Black Carbon as a Short-Lived Climate Forcer: A Profile of Emission Sources and Co-Emitted Pollutants

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ABSTRACT:

A growing body of research in the past few years points to a potent source of human-induced climate warming: airborne aerosol particles, and especially the black carbon component of aerosols. The science behind black carbon and its role in climate warming is evolving but the evidence is mounting. Unlike CO_2, which can remain in the atmosphere for centuries, black carbon has a relatively short life span (and thus is known as a "Short-Lived Climate Forcer" (SLCF)). It remains in the air just a few weeks before deposition and removal. That's important, because if we can reduce black carbon emissions now, it would help blunt warming almost instantly and help offset some of the effects felt by the longer lived greenhouse gases like CO_2. Congress has mandated a Report from EPA that explores the science of black carbon and potential mitigation options, both domestically and globally, as it relates to climate impacts and air quality. The objective of this paper is to discuss SLCFs, and black carbon, in particular, and to summarize the status of the Black Carbon Report to Congress (RtC) with a focus on its description on the development and characterization of domestic and global black carbon emission inventories.

It should be noted that throughout this paper, "black carbon" and "elemental carbon" will be used interchangeably although many if not most of the measurements are EC. The RtC specifically has a detailed discussion of how one is different from the other, and the ramifications of nomenclature when it comes to climate- and health-based impacts

This paper presents black carbon inventories for the US for the year 2005 giving a segmented breakdown. The largest contributor to black carbon emissions in the US is mobile sources, specifically diesel vehicles/engines.

As a general note, the numbers in this paper are currently being reviewed and will be updated as necessary. They are still draft and may change in the future.

INTRODUCTION:

The most important long-term strategy for combating climate change is to slow and eventually reduce atmospheric build-up of CO_2 concentrations. Some investigators conclude that shorter-lived pollutants, including black carbon, methane and tropospheric ozone, may be collectively responsible for as much temperature impact as CO2, especially in eco-sensitive areas like the Arctic (Quinn et al., 2008). Addressing these pollutants has the advantage that emission reductions might be felt much more quickly than reductions of longer-lived gases.

Black carbon or "soot" is the shortest-lived of these pollutants, remaining in the atmosphere only days to perhaps weeks (Prather, 2005). It is comprised of small dark particles that remain after incomplete combustion of fossil fuel or biomass. Black carbon warms primarily by absorbing solar radiation, heating the atmosphere and contributing to overall global warming. In addition, in areas covered with snow and ice, such as the Arctic and Himalayan regions, the deposition of BC onto ice and snow darkens the surface, increasing the absorption of radiation (Flanner, 2007). This BC-ice albedo effect, leads to warming of the lower atmosphere and melting of snow and ice.

In addition to the climate impacts of black carbon emissions, there are also health impacts as well since black carbon is a contributor to PM2.5. Numerous scientific studies have linked levels of PM2.5 to a series of health problems, including: premature death in adults with heart and lung disease, heart attacks, low birth weight, aggravated asthma and other respiratory symptoms. BC, a primary pollutant, has also been associated with respiratory (Kulkarni, 2006) and cardiovascular (Peters et al., 2000) health effects.

Black Carbon is always co-emitted with a range of other pollutants during combustion of fossil fuel and biomass burning. Depending on the emission sources, BC may be a smaller or larger fraction of the total PM directly emitted. Current emission inventories do not always appropriately characterize BC fraction of the PM inventory, especially its spatial and temporal distribution.. When understanding the overall climate impacts of any control measure, it is very important to understand the location and type of the source and the characteristics of ALL the co-emitted pollutants. For example, BC's warming can be offset to varying degrees by cooling from reflective pollutants emitted by the same sources, especially organic carbon (OC), which is generally co-emitted from most of the sources that emit BC. A scientific investigator concluded that BC warms much more than OC cools per ton (Lesins et al., 2002) and, in an internal mixture, is expected to exert an increased warming effect (Chung et al., 2002; Ramanathan et al., 2008). Some sources (such as diesel engines) can have 80% of the PM2.5 being black carbon while others (such as biomass burning) can have a much smaller fraction such as 20% or lower. The ratio of OC to BC and emissions of other climate forcers varies greatly by source sector, and must be taken into account when designing mitigation strategies. From a climate perspective, it is advantageous to reduce EC from sources emitting significant EC that emit relatively smaller quantities of OC. It is also noteworthy that over reflective surfaces such as snow and ice, even OC (and sulfates) are "warming" because they are less reflective than the surface below. As a result, some sources and aerosol mixtures that might be cooling in other regions result in warming over the Arctic and Himalayan regions.

Given all of the mounting scientific evidence, Congress, as part of the October 2009 Interior Appropriations Bill, required EPA to conduct a comprehensive study on black carbon. Congress has required "not later than 18 months after the date of enactment of this Act, the Administrator, in consultation with other Federal agencies, shall carry out and submit to Congress the results of a study on domestic and international black carbon emissions that shall include

- an inventory of the major sources of black carbon,
- an assessment of the impacts of black carbon on global and regional climate,
- an assessment of potential metrics and approaches for quantifying the climatic effects of black carbon emissions (including its radiative forcing and warming effects) and comparing those effects to the effects of carbon dioxide and other greenhouse gases,
- an identification of the most cost-effective approaches to reduce black carbon emissions, and
- an analysis of the climatic effects and other environmental and public health benefits of those approaches."

