Atmospheric Brown Clouds

Long-Range Transport and Climate Impacts

by V. Ramanathan and M.V. Ramana

Although mistakenly assumed to be restricted to urban regions, atmospheric brown clouds, such as the Denver Brown Cloud, are frequently occurring phenomena in many regions of the world. Recent field studies and satellite data have revealed that, due to long-range transport, the brown cloud (or haze) covers vast areas of the world, including an entire continent and ocean. This article considers the environmental and climate influences of atmospheric brown clouds.

INTRODUCTION

In February 1999, more than 200 scientists from Europe, India, and the United States gathered in the Maldives to conduct the Indian Ocean Experiment (INDOEX).[1] The study data were collected using several aircraft, ships, surface stations, and satellites[2,3] and helped forewarn of a potentially major environmental problem facing Asia. The findings from INDOEX[2,3] revealed that the so-called "brown cloud phenomenon" in Asia is spreading from the Himalayas over the North Indian Ocean region (see Figure 1) and that, due to the long-range transport of air pollution, the brown haze that is normally associated only with urban regions now spans an entire continent and ocean basin (see Figure 2). Both fossil fuel and biomass burning contribute to the aerosols (or particles) that form the brown haze, which has a potentially large impact on both the radiative heating[2] and the regional gas phase chemistry of the region.[4] The persistence of the haze during the long dry season in Asia from November to May, its black carbon content, the negative effect on the radiative energy budget of the region, and its simulated impact on the monsoon rainfall distribution have significant implications for the regional and global water budgets, agriculture, and human health.[3] The logical implication is that air pollution and climate changes are intricately linked and should be addressed under one common framework.[2]

In addition to Asia, recent National Aeronautics and Space Administration (NASA) Terra Satellite results show that pollution aerosols in the form of haze are found in and downwind of all inhabited regions of the world and that each of these hazes is spread over a vast region. For example, Figure 3 shows the following phenomena for the month of April (April–May, 2001 and 2002 average): anthropogenic aerosols extending from the eastern half of North America to the mid-Atlantic ocean and Europe, heavy aerosol loading in northern and eastern Europe and the Mediterranean sea, dust and anthropogenic haze from Mongolia and China extending across the Pacific, and the biomass burning aerosols from Africa and the Amazon region extending into the Atlantic. These phenomena can be thought of as the "South and North American Brown Cloud," "Asian Brown Cloud," and "African Brown Cloud." Furthermore, air parcels carrying the aerosols can travel across an entire ocean basin or continent within five to seven days, as shown in Figure 4. The particle trajectory in Figure 4 is shown for an altitude of approximately 3 km since peak concentrations of anthropogenic particles are found even at 3 km.[2,3] Through the sponsorship of the United Nations Environmental Programme (UNEP) and the National Oceanic and Atmospheric Administration (NOAA), an international program called Atmospheric Brown Clouds (ABC) has been created to come to grips with this complex problem. ABC will focus first on the haze problem in Asia.[5] Many government agencies in Asia will soon join ABC. Using INDOEX and ABC data, this article considers the environmental and climate influences of atmospheric brown clouds.

THE INDIAN OCEAN EXPERIMENT

The South Asian Brown Cloud, which covers most of the Arabian Sea, the Bay of Bengal, and southern Asia (an area roughly equivalent to the continental United States), occurs every year and typically extends from November through May. The brownish haze is composed of a 3-km-thick mixture of anthropogenic sulfates, nitrates, organics, black carbon, dust and fly ash particles, and natural aerosols such as sea salt and mineral dust. Its brownish color is due to the absorption and scattering of solar radiation by the anthropogenic black carbon, fly ash, soil dust particles, and nitrogen dioxide. Radiation observations taken from an observatory stationed in the Maldives[6] revealed that the haze absorbs a large amount of radiation within the atmosphere, which results in a significant decrease in the amount of radiation that reaches the surface. In-situ measurements of aerosol chemistry from aircraft, ships, and surface stations found that anthropogenic sources (e.g., biomass burning, fossil-fuel combustion) contribute as much as 75% to the observed aerosol

concentration.[2,4] The main sources of the Asian brown haze are the northeast trade winds over the western Arabian Sea and northern Indian Ocean, carrying pollution from southwest and central Asia; the northwest–northeast flow along the western coast of India; and northeast trade winds over the Bay of Bengal from the eastern half of southeast Asia.

