varies interannually, but this study analyzes five-year averages of observed and predicted concentrations. To examine the stability of atmospheric characteristics during the study period, we retrieved historical PBLH observations in California from NASA MERRA-2 database for

16

1979-2005. The coefficient of variation of seasonal, five-year seasonal average and five-year annual average PBLH is 6%, 3%, and 1%. Also, the five-year annual average precipitation exhibits ±20% fluctuation in California and ±7% fluctuation in New Jersey during 1965 to 2000 (NOAA National Centers for Environmental information, 2018). Moreover, the 150-year simulation (Section 2.3.3) shows quite small variability in deposition (see Figure S1). The coefficient of variation of the decadal deposition rate during this 150 years is less than 0.3 in each grid box, except in a small part of the South Pacific Ocean, which is outside the study region. These variations in meteorology are much smaller than the threefold decrease in annual average BC concentration (Kirchstetter et al., 2017), which justifies using the same transport factor for the 50 years of this study.

3.1.2 Adjustment for model meteorology

We investigated the relationship between monthly-averaged meteorological parameters and BC in the model. Regressions showed that PBLH is a statistically significant predictor of BC concentrations for both California and New Jersey ($p<0.05$) but wind speed is not ($p>0.3$). This distinction is physically plausible, as BC concentrations may be transported from upwind without demonstrating apparent dilution, while an increase in PBLH always has a diluting effect. Y. Wang et al. (2011) also found no significant correlation between local wind speed and rural BC concentrations in remote sites of Beijing, which has similar size to the 2° grid box in the simulation.

Because PBLH has a strong effect on concentration, incorrectly modeled PBLH could lead to error in predicted concentration. The model underpredicts measured summer PBLH by about 60% in both states. For California, the ratio of measured summer and winter PBLH is 1.6, whereas the modeled ratio is 1.1, so the modeled seasonality is also underpredicted. For New Jersey, the ratio between measured summer and winter PBLH is 1.2, while the modeled ratio is 0.5, incorrectly suggesting greater pollutant dispersion in winter. Comparisons for modeled and measured PBLH are shown in Figure S2. These differences illustrate the need to correct the model's predicted PBLH.

17

We accounted for error in modeled PBLH by applying the regression relationship (Supporting Information Text S4) to the modeled and measured values to infer correction factors for BC concentration in each grid box. These adjustment factors for each state are shown in Table S4. Based on the regression coefficients, BC concentrations in California are more sensitive to PBLH than in New Jersey. Therefore, the adjustment factors introduce more seasonality to BC concentrations in California.

Neither PBLH nor its adjustment has great spatial heterogeneity. The difference between PBLH adjustments for the two grid boxes in New Jersey is less than 1%, and in California, the coefficient of variation of the adjustment is less than 0.1. Therefore, we use the state-averaged transport factor and PBLH adjustment factor.

After adjusting for modeled meteorology, the calculated transport factors for California receptor show seasonal variations of ±30%, with greater dispersion in summer. This seasonality is due to winter conditions in California that cause higher pollutant concentrations, as also found by Kirchstetter et al. (2008). The transport for New Jersey, however, shows only weak monthly fluctuations of about 10%. This difference of seasonality in atmospheric transport between the eastern and western US has also been observed in CO measurements by Glen et al. (1996).

We did not adjust or account for trends in precipitation. Urban monitoring sites are close to the sources, so removal between source and receptor is limited. We found that seasonality of concentrations was opposite to that predicted from the variation of wet removal.

3.2 Discrepancy Factors and Normalization

Discrepancy factors are the ratio of observed to predicted BC (Equation 4). Raw DFs, without the normalization that accounts for spatial heterogeneity, are given in Table S5 of the Supporting Information, and normalized DFs are shown in Figure 3 separately for baseline and seasonal emissions.

During the year-2000 period used for normalization, the ratio between observed and predicted baseline concentrations averaged about two in California and six in New Jersey. In California, the ratio between observed and predicted seasonal concentrations is greater than six

18

in 2000, much greater than discrepancies expected from spatial heterogeneity in the baseline emission. This large additional difference might be explained by interference by brown carbon. Kirchstetter et al. (2004) and Chen and Bond (2010) showed that biomass burning can produce brown carbon that contribute to light absorption measurements. Wood combustion is the dominant source for California winter emissions of BC (>85%), and the historical COH monitor used a broadly emitting incandescent lamp that cannot distinguish BC and brown carbon. Thus, predicted residential emissions may not match observations until absorption by brown carbon is included.

The seasonal emissions in New Jersey that appeared in the early observations vanished in 1990, and no seasonality of BC concentration was observed (Figure 2, black line). Therefore, those years cannot be used for adjustment of seasonal emissions in New Jersey. For presentation, we use an assumed normalization factor of two. We rely on the trend of the normalized DFs and emission measurements to determine source categories that need adjustments, as we do for other sectors.
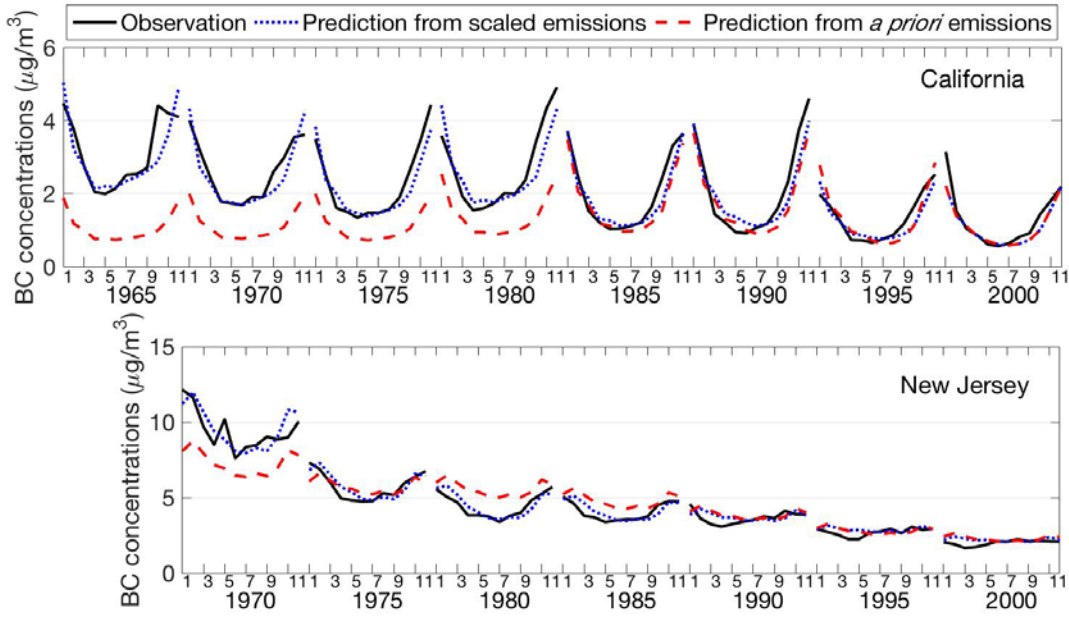
19

Figure 2. Comparison of monthly BC concentrations for California and New Jersey. BC concentrations observations (black line) are averaged over each five-year period (e.g. January BC concentrations centered in 1965 is based on January observations from 1963 to 1967). BC concentrations are predicted from *a priori* emissions (red line), and predicted from scaled emissions (blue line). See Table 1 for terminology.