This report is due in April 2011. While the Report to Congress (RtC) will have detailed sections to address each of the topics shown above and other related topics such as the definition of "black carbon," the objective of this paper is to focus on the draft emission inventory pieces of the RtC, including sources that are dominant emitters of black carbon domestically and globally, the spatial distribution of black carbon emissions as well as a look at co-emitted organic carbon, which reflects radiation and thus, generally, cools the planet.

BODY:

U.S. Black Carbon Emissions

The U.S. emissions inventory uses estimates of primary $PM_{2.5}$ emissions to derive information on direct emissions of carbonaceous particles, including EC and OC. Details on how $PM_{2.5}$ emissions are estimated and how they are chemically speciated (into black carbon, organic carbon, and other primary PM2.5 chemical constituents) is available on EPA's websites (EPA, 2005/2010). Therefore, all of the available emissions inventory information on light-absorbing carbon emissions in the U.S. is restricted to those source categories with sufficient $PM_{2.5}$ emissions estimates to support this estimation. Separate estimates of BC, Brown Carbon (BrC), and total Light-Absorbing Carbon ("LAC") — the categorization most relevant to climate — are not currently available and are beyond the scope of this paper. The U.S. emits approximately 6% of the total global EC emissions each year, or about 540,000 tons out of more than 9 million tons globally, making it the 7th largest emitter. The majority of "current" U.S. emissions come from mobile sources and open biomass burning. From 1990 to 2005, EC emissions in the U.S. have declined by about 30%, and are expected to decline by an additional 80% by 2030 compared to 2005 levels, largely due to PM regulations on emissions from mobile sources, specifically diesel vehicles/engines.

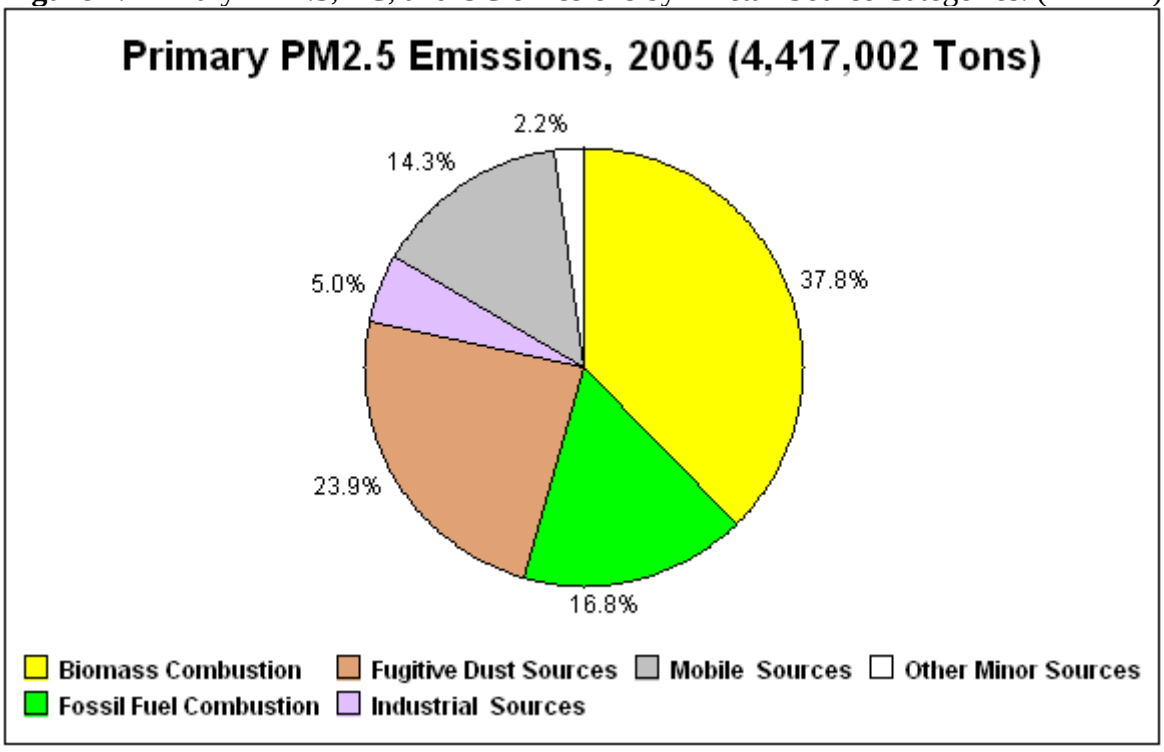
Total primary $PM_{2.5}$ emissions in the U.S. in 2005 are estimated to be about 4,417,002 tons (or about 4,008 Gg), of which approximately 536,098 tons (12%) is EC and about 1,108,560 tons (25%) is primary OC. Thus at a national level, there is approximately twice as much OC emitted

