Use of Black Carbon and Organic Carbon Inventories
for Projections and Mitigation Analysis

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ABSTRACT

Black carbon (BC) and organic carbon (OC) aerosols have been identified as having potentially significant impacts on climate change, particularly at regional scales. However, the appropriate role for BC and OC emissions in any climate change mitigation strategy remains uncertain. We present emission inventories and projections for BC and OC in the United States circa 2000, and projections for the years 2015 and 2020. The projected U.S. emission inventories for 2015 and 2020 take into account recently promulgated regulations that target $PM_{2.5}$ (of which BC and OC are essentially subcomponents), such as the Clean Air Highway Diesel Rule (or 2007 Highway Rule), the Clean Air Nonroad Diesel Rule, the Clean Air Interstate Rule, and others. BC emissions in the United States are projected to decline by 42 percent from 2001 to 2020, primarily as a result of diesel vehicle regulations. In contrast, OC emissions are projected to decline by only 9 percent, because open biomass burning emissions are assumed to be largely unaffected by these air quality regulations. We also briefly present the results of global assessments of 1996 and 2030 emissions of BC and OC, and contrast these results with the inventory results for the United States.

INTRODUCTION AND CLIMATE CHANGE BACKGROUND

Purpose

The purpose of this research is to report and assess different black carbon (BC) and organic carbon (OC) primary emission inventories in the United States, to report and characterize the effects of air quality regulations that target $PM_{2.5}$ on BC and OC emissions, and to identify and evaluate emission mitigation measures that can lower emissions beyond levels achieved by full implementation of existing and forthcoming regulations.

Background

BC and OC are particulate aerosols formed by incomplete combustion. The co-emission ratio of BC to OC varies by fuel type, combustion efficiency, and the extent of emissions control. When fossil fuels, such as oil and coal, are incompletely combusted (i.e., not completely oxidized to carbon dioxide [CO_2]), BC tends to be formed in much larger amounts than OC. Diesel engines, for example, are estimated to be the largest source of BC in the United States.[1,2,3] When biomass fuels, such as wood, are incompletely combusted, OC is formed in greater amounts than BC.

The net effects of BC and OC on climate are difficult to quantify, and not all aspects of their climate effects are well understood. BC is generally thought to have both a direct warming effect, by absorbing incoming solar radiation in the atmosphere, and an additional warming effect, by reducing the albedo (reflectivity) of snow and ice.[4] The Intergovernmental Panel on Climate Change (IPCC) recently estimated these cumulative (since 1750) BC radiative effects to be roughly +0.3 Watts per square meter (W/m^2).[5] OC is generally thought to have a direct cooling effect, by reflecting incoming sunlight. The direct negative forcing effect of OC emitted from fossil fuels is estimated to be roughly -0.1 W/m^2. When the direct radiative forcing of BC and OC are combined with other aerosols (sulfates, nitrates, mineral dust), the total net aerosol direct effect is negative and estimated to be approximately -0.5 W/m^2. Like other aerosols, BC and OC have indirect radiative effects on clouds by altering their albedo and lifetime. The indirect cloud albedo effect of *all* aerosols (BC, OC, sulfates, nitrates, mineral dust) is roughly -0.7 W/m^2. By contrast, the total cumulative radiative forcing effect of the key anthropogenic greenhouse gas emissions (CO_2, methane, and nitrous oxide) is estimated to be +2.3 W/m^2.[5]

The net effects of BC and OC on temperature and precipitation are potentially significant, especially at regional scales, because BC and OC have relatively short atmospheric lifetimes (days to week) and are therefore not well-mixed in the atmosphere, unlike greenhouse gases which are well mixed due to their very long atmospheric lifetimes (decades to centuries). Some researchers have postulated that reductions in BC may be a relatively inexpensive and achievable method to reduce future temperatures.[6,7] However, any climate benefits of BC reductions (less warming) need to be weighed against the co-effects of associated OC reductions (potentially more warming). Koch et al. show, for example, that different sectors generate different net positive or negative forcing,[8] based on the differences in relative amounts of BC versus OC, implying that targeting BC reductions as a means of climate change mitigation may be viable only for certain sectors (in their study, the residential and transportation sectors have the highest positive forcing due to BC).

Regardless of the role of BC and OC emissions in climate change, reductions in those emissions are expected to have significant health benefits. For example, BC is estimated to account for a significant fraction of the total particulate mass from diesel vehicles, which has been associated with an estimated 125,000 annual U.S. cancer cases.[9]

Scope and Limits

This project focuses on primary BC and OC emission inventories and mitigation measures for the United States. The results of global BC and OC emission inventories will be presented; however, a thorough review of existing and projected future global inventories of BC and OC is beyond the scope of this project. In particular, this project does not address potential future trends of BC and OC emissions in developing countries (e.g., China and India). These developing country trends may mean that conclusions reached for the United States can not be extended to global emissions.

