Interactions between Global Change and Human Health
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ATMOSPHERIC BROWN CLOUDS: HEALTH, CLIMATE AND AGRICULTURE IMPACTS

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1. Aerosols or Particles in the Air

Aerosols or atmospheric particles are either in solid form, such as dust or sea salt, or in liquid form, such as sulfates, nitrates or organics dissolved in water. Nature produces aerosols in the form of sea salt, mineral dust, sulfates and nitrates. Human activities either emit gaseous precursors, such as SO_2 and NO_x which get converted into particles through chemical processes, or directly emit particles such as soot, which is a mixture of elemental carbon and organics. The concentrations of natural aerosols in pristine air are typically around 100 to 1000 particles per cc in continental air and about 100 to 500 per cc in marine air. It is now increasingly difficult, if not impossible, to find such pristine air. In most regions of the northern hemisphere, including over the oceans, the concentrations are larger by factors ranging from two to ten. Humans add particles to the air by a variety of actions, including the burning of biofuels and fossil fuels for cooking purposes, the consumption of fossil fuel for energy, the clearing of forests, and land use modification, to name a few.

2. Atmospheric Brown Clouds

Coincident with greenhouse gas warming is the appearance of atmospheric brown clouds. If greenhouse gases, such as CO_2, are the ultimate end product of fossil fuel burning, then particulates in the air represent an intermediate phase. The brownish color in the haze (Fig. 1, see page 397) is due to the absorption and the scattering of visible solar radiation by black carbon, fly ash, soil dust and NO_2. During the Indian Ocean

Experiment (INDOEX), widespread brown clouds, about 3 km thick, were found over most of the Arabian Sea, Bay of Bengal and N. Indian Ocean (Ramanathan *et al*., 2001a). Subsequently, a new NASA satellite instrument (MODIS) has identified (Kaufman *et al*., 2002 and Ramanathan and Ramana, 2003) such widespread brown clouds over most industrialized regions of the world (see Figure 2, page 397; also see Molina and Molina, 2002).

Particle trajectories using observed winds clearly reveal how particles emitted in one continent can travel to another continent across the oceans in about a week (Fig. 3, see page 398).

This is significant because the lifetimes of most aerosols are in the order of one to two weeks. For example, model simulations (Fig. 4, see page 398) show that black carbon emitted in East Asia can travel to N. America and increase BC concentrations over N. America by as much as 75%. The fact that indeed such transport was occurring was shown by Roberts *et al*., (2006) using aircraft data (Fig. 5). The aircraft data for the

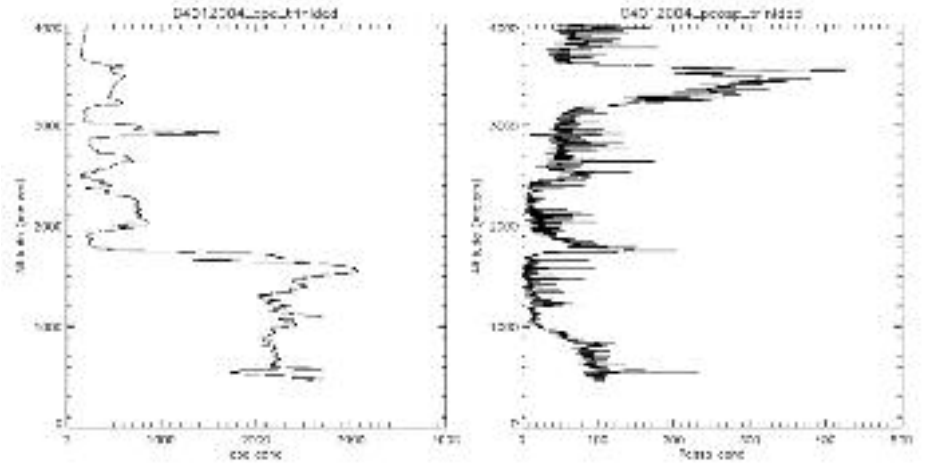

Figure 5. Long-range transport of E. Asian aerosols as measured off the coast of Trinidad in N. California on April 1, 2004 from a research aircraft during a campaign conducted by the author. The left hand panel shows the total aerosol number concentration, while the right hand panel shows the aerosol concentration between 0.1 micrometer to 3.5 micrometer. The unit for the horizontal axis is number/cm3. The vertical axis is altitude in meters. The particles due to long-range transport are seen in the layer between 3000 m to 3700 m altitude.