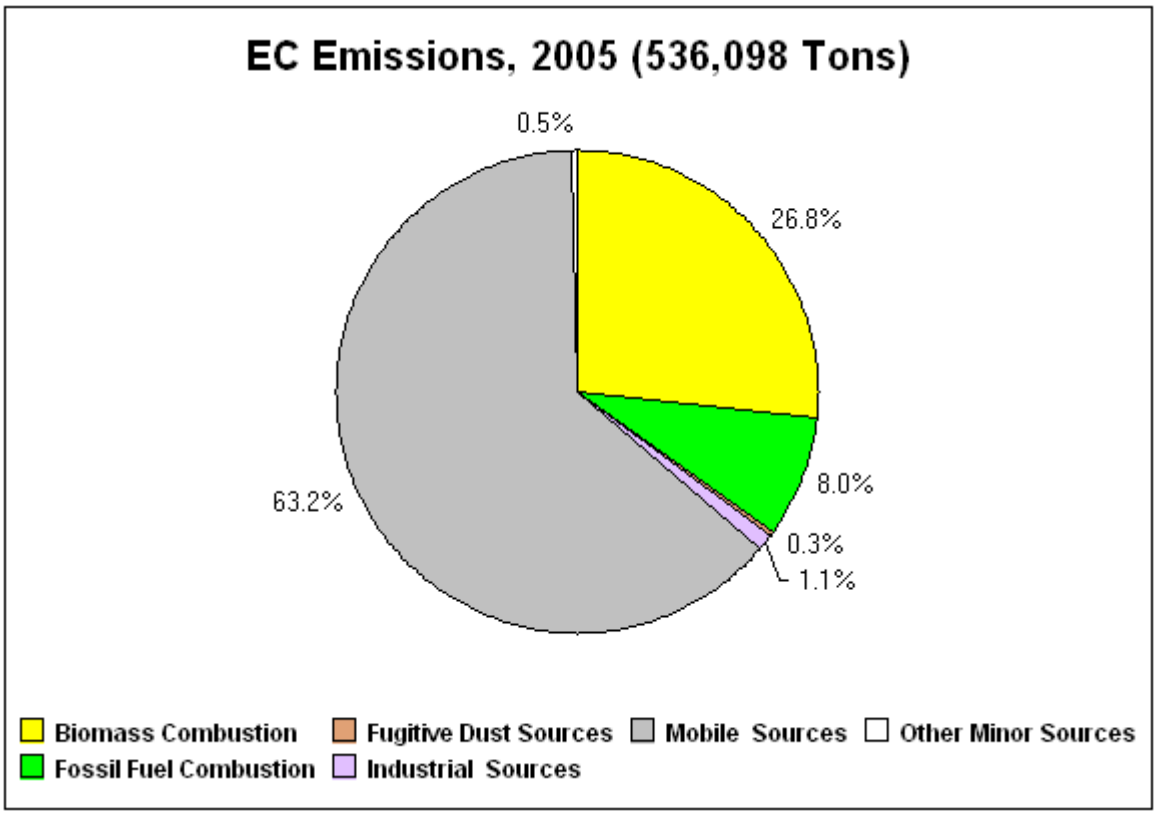
from domestic sources as EC. Figure 1 displays the percentage of total U.S. primary $PM_{2.5}$, EC, and OC emissions for six meta source categories: biomass combustion, fossil fuel combustion (which includes natural gas, coal and oil from residential, industrial, commercial, and electric generation), fugitive dust sources, industrial sources, mobile sources, and other minor sources. Table 1 shows the actual tons per year of EC, OC, and direct $PM_{2.5}$ for these source categories, as well as some key emissions ratios

The EC pie chart in Figure 1 clearly shows mobile sources (which include exhaust emissions plus tire and brake wear) to be the dominant contributor of the total EC emissions in the US in 2005. Mobile sources contribute 63% of the total EC emissions, followed by biomass burning (27%), and fossil fuel combustion (8%). All other categories are about 1% or less.

As shown by the ratios in Table 1 (EC/OC and EC/$PM_{2.5}$), the composition of primary $PM_{2.5}$ emissions varies significantly between source categories, and such differences have important implications for climate. For example, mobile sources emit significantly more EC than OC, while the opposite is true for biomass burning. In addition, the data indicate that for some source categories, EC and OC together make up less than 50% of total $PM_{2.5}$ emissions, indicating that there are significant amounts of other co-pollutants in the emissions mixture.