In this paper, BC and elemental carbon will be treated as equivalent. That is, we assume that any inventory that estimates the mass of elemental carbon emitted would produce the same mass emission estimate for BC. These definitional differences for carbonaceous aerosols introduce uncertainties about the climatic effects of "black carbon." These issues will not be addressed here but are receiving attention.[10] A similar assumption is made for OC and primary organic aerosol.

Other Relevant Literature

Many researchers are developing global and regional estimates of current and future emissions of BC and OC, as well as estimates for specific sectors and countries. This research is being conducted in

both a bottom-up manner (i.e., emissions estimates based on emission factors and activities levels for various sources) and in a top-down manner (i.e., emissions estimates based on field or satellite measurements and transport modeling).

Bond et al. developed bottom-up global inventories for BC and OC for the year 1996,[2] which Streets et al., Rao et al., and Smith and Wigley use as a basis for projecting future emissions. [3,11,12] Top-down analysis to estimate BC emissions for South Asia based on ambient concentrations has produced a significantly higher estimate of emissions than produced by bottom-up methods, and direct testing of sources in India has been recommended.[13] All the emissions inventories presented in this paper are for primary emissions and were produced by bottom-up analyses; however, it is important to resolve differences between inventories obtained by bottom-up versus top-down methods.

In the United States, regional planning organizations are combining resources to estimate particulate emissions from open biomass burning (i.e., wildfires, prescribed burning, and field burning of agricultural wastes).[14] These results are important, because open biomass burning is a significant source of OC and BC emissions in the United States and around the world.

INVENTORY AND MITIGATION

Methods

Our objectives are to show (1) existing U.S. and global inventories of BC and OC; (2) future projections of BC and OC based on current inventories; (3) the extent to which air quality regulations in the United States are expected to change BC and OC emissions; and (4) what mitigation options are available, and at what cost, to reduce residual BC and OC emissions that may not be addressed by air quality regulations.

We review emission inventories for BC and OC circa 2000 (specifically, inventories were identified for 1996, 2000, and 2001) and identify the effects of existing and forthcoming regulations on those pollutants for years 2015 and 2020. The mitigation analysis covers this same timeframe and incorporates the existing and forthcoming regulations into our baseline against which emission reductions and costs are measured.

Results and Discussion

<u>Available U.S. Inventories</u>

We examined three U.S. inventories of BC and two U.S. inventories of OC. The specific inventories examined included that of Streets et al. for the year 1996, Battye et al. for the year 2000, and the U.S. Environmental Protection Agency (EPA) for the year 2001. [3,1,15, 16, 17] The Streets inventory was generated specifically for the purpose of producing global bottom-up (i.e., technology-based) BC and OC inventories. The Battye inventory was specifically for BC (and did not include OC), and was generated using $PM_{2.5}$ data from EPA's 1999 National Emissions Inventory (NEI) and BC speciation factors for that $PM_{2.5}$ data. The EPA inventories were for elemental carbon and primary organic aerosol, which we assume to be equal in mass to BC and OC. The EPA inventories for 2001 were completed as part of the EPA Office of Air Quality Planning and Standards Regulatory Impact Analysis (RIA) measures needed to achieve compliance with the $PM_{2.5}$ National Ambient Air Quality Standards (NAAQS) (15 µg/m^3 annual average, 35 µg/m^3 average over 24 hours) by the year 2020.

Table 1 and Figure 1 present the results of these three inventories of BC emissions. Table 1 shows good agreement for U.S. BC emissions among the three inventories. Table 2 and Figure 2 show the two available OC inventories. Table 2 shows reasonably good agreement for the two OC inventories.

Review of U.S. Regulations Affecting PM$_{2.5}$ (and Therefore BC and OC)

This section identifies and describes selected regulations that will come into effect between 2000 to 2020, and qualitatively expresses their general effect on PM$_{2.5}$, BC, and OC.

Clean Air Nonroad Diesel Rule. This rule regulates sulfur content, nitrogen oxides (NO$_x$), non-methane hydrocarbons (NMHC), and particulate matter (PM) emissions for nonroad diesels. On May 11, 2004, EPA signed the final rule introducing Tier 4 standards, which are to be phased in over the period from 2008 to 2015. At the time of signing, average in-use sulfur content was approximately 3,000 ppm (0.3%). To enable the sulfur-sensitive control technologies required to meet the Tier 4 emission limits—such as catalytic particulate filters and NO$_x$ absorbers—EPA mandated reductions in sulfur content in nonroad diesel fuels. The limit is 500 ppm, effective June 2007 for nonroad, locomotive, and marine diesel fuels. The limit will be further reduced to 15 ppm (ultralow sulfur diesel) effective June 2010 for nonroad fuel, and June 2012 for locomotive and marine fuels.[18]

