II. Black Carbon, Atmospheric Brown Clouds and Greenhouse Effect: Background

1. **Origin of Black Carbon:** Black carbon (BC), a major component of soot, is emitted through cooking with solid fuels (coal, wood, cow dung and crop residues), by biomass burning and fossil fuel combustion (solid coal or combustion of diesel fuel). These sources also emit organic aerosols and the mix of BC and organics is popularly referred to as soot. In the atmosphere, BC is mixed (Moffet and Prather, 2009) with other particles such as sulfates, nitrates, dust and other pollutants. A single particle can contain a mixture of BC and one or more of these other chemical species, in which case, the particle is referred to as internally mixed. BC can also exist as a separate particle, coexisting with other aerosols side by side, and in this instance BC is referred to as externally mixed. BC and these other particles remain in the atmosphere for several days to few weeks, during which they can be transported thousands of kilometers away from their source.

2. **Atmospheric Brown Clouds and BC Hotspots:** Such vast plumes of pollution aerosols containing BC are sometimes referred to as Atmospheric Brown Clouds (ABCs). Hot spots of ABCs with large concentrations of BC as well as other man-made aerosols such as sulfates, organics, nitrates and others have been identified by synthesizing satellite observations with ground base and aircraft observations: The following regions fall under the hot spot category: (1) east Asia (eastern China, Thailand, Vietnam and Cambodia), (2) Indo-Gangetic Plains in south Asia (the northwest to northeast region extending from eastern Pakistan, across India to Bangladesh and Myanmar), (3) Indonesian region, (4) southern Africa extending southward from sub-Saharan Africa into Angola and Zambia and Zimbabwe, and (5) the Amazon basin in South America. However, ABCs are a world wide problem, including developed nations. For example, per capita emissions of black carbon in US is comparable to that in E. Asia.

3. **Policy Implications of the Regional Nature of BC Effects:** The regionally concentrated nature of BC concentrations is a potential advantage for policy makers. While one of BC’s major effect is on global and regional climate change, the immediate effect of BC reductions will be felt locally (wherever mitigation actions are taken) as an improvement in air quality and visibility accompanied by mitigation of the impacts of BC on human health, agriculture, and local precipitation.
4. **Black and Brown Carbon Terminology:** Black carbon is not a ‘greenhouse gas’. It is a particle and it is the strongest absorber of solar radiation in the atmosphere. It also absorbs and emits infra red or heat radiation and contributes to the greenhouse effect. However, the latter effect is much smaller than the solar warming effect. The term "black carbon" is not rigorously defined. Climate models have largely assumed black carbon is the same as elemental carbon. All man-made sources of black carbon also emit hundreds of organic aerosols and gases (which later become aerosols). Most climate models treat these organic aerosols as purely reflecting (and not absorbing) aerosols, i.e., they have a cooling effect. Recent experimental work has given compelling evidence that some of these organic aerosols also absorb sunlight in UV and visible wavelengths and thus enhance the warming effects of BC (Magi, 2009). These absorbing organic aerosols are popularly referred to as ‘Brown Carbon’ (Andreae and Gelencser, 2007). For the purpose of this report, absorption of solar radiation by BC and brown carbon are treated together, since they occur in the same wavelength region.

5. **How Does BC contribute to Global Climate Change?**

BC warms the climate in at least 5 different ways (Jacobson, 2010): i) It traps (absorbs) solar radiation in the atmosphere, directly heats the air and thus contributes to climate warming. There is now strong experimental evidence that internally mixed BC absorbs significantly more solar radiation than externally mixed BC (Moffet and Prather, 2009). ii) When BC is deposited on sea ice, snow packs and glaciers, it darkens the snow and ice surfaces, enhances absorption of sunlight and contributes to melting of snow and ice. iii) BC also absorbs and emits heat radiation (Infra red radiation) and adds to the atmospheric greenhouse effect. This effect, although much less than the warming from the solar heating effect, can be important in the arctic and during nights. iv) BC gets into cloud droplets (by nucleation or scavenging) and enhances absorption of solar radiation by drops. v) The day time warming of the lower layers of the atmosphere, first few kilometers, by BC can suppress the relative humidity and evaporate low level clouds, which will allow more solar radiation to reach the ground and amplify the warming.