Figure 1. Primary PM2.5, EC, and OC emissions by "Meta" Source Categories. (DRAFT)




EC Emissions, 2005 (536,098 Tons)
0.5%
26.8%
8.0%
0.3%
1.1%
63.2%
Biomass Combustion
Fugitive Dust Sources
Mobile Sources
Other Minor Sources
Fossil Fuel Combustion
Industrial Sources

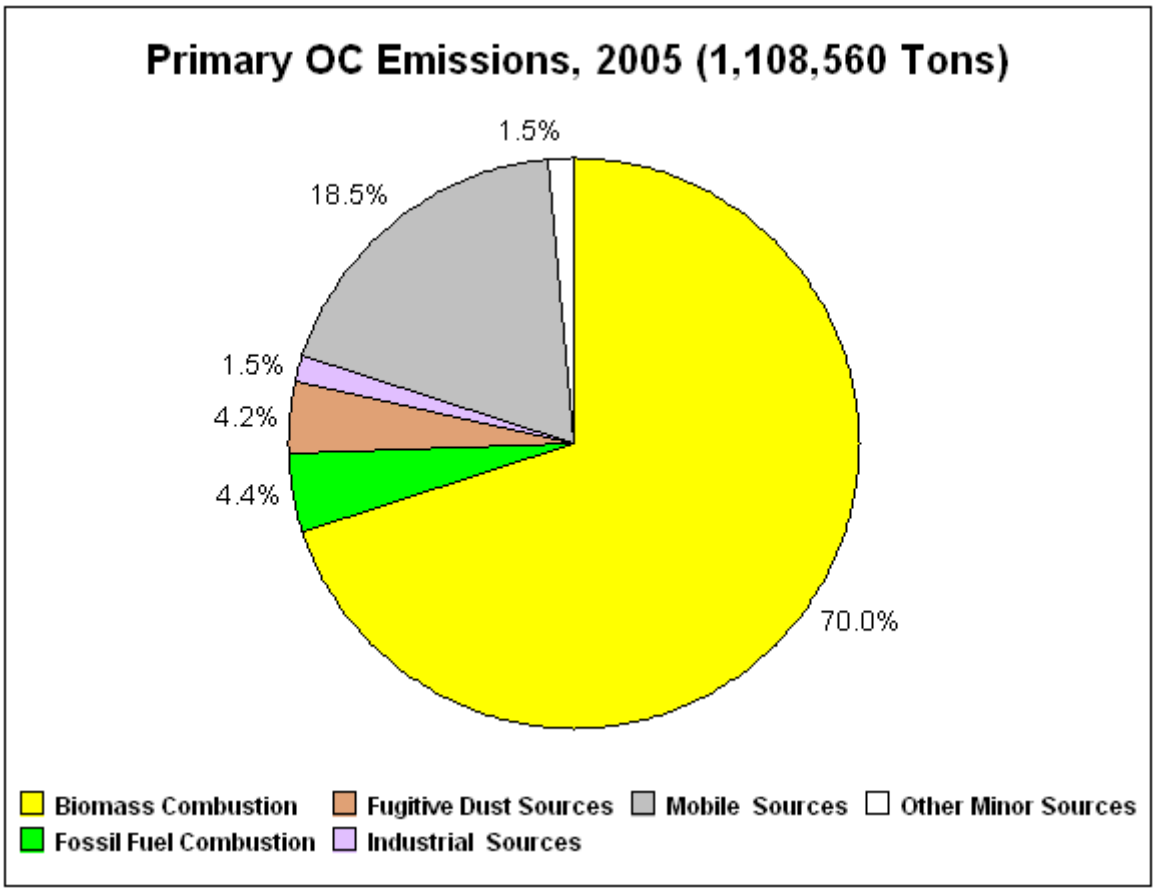

Primary OC Emissions, 2005 (1,108,560 Tons)
1.5%
18.5%
1.5%
4.2%
4.4%
70.0%
Biomass Combustion
Fugitive Dust Sources
Mobile Sources
Other Minor Sources
Fossil Fuel Combustion
Industrial Sources