3.3 Observed and Predicted Concentrations

Figure 2 shows observed BC concentrations (black line) averaged over each five-year period and compares them with concentrations predicted from *a priori* emissions (red dashed line). After the meteorology adjustment described in Section 3.1 and the normalization described in Section 2.5.2 and 3.2, the *a priori* predictions still differ from observations. Differences between predictions and observations during baseline months can be seen by comparing June, July, and August, and differences in the seasonal component are shown by comparing the heights above baseline.

Scaled emissions are calculated by applying the normalized DFs shown in Figure 3 to the *a priori* emissions. The resulting concentrations predicted from scaled emissions (blue line) match the observed BC well except for a small overestimation of seasonality in 1965 and 1970 for California. Although the method guarantees that the scaled emissions would agree with observations on average, a match in the monthly variation is not required.
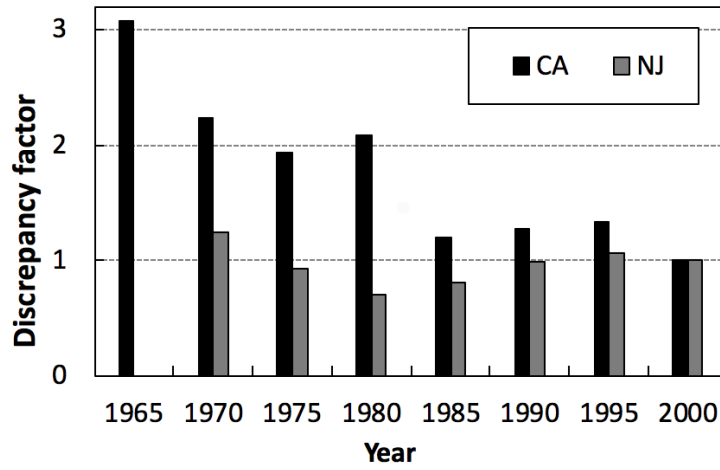
(a)
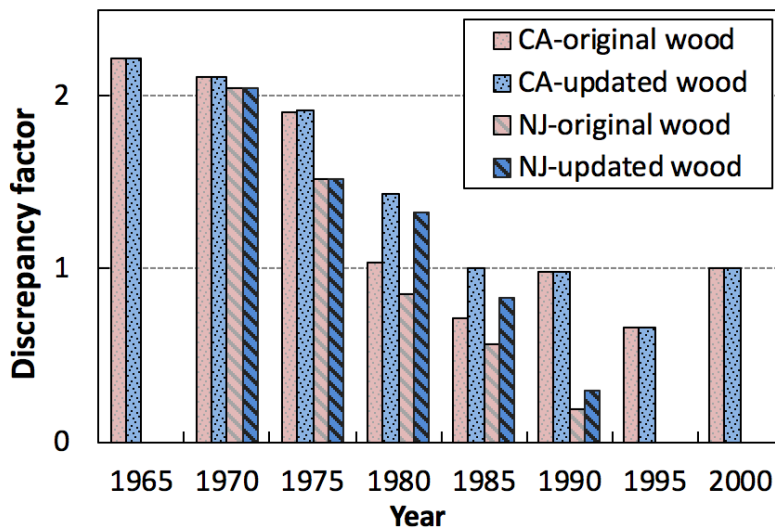
(b)

Figure 3. Normalized discrepancy factors (DFs) for baseline emissions (a) and seasonal emissions (b) for California and New Jersey. Discrepancy factor is the ratio of observed BC to BC predicted from the *a priori* emission, normalized to the value in the year 2000. A factor of two is used to normalize DF for seasonal emission in New Jersey in this figure (see section 3.2 and 4.2). Seasonal normalized DFs before the update in wood usage (Section 4.2.1) are labeled "original wood". Normalized DFs after that modification are labeled "updated wood".

4. Causes of Discrepancy

This section discusses a search for the emission sources responsible for the disagreement between observed BC concentrations and BC concentrations predicted from the *a priori* emission inventory (Figure 2), as indicated by normalized DFs (Figure 3). We rely on the trend and magnitude of normalized DFs to determine source categories, and use emission measurements to support any adjustment. EFs (grams of BC emitted per kilogram of fuel burned) decrease with improvements in combustion and emission control technologies, and activity (kilograms of fuel burned) changes annually. Either or both could contribute to the discrepancies. Section 4.1 discusses the causes of discrepancies in baseline emissions. Section 4.2 focuses on the seasonal emissions from the residential sector.

4.1 Discrepancy in Baseline Emissions

A four-fold decreasing trend in baseline concentrations is observed in both California and New Jersey between 1965 and 2000. In BC predicted from the *a priori* emission inventory, the New Jersey decrease is smaller than the observed one and California does not have a decrease (Figure 2). For California, the *a priori* inventory underpredicts emissions (normalized DF>1), especially in 1980 and before. For New Jersey, the *a priori* emission generally agrees with the observation (normalized DF averaged 0.96 ± 0.18) except for an underestimation in 1970. This suggests that some baseline emissions dominant in California but not in New Jersey are underestimated before 1980. Emissions after 1980 are underestimated by 20% in California.

22

Consumption in three fuel-technology groups have magnitudes and decadal trends that would allow them to explain the baseline normalized DF before 1980. These are pre-regulation LDGV, pre-regulation HDDV, and domestic shipping using heavy fuel oil. We define the pre-regulation HDDV and LDGV as those before rudimentary regulations in 1970s. Other categories are not likely to be the source of the discrepancy. They would either require order-of-magnitude changes in EF, or no plausible changes in EF would reproduce the decreasing trend. Because the *a priori* inventory is predicted by a vehicle fleet model that represents shifts in the vehicle population as technologies change, the comparison can target fuel-technology combinations built to different standards. A greater EF for pre-regulation LDGV would increase emissions in 1965 and 1970, and a greater EF for pre-regulation HDDV would increase emissions in 1970 and 1975. By 1985, 99% of pre-regulation HDDVs and LGDVs had retired, so increasing emissions of these vehicles would not change the fleet emission after that year. An increase in the BC EF for ships would contribute mainly to the emissions for California in 1965 when the fuel consumption peaked, without much effect on New Jersey.

The apparent dip in the baseline normalized DFs for California moving from 1980 to 1990 is caused by a peak in the *a priori* emission inventory that is not reflected in the observations. This peak results from middle distillate emissions in the industrial sector. New technology with lower EFs could have been phased in, causing a decreasing trend of EFs from 1980 onward. For example, off-road diesel engine EFs differ by a factor of three for pre-1980 and post-1988 engines (US EPA, 1991a, 1998).

We first investigated whether a linear regression over all years and both sites (Section 2.5.4) could identify fuel-technology groups that required adjustment. Only pre-regulation LDGV (indicated EF: 1.41 g/kg, p=0.003) and pre-regulation HDDV (indicated EF: 6.15 g/kg, p=0.058) have regression coefficients that are statistical significant at the 10% level. The remaining fuel-technology groups were not statistically significant when isolated, but as a whole indicated a required increase of about a factor of 1.65 (p<0.001). The regression constant was not statistically significant. These results should be interpreted with caution as many fuel-technology

23

groups change concurrently, and thus smaller groups without statistically significant results could be subsumed into the most influential groups. For each fuel-technology group, including some without statistical significance in the regression, we sought additional evidence to confirm or deny the inferred emission increase, as discussed in the following sections.