Clean Air Highway Diesel Rule (or 2007 Highway Rule). This rule requires the sulfur content of highway diesel vehicles to be lowered from 500 ppm to 15 ppm, starting in June 2006; the conversion to 15 ppm must be complete by 2009. The rule also requires lower emissions for NO$_x$ and NMHC, which will be phased in from 2007 to 2010. Finally, the rule contains a PM limit of 0.01 grams per brake horsepower-hour, coming into full effect in the 2007 heavy-duty engine model year.[19]

Clean Air Interstate Rule. This rule requires lower emissions of sulfur dioxide (SO$_2$) and NO$_x$ from coal-fired power plants and is expected to significantly lower sulfate and nitrate secondary particulate (i.e., particulate aerosols formed in the atmosphere after the flue gas has exited the stack). However, nationwide emissions of BC and OC are not expected to be significantly affected by the Clean Air Interstate Rule, particularly because emissions from coal-fired power plants are already estimated to contribute less than 3 percent of nationwide BC emissions and less than 1 percent of nationwide OC emissions. Therefore, the limited effects of the Clean Air Interstate Rule on BC and OC inventories can be contrasted to other regulations (e.g., the Clean Air Nonroad Diesel Rule and the Clean Air Highway Diesel Rule, which significantly reduce emissions of BC).

PM$_{2.5}$ NAAQS. In September 2006, EPA revised the daily PM$_{2.5}$ NAAQS from 65 to 35 µg/m^3, while retaining the yearly standard of 15 µg/m^3. It is expected that the regulations mentioned above (the diesel rules and Clean Air Interstate Rule) will play a significant role in helping most areas of the United States comply with the revised PM$_{2.5}$ NAAQS. However, there will still be some localities in non-attainment for the revised PM$_{2.5}$ NAAQS. In the PM$_{2.5}$ NAAQS RIA, EPA identified various measures that could be used to bring localities into attainment with these revised NAAQS. These measures produced reductions in BC and OC, as well as PM$_{2.5}$. However, because these measures are implemented locally, they are not expected to produce large reductions in nationwide emissions of BC and OC.

U.S. BC and OC Projections for 2015 and 2020

EPA developed inventories for PM$_{2.5}$, BC, and OC as part of the NAAQS RIA. These inventories are designated A through F. This paper deals with scenarios A, C, and F only. Scenario A represents the emission inventory in 2001. Scenario C represents the emission projections for 2020, taking into account

such national regulations as the Clean Air Nonroad Diesel Rule, the Clean Air Highway Diesel Rule, the Clean Air Interstate Rule, and others. When the emission inventory for Scenario C was modeled to determine resultant $PM_{2.5}$ ambient air concentrations, some localities were found to be above the NAAQS limits of 15 µg/m^3 as an annual average, and 35 µg/m^3 as a 24-hour average. Therefore, additional local measures to further reduce emissions were considered for those localities, until those localities came into compliance with the 15/35 µg/m^3 ambient concentration limits. The resultant emissions scenario was labeled Scenario F.

Table 3 and Figures 3 and 4 depict EPA estimates for BC and OC, respectively, for the years 2001, 2015, and 2020. The estimates for the year 2020 are for Scenario C (U.S. federal regulations implemented, but some local areas still in non-attainment for the $PM_{2.5}$ NAAQS) and Scenario F (local measures implemented such that all localities in the United States are in $PM_{2.5}$ attainment). The impacts of the local measures from Scenario C to Scenario F are limited, with reductions of approximately 6 percent and 2 percent for BC and OC, respectively.

Table 3 indicates that U.S. BC emissions are projected to decline from 436 Gg in 2001 to 255 Gg in 2020 Scenario F; this represents a reduction of approximately 42 percent. In contrast, OC emissions are projected to decline from 1,326 Gg in 2001 to 1,206 Gg in 2020 Scenario F; this represents a reduction of 9 percent. One reason the analysis results presented in Table 3 indicate a greater percentage reduction from 2001 to 2020 Scenario F for BC than for OC is that mobile sources represent the largest fraction of U.S. BC emissions in 2001, and this source is reduced significantly by the combined effects of the diesel regulations. Mobile source BC emissions are estimated at 234 Gg in 2001, representing 54 percent of the nationwide BC emissions of 436 Gg. Under 2020 Scenario F, mobile source BC emissions are projected to decline to 71 Gg, a reduction of 163 Gg. In fact, because the total reduction of BC emissions from all sources from 2001 to 2020 Scenario F is 181 Gg, the reduction of 163 Gg from mobile sources represents approximately 90 percent of the nationwide BC reduction.

Table 3 shows a smaller percentage decrease for OC than for BC. This smaller reduction for OC is largely due to the assumption that the biomass burning emissions are constant from 2001 to 2020 Scenario C. This assumption resulted in a generally low percentage reduction in nationwide OC emissions, because biomass burning is the largest source of OC emissions in the United States.