concentration of aerosol number concentration was collected over the west coast of the USA adjacent to the Sierra mountain range (shown on the background photo) and reveals that the transport occurs in narrowly confined layers (see the concentration spikes seen at 3 km in the left panel and at 3.5 km in the right hand panel).

In February 1999, over 200 scientists from Europe, India and the USA gathered in the islands of the Maldives to conduct the Indian Ocean Experiment (INDOEX; http://www.indoex.ucsd.edu/publications/). The interdisciplinary and comprehensive data gathered from several aircraft, ships, surface stations and satellites (Ramanathan *et al*., 2001a) helped forewarn the world (UNEP Report 2002) of a potentially major environmental problem facing the Asian region and the world. INDOEX revealed the so-called brown cloud phenomenon (see Fig. 1 from Ramanathan and Ramana, 2002) spreading from the Himalayas all the way over the N. Indian Ocean region. INDOEX collected direct evidence that manmade particles travel several thousand kilometers across the ocean (Fig. 6, see page 399). Particles were captured on filter papers from aircraft flying into the plume and subsequently analyzed in the laboratory (courtesy of J. Anderson, Arizona State Univ.). Photographs from corresponding regions were taken from the aircraft. Fig. 6 shows clearly that the haze layer extends from the Arabian sea (7°N in the top panel) into the southern Indian Ocean up to about 6.6°S, and microscope pictures reveal soot attached to the particles in the brown clouds, whereas particles from the pristine southern Indian Ocean (bottom panel) south of 8°S, which is free of haze (as revealed by the blue skies seen in the photo) is also free of soot as revealed by lack of irregular soot clusters on the particles. The particles on this electron microscope image are natural sea salt particles from the sea surface.

The major finding of INDOEX was that the brown clouds reduced the seasonal averaged solar radiation reaching the sea surface by as much 10% (Satheesh and Ramanathan, 2000) and this phenomenon is discussed later in this paper under the 'Dimming Section'. The brown cloud induced dimming has a large impact on the radiative heating of the region (Ramanathan *et al*., 2001a and b). The impact on the regional gas phase chemistry of the region (Lelieveld *et al*., 2001) was also large. The fundamental message from INDOEX is that due to long-range transport, what we normally associate with urban haze can span an entire continent plus an ocean basin (see Fig. 2, page 397). Both fossil fuel and biomass burning contribute to the particles in the haze. The persistence of the haze during the long dry season from November to May, its black carbon

content, the large perturbation to the radiative energy budget of the region and its simulated impact on the monsoon rainfall distribution have significant implications to the regional and global water budget, agriculture and health (Ramanathan *et al.*, 2001b). The logical implication is that air pollution and climate changes are intricately linked and should be addressed under one common framework, which gave rise to the creation of the Atmospheric Brown Clouds project by UNEP (see Ramanathan and Crutzen in http://www.atmosphericbrowncloud.ucsd.edu/).

Why is the air pollution problem more widespread and acute in the Tropics? The brown haze is a particularly severe problem over the tropics due to a variety of reasons. The large increase in emissions of aerosols and their precursors is an important reason. For example emissions of SO_2 in South Asia have increased by a factor of 3 to 4 since 1970. However, SO_2 emissions from S. Asia are only 25% of the US emissions. Hence other factors have to be invoked to account for the thickness and extent of the haze. Organic and black carbon and fly ash contribute more than SO_2 to the haze in Asia. The other important contributor is the unique meteorology of the tropics and the subtropics (including S. Asia) which leads to a long dry season extending from late fall and winter until spring. The dryness is caused by subsidence, which precludes the wet removal of haze particles by rain. In the mid and high latitudes, the absence of a long dry season, and seasonally distributed rainfall (and snow fall) cleans the atmosphere more efficiently.