4.1.1 Heavy-duty and light-duty vehicles

Over the period from 1960 to 2000, HDDV BC emissions contributed 15% to 50% of total US emissions and 35% to 90% of transportation-sector emissions. LDGV accounted for 5%-20% of total US emissions and 10%-60% of transportation emissions in the *a priori* emission inventory (Bond et al., 2007). The pre-regulation EFs used in the *a priori* inventory had little measurement support. For HDDV, the value had been estimated by ratios between subsequent standards (Yan et al., 2011; Yanowitz et al., 2000), since no direct measurement was available before 1974 when the first regulation was published. For LDGV, it was based on only one available measurement (Williams et al., 1989).

We sought additional evidence to apportion HDDV versus non-HDDV increases using the weekly cycle of BC observations in California for 1965 to 2000 (Section 2.5). This analysis isolates HDDV emissions from others because of its strong weekly patterns (Dreher & Harley, 1998; McDonald et al., 2014). Because LDGV emissions do not have such a distinct timing, they cannot be separated from other fuel-technology combinations within the baseline group, although they likely dominate that group. As discussed in Section 2.5, weekly cycle analysis does not produce an independent discrepancy factor, but gives different adjustments for HDDV and non-HDDV baseline emissions in each year by allocating the total discrepancy. Figure 4 shows the allocated normalized DFs, indicating that, for 1965, 1970, and 1975, the emissions for HDDV fleet should increase by a factor of 2.1, 1.4 and 1.1, and other baseline emissions including LDGV should increase by factors of 3.5, 2.7 and 2.5. Since pre-regulation HDDVs dominated the fleet of 1970 and before (>90%), an EF of 5.0 g/kg and 3.3 g/kg is inferred for pre-regulation HDDVs in 1965 and 1970. We attribute this large reduction in HDDV EF to the introduction of turbocharged diesel engines. These engines were introduced in the late 1960s, and gained favor

24

after the first oil crisis in 1973 due to their fuel efficiency (Pettitt, 2004). However, we could not further divide pre-regulation HDDVs into separate technologies due to lack of information on both the market share and emission measurement.

Unregulated HDDV trucks and buses, which may have been built to similar specifications, were measured by Subramanian et al. (2009) in Thailand, with a reported average PM EF of 4-10 g/kg. Several studies also measured smoke opacity from these vehicles to be around 20% (Springer et al., 1974; Stahman et al., 1968; Williams et al., 1989) which can be converted to a PM EF of 5-7 g/kg as described in Supporting Information Text S5. We chose an average PM EF of 7.7 g/kg, which gives a BC EF of 4.4 g/kg for pre-regulation HDDVs, assuming a 57% BC fraction (Durbin et al., 1999; Gillies & Gertler, 2000).

Considering the measurements and the weekly-cycle analysis, we adopt a BC EF of 4.4 g/kg (a factor of 1.8 increase) for pre-regulation HDDVs with an uncertainty of 50%. The chosen value of uncertainty encompasses both the variation in weekly-cycle analysis and the EF estimated by the overall regression.

Updating the EF for pre-regulation HDDVs in the vehicle fleet model alters fleet-average PM EF for HDDV in each year that those vehicles remain in operation. The fleet-average PM EF inferred from weekly cycle analysis (open circles), and from regression (X) are shown in Figure 5. It also includes the EF from dynamometer measurements, as used in the *a priori* emission inventory (open triangles) and EFs estimated from tunnel measurements (black diamonds) summarized by McDonald et al. (2015) The tunnel measurements relied on information from Pierson and Brachaczek (1982), Pierson et al. (1996), Gertler and Gillies (2002), Fraser et al. (2003), Geller et al. (2005), Grieshop et al. (2006), Ban-Weiss et al. (2008), Gentner et al. (2013), and Dallmann et al. (2014). The fleet-average EFs are greater in early years, decrease more quickly, and agree better with the tunnel measurements when the pre-regulation EFs come from weekly-cycle or regression analysis, as compared with EF inferred from dynamometer measurements.

For LDGV, pre-regulation vehicles dominated the fleet in 1970 and before, with the fraction dropping to 10% by 1975, according to our fleet model. This transition required the introduction of catalysts around 1975 and the removal of lead from gasoline (Cadle et al., 1999), so that performance without catalysts and with leaded gasoline is the best representation of pre-regulation vehicles. Only a few dynamometer studies measured this type of engine. They reported an averaged BC EF of 0.13 g/kg with an range of 0.28 to 0.01 g/kg (Cadle et al., 1979; Lang et al., 1981; Rogge et al., 1993; Williams et al., 1989). Chassis dynamometer studies measure individual vehicles with specific vehicle type, fuel and driving cycles (Smit et al., 2010), and are sensitive to testing conditions (Yanowitz et al., 2000). Therefore, dynamometer measurements may differ from average on-road emissions from the entire fleet. Lower emission values are obtained from dynamometer studies as compared to fleet-average, traffic-tunnel measurements of diesel trucks for both PM (Canagaratna et al., 2004) and BC (May et al., 2014; Tunnel measurements summarized in McDonald et al., 2015; Shah et al., 2004). However, the same comparison has not been done for gasoline vehicles.

Westerdahl et al. (2009) reported an fleet-averaged LDGV BC EF of 0.3 g/kg in Beijing during 2007. Beijing adopted Euro I, II and III standards for new vehicles in 1999, 2003 and 2005 respectively, leaving only 10% of pre-regulation vehicles ("yellow label vehicles") in the fleet around 2007 (X. Wang et al., 2009; Wu et al., 2010). Beijing also banned leaded gasoline in 1997 (Wu et al., 2010). Therefore, the Westerdahl et al. (2009) value should be lower than the pre-regulation vehicle EF in the U.S. before 1975. Also, the fleet-averaged BC EF dropped to 0.2 g/kg when the yellow label vehicles were temporarily banned during the 2008 Olympic Games (X. Wang et al., 2009). Assuming 10% of fleet was yellow label vehicles and that their removal caused the reduction results in a BC EF of 1g/kg for pre-regulation vehicles, which is much higher than the previous estimates used in any inventory.

EF of pre-regulation LDGV is also estimated based on the weekly cycle analysis. Using the normalized DFs for non-HDDV baseline emission (Figure 4, white bar), and the updated EFs of identified fuel and technologies (heavy-fuel oil domestic shipping and middle distillate industry,

26

see Section 4.1.2 and 4.1.3 respectively) gives a prediction for pre-regulation LDGV of 1.50 g/kg in 1965 and 0.96 g/kg in 1970.

Considering the fleet-averaged BC EF in Beijing 2007, weekly cycle analysis result, and the regressed result, we recommend pre-regulation LDGV BC EF to be 1.0 g/kg, which is a factor of 2.5 higher than the *a priori* estimate.