Mitigating BC/OC Beyond Baseline 2020

For the mitigation analysis, we used the EPA inventories for BC and OC that were associated with the RIA for the $PM_{2.5}$ NAAQS. These inventories were chosen because detailed BC and OC emissions projections were available for 2015 and 2020, and these projections were closely tied to analysis of the effects of U.S. regulations on particulate emissions

Our research is currently looking beyond the emission reduction measures that produced the BC and OC inventories in 2020 Scenario F. Two key sectors are being analyzed to achieve BC and OC reductions beyond those in Scenario F:

- In some cases, regulations that are included in Scenario F will not be fully implemented in the year 2020. For example, since the Clean Air Highway Diesel Rule and the Clean Air Nonroad Diesel Rule will apply to new vehicles, the entire fleet of vehicles will not have been built to meet these regulations by 2020. Therefore, there are additional reductions that will accrue until 2030 or beyond, when the entire fleet will eventually meet the applicable regulations. These reductions could be accelerated at additional cost.

- Some additional measures are not even partially included in the 2020 Scenario F inventory. For example, nationwide open biomass burning emissions were assumed to be constant from 2001 to 2020 Scenario C, and declined by less than one percent to Scenario F. Therefore, open biomass burning mitigation measures will be examined in depth in the mitigation assessment.

The mitigation assessment is evaluating the emission reduction potentials and costs of these additional measures. AirControlNET is being used to determine reduction potentials and costs, where appropriate.

Global Inventories for BC and OC

Figures 5 and 6 show global emissions estimates for 1996 for BC and OC, respectively.[3] Figure 5 shows much larger BC emissions from East Asia (predominantly China) than from the United States. The residential sector is the largest source of BC emissions in East Asia and South Asia (predominantly India), whereas the transportation sector is the largest source of BC in the United States (and in European Union member countries of the Organisation for Economic Co-operation and Development [OECD Europe]). Figure 6 shows that open biomass burning is the largest source of OC emissions in most areas of the world, including the United States. However, the residential sector is the largest source of OC emissions in East Asia and South Asia.

Streets et al.[3] used the global emissions estimates for 1996 to project regional and global BC emissions for 2030 and 2050 under various IPCC scenarios. Their results for BC in the United States in 2030 ranged from 246 to 308 Gg. These results are generally compatible with the EPA 2020 Scenario F estimate of 255 Gg, although additional reductions from the EPA 2020 Scenario F estimate would be expected from 2020 to 2030. Similarly, Streets et al. project OC emissions in the United States in 2030 to range from 1,262 to 1,390 Gg. These results are generally compatible with the EPA 2020 Scenario F estimate of 1,206 Gg, although additional reductions in OC would also be expected from 2020 to 2030.

Globally, Streets et al. estimate that BC emissions will decrease anywhere from 9% to 34% from 1996 to 2050. OC emissions are projected to decrease 12% to 29% over the same time period.

One particularly interesting set of conclusions in the Streets et al. paper deals with the difficulty of control of transportation BC emissions and the trend in the ratio of BC to OC emissions. Streets et al. conclude that "[p]articularly difficult to control are BC emissions from the transport sector, which are projected to increase under most scenarios. We also expect that the global BC/OC emission ratio for energy sources will increase in the future; this signifies that the trends in carbonaceous aerosol emissions will probably shift toward increased net warming of the climate system."[3] When biomass emissions are included, however, the BC to OC ratio remains relatively constant over time at the global scale.

These conclusions for global emissions can be contrasted with the expected trends for the United States. As previously discussed, transportation BC emissions in the United States are expected to be substantially reduced (by almost 70 percent) from 2001 to 2020 Scenario F. The combined U.S. ratio of BC to OC is projected to decrease from 2001 to 2020 Scenario F, with an expected shift towards a net cooling effect, but only as far as carbonaceous aerosols are concerned. The differences in apparent results for global versus U.S. emissions is probably due to trends in the developing world.

CONCLUSIONS

This paper examines three U.S. BC and two U.S. OC inventories circa the year 2000. The three BC inventories show good agreement. The U.S. BC emissions for the three inventories range from 415 to 436 Gg. The two available U.S. OC emissions inventories also show fairly good agreement, with values of 1,476 and 1,326 Gg. The effects of U.S. regulations on $PM_{2.5}$, BC, and OC from circa 2000 to the year 2020 were examined. Regulations related to diesel mobile sources (e.g., the Off-Road Diesel Rule and the Clean Air Highway Diesel Rule) are expected to produce large reductions in BC emissions. (The diesel transportation regulations also reduce OC emissions, but because transportation is not as significant a source of OC emissions, the nationwide impact was not as significant for OC compared to BC.) Some regulations that target large reductions in $PM_{2.5}$ are not expected to produce significant reductions in BC or OC (e.g., the Clean Air Interstate Rule and Clean Air Mercury Rule) because these regulations do not address the carbonaceous fractions of $PM_{2.5}$.