3. Direct Impacts of ABCs

3.1. *Human Health*

The most direct and important influence of aerosols is on human health. Aerosols are inhaled and ingested in the lungs and cause acute respiratory infections, chronic obstructive pulmonary disease and lung cancer. Public health experts have long suspected that particulates cause respiratory and other health problems. What is new is that they are now linked convincingly with fatalities. Worldwide, aerosols resulting from indoor and outdoor air pollution lead to about 1.6 million premature deaths annually (Smith, 2002). In India alone about 400 to 550 premature deaths are attributed to inhalation of air pollution and particles from the use of biomass fuels for cooking (Smith, 2000).

3.2. *Reduction of Photosynthetically Active Radiation (PAR) at the Surface*

Another potentially important environmental effect of ABCs is their large effect in reducing the total (direct + diffuse) PAR (Fig. 7 from Meywerk and Ramanathan, 2002). The brown clouds over the Arabian Sea decreased direct PAR by 40% to 70%, but enhanced the diffuse PAR substantially, with a net reduction in total PAR by as much 10% to 30%. The potential impact of large reductions in direct PAR and corresponding enhancements in diffuse PAR accompanied by net reduction in total (direct + diffuse) PAR on marine and terrestrial photosynthesis and on agriculture productivity (Chameides *et al*., 2002; Stanhill and Cohen, 2001) have not been adequately studied. Similar changes in the UV spectrum due to ABCs need to be established which may have health effects due to the potential importance of UVB in producing Vitamin D (Garland and Garland, 1980; Gorham *et al*., 1989).

Nucleation of more cloud drops and suppression of precipitation efficiency: Aerosols, in particular sulfates, larger than 0.1μm provide the necessary nuclei for all cloud drops and ice crystals. Organic aerosols also serve as cloud condensation nuclei in par with the role of sulfate particles. Trace amounts of soluble gases and organic substances in air pollution can ampli-

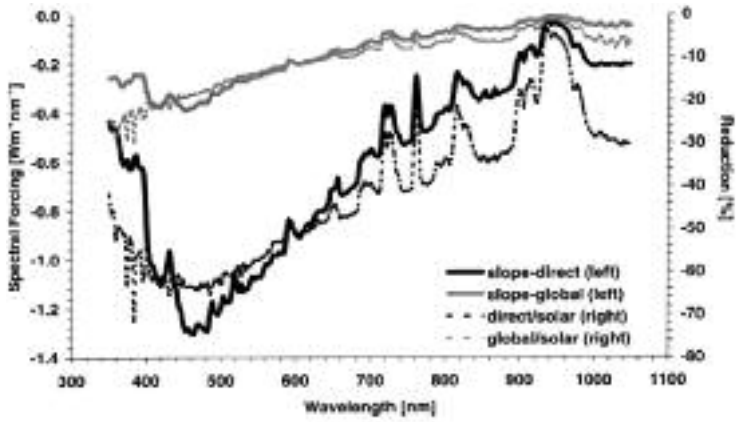

Figure 7. Decrease in solar radiation as a function of wavelength due to brown clouds as measured directly by a grating spectrometer in the Arabian sea during INDOEX. As shown the maximum decrease is seen in the PAR (400 to 700 nm) region. Source: Meywerk and Ramanathan, 2003.