BC has also a potential cooling effect. When aged and mixed with other aerosols such as sulfates and oxidized organics, BC can also be efficient cloud nuclei. Formation of new cloud drops through BC nucleation, in low level stratus and cumulus clouds, can make the clouds brighter and shield the surface from solar radiation and cause surface cooling.
6. **Recent Estimates of BC Forcing:** Compared with the climate forcing due to carbon dioxide which has been studied intensely for several decades, the science of BC and its climate effects is relatively new. Our understanding of the impact of black carbon (BC) aerosols has undergone major improvements and revisions during the last 10 years. The major contributing factors are listed below: i) Experimental findings from field and aircraft observations (e.g. INDOEX, ACE-Asia and others) in Asia, Africa, Arctic, Europe, and N America. ii) new satellite observations [e.g., MODIS, CALIPSO]. iii) surface observatories such as the IMPROVE network in USA; worldwide AERONET network by NASA and Atmospheric Brown Cloud observatories for the Indo-Asian-Pacific region by UNEP, NOAA and others; iv) Scripps’ Unmanned Aircraft Observing systems funded by NSF and NOAA; v) UCSD’s Time of flight mass spectrometer single particle measurements; vi) observationally constrained emission inventories; vii) aerosol chemical-transport models developed at Stanford, Caltech, NASA, NOAA and NCAR laboratories. We now have direct UAV measurements for the large enhancement of atmospheric solar heating by BC (Ramanathan et al, 2007b).

Global averaged estimates for the radiative heating (or radiative forcing) of the surface-atmosphere system by BC as of now (year 2005) is in the range of 0.3 Wm\(^{-2}\) to 0.9 Wm\(^{-2}\) for the direct solar absorption by atmospheric BC; 0.05(±50%) Wm\(^{-2}\) for the BC solar heating of ice and snow; 0.03 (±50%) Wm\(^{-2}\) for the greenhouse effect of BC. The combination of these three warming effects of BC is referred to as direct radiative forcing. The direct radiative forcing of BC (0.4 to 1 Wm\(^{-2}\)) is about 20% to 60% of the pre-industrial to year 2005, CO\(_2\) greenhouse forcing. We have adopted Forster et al’s (2007) IPCC estimates of 1.66 Wm\(^{-2}\) for the pre-industrial to year 2005, CO\(_2\), radiative forcing. The heating due to BC in clouds (items iv and v above) and the cooling effect due to BC nucleation of cloud drops are not yet firmly established.

7. **BC forcing in the context of the Greenhouse Blanket:** The greenhouse gases (water vapor, carbon dioxide, ozone and others) surround the planet like a blanket. A blanket keeps us warm on a cold winter night by retaining (or trapping) our body heat. Similarly, the GHGs retain much of the infrared radiation (or heat radiation) given off by the surface and the atmosphere (including clouds) within the planet. The energy retained (thickness of the blanket) by the atmosphere has been determined from satellite radiation budget data and other correlative surface temperature data, to be about 163 Wm\(^{-2}\) (5%) for the 1985 to 1989 period. H\(_2\)O in the form of...
vapor, cloud drops and ice crystals provide about 2/3 of the total effect, CO\textsubscript{2} about 20% and the balance is due to other GHGs including ozone, methane, nitrous oxide.

Since pre-industrial times, the increase in GHGs (CO\textsubscript{2}; methane; CFCs; nitrous oxide; and others) by human activities has thickened the blanket and retained more energy. CO\textsubscript{2} alone has increased from 280 ppm to 387 ppm. This increase as well as increase in the other anthropogenic GHGs has retained additional energy of about 3 (15%) Wm\textsuperscript{-2} or thickened the blanket by about 1.8% (3 Wm\textsuperscript{-2} out of 163 Wm\textsuperscript{-2}). The 1.8% may seem small, but it should be noted that, the increase in global mean surface temperature from glacial to the current interglacial period is about 5° K (5°C) and in the absolute Kelvin temperature scale, 5°C is only 1.7 % of the global mean temperature of 289° K (15.5°C). Reverting back to the effect of BC, we can think of BC as smoke in the blanket that traps sunlight (0.3 to 0.9 Wm\textsuperscript{-2}) directly into the blanket and into snow and sea ice (0.05 Wm\textsuperscript{-2}) as well as retaining infra-red radiation (0.03 Wm\textsuperscript{-2}) in the blanket.

8. Role of Non-CO\textsubscript{2} Climate Warmers in Mitigation: BC is but one of several non-CO\textsubscript{2} climate warmers. Human activities have added several other GHGs. These include, methane, nitrous oxide, halocarbons and tropospheric ozone. Compared with the centuries to millennial time scales of CO\textsubscript{2}, the life times of several non-CO\textsubscript{2} warmers are much shorter. BC's life time is less than a few weeks; Ozone molecule’s life time is few months, methane less than 15 years and halocarbons range from a year to a decade. Because of their shorter life times, reductions in the emissions of short lived warmers will lead to quicker reductions in the concentrations and their radiative warming of climate. In fact, such a mitigation action, will also help the science of climate change test its model predictions of cause and effect. Several studies have estimated that, with continuation of current trends in GHGs emissions, there is more than a 50% probability of surpassing the 2°C warming threshold during this century. A IIASA study estimates that with currently available technology and stringent implementation of current air pollution laws, it is possible to achieve about 30% reductions in methane, ozone, HFCs and more than 30% for BC. Such reductions in non-CO\textsubscript{2} warmers, can postpone the time for crossing the 2°C threshold by few decades or more.