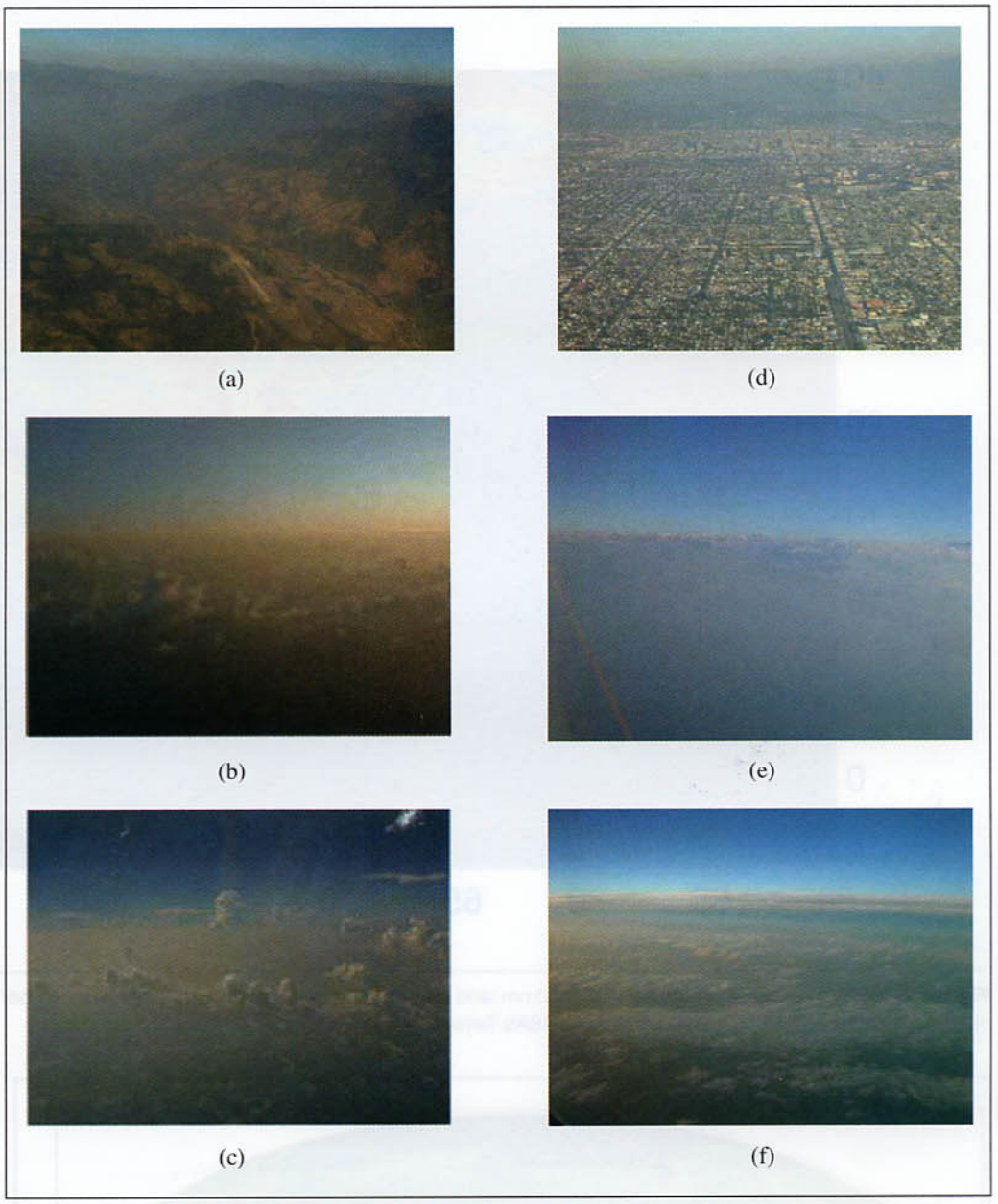

Figure 1. (a) Haze over the lower Himalayas, south of Mt. Everest[2]; (b) Haze over the Arabian Sea, March 25, 1999 (3.0°N, 74.5°E)[4]; (c) Haze over the Indian Ocean, February 24, 1999, just north of ITCZ (0.5°N, 73.3°E)[4]; (d) Haze over Los Angeles, December 27, 2002 (34°N, 118°E); (e) Haze over the Alps, Geneva, February 2003 (46°N, 10°E); and (f) Haze over the South China Sea, December 24, 2002 (22°N,113°E) (photos d, e, and f courtesy of V. Ramanathan).

AIR POLLUTION IN THE TROPICS

Brown haze is a particularly severe problem in the South Asian (tropics) region and is, in part, due to significant increases in emissions of aerosols and their precursors. Emissions of sulfur dioxide (SO_2), for example, have increased by a factor of 3–4 in South Asia since 1970. However, these emissions account for only 25% of U.S. SO_2 emissions. Hence, other factors have to be invoked to account for the thickness and extent of the brown haze. Observatory and satellite data revealed that organic and black carbon and fly ash contribute more to the haze in Asia than SO_2. Another important contributor to the brown haze is the unique meteorology of the tropics and subtropics regions (including South Asia), which leads to a long dry season that extends from late fall to spring. The dryness in the atmosphere is caused by subsidence, which precludes the wet removal of haze particles by rain. By contrast, in the mid- and high latitudes, the absence of a long dry season, and the occurrence of seasonally distributed rainfall (and snow fall), helps clean the atmosphere more efficiently.

IMPACTS ON RADIATION

Aerosols, by scattering and/or absorbing solar radiation and by emitting and/or absorbing long-wave (infrared) 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 "direct forcing." At the surface, aerosols decrease the direct solar beam and enhance the diffuse solar radiation (both of these effects have been measured and included in INDOEX forcing values[6]). Black carbon over the northern Indian Ocean and the Arabian Sea contributes as much as 10–14% to aerosol mass (measured during the INDOEX measurement campaign), compared to 5% in the suburban regions of