fy the CCN activity of small aerosols and have led to an increase in the number of cloud drops. There is now a substantial body of in-situ aircraft observations of continental and marine clouds that show that anthropogenic aerosols enhance CCN and cloud drop number density (Fig. 8, see page 399). By nucleating more cloud drops, aerosols increase the reflection of solar radiation by clouds, which adds to the surface cooling effect. This effect is known as the *indirect forcing*. In INDOEX it was shown by direct aircraft measurements that the trade cumulus and strato-cumulus clouds in the polluted Arabian Sea had six times as many cloud drops as the pristine clouds south of the ITCZ. Another major effect of increasing the concentrations of cloud droplets is a reduction in the size of drops since they are competing for the water available for condensation. Increase in aerosols can nucleate copious amounts of small droplets. The small drops coalesce very inefficiently into raindrops, which can inhibit the formation of larger raindrops and decrease precipitation efficiency (Andreae *et al*., 2004), one consequence of which is suppression of rain over polluted regions. This was recently confirmed by satellite observations from the Tropical Rainfall Measuring Mission (TRMM), showing tracks of reduced cloud particles emanating from forest fires and from pollution sources such as coal power plants, refineries, smelters and urban areas (Rosenfeld, 2000).

3.3. *Wet Deposition (Acid Rain)*

Aerosols such as sulfates, nitrates and organic acids are incorporated into raindrops either as nuclei or by scavenging, and are removed from the atmosphere during rainfall. A raindrop in a clean atmosphere has a pH of about 5.6, but during wet deposition of pollution aerosols the pH drops, i.e., rain becomes acidic. There is a vast literature on environmental damages due to acid rain (e.g., see review in Seinfeld and Pandis, 1998), and the acid rain problem constitutes another major environmental threat from brown clouds (e.g. see Molina and Molina, 2002).

3.4. *Global Dimming*

Aerosols, by scattering/absorbing solar radiation and emitting/absorbing long wave (IR) radiation, change the radiation fluxes at the surface and the top of the atmosphere, thereby significantly perturbing the atmospheric absorption of solar radiation. These aerosol-induced changes in the radiation budget are referred to as the *direct forcing*. At the surface, aerosols decrease

the direct solar beam and enhance the diffuse solar radiation, and both of these effects have been measured during INDOEX (Ramanathan *et al.*, 2001a). Black carbon, which strongly absorbs solar radiation, plays a major role in the forcing by partially shielding the surface from the intense tropical solar radiation. This shielding effect of BC amplifies the surface radiative forcing due to all other manmade aerosols (sulfates, organics, nitrates, fly ash) by a factor of two or more in cloudy skies. BC over the northern Indian Ocean and the Arabian Sea (during the INDOEX measurement campaign) contributed as much as 10% to 14% to aerosol mass (Mayol-Bracero *et al.*, 2002), compared with about 5% in the suburban regions of Europe and N. America. The black carbon and other species in the haze reduce the average solar radiative heating of the ocean by as much as 10% and enhance the atmospheric solar radiative heating by 50 to 100%. For comparison, the perturbation to the regional IR radiation budget (the greenhouse effect) by the observed increase in CO_2 and other greenhouse gases is about 3 W m^{-2}, equivalent to a 1.5% increase in the solar heating of the ocean.

Large reduction of the seasonal averaged solar radiation of the order of 10% or larger (due to anthropogenic aerosols) is not restricted to S. Asia. It has now been observed in many regions of the planet including E. Asia, the Atlantic, Western Pacific, Mediterranean, in Europe, N. and S. America and Africa (see summary in Ramanathan and Ramana, 2003). More recently we have observed reduction of 10 to 15% in the Himalayas. Given such large observed reduction in solar radiation due to brown clouds, we would anticipate that the planet has become dimmer with time, since emissions of brown cloud particles (notably sulfates and black carbon) have changed substantially with time (Fig. 9, see page 400). Long-term negative trends in surface solar irradiance have been observed by surface radiometers worldwide over land. The reported trends in the annual mean irradiance vary from -5% (10 W m^{-2}) between 1958 to 1985 for all land stations (*48*) to about -1 to -3% per decade for the last four decades over many of the 1500 stations in the GEBA data sets (Stanhill and Cohen, 2001). More recently it has been observed (Wild *et al.*, 2005) that several stations in N. America and western Europe have seen a reversal in the dimming trend since the 1980s, consistent with reductions in sulfur and BC emissions (Fig. 9). However, India and China are witnessing an unabated continuation of the dimming trend into the twentieth century. Surface solar insolation over Indian stations has decreased by 10 W m^{-2} from 1970 to 2004 (Ramanathan *et al.*, 2005), and in China it has decreased by about 15 W m^{-2} from 1954 to 2001 (Qian *et al.*, 2006).