For baseline emissions other than HDDV, applying the regressed EFs of identified fuel and technologies (heavy-fuel oil domestic shipping and middle distillate industry) gives a prediction that is 31%, 7%, and 38% lower than the total adjustment estimated by weekly-cycle analysis in the 1960s, 1970s and 1980s, respectively.
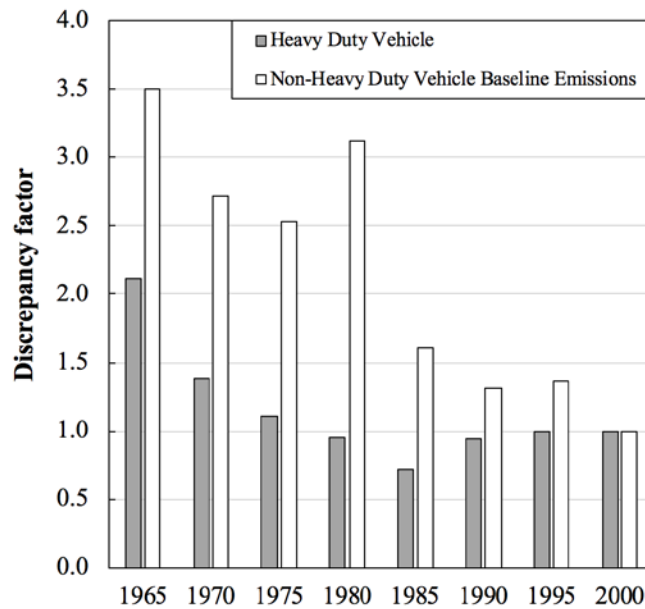

Figure 4. Normalized discrepancy factor (DF) for heavy-duty diesel vehicles and for all other baseline emissions in California, allocated using weekly cycle observations. All values are normalized to year 2000.
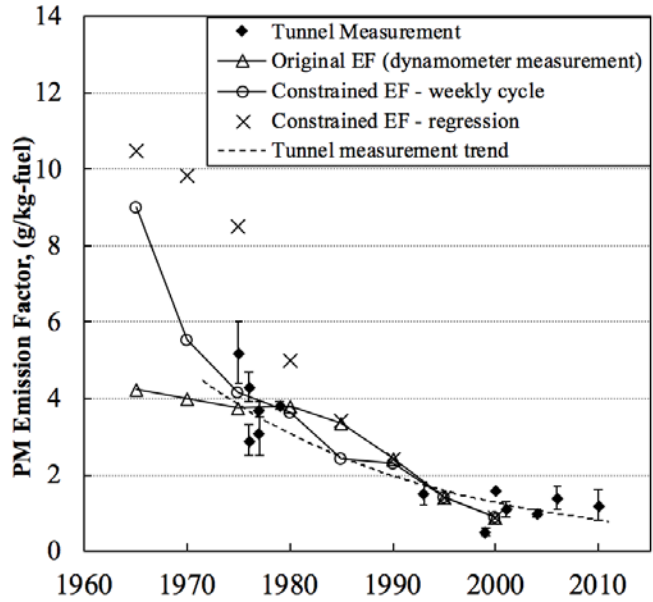
Figure 5. Heavy-duty diesel vehicle fleet-averaged PM emission factors. Open triangles: original emission factors in the *a priori* emission inventory; open circles: emission factors inferred from the BC weekly cycle; X: emission factors by applying adjustment for pre-regulated vehicles from regression into the whole fleet; black diamonds: tunnel measurements summarized by McDonald et al. (2015) Data shown are PM_{10}, except tunnel observations after 1990 which measured $PM_{2.5}$.

4.1.2 Domestic shipping using heavy fuel oil

The regression analysis did not give a statistically significant estimate of EF for heavy fuel oil used in domestic shipping. The EF of 0.41 g/kg in the *a priori* inventory was taken from low-speed diesel engine measurements in Lack et al. (2008). Low-speed diesel engines are typical for vessels that are used in long distance international shipping, such as tanker, container, cargo and bulk ships (Lack et al., 2008). Busy ports near populated areas, which would be close to the monitoring network, are usually dominated by vessels with medium speed diesel engines, such as tug and large fishing boats (Lack et al., 2008). The EF for those engines is 0.97 ± 0.66 according to Lack et al. (2008). We adopted an EF of 1.0 g/kg, with an uncertainty of 50% as indicated by the measurements.

28

4.1.3 Industry

The regression analysis did not provide a statistically significant estimate of EF for middle distillate industry, including tractors used in agriculture, off-road construction equipment in industry and boilers as a group. Emissions from off-road engines account for over 99% of middle distillate emissions from industry and agriculture (Bond et al., 2004; Kean et al., 2000; US EPA, 1996). The EFs for off-road engines in the *a priori* inventory are taken from Kean et al. (2000), which is based on US EPA measurements of post-1988 engines (US EPA, 1998).

Standards for off-road engines began in 1996 (US EPA, 2001), but technology had evolved slowly prior to that time from the late 1960s to the 1980s, with the advancements of turbocharging and charge air cooling (Bennink, 2015; Lloyd & Cackette, 2001). Both technologies improve fuel economy and reduce PM emissions (Bennink, 2015; Jääskeläinen & Khair, 2012; Lloyd & Cackette, 2001). Before 1970, off-road engines were naturally aspired and had higher emissions (Bennink, 2015); these naturally-aspired engines are missing from the *a priori* inventory. We estimate these EFs as follows. The Nonroad Engine and Vehicle Emission Study (US EPA, 1991a) provides PM EFs for agricultural tractors and construction equipment of 12 g/kg and 6-10 g/kg based on measurements in 1973 and 1982, respectively. US EPA (1998) also recommended multiplying these EFs by 0.64 and 1.2-2.0, respectively, to account for in-use operations. Assuming 57% of PM is BC (Durbin et al., 1999; Gertler & Gillies, 2002) provides EFs of 4.3 g BC/kg for early tractors and 8.9 g BC/kg for construction equipment.

Weighting these EFs by the amount of fuel used in agriculture and construction in the US, IEA (2004b) gives a net BC EF of 5.9 g/kg for combined industry sources in 1970, with an uncertainty of about a factor of two. The technical advancement in off-road engines, turbocharging, mainly occurred from late 1960s to 1970s (Bennink, 2015). We use the transformed normal function (Geroski, 2000; Grübler et al., 1999; Streets et al., 2004) with a transformation time of nine years to represent this transition from naturally aspired to turbocharged engines. This gives a naturally-aspired engine fraction of 86% in 1970 and 25% in 1980.

29

4.2 Discrepancy in Seasonal Emissions

The monthly variation in observations is well reproduced by the BC concentration predicted by the scaled emission, based on the assumption of distributing seasonal emissions using HDD (Section 2.2) when a single scaling factor is applied to all heating emissions. Therefore, the assumption that seasonal emissions are caused by heating fuels in the residential sector is consistent with observations.

Seasonal BC in New Jersey decreased beginning in 1970 and disappeared in 1990 (Figure 2). For California, observations show a persistent seasonality in BC, caused by both emissions and atmospheric transport (Section 3.1.3). Discrepancies in both California and New Jersey show a steep drop from 1975 to 1985 in the *a priori* inventory (Figure 3b), which cannot be explained by any plausible adjustment of EFs. We therefore investigated both EFs and fuel consumption.