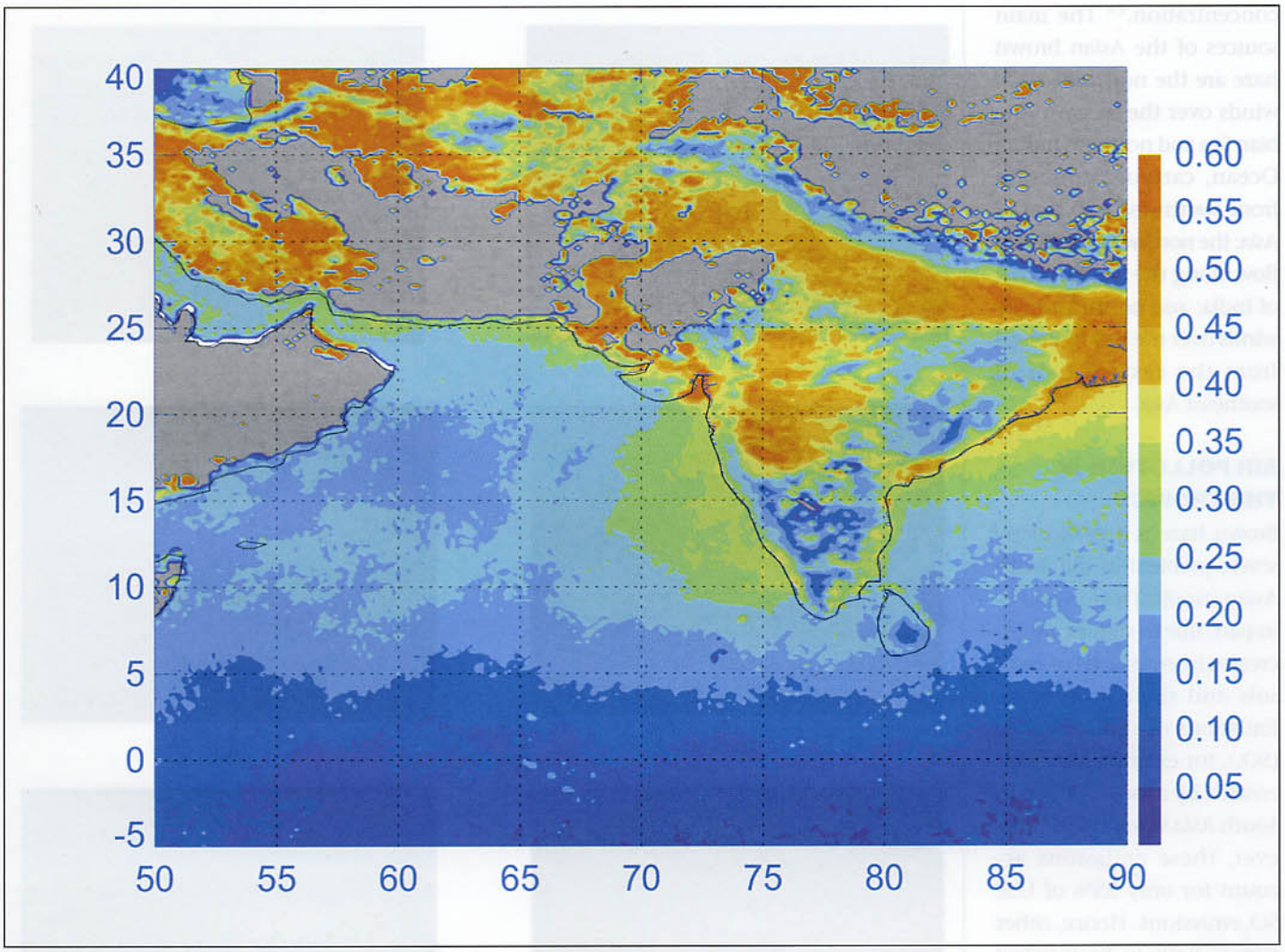

Figure 2. Mean aerosol optical depth (AOD; 550 nm land and ocean) at visible wavelength from December 2001 to May 2002. The data were obtained from the MODIS instrument onboard NASA's Terra Satellite.[9]