The effect of ABCs on surface solar radiation is prevalent over most parts of the globe today and has been estimated by us for the period 2000 to 2004 using modern satellite and surface data gathered around the world (Fig. 10, page 400). It ranges from a reduction of 3 to 12 W m^{-2} over many parts of N. America and Europe and from 6 to 25 W m^{-2} over Asia, Africa and S. America. The potential consequences of this dimming effect on climate are discussed next.

4. Impact on Climate and Water Budget

4.1. *Masking of Global Warming*

The surface dimming due to ABCs has a surface cooling effect, which should have masked some of the surface warming due to greenhouse gases. The magnitude of this masking effect is one of the central problems in climate change with significant implications for future climate changes and policy responses to global warming (Andreae *et al*., 2005). The extent of global warming is not fully reflected in the Earth's observed surface temperatures. The additional heat trapped by the increase in greenhouse gases from the late nineteenth century to the present time should have committed the planet to a global warming in the range of 1ºC to 3ºC (see Ramanathan, 1988 for an explanation of the term *committed warming*). The observed global surface warming is only about 0.6 K, i.e., only about 20% to 60% of the committed warming (depending on whether we use 3 K or 1 K for the committed warming). Some of this warming has been masked by the dimming due to brown clouds and the remaining heat is stored in the depths of the ocean to be released in the coming decades to centuries. Through the process of convective overturning, oceans transfer infrared energy to their deepest layers and hold the heat, delaying the impact of global warming. Whether this stored heat will warm the atmosphere in a few decades or a few centuries is unknown.

Current estimates of the ABC masking effect range from 30% to as high as 75% (Crutzen and Ramanathan, 2003; also see Andreae *et al*. in this issue for a more detailed consideration of the aerosols' masking effect). The fundamental issue with this large range of uncertainty of the masking effect is that, policies to reduce ABCs (due to their effects on health, acid rain and agriculture) will unmask the cooling effect of ABCs quickly and have a potentially large effect in the acceleration of global warming. This is

because the lifetime of ABCs is a few weeks and hence their masking effect will diminish as soon as their emissions are curtailed, whereas GHGs will respond to emission reduction policies on decadal to century time scales. Without a better quantification of the masking effect of ABCs, we would not know whether warming in the coming decades (if it were to occur) is due to the unmasking effect of ABCs (by efforts of cleaning up in Asia for example), or due to release of stored greenhouse heating in the oceans, or continued increase in emission of greenhouse gases. In the meantime, every decade we delay in taking action, we are committing the planet to additional warming that future generations have to deal with.

4.2. *Global Scale Drying*

Global mean land average precipitation (P) decreased by about 2.4% from 1947 to 1996 (Hulme *et al*., 1998). The long-term precipitation trends for the global oceans are unknown due to lack of data. The large warming witnessed during the last 50 years was not accompanied by an increase in precipitation. On the other hand, climate models which include greenhouse forcing suggest an increase in global rainfall with surface warming since the warmer oceans evaporate more moisture which subsequently reaches the surface as rainfall. Increased rainfall does not automatically imply increased availability of surface water, however, since in a warmer climate evaporation of surface moisture is also accelerated.

The primary mechanism by which aerosols reduce global mean rainfall is by reducing the solar radiation reaching the surface of the earth (supporting online material). Direct observations revealed that the presence of absorbing black carbon magnified by a factor of 2 to 3, the reduction of solar radiation due to sulfates and other scattering aerosols. A reduction in the surface solar radiation can slow down the atmospheric hydrological cycle and reduce rainfall because roughly 75% of the globally averaged radiative heating of the surface is balanced by evaporation.