4.2.1 Inconsistencies in residential wood fuel data

A large part of seasonal emissions is due to wood combustion: 57% in New Jersey and 85% in California for 1965 to 2000 in the *a priori* emission inventory. That inventory relies on state wood consumption data from US EIA (2010) (Section 2.3). For 1960 to 1979, US EIA used the state wood consumption data in Estimates of US Wood Energy Consumption from 1949 to 1981 (US EIA, 1982) (hereafter referred as "Estimates of Wood"). Starting in 1980, US EIA switched its data source to US EIA Residential Energy Consumption Survey (RECS) (US EIA, 1981, 1984, 1987, 1990, 1993, 1997, 2001). "Estimates of Wood" relied on stove inventory and heating load data for each state. RECS provided wood use data by census region, each of which consists of several states, and US EIA distributed those totals among states according to total housing units. We assume that "Estimates of Wood" is a more accurate report of state wood consumption because of its detail.

The two data sets overlap in 1980 and 1981. In those years, data from "Estimates of Wood" is lower than that in RECS, by 45% in New Jersey and 29% in California. We therefore reduced the wood use by those fractions for New Jersey from 1980 to 2000, and for California from 1980 to 1985. California has its own state wood report from 1990 onward. This adjustment reduces the

30

discrepancy between measured and modeled wood use by about 70% in 1985. Even with the adjustment, wood fuel use still peaked around 1980s due to renewed heating use during the energy crisis in 1970s.

4.2.2 Residential emission factor updates

Figure 3b includes normalized DFs after the wood fuel adjustment. For California, the normalized DF decreases from 2.1 in 1970 to 1.4 in 1980, and remains around unity from 1990 onward, since the increasing trend in *a priori* emission is steeper than the observed trend. For New Jersey, the *a priori* emission shows a flatter decreasing trend than the observation during 1970-2000. In the following sections, we review EFs for residential coal, wood, and fuel oil, as each of them contribute more than 20% in at least one of the two states.

4.2.2.1 Coal heating stoves

EFs for coal heating stoves in the *a priori* inventory were taken from Bond et al. (2004). The PM emission for small stoker-fired and handfeed coal combustion sources has been decreased by better design of air delivery systems and proper combustion practice (US EPA, 1993). In the work of Bond et al. (2004), several measurements made in the early 1980s were tabulated but not used because they did not represent the technology in the inventory year of 1996. These measurements were 10-22 g/kg (Hughes & DeAngelis, 1982), 17–79 g/kg (Jaasma & Macumber, 1982), 0.6–65 g/kg (Sanborn & Montpelier, 1982). Additional measurements were 10.4 g/kg (Butcher & Ellenbecker, 1982) and 7.6 g/kg (Truesdale & Cleland, 1982). If all the measurements represent a lognormal distribution, the expected value would be 23 g-PM/kg; assuming the BC fraction given by Bond et al. (2004) (0.4) gives 10 g-BC/kg fuel. We choose the expected value as EFs for coal heating stoves before 1980.

4.2.2.2 Middle distillate and heavy fuel oil burner

Both middle distillate and heavy fuel oil were consumed mainly by boilers and furnaces in the US (Bond et al., 2004). The PM emissions from these heating units have been reduced between 1960 and 2000 by upgrading oil atomization and combustion aerodynamics (Offen et al., 1976; US EPA, 1996).

31

In the *a priori* inventory, EFs were from US EPA AP-42 (1996), giving a PM EF of 1.1 g/kg for heavy fuel oil boilers. Earlier measurements in 1970s, however, show higher EFs: Barrett et al. (1973) reported a PM EF of 2.5 g/kg, and Offen et al. (1976) presented an EF of 4.5 g/kg for this source. We choose an EF of 3.5 g/kg for heavy fuel oil boiler in 1975 and before, with an uncertainty that includes both measurements. The BC EF is then 0.13 g/kg assuming a 3.6% BC fraction in PM (US EPA, 1991b, 1996), a factor of 2 higher than the original EF.

For middle distillate emission, AP-42 gives 0.05 g/kg PM EF for new design furnaces and 0.40 g/kg for pre-1971 devices (US EPA, 1996), and a 0.25 g/kg was chosen in the *a priori* inventory (Bond et al., 2004). We choose 0.40 g-PM/kg EF from AP-42 (US EPA, 1996) for devices used in 1970 and before to represent the old burner technology. The corresponding BC EF is 0.10 g/kg for middle distillate burners, assuming a 26% BC fraction in PM (Hildemann et al., 1991; US EPA, 1996).

4.2.2.3 Wood heating stoves

The EF for wood heating stoves in the *a priori* inventory came from the Houck and Tiegs (1998) review of conventional stoves. These exclude Phase I and Phase II wood stove standards published by EPA in 1988 and 1990 (US EPA, 1988, 2000). We added EPA-certified wood stoves as a possible technology.

The following PM EFs have been reported in literature for EPA certified stoves: 3.4g/kg by Tiegs and Houck (2000), 7.2 g/kg by Gullett et al. (2003), 1.7 g/kg by Fine et al. (2004), and 2.2 g/kg by Houck et al. (2008). We chose the average of these measurements for the updated inventory. Assuming 16% of PM is BC (Fine et al., 2004) provides a BC EF of 0.58 g/kg for EPA certified wood stoves. EPA standards are set in g/h and were not used to determine emission rates.

A wood use survey conducted in Vermont, Delaware, Maine, and Pennsylvania (Pittsburgh) reported that 30% to 40% of wood stoves were EPA-certified in the 1990s (Broderick et al., 2005). We use the transformed normal function (Geroski, 2000; Grübler et al., 1999; Streets et al., 2004) to represent a transition to EPA-certified stoves between 1980 and 2000. The

32

transformation time of seven years gives a prediction of 10% EPA-certified stoves in 1985 and 44% in 2000.

4.2.1 Caveat

When calculating seasonal emissions, we do not account for sources other than residential sector. In winter, vehicle emissions are 11% to 19% higher during a cold start since catalysts may not function properly (H. Wang et al., 2008; Weilenmann et al., 2009; Yanowitz et al., 1999). However, higher winter vehicle emissions can be offset by lower on-road, recreational, or agricultural activities. In California, winter vehicle activity was lower than other seasons by 10% and 25% for LDGV and HDDV respectively (McDonald et al., 2014). Vehicles manufactured before 1975 did not have differing cold-start emissions because they were not equipped with catalytic converters (Cadle et al., 1999). The seasonality then depends on vehicle activity. If the activity of LDGV and HDDV is assumed to be 10% and 25% lower in winter, as observed in California in 2010, then seasonal emissions from vehicles would be 30% and 10% of the seasonal emissions from residential sector in California and New Jersey, respectively, based on the *a priori* emission inventory for 1975. Overall, the seasonal emissions from vehicles are small compared with the residential sector.

4.3 Updated Emission Trend

In the preceding sections, we described how normalized DF trends derived from observations, along with fuel and technology evolution in the *a priori* inventory, were used to identify likely causes of discrepancies. We then examined the evidence for the EFs used in the inventory and adopted plausible EF updates, which are summarized in Table 2.

Figure 6 shows the emission inventory with the new EFs ("updated emission") and compares it with the emission inventory using scaling based on normalized DFs ("scaled emission"). The updated emissions reproduce the decreasing trend in the observations, but they are not identical to the scaled emissions, and include attribution to particular causes.