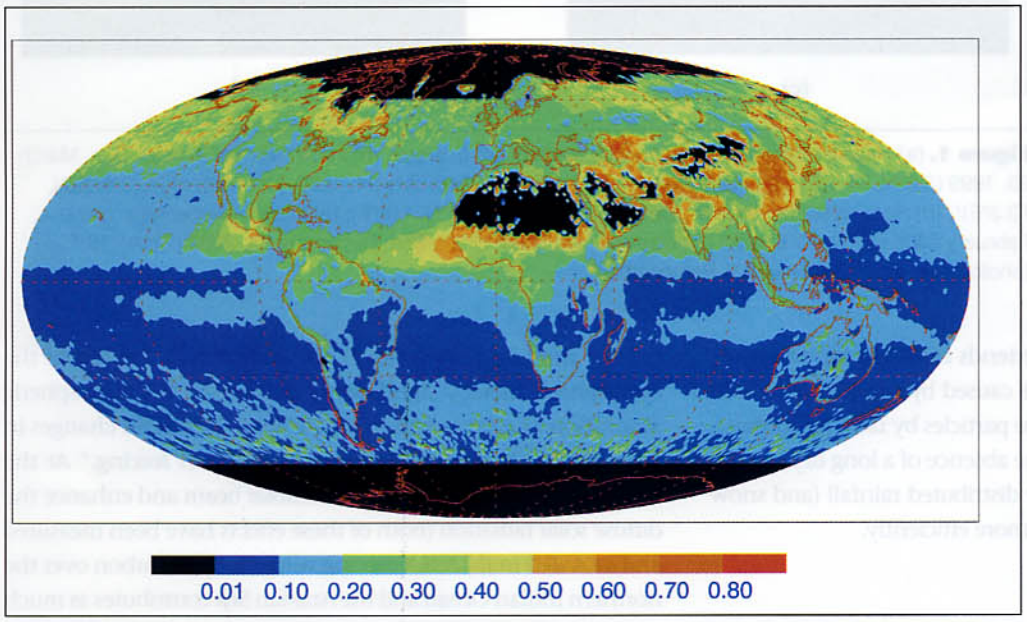

Figure 3. Global distribution of mean AOD (550 nm) for April and May, 2001 and 2002. The data were obtained from the MODIS instrument onboard NASA's Terra Satellite.[9] Black area represents no data.

Europe and North America. Black carbon, which strongly absorbs solar radiation, plays a major role in direct forcing by partially shielding the surface from the intense tropical solar radiation. This shielding effect amplifies the surface radiative forcing contributed by other man-made aerosols (e.g., sulfates, organics, nitrates, fly ash) by a factor of two or more in cloudy skies. In addition, black carbon and other species found in the brown haze reduce the average radiative heating of the ocean by as much as 10% and enhance atmospheric solar radiative heating by as much as 50–100%.

By nucleating more cloud drops,[3] aerosols increase the reflection of solar radiation by clouds, which adds to the surface cooling effect. This is known as "indirect forcing." The INDOEX findings using direct aircraft measurements[7] showed that the trade cumulus and strato-cumulus clouds over the polluted Arabian Sea were found to have six times as many cloud drops as the pristine clouds south of the Inter Tropical Convergence Zone (ITCZ). However, large reductions of seasonal averaged solar radiation on the order of 10% (or larger) due to anthropogenic aerosols are not restricted to South Asia. This phenomenon has been observed in many regions of the world, including the Atlantic, Western Pacific, Mediterranean, Europe, North and South America, and Africa.[8-13] More recently, the authors have observed reductions of seasonal averaged solar radiation of 10–15% in the Himalayas. The potential negative consequences of this dimming effect of aerosols are discussed below.

IMPACTS ON CLIMATE

One of the major accomplishments of INDOEX is that it integrated field measurements with satellite data and aerosol assimilation models to estimate the seasonally and regionally average direct and indirect forcing for anthropogenic aerosols. The findings showed that the regional radiative perturbation by the anthropogenic aerosols at the surface and within the atmosphere is an order of magnitude greater than that due to anthropogenic greenhouse gases (GHGs). This does not imply that GHGs are not an important factor, but that regional climate changes may be strongly influenced by absorbing aerosols. GHG forcing is distributed globally and is cumulative with time, while the aerosol forcing is concentrated regionally. In addition, absorbing aerosols may have a negative impact on the regional hydrological cycle.[2,3] This is because approximately 50–80% of the solar heating of the ocean is balanced by evaporation. The reduction in solar radiation reaching the surface (approximately 20 Wm^{-2}, or 10% of the absorbed solar radiation) will lead to a reduction in evaporation, which, in turn, will lead to a reduction in precipitation.