4.3. *Regional Climate, Monsoon Rainfall and Water Budget Effects*

In addition to their effect on global mean rainfall, ABCs alter the spatial gradient in surface and atmosphere solar heating (see Fig. 10) which leads to significant regional impacts, particularly on the S. Asian monsoon impacting over 2 billion Asians (Ramanathan *et al*. 2005, hereafter referred to as R2005). The link between aerosols and rainfall has been

suggested for selected regions, including the well-known Sahelian drought (Rotstayn and Lohmann, 2002), the north-south shift in the East Asian monsoon (Menon *et al*., 2002), and the South Asian region (Chung *et al*,. 2002; R2005). I will focus on the impact of ABCs on the S. Asian monsoon since it has not only been examined extensively by us (Chung *et al*., 2002; Chung and Ramanathan, 2003; R2005; Chung and Ramanathan 2006) but also impacts over a billion S. Asians. ABCs impact the monsoon circulation and climate through several distinctively different processes.

1) The dimming of the N. Indian Ocean and S. Asia decreases surface evaporation from the ocean (see Fig. 2 of R2005) and since the primary fuel for the monsoon rainfall is evaporation of moisture from the N. Indian Ocean (including the Arabian Sea and Bay of Bengal), this has a direct influence in reducing monsoon circulation and rainfall.

2) The dimming by ABCs is accompanied by a large increase in atmospheric solar heating (by 50% to 100%) which tends to stabilize the surface-atmosphere system by cooling the surface and warming the atmosphere. This redistribution of sunlight causes warmer air to overlie the colder surfaces. This increase in atmospheric stability, which has been observed using microwave satellite data (see Fig. 4 of R2005), tends to suppress convective cloud formation and rainfall. The increased atmospheric stability during the dry season can also decrease the ventilation of the pollution away from the boundary layer leading to increased occurrences of pollution events and fog formation.

3) Since ABCs and their dimming effects are predominantly concentrated in the N. Indian Ocean (NIO), Arabian Sea and Bay of Bengal (due to their proximity to pollution sources in S and SE Asia) with minimal effects in the S. Indian Ocean (see Fig. 10), ABC induced dimming suppresses the sea surface temperatures (SSTs) north of the equator while allowing the S. Indian Ocean (SIO) to warm in response to the greenhouse warming (R2005). Observed sea surface temperatures reveal that this differential masking of the N. Indian Ocean SSTs is indeed happening (Chung and Ramanathan 2006). For example, 50 year trends (from 1950 to 2000) in observed SSTs reveal tropical SIO warming by as much as 0.7 K while the trend is much smaller in the NIO with an almost zero warming trend in the N. Arabian Sea and N. Bay of Bengal. Since deep convection and tropical rain bands gravitate toward warmer oceans, climate model simulations suggest that the differential warming trend has led to a slowing down of the monsoon circulation resulting in decreased rainfall over land regions (S. Asia) and increase in oceanic rainfall over the southern tropical Indian Ocean.

The monsoon rainfall over India has decreased since the 1950s and our studies suggest that ABCs may be the primary driver of this negative trend. Clearly more studies of this important influence of ABCs are required to confirm these findings. Impact on the monsoon circulation has potentially a large impact on S. Asian agriculture, since monsoon rainfall correlates linearly with rice production (Webster *et al.*, 2002). The impact of the haze on monsoon rainfall provides another strong rationale for reducing air pollution in the developing nations.

SUMMARY

Manmade particles in ABCs lead to about 1.6 million deaths annually, decrease photosynthetically active solar radiation, have direct agricultural impacts, lead to a large dimming of the planet, decrease the precipitating efficiency of clouds, stabilize the surface-atmosphere system during dry seasons, alter sea surface temperature gradients, alter regional rainfall patterns, and lead to global cooling and drying. In addition, the ASIAN monsoon system affecting the lives of over 2 billion people is vulnerable to ABCs. The only positive thing that can be said about ABCs is that their lifetime is only about a few weeks long which implies that actions to regulate their emissions will result in immediate benefits.

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