Compared with the *a priori* emissions, baseline emissions are increased before 1985. Baseline emissions decrease by a factor of seven and three for New Jersey and California,

33

respectively, over the period from 1965 to 2000. For seasonal emissions, updates to wood stove EF decrease California emissions between 1980 and 2000, and updated EFs for coal and oil boilers increase seasonal emissions for New Jersey in the 1970s.

Table 2. BC Emission Factor Updates and Technology Additions.

Sector	Fuel	Technology	Original Emission Factor, g/kg	New Emission Factor, g/kg
Vehicle	Light distillate	Pre-regulation LDGV	0.28[a]	1.0[b]
Vehicle	Middle distillate	Pre-regulation HDDV	2.38[c]	4.4[b]
Transportation	Heavy fuel oil	Domestic shipping	0.41[d]	1.0[d]
Industry	Middle distillate	Off-road equipment and tractor, naturally aspired engines*	2.0[e]	5.9[f]
Residential	Coal	Heating stove, 1980 and before*	5.4[g]	10[h]
Residential	Middle distillate	Boiler, 1970 and before*	0.06[i]	0.10[j]
Residential	Heavy fuel oil	Boiler, 1975 and before*	0.04[i]	0.13[i]
Residential	Wood	Heating stove, EPA certified*	1.35[k]	0.58[l]

[a]Williams et al. (1989).
[b]See discussion in section 4.1.1.
[c]Yan et al. (2011).
[d]Lack et al. (2008), see discussion in section 4.1.2.
[e]Kean et al. (2000).
[f]US EPA (1991a) and (1998). 70%, 50%, 20% and 10% engines are naturally aspired in 1970, 1975, 1980 and 1985.
[g]Bond et al. (2004).
[h]See discussion in section 4.2.2.1.
[i]AP-42 US EPA (1991b, 1996), see discussion in section 4.2.2.2
[j]See discussion in section 4.2.2.2
[k]Houck and Tiegs (1998).
[l]See discussion in section 4.2.2.3. 10%, 29%, 40% and 44% stoves are certified by EPA in 1985, 1990, 1995 and 2000.
* New technology added in this work

The correlation coefficients between updated emissions and scaled emissions are 0.98 for both baseline and seasonal emissions in New Jersey, 0.92 for baseline emissions in California, and 0.59 for seasonal emissions in California. However, there is some remaining discrepancy.

34

For California, the updated seasonal emissions are 50% lower than the scaled emissions from 1965 to 1980 and also show a greater increase than the scaled emissions for 1965 to 1985. This discrepancy could be caused by emission of brown carbon from wood combustion as discussed in Section 3.2.

For New Jersey, seasonal emissions vanish in observations in 1990, and decrease but do not vanish in the updated emissions. These discrepancies could be caused by the spatial heterogeneity of seasonal emission reduction. The COH measurements were mainly made in urban areas, where emission reduction from improved stove technology could have occurred more quickly in urban area than in rural areas. We found no records of this difference for New Jersey, but residents in rural areas of Oregon used 30% more non-certified wood stoves than in urban areas in 2014 (Swab, 2015).

This hypothesis is supported by the fact that rural winter BC concentrations are 25% higher than those in non-winter seasons during the 2000s, based on data of IMPROVE network (http://vista.cira.colostate.edu/improve/, 2017) (Malm et al., 1994), whereas the seasonal difference in the urban COH measurement used here is less than 1%. We are not able to apply IMPROVE data to infer long-term trends, because that data set began in year 1985. However, this rural-urban difference would mainly influence the residential sector. Other sectors, like vehicle, shipping, and industry, are subject to nationwide standards and have more consistent EFs among regions.
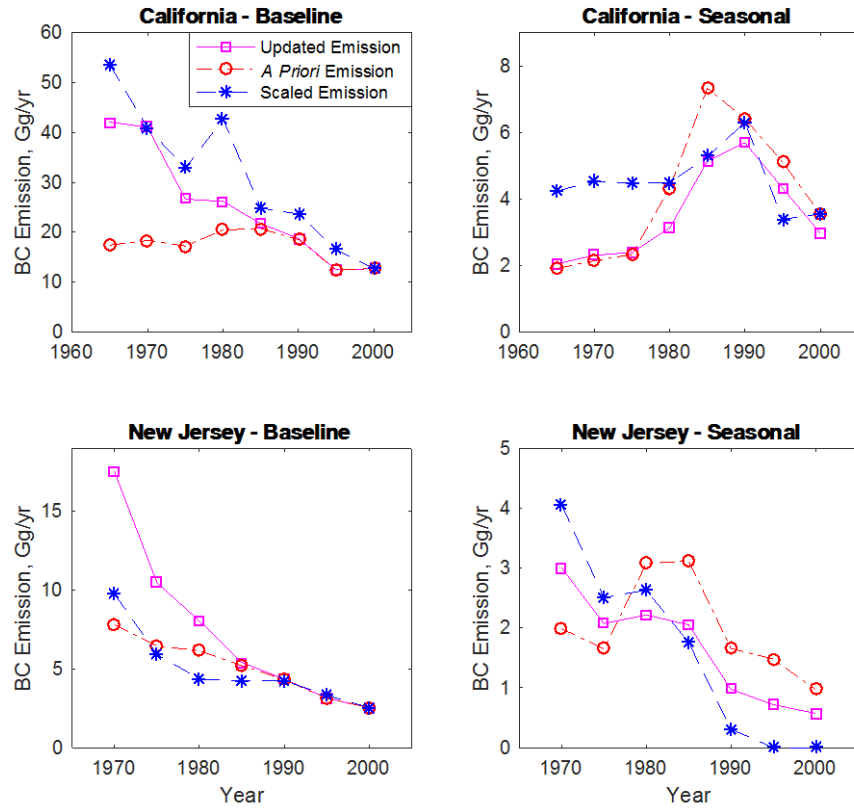
35

Figure 6. Comparison of *a priori* emissions (red line), scaled emissions (blue line) and new emissions calculated with the updated emission factors (purple line). The *a priori* emissions use the original wood fuel, and the scaled and updated emissions are calculated with updated wood fuel (Section 4.2.1).
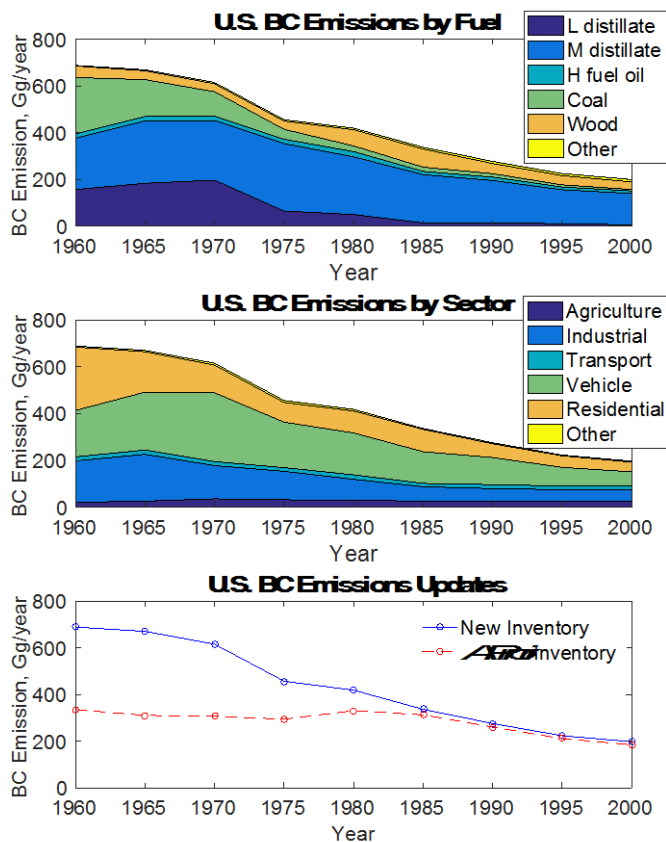
36

5. New US Emission Inventory

Figure 7. Updated US emission inventory by fuel (top) and by sector (middle), and comparison of the updated inventory and the *a priori* inventory (bottom), excluding open burning. L: Light, M: Middle, H: Heavy.