The 50–100% enhancement in solar heating of the lower atmosphere may affect the monsoonal circulation, since the aerosol heating is distributed nonuniformly with latitude and longitude. Indeed, when the INDOEX haze data are inserted into global climate models,[14,15] the results show a significant perturbation of the region's rainfall patterns, with more rain in some regions and drought in others. The haze may also lead to a cooling effect of the land surface during the dry season, strengthening of low-level inversion, and a cooling of the North Indian Ocean, which has been confirmed by observation data.[16] It is interesting that the North Indian Ocean is one of the few oceanic regions with very little increase in heat content as shown by Levitus et al.[17] It is likely that the global warming effect over the North Indian Ocean has been nearly balanced by the haze cooling with no net change in the ocean

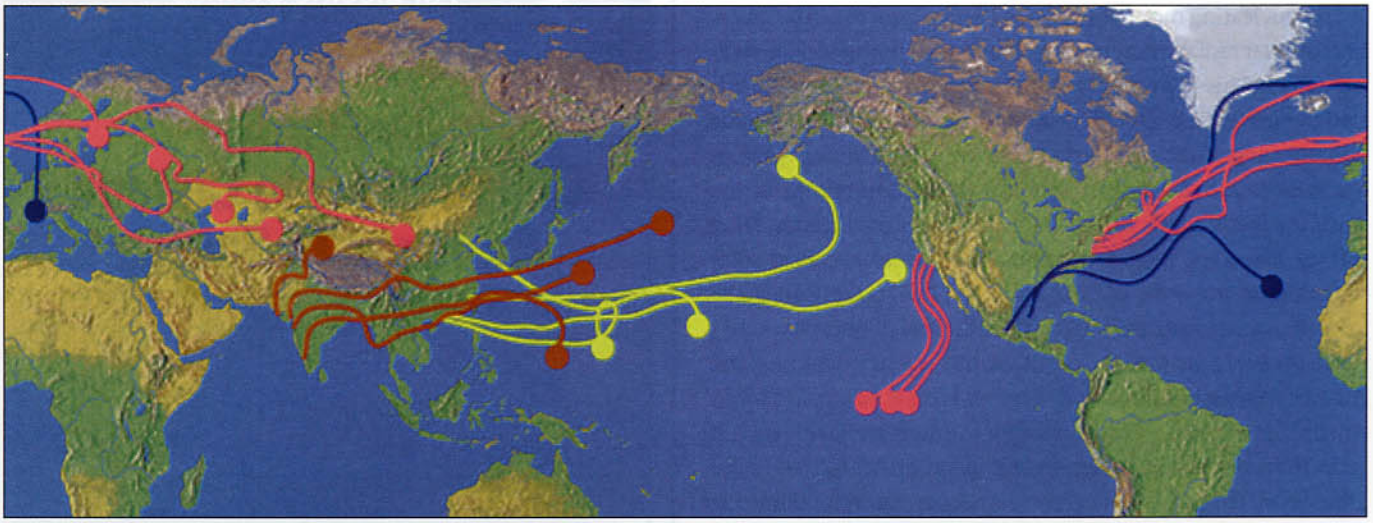

Figure 4. Seven-day forward particle trajectories at 3-km altitude during April originating from India, China, Mexico, the East and West Coasts of the United States, London, Paris, and Berlin, showing potential transcontinental nature of the "brown haze" (courtesy of T.N. Krishnamurti).

heat content. Recent studies have also demonstrated that the anthropogenic haze produces copious amounts of smaller drops in convective clouds, thus suppressing precipitation over polluted areas and favoring higher aerosol concentrations in the upper troposphere.[18]

LINKS BETWEEN AEROSOLS AND GLOBAL MEAN PRECIPITATION

The global mean precipitation level has been decreasing since the 1950s.[19] This is largely due to a pronounced decrease in precipitation in the tropics region (between 25°S and 30°N) and a smaller increase in the extra-tropics region (>=30°N and S). Please note that precipitation data are typically collected using land stations. Climate models that reflect only GHGs do not simulate this negative trend, but when sulfate aerosols are added to the models, some of the models are able to simulate a portion of the observed drying trend in the tropics.[19,20] However, sulfates are only part of the aerosol forcing problem. The addition of black carbon can enhance the sulfate surface forcing by a factor of two or more.[2,6]