Inventories for 2015 and 2020 show percent reductions in emissions from 2001 (Scenario A) to 2020 (Scenario F) of 42% for BC and 9% for OC. The relatively large percentage reduction in BC emissions comes primarily from diesel mobile source regulations. One reason for the relatively smaller reduction in OC emissions is the assumption that nationwide biomass burning emissions (i.e., combined emissions from wildfires, prescribed fires, and agricultural residues burning) remain essentially constant from 2001 to 2020 Scenario F.

Mobile source subsectors are considered for a mitigation assessment beyond the 2020 Scenario F. These subsectors include diesel engines powering non-road vehicles, line haul locomotives, and marine vessels. Open biomass burning was also identified as a large source of OC (and BC) emissions, and therefore also an appropriate target for mitigation beyond 2020 Scenario F.

Worldwide estimates of BC and OC emissions as calculated by Streets et al. were presented to provide contrast to and context for U.S. BC and OC emission estimates circa 2000. Emissions of BC in East Asia (predominantly China) are larger than in the United States. The residential sector dominates BC emissions in East Asia, while the transportation sector dominates in the United States and OECD Europe. The largest source of OC emissions for the United States and most of the world is open biomass burning, although the residential sector is the largest source of OC emissions in China and India.

BC and OC emissions projections for the United States in the year 2030 from the global assessment by Streets et al. were presented. There was generally good agreement with the 2020 Scenario F estimates. Conclusions from the Streets et al. global assessment were presented regarding the difficulty of controlling transportation BC emissions and the trend in the future ratio of energy-sector global BC/OC emissions. These global assessment conclusions were not similar to the conclusions that are obtained for the United States alone. This difference is probably due to different emission trends in the developing world compared to U.S. and other OECD trends.

The next steps in this research will include quantifying the emission reductions and costs for BC and OC mitigation measures in the United States that go beyond 2020 baseline projections.

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KEY WORDS

Black Carbon
Organic Carbon
Aerosols
$PM_{2.5}$
Climate Change
Air Quality Regulations

Table 1. United States BC inventories, circa 2000 (Gg).

	Biomass Burning	Residential/ Area	Industry/ Non-Power Point Sources	Mobile	Power	Other (non-combustion sources)	Total
Streets[a,b]	61	79	66	203	6		415
Battye[a]	93	41	29	236	4	29	433
EPA	110	54	18	234	14	7	436

[a] Estimated "residential" emissions rather than "area" emissions. "Area" emissions include sources such as commercial charbroilers that are in many sites within a locality.

[b] Did not estimate emissions for non-combustion sources.

Table 2. United States OC inventories, circa 2000 (Gg).

	Biomass Burning	Residential/ Area	Industry/ Non-Power Point Sources	Mobile	Power	Other (non-combustion sources)	Total
Streets[a,b]	954	348	31	133	10		1,476
EPA	658	298	121	148	12	90	1,326

[a] Estimated "residential" emissions rather than "area" emissions. "Area" emissions include sources such as commercial charbroilers that are in many sites within a locality.

[b] Did not estimate emissions for non-combustion sources.

Table 3. United States BC and OC inventories for 2001, 2015, and 2020 (Gg).

| Sources	EPA Inventories, from PM$_{2.5}$ NAAQS Regulatory Impact Assessment			
	2001	**2015**	**2020 Scenario C[a]**	**2020 Scenario F[b]**
BC				
Mobile	234	109	83	71
Fugitive dust	7	7	7	6
Area	54	48	46	43
Industry	18	17	17	17
Power	14	8	8	8
Biomass burning	110	110	110	110
Total	436	298	271	255
OC				
Mobile	148	104	87	85
Fugitive dust	90	90	90	89
Area	298	267	256	241
Industry	121	118	118	116
Power	12	18	21	21
Biomass burning	657	657	657	655
Total	1,326	1,256	1,230	1,206

[a] Scenario C: Base case projection with Clean Air Visibility Rule, Clean Air Mercury Rule, Clean Air Visibility Rule.