New estimates of US anthropogenic BC emissions, using the updated EFs (Table 2) are shown in Figure 7 for 1960 to 2000. Emissions decreased by a factor of three from 1960 to 2000, totaling about 690 Gg/yr in 1960 and 620 Gg/yr in 1970 (excluding open burning).

Compared with the *a priori* inventory, the updated US BC emission inventory is 110% higher in the 1960s and 77% higher in the 1970s (Figure 7, bottom panel). The new emissions decrease from 1960 to 1980, while the *a priori* inventory peaks in 1980. The altered trend is mainly produced by decreasing vehicle EFs caused by standards implemented during the 1970s

and 1980s, and technology improvements in off-road engines, residential stoves and boilers as discussed in Section 4.

In the new estimates, as before, BC emissions from 1960 to 2000 are dominated by on-road and off-road vehicles using middle distillate fuel (Figure 7, top panel). BC from middle distillate is 130% greater in 1960s and 1970s than the previous estimate, accounting for 44% of the total emissions. Compared with the *a priori* inventory, BC emission from coal is 58% higher before 1980, exhibiting a greater fractional decline as the coal consumption shifted from residential heating use to power generation. BC emissions from wood peaked in 1980 in both *a priori* and the new estimates, but the updated wood emission reduced by 11% after 1980. BC from light distillate (gasoline) fuels produced 25% of emissions in the 1960s, decreasing to less than 5% after 1985 when more stringent standards were implemented. The new estimate for light distillate is 180% higher before 1980.

Vehicle, industrial and residential sectors contribute about 85% of US BC emissions from 1960 to 2000. Because of continuous implementation of vehicle standards, BC emissions decreased even when the consumption of light distillate and middle distillate increased by factors of two and eleven, respectively. From 1960 to 2000, the effective EF for the LDGV fleet decreased from 1.0 g/kg to 0.01 g/kg, and the EF for the HDDV fleet decreased from 4.4 g/kg to 0.5 g/kg.

The residential sector also decreased its emissions by switching away from coal consumption, which dropped by 95% from 1960 to 1980. For industrial sectors, the emission reduction is associated with the technology improvement of off-road vehicles, resulting in a threefold decline in EFs.

6 Summary

This work provides the first long-term observational constraints on a US BC emission inventory in which activity and technology are explicitly represented and adjusted. We combine top-down constraints (ambient observation in California and New Jersey) and bottom-up

38

constraints (emission measurement) to improve representation of emission sources and EFs. The method developed in this work is also applicable to other short-lived tracer species when the influence of primary emissions dominates observed concentrations. The updated US BC emission inventory for 1960 to 2000 better agrees with observations, is higher than the *a priori* estimation by about 80% between 1960 and 1980, and has a decreasing trend that was missing in the *a priori* inventory. Technology improvements and emission standards have driven the emission reduction in vehicle and industrial sectors, while fuel transition and reduction caused the decline in residential emissions. Based on both observational constraints and emission measurements, we recommend the following updates: (1) EFs for pre-regulation vehicles should be 80%-250% higher; (2) EFs for residential heating stove and boilers should be 70% to 200% higher for the 1980s and before; (3) technologies for naturally aspired off-road engines should be added before 1980; and (4) technology for certified wood stove after 1985 need to be included.

This work is subject to some limitations. It assumes that spatial heterogeneity is relatively constant throughout the study period, and that seasonality in concentrations due to differing atmospheric transport is adequately corrected. Also, comparing a bottom-up emission inventory, which consists of many fuel-technology groups, with top-down constraints from measurements of two states, is an underdetermined problem. If long-term observations from other states were available, they could further constrain sources dominant in those states; the availability of data from only two states leaves the dominant sources relatively unconstrained. Although we used fuel usage trends and differences between the two states to identify groups of the emission sources that required updates, there is insufficient information to unambiguously identify specific technologies that are poorly represented.

Despite these caveats, the combination of observational constraints and evidence review provides the most detailed and supportable representation of US historical emissions to date. These updated EFs better represent historical technology transitions and can be applied to the same technology in emission inventories for other regions. The new trend indicates a decreasing climate forcing from BC in the US for 1960 to 2000, an important input for climate studies.