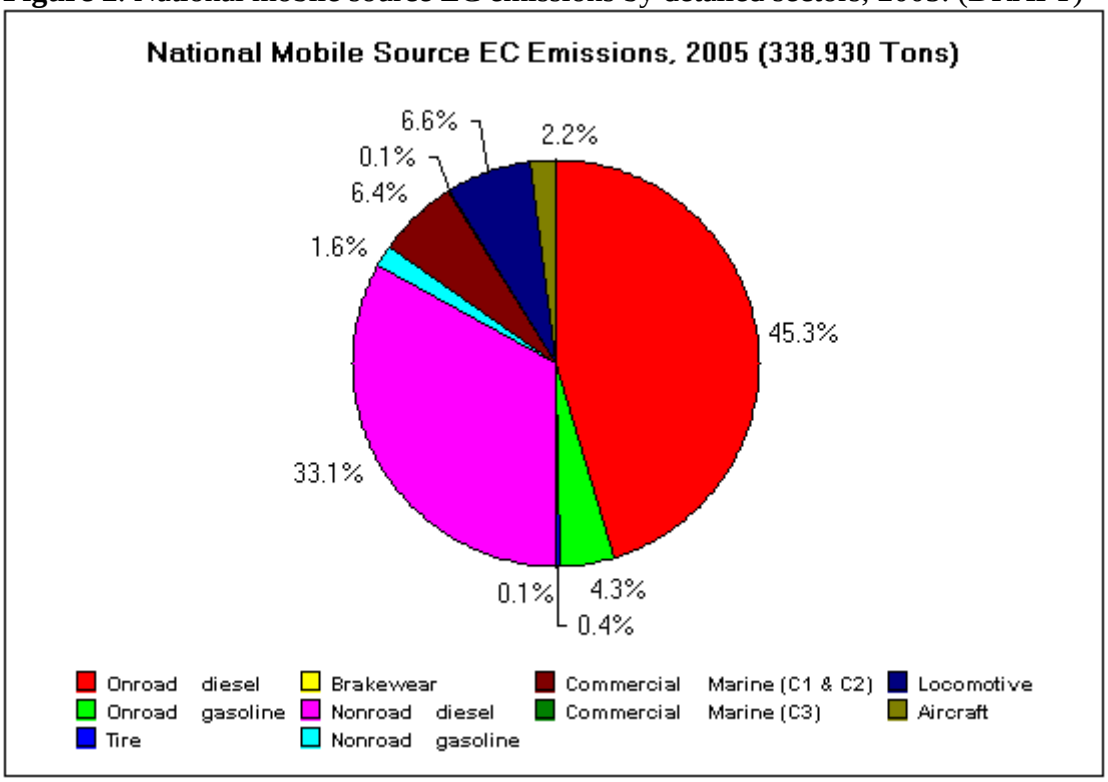
Table 1. 2005 emissions and ratios of emissions by "Meta" source category. (DRAFT)

"Mega" Source Category	$PM_{2.5}$	EC	OC	EC/OC	$EC/PM_{2.5}$
Biomass Combustion	1,668,395	143,902	775,724	0.19	0.09
Fossil Fuel Combustion	740,335	43,049	48,888	0.88	0.06
Fugitive Dust Sources	1,056,877	1,609	46,221	0.03	0.00
Industrial Sources	219,460	6,083	16,232	0.37	0.03
Mobile Sources	633,222	338,930	204,855	1.65	0.54
Other Minor Sources	98,714	2,525	16,640	0.15	0.03
	4,417,002	**536,098**	**1,108,560**		
Totals:	**(4,007 Gg)**	**(486 Gg)**	**(1,006Gg)**	**0.48**	**0.12**

Emissions from Mobile Sources and Biomass Burning Categories

As noted above, mobile sources accounts for 63% of the total US EC emissions in 2005. Within this category, emissions from diesels (both nonroad and onroad) dominate, accounting for about 92% of EC. Gasoline vehicles/engines are responsible for the remaining 8% of EC emissions from the mobile source category. The pie chart in Figure 2 shows this more detailed breakout of mobile source EC emissions. In general, diesel $PM_{2.5}$ from vehicles/engines without diesel particulate filters (e.g., pre-2007 diesel trucks) consists of about 70-80% EC and about 20% OC. Gasoline $PM_{2.5}$, in contrast, consists of about 20% EC with the remainder being mostly OC. Diesel PM is thus unique in having a very high ratio of EC to OC. Mobile sources are also significant emitters of polycyclic aromatic hydrocarbons (PAHs), which are precursors to EC and light-absorbing in nature. Total PAH emissions from mobile sources in 2005 are estimated at about 4,550 tons. While this amount is significantly smaller than mobile source EC and OC emissions, it represents about 21% of the total PAH inventory in 2005. Gasoline engines contribute significantly more to PAH emissions than diesel engines (approximately 89% vs. 8%), which is exactly the opposite of how the EC emissions are distributed across gas and diesel engines. The organic carbon (OC) associated with PM consists of many higher molecular weight compounds with PAH compounds themselves constituting a small fraction of the OC by mass. The major point is that diesels versus other sources emit relatively little OC compared to EC. The light absorbing capability (LAC) of the specific compounds and the resultant mixture emitted in diesel or gasoline exhaust is not known.

Figure 2. National mobile source EC emissions by detailed sectors, 2005. (DRAFT)



The general category of biomass combustion in the 2005 inventory includes both open burning (agricultural burning, prescribed burning, slash burning, and wildfires) and other categories such as charbroiling, potato-deep frying, meat frying, residential wood combustion, and wood-fired boilers. **Figure** 1 and Table 1 indicates that this group of sources contributes about 26% of the total domestic EC emissions, second only to mobile sources in terms of contribution to total domestic EC. However, unlike for mobile sources, EC:OC ratios for this source category are generally less than one, indicating a predominance of OC emissions (about 80% on average). Major "open burning" categories frequently show higher EC/OC ratios than biofuel burning categories (such as wood fired boilers and residential wood combustion). However, literature suggests (Chakrabarty et al., 2010; Hecobian et al., 2010) that, in general, OC from biomass burning may contain more light-absorbing organic carbon compared to other sources. Total PAH emissions from certain types of Residential Wood Combustion sources, such as fireplaces, are also very high. Quantification of exactly how much of the inventoried OC is LAC is not known at this time.