Pollution from aerosols (particularly black carbon) is generally greater in the tropics than other regions, and so aerosols likely played a significant role in the observed tropical drying effect. For the Indian Subcontinent, the climate models containing only GHG increases predicted an increase in precipitation during the wet season (June, July, August), while all of the models with GHG and sulfate aerosols showed either reduced rainfall increases or strong rainfall decreases.[20] The link between aerosols and reduced rainfall has also been noted for other tropical regions, including the Sahelian drought,[21] the north–south shift of the East Asian monsoon,[14,22] and rainfall changes in the southwestern Asia region.[15] Xu[22] shows that the East Asian monsoon has gradually moved southwards in recent decades, a movement that began in the late 1970s, leading to a severe drying effect in the north and flooding in the south. Xu suggests that the main reason for this shift in direction may be the negative solar forcing caused by pollution aerosols. Indeed, several recent studies, including Chung et al,[14] have suggested that anthropogenic aerosols may have inversely affected precipitation in the tropical regions.

CONCLUSIONS

The negative forcing (cooling effect) from aerosols can be compared to the positive forcing from GHG emissions.[20] The major difference is that the GHG forcing is distributed globally, whereas the aerosol radiative forcing is concentrated regionally. For example, the GHG radiative forcing in the northern and southern hemispheres is comparable, while the aerosol forcing is a factor of 1.5 (or more) greater in the northern hemisphere than it is in the southern hemisphere.[23] In addition, the aerosol forcing is more concentrated over land than water. Another difference is that the aerosol forcing is negative at the surface and positive in the atmosphere (due to the absorption of solar radiation by black carbon). Therefore, aerosols could have contributed to asymmetric climate changes between the northern and the southern hemisphere, between the land and the oceanic regions, and between the atmosphere and the surface.

Furthermore, while global warming is expected to increase precipitation,[20] the large negative forcing effect due to aerosols consequently may have led to a decrease in precipitation (drying effect).[3] Coupled ocean–atmosphere simulations (completed by the authors in collaboration with Drs. Washington, Bettge, and Kiehl of the National Center for Atmospheric Research) reveal that the brown haze forcing in Asia may have slowed the summer monsoonal circulation, resulting in a reduction in monsoonal precipitation over South Asia.[24] If the observed increases in black carbon emissions from South Asia continue for the next several decades, model calculations suggest that much larger decreases in summer precipitation may occur as a result.[24] At the same time, the soot-induced solar heating of the boundary layer may strengthen the low-level inversion during the long dry season, which can positively feed back into aerosol life times and increase their concentration. In addition, the soot lodged within clouds may enhance cloud solar absorption and influence cloud life times. In summary, the absorbing aerosols can have major impacts on all aspects of the atmospheric hydrological cycle.

It is important to note that the role of GHGs will increase in the future because of their long lives in the atmosphere (on the order of 100 years or more). By the latter half of this century, global warming due to GHGs will likely dominate the aerosol impact. The developed nations share the major responsibility for past emissions of GHGs, black carbon, and aerosol precursor gases, such as SO_2. Developing nations in Asia, Africa, and South America are now becoming major contributors of aerosol problems, particularly black carbon emissions. Of particular concern is Asia, where the brown cloud is thick and widespread during the long dry season. More than 50% of the world's population inhabits the region, which is experiencing impressive industrial growth rates and could therefore be vulnerable to unexpected negative impacts from the brown haze on health,[25] the hydrological cycle, and agriculture.