Non-CO\textsubscript{2} Climate Warmers

<table>
<thead>
<tr>
<th>Greenhouse Gases</th>
<th>Contribution to 2005 forcing relative to CO\textsubscript{2} (1.66 Wm\textsuperscript{-2})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ozone (troposphere)</td>
<td>20%</td>
</tr>
<tr>
<td>Methane</td>
<td>30%</td>
</tr>
<tr>
<td>Halocarbons</td>
<td>20%</td>
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</tbody>
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<table>
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<tr>
<th>Particles (Aerosols)</th>
<th>Contribution to 2005 forcing relative to CO\textsubscript{2} (1.66 Wm\textsuperscript{-2})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Black Carbon (soot/smoke)</td>
<td>27% to 55%*</td>
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</table>

Total Non-CO\textsubscript{2}: 97% to 125%

All numbers except the red are IPCC values; Long lived N\textsubscript{2}O not included

* Ramanathan & Carmichael; 2008

Deposition of BC on Snow in California: Direct Measurements

Measured Black Carbon Deposition on the Sierra Nevada Snow Pack
© Hadley, C. Corrigan, T. W. Kirchstetter, C. S. Davis, V. Ramanathan

California CEC Project

<table>
<thead>
<tr>
<th>Year</th>
<th>Location</th>
<th>BC (\text{pg} m\textsuperscript{-2})</th>
</tr>
</thead>
<tbody>
<tr>
<td>2008</td>
<td>Lake</td>
<td>5.6 (± 0.3)</td>
</tr>
<tr>
<td></td>
<td>CR</td>
<td>3.1 (± 1.3)</td>
</tr>
<tr>
<td></td>
<td>Snow</td>
<td>6.7 (± 0.1)</td>
</tr>
</tbody>
</table>

308 BC (pg m\textsuperscript{-2})
9. **Regional Climate Effects of BC**: The regional effects of BC are estimated to be particularly large over Asia, Africa and the Arctic. In these regions its effects include alteration of surface and atmospheric temperatures, disrupting monsoon circulation and rainfall patterns (Menon et al, 2002; Ramanathan et al, 2005; Lau et al, 2008). The interaction of the regional climate effects of greenhouse gases and ABCs deserve more attention. For example, a recent study (Ramanathan et al, 2007b) employing unmanned aerial vehicles suggests that BC enhances atmospheric solar heating by as much as 50%. When these data are combined with CALIPSO and other satellite data over S, SE Asia and the Indian Ocean and employed in a climate model, the simulations suggest that the elevated atmospheric warming over the S and SE Asian region, (including the elevated Himalayan regions) by ABCs is as much as that due to the greenhouse warming. Thus the atmospheric solar heating by BC may be intensifying the effects of greenhouse gases on the Himalayan-Tibetan glacier region. Climate model studies also suggest that fossil fuel and biofuel BC emissions in Asia and Europe induce as much springtime snow cover loss over Eurasia as anthropogenic carbon dioxide, a consequence of the darkening of the snow by deposition of snow and strong snow-albedo feedback. (Flanner et al, 2009). We now have direct measurements of efficient removal of BC by snow over the Sierras in California.

10. **Effects of BC and ABCs on Regional Water Budget**: Reverting to the general effects of all aerosols (and not just BC), ABCs enhance scattering and absorption of solar radiation and also produce brighter clouds (IPCC, 2007) that are less efficient at releasing precipitation (Rosenfeld et al, 2000). These in turn lead to large reductions in the amount of solar radiation reaching the surface (also known as dimming), a corresponding increase in atmospheric solar heating, changes in atmospheric thermal structure, surface cooling, atmospheric warming, alterations of north-south and land-ocean contrast in surface temperatures, disruption of regional circulation systems such as the monsoons, suppression of rainfall, and less efficient removal of pollutants (Ramanathan et al, 2001b, 2005, 2007a; Menon et al, 2002). Together the aerosol radiation and microphysical effects can lead to a weaker hydrological cycle and drying of the planet. This connects aerosols directly to availability of fresh water, a major environmental issue of the 21st century (Ramanathan et al, 2001b). For example, the Sahelian drought during the last century is attributed by some models to the north-south asymmetry in aerosol forcing (Rotstayn and Lohman, 2002). In addition, new coupled-ocean atmosphere model studies suggest that aerosol radiative forcing may be
the major source for some of the observed drying of the land regions of the planet (e.g. India & northern China) during the last 50 years (Ramanathan et al, 2005 and Meehl et al, 2008). Regionally aerosol induced radiative changes (forcing) are an order of magnitude larger than that of the greenhouse gases, but because of the global nature of the greenhouse forcing, its global climate effects are still more important. However there is one important distinction to be made. While the warming due to the greenhouse gases is projected to increase global average rainfall, the large reduction in surface solar radiation due to absorbing aerosols would offset it.