US EC Emissions North of the 40th Parallel

Emissions in the northern hemisphere affect most the deposition of BC onto snow and ice in the Arctic, thereby causing warming and melting of the ice. Due to these effects, and because BC in the atmosphere causes more warming when it is present over reflective surfaces such as ice, BC has impacts in the Arctic and over snow and ice covered areas. More recently, literature has indicated that emissions north of the 40th parallel particularly effect Arctic impacts. The 40th parallel north is a circle of latitude that is 40 degrees north of the Earth's equatorial plane.

Globally, it crosses Europe, the Mediterranean Sea, Asia, the Pacific Ocean, North America, and the Atlantic Ocean. In the U.S., the 40th parallel approximately bisects New York City in the East and San Francisco in the West.

For the national inventories developed and described above, totals can be estimated for the counties that fall north of the 40th parallel in the U.S. In 2005, out of a total of 3,141 counties in the U.S., about 1,180 counties (or about 38% of the counties) are north of the 40th parallel. EC emissions have been summed for these 1,180 counties for at the "meta" source categories shown earlier and displayed below in Table 2**.**

Table Error! No text of specified style in document**.** A comparison of national to "north of 40th parallel" EC emissions in 2005 (in tons). (DRAFT)

	Total US EC Emissions	EC Emissions Estimated North of 40th Parallel	Fraction of EC Emissions above 40th Parallel
Biomass Combustion	143,902	39,370	0.27
Fossil Fuel Combustion	43,049	2,794	0.06
Fugitive Dust Sources	1,609	483	0.30
Industrial Sources	6,083	1,573	0.26
Mobile Sources	338,930	128,142	0.38
Other Minor Sources	2,525	755	0.30
Totals:	**536,098**	**173,116**	**0.32**

With the exception of the Fossil Fuel Combustion category, most of the other categories show that the amount of EC emissions north of the 40th parallel are proportional to the number of counties that are north of the 40th parallel in the U.S. Figure 3 shows the fractional contribution of sources to emissions north of the 40th parallel in the U.S. A comparison of Figure 3 to Figure 1 shows that mobile sources contribute even more when considering emissions north of 40, due to a proportionally lower amount of EC emissions from other source categories north of 40.

Global Black Carbon Emissions

All of the information presented in the RtC on global emissions draws extensively on available literature. The most widely used and cited global black carbon inventories are those developed by Bond and Streets, and the reader is referred to those references for additional details (Bond et al., 2004; Streets et al., 2004) in the summaries presented below.

Figure 4 shows the amount of EC estimated to be emitted by each of the world regions. Asia accounts for nearly 37% of total global EC emissions, Africa for about 25%, and Latin America for about 16%. Figure 6 shows that, unlike in the U.S., where transportation sources dominate the EC inventory, global BC totals are dominated by open biomass burning and residential cooking sources. However, it is noteworthy that while places like OECD Europe, the Middle East, and Japan have very low amounts of BC emissions, transportation is the dominant contributor to those emissions, like in the U.S. Japan also has a significant contribution from industrial sources.

Figure 3. US EC Emissions north of the 40th Parallel, 2005. (DRAFT)