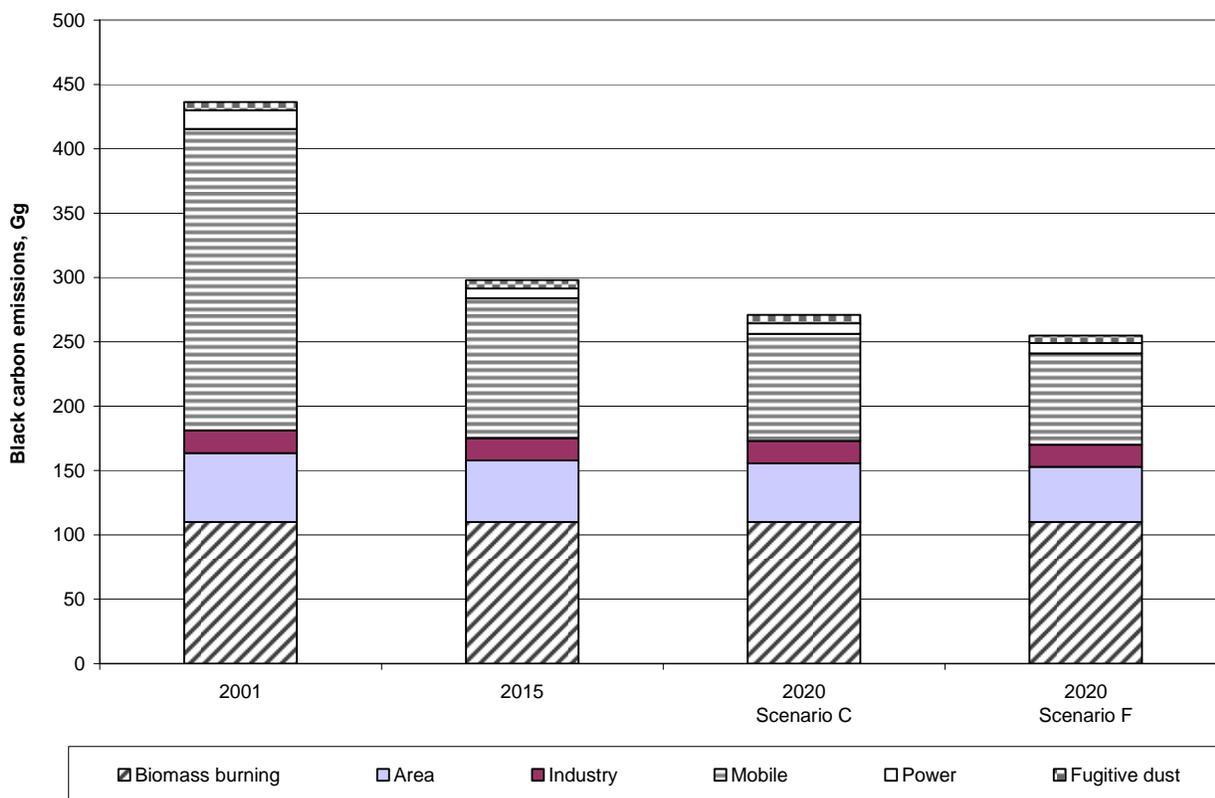[b] Scenario F: Compliance with PM$_{2.5}$ NAAQS: 15 µg/m^3 annual average, 35 µg/m^3 24-hour average.