11. Challenges and Opportunities for Mitigation:

BC’s warming effect presents an opportunity to reduce projected warming in the short term (as also suggested by others, e.g. Hansen and Sato, 2001; Jacobson, 2002; Bond and Sun, 2005). My thesis is that, BC reductions have the potential to forestall the onset of the so-called dangerous climate change. For example, a reduction of BC emissions by about 50%, may reduce the radiative forcing by about 0.2 Wm\(^{-2}\) to 0.5 Wm\(^{-2}\). In comparison, if CO\(_2\) continues to increase at the current rate of increase, it will add about 0.2 to 0.3 Wm\(^{-2}\) per decade. Thus a drastic reduction in BC has the potential to offset CO\(_2\)-induced warming for a decade or two. Effectively, BC reduction may provide a possible mechanism for buying time to develop and implement effective steps for reducing CO\(_2\) emissions. The following issues need to be factored in further consideration of this proposal:

i) The life time of BC is of the order of days to several weeks, depending on the location. Thus the BC concentration and its global heating will decrease almost immediately after reduction of its emission;
ii) Inhalation of soot is a major public health issue. For example, in India, alone it is estimated inhalation of indoor smoke is responsible for over 400,000 deaths annually (mostly among women and children; Smith, 2000). Air pollution related fatalities for Asia is estimated (Pachauri and Sridharan, 1998) to be over one million (indoor smoke inhalation and outdoor brown clouds). Thus reduction of BC emissions may be warranted from public health considerations alone.
iii) The developed nations have reduced their BC emissions from fossil fuel sources significantly since the 1960s. Thus the technology exists for a drastic reduction of fossil fuel related BC. With respect to bio-

fuel cooking, it can be reduced if not eliminated, by providing alternate cooking methods in rural areas in Asia and Africa. But we need to conduct a careful and well documented scientific study of the impact of BC reduction on radiative forcing and its cost effectiveness. Towards this goal, this author along with a team of NGOs, public health experts and alternate energy experts, has proposed Project Surya (http://www-ra
manathan.ucsd.edu/ProjectSurya.html), that will adopt a large rural area of about 50,000 population, in India, and provide alternate cooking with biogas plants, smoke free cookers and solar cookers. The objective of this experiment is to estimate from observations the warming potential of BC. Surya will also assess the impact of BC reduction on human health and the cost of reducing BC emissions from biofuels. Results from this pilot experiment will be used to scale up similar efforts throughout the subcontinent.
iv) Long range transport of BC is an important factor for policy discussion of BC mitigation. For example, studies have shown that transport of BC from E. Asia across the pacific is a major source of BC above one km in altitude over California (Hadley et al, 2007). Likewise, BC from N. America and Europe deposits on snow and sea ice in the Arctic. BC from S. Asia and E. Asia surrounds the Himalayan-Tibetan mountain ranges.
v) The notion that we may reach a level of dangerous climate change during this century is increasingly perceived as a possibility. Given this development, options for mitigating such dangerous climate changes are being explored worldwide. The present BC reduction proposal should also be considered in this context, and by no means is BC reduction being proposed by this author as an alternative to CO\(_2\) reduction. At best, it is a short term measure, to buy a decade or two time for implementing CO\(_2\) emission reduction.

Major Uncertainties: Our ability to model the effects of BC in climate models is severely limited. One of the main reasons is the large uncertainty (factor of 2 or more) in the current estimates of the emission of organic (OC) and elemental carbon (EC) (See Bond et al, 2004; 2007). Furthermore, biomass burning contributes significantly to the emissions of OC and EC and the historical trends (during the last 100 years) in these emissions are unknown. Models currently resort to adhoc methods such as scaling the present day emissions with past trends in population.
BC has two competing effects inside clouds. Mixtures of BC with sulfates or organics can become cloud nuclei and in turn enhance the number of cloud drops. This in turn can lead to a decrease in drop size, an increase in cloud lifetime followed by an increase in reflection of solar radiation. In addition, the decrease in drop size can suppress rain formation. On the other hand, the large solar heating of the cloudy skies by BC can decrease relative humidity and evaporate clouds, which can lead to increased penetration of sunlight to the ground and warming of the surface. These two competing effects have been examined with surface data (Kaufman and Koren, 2006) over several continental and marine locations. These data suggest that the warming effect of BC dominates the cooling effect of BC-Organic-Sulfate mixtures. Satellite data over Amazon have been used to examine the net interaction of BC laden biomass burning smoke with clouds. This study (Koren et al, 2004) also showed that the smoke lead to dissipation of low clouds. Similarly, cloud coverage in highly polluted E Asia exhibited a long term declining trend (Qian et al, 2006).

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