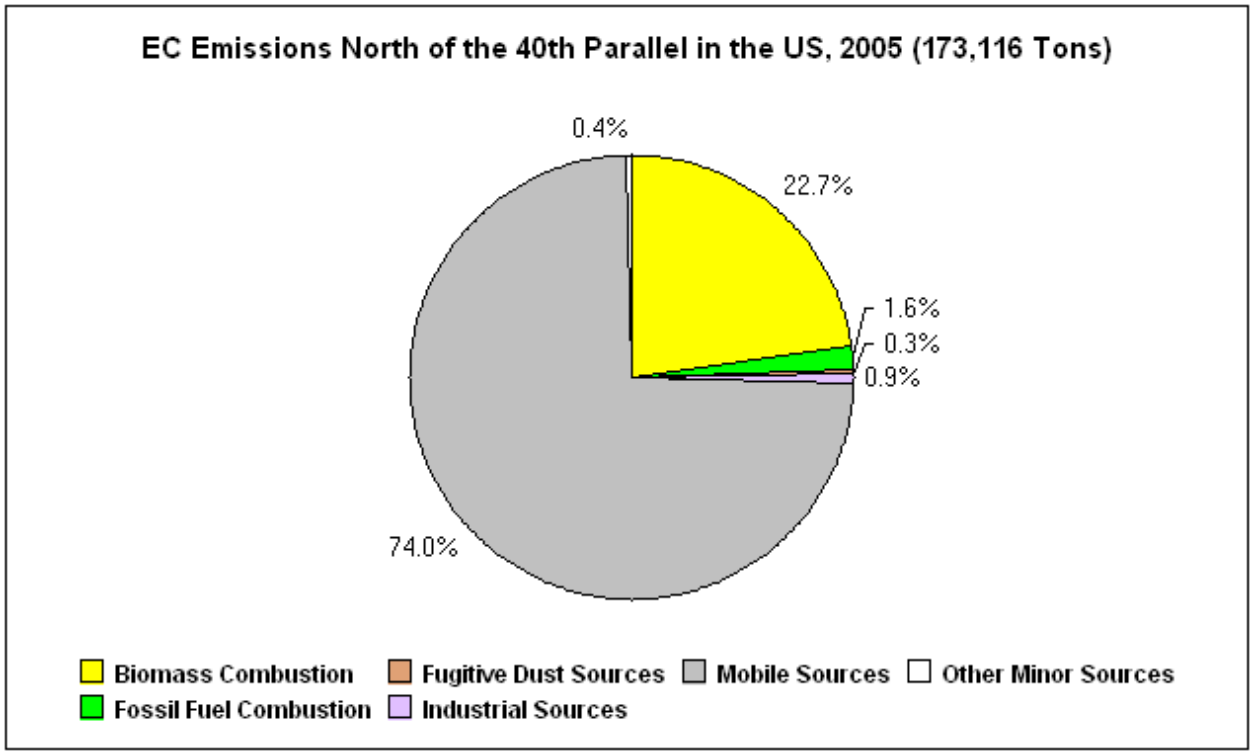


Figure 4. BC Emissions by world region. (DRAFT)

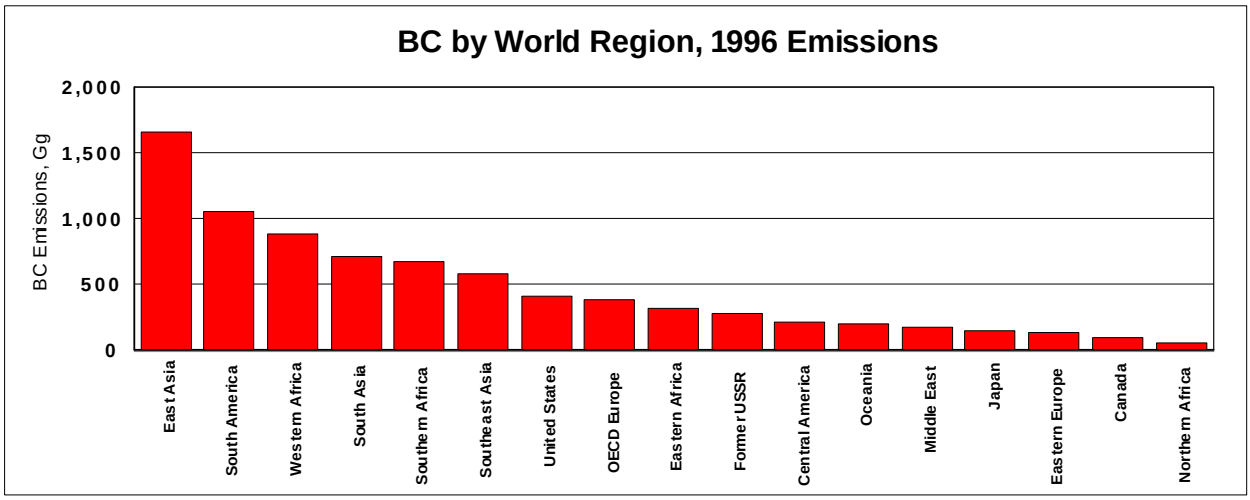
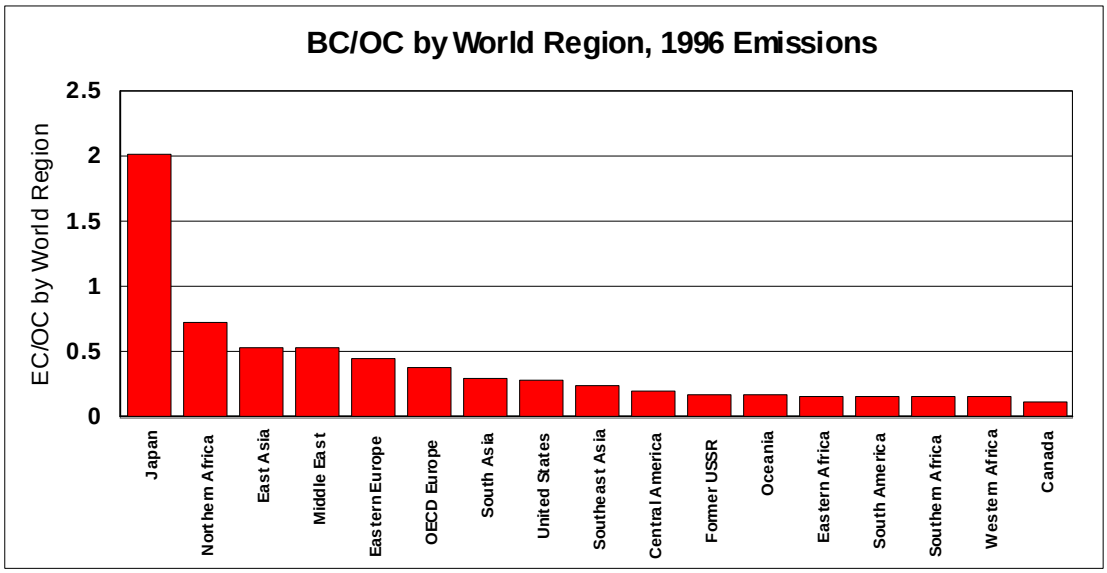