Figure 1. United States BC inventories, circa 2000.
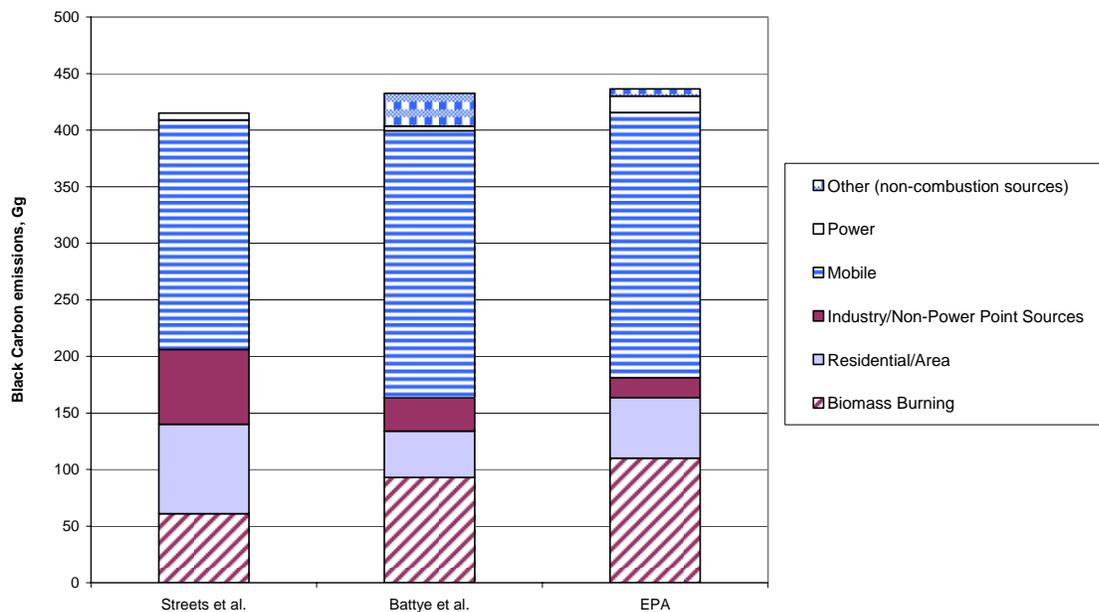
**Figure 2.** United States OC inventories, circa 2000.
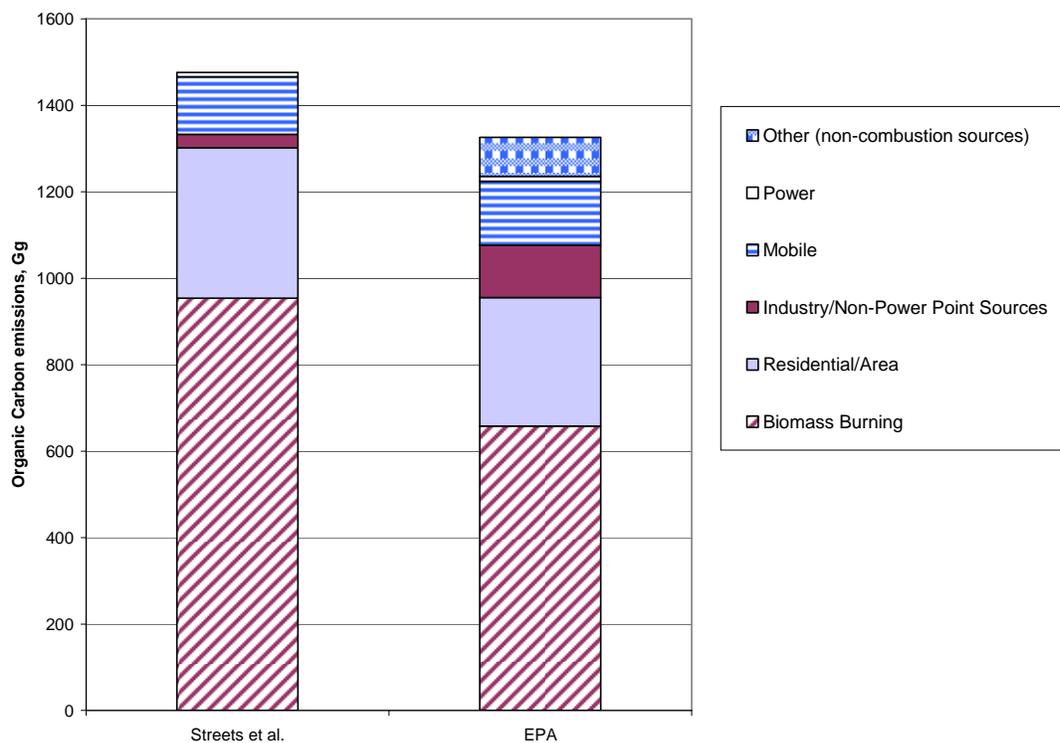
**Figure 3.** United States BC inventories for 2001, 2015, and 2020.

Scenario C: Base case projection with Clean Air Visibility Rule, Clean Air Mercury Rule, Clean Air Visibility Rule.
Scenario F: Compliance with PM$_{2.5}$ NAAQS: 15 μg/m^3 annual average, 35 μg/m^3 24-hour average.