39

Acknowledgments

40

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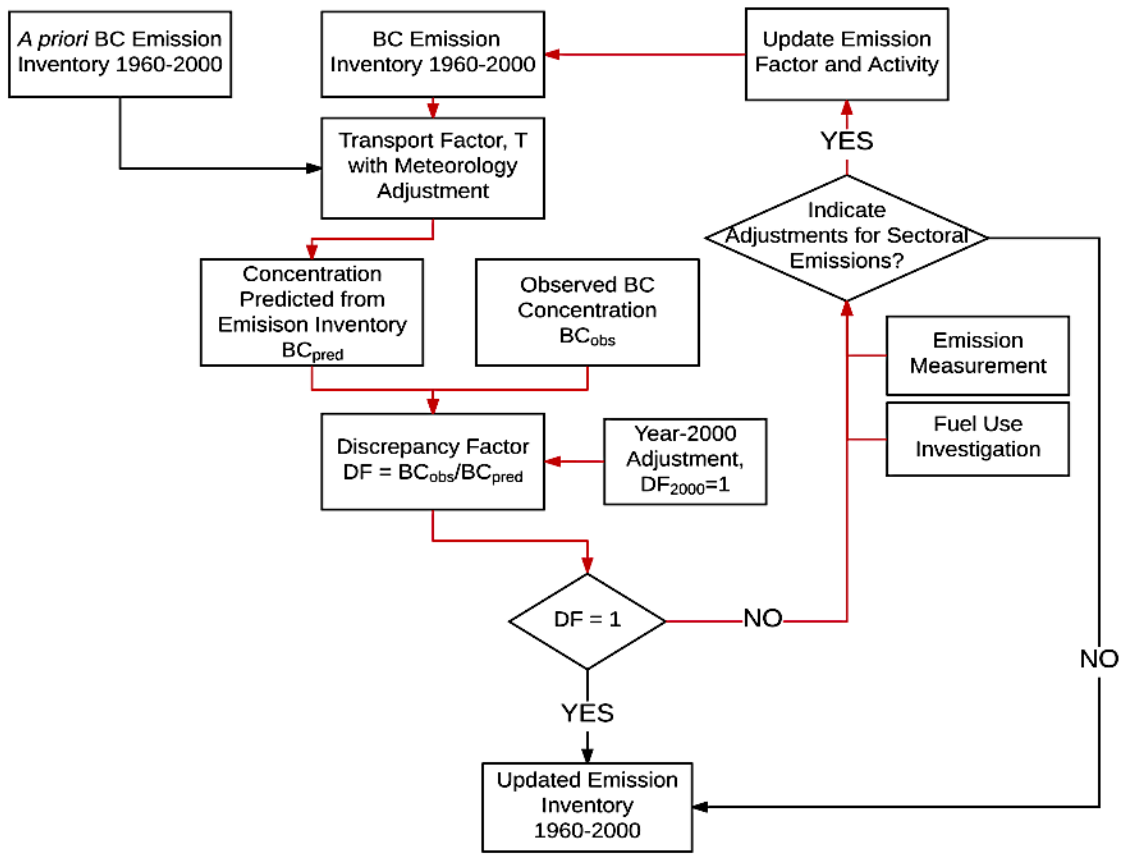
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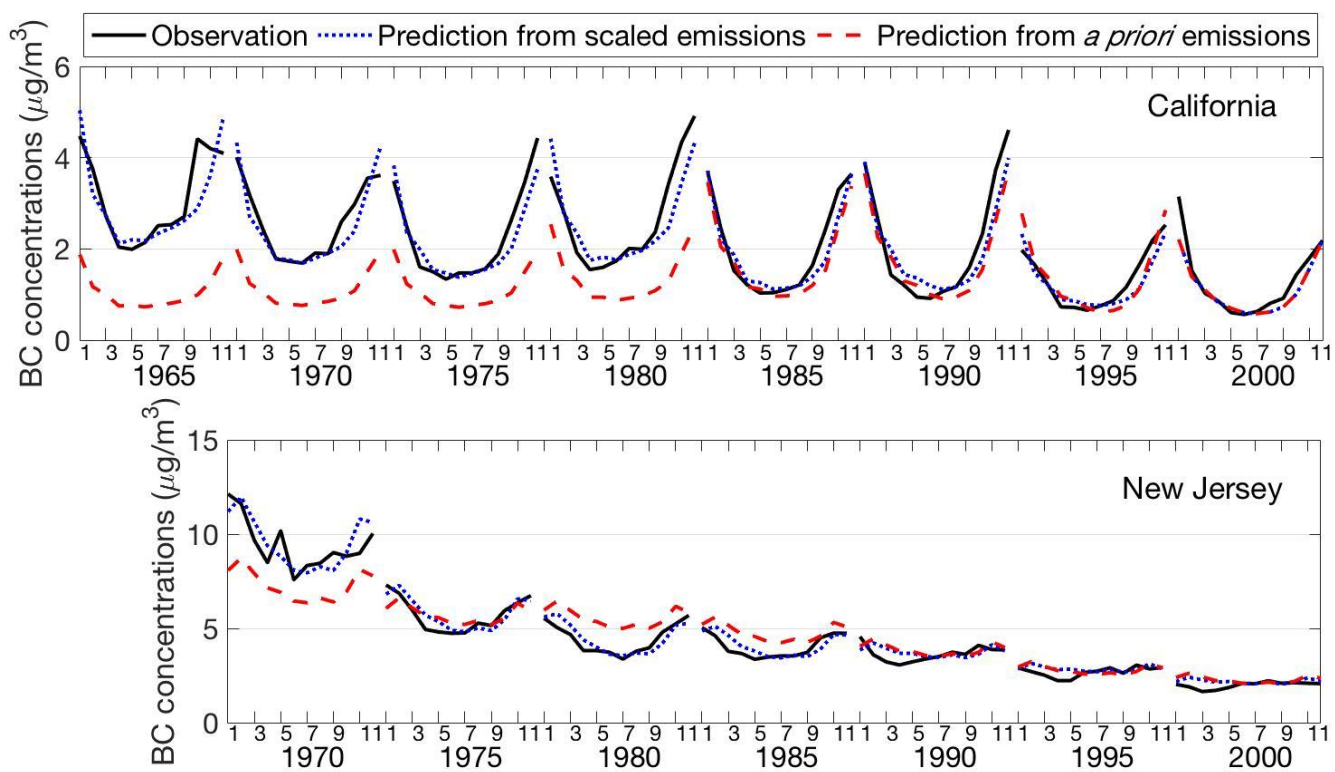
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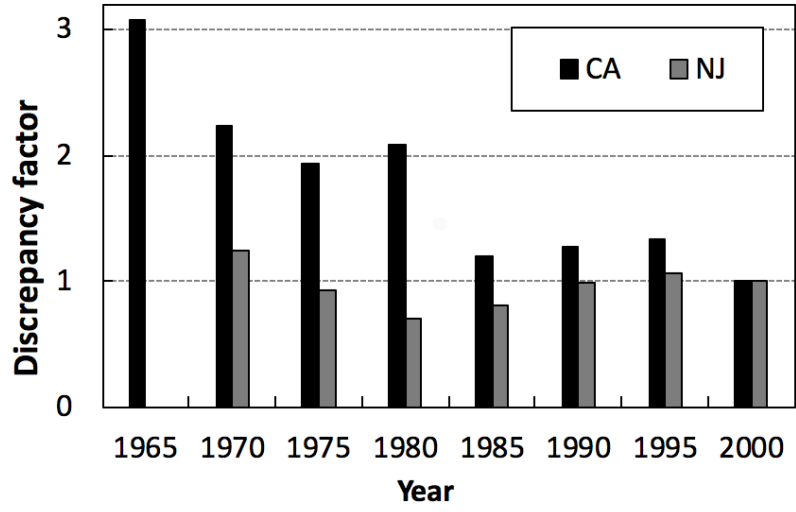
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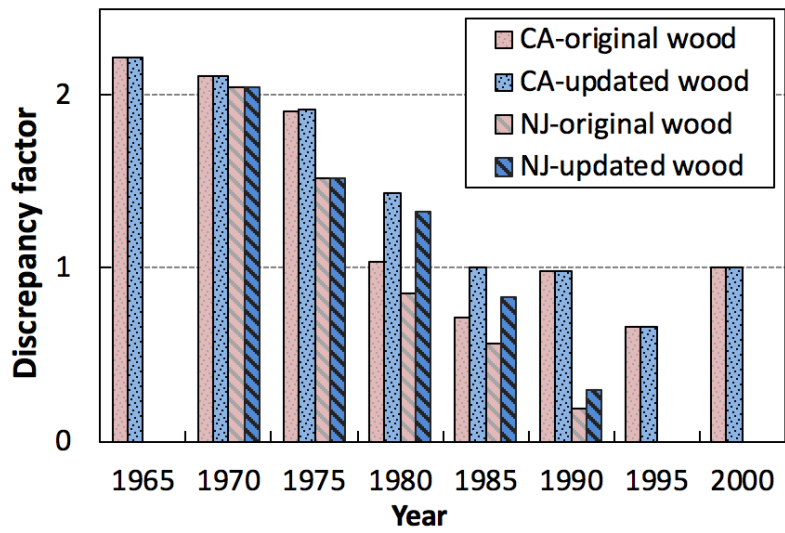
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(a)

(b)

California - Baseline

California - Seasonal

New Jersey - Baseline

New Jersey - Seasonal

U.S. BC Emissions by Fuel

U.S. BC Emissions by Sector

U.S. BC Emissions Updates