Figure 5. BC:OC ratios by world regions. (DRAFT)



Another interesting way to look at BC/OC ratios is by world region, which is done in **Figure 5**. Here, total BC/OC ratios are plotted by world region. It is interesting to note that only in Japan, where the total BC emissions is very small, is there more estimated BC emissions than OC emissions. This is likely due to the fact that there is very little open burning or emissions from the residential sector in Japan. As with US inventories, global inventories show that transportation and industrial sources seem to matter the most for global BC emissions north of the 40th parallel, which globally bisects New York City and Beijing. These patterns have important implications fro assessing the contribution of source regions to snow melt in the Arctic as well as total BC-related forcing in the Northern Hemisphere.

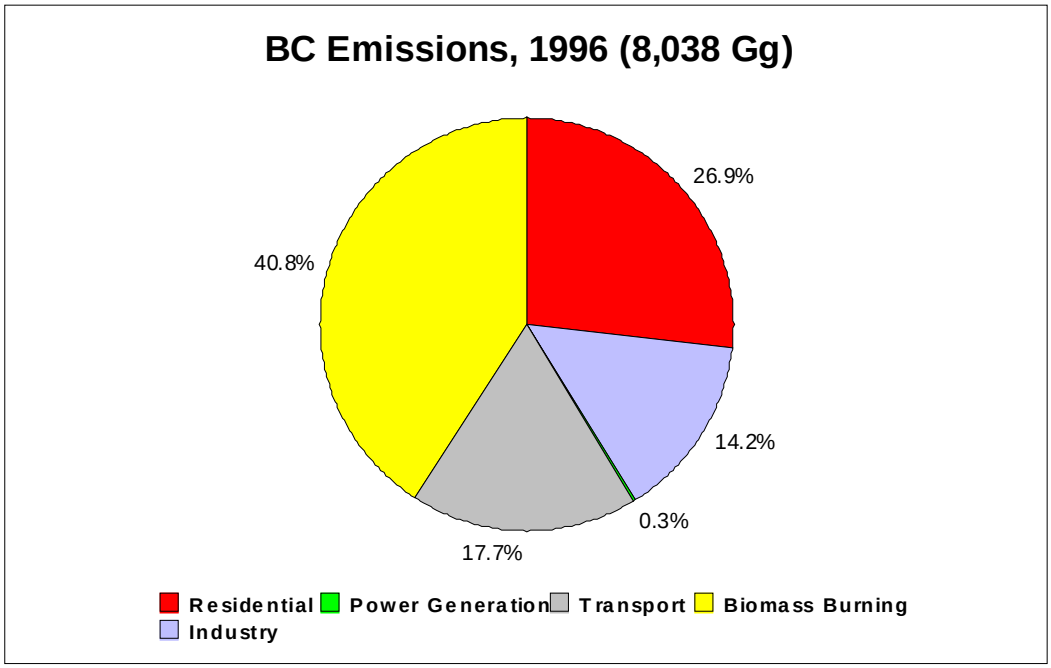
Conclusions

Like CO_2, SLCFs contribute to climate change, but unlike CO_2, they don't stay in the atmosphere for hundreds of years. Many SLCFs last for just a few hours to a few days. The most important short-lived forcers include black carbon, methane, and tropospheric ozone. The product of incomplete combustion in engines and cooking fires, black carbon is essentially heat-absorbing soot; it is harmful to human health, heats the air around it, and, when deposited on snow and ice, accelerates the melting process. Black carbon is believed to contribute significantly to the rapid melting of the Arctic and Himalayan glaciers.

Emissions of black carbon, to date, has received relatively little attention in US inventory development. Due to black carbon's role as an SLCF, and more recent efforts to understand sources of global warming, more attention has been given to better understanding domestic emissions of black and organic carbon emissions. In the US, for the year 2005, mobile sources (and especially diesel sources) are seen to be the predominant contributor to the black carbon

inventory. Biomass burning is the second biggest source category of EC even though there are a lot of co-emitted organic carbon emissions. In the future, as mobile source diesel regulations reduce EC emissions from those sources, other sources, like biomass burning, will represent a larger portion of the EC emissions in the United States then.

Figure 6. Global BC emissions by source category. (DRAFT)



Globally, residential sources and biomass burning are the major sources of black carbon. In contrast to expected mobile source reductions in EC in the United States in the future, projected global BC trends seem to indicate an increase in BC emissions from the transport sector for developing countries. Prediction of future global BC emissions is a very uncertain area and needs more research.

Future research efforts must also focus on improving the most uncertain parts of the current black carbon inventories, including those sources that are expected to be a major player in the future.

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Disclaimer

All of the thoughts and comments expressed in this paper are those of the authors', and should not necessarily be considered as US EPA views.