Figure 4. United States OC inventories for 2001, 2015 and 2020.
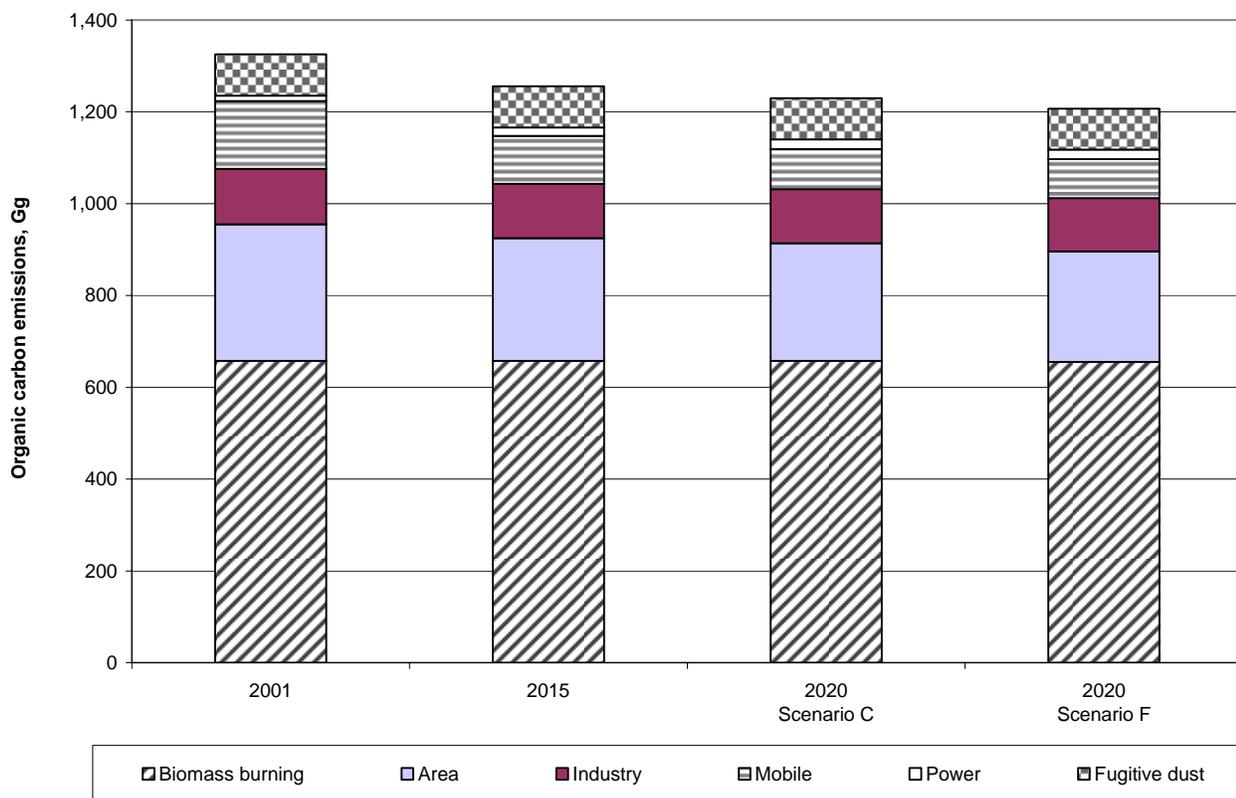
Scenario C: Base case projection with Clean Air Visibility Rule, Clean Air Mercury Rule, Clean Air Visibility Rule.
Scenario F: Compliance with PM$_{2.5}$ NAAQS: 15 µg/m^3 annual average, 35 µg/m^3 24-hour average.

Figure 5. Worldwide BC emissions estimates (Streets et al.).
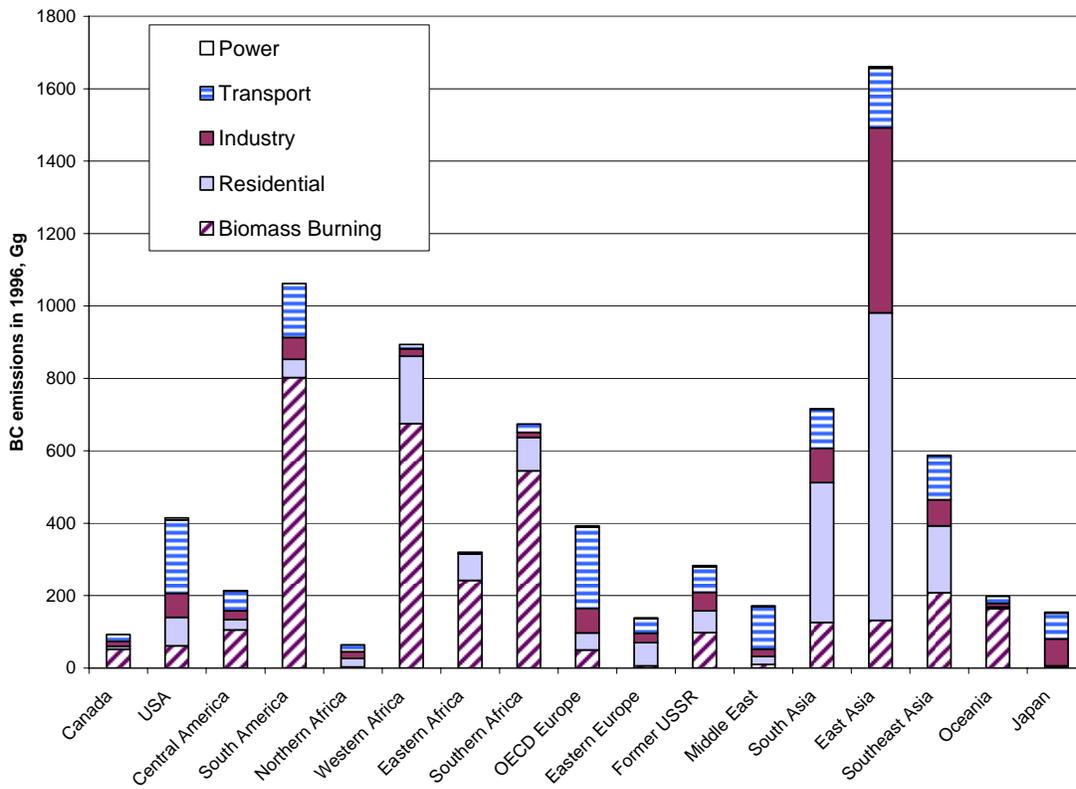
Figure 6. Worldwide OC emissions estimates (Streets et al.).