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Regional Haze Trends in Alaska

Implications for Protected Class I Visibility Areas

by Walter J. Wilcox II and Catherine F. Cahill

Mt. Denali at the Denali National Park and Preserve (NPP) in Alaska. Photo by Chris LeDoux

Visibility degradation during winter and spring in Alaska's Denali National Park and Preserve is primarily the result of airborne pollution transported from Russia and Western Europe. While the severity of the so-called "Arctic Haze" abated in the 1980s and 1990s, scientists fear that this trend may soon reverse. This article looks at recent trends in regional haze in Alaska.

BACKGROUND

During winter, the Arctic atmosphere becomes contaminated with anthropogenic pollution transported from sources in Western Europe and North-Central Russia.[1] This unusual form of regional air pollution consists of approximately 90% sulfate (SO_4^{2-}) aerosols and 10% soot (a wide variety of metals and organic compounds are also present[2]) and is commonly referred to as "Arctic Haze." Arctic Haze is a seasonal phenomenon, absent in summer and most conspicuous in spring. The absence of moisture and sunlight during winter means that very little sulfur dioxide (SO_2) is oxidized into SO_4^{2-} by aqueous- or photochemistry. Consequently, the majority of the sulfur that reaches interior Alaska during mid-winter is in the form of SO_2. Sulfur dioxide is not a component of regional haze per se because it does not scatter light. Arctic Haze reaches its peak intensity around March, as sunlight returns to the Arctic and aerosols of ammonium sulfate ($(NH_4)_2SO_4$), ammonium bisulfate (NH_4HSO_4), and sulfuric acid (H_2SO_4) are formed by the photo-oxidation of SO_2. These particles scatter light very effectively and cause a significant reduction in visual range.[3]

In the high Arctic region, the wintertime visual range is often reduced to 10 km, even with the absence of fog or blizzard conditions, from a typical summer visual range of 270 km.[3] The observed concentrations of SO_4^{2-} aerosols alone cannot account for such an extreme reduction in visibility. Previous research suggests that at temperatures below -25 °C, the nucleation of ice crystals onto acidic aerosols further contributes to the reduction in visibility.[4] Other aerosols (especially soot) are also known to contribute to the reduction in visual range. Although the effects are not as striking as in the high Arctic region, Arctic Haze also impacts visibility at the Denali National Park and Preserve (NPP) in Alaska. Denali NPP is a Class I Visibility Area and is therefore protected from visibility degradation under the Clean Air Act. For seven months of the year (November–May), SO_4^{2-} aerosols are the dominant fine particulate matter ($PM_{2.5}$) species found in the atmosphere at Denali NPP (see Figure 1). Visual range can be calculated from the Interagency Monitoring of Protected Visual Environments (IMPROVE) program's data set using the methods outlined in the Federal Land Managers' Air Quality Related Values Workgroup Phase I report.[5] These calculations suggest that during periods of high SO_4^{2-} concentration between January and April (i.e., during Arctic Haze episodes), the visual range in Denali NPP drops to 100 km from a seasonal maximum of approximately 280 km. However, these calculations may underestimate the impact on visual range at very low temperatures because the nucleation of ice crystals onto acidic aerosols below -25 °C is not accounted for by this method.

Europe and Russia are the main contributors of long-range transport of sulfur to the Arctic airmass. Barrie et al.[6] created the most detailed model to date of the transport of anthropogenic sulfur into the Arctic by analyzing the meteorology of the winter 1979–1980 in 6-hr intervals. According to this model, 52% of sulfur that entered the Arctic came from Europe, 42% from the former Soviet Union (FSU), and 6% from North America. Transport patterns and proximity caused pollution from Europe to have a much greater impact on the Arctic airshed than pollution from North America, while pollution from the FSU had a greater impact than European pollution (i.e., 8.5% of the sulfur produced in the FSU reached the Arctic, as opposed to 5.7% from Europe and 0.8% from North America).[6]

ARCTIC HAZE DECLINE

Although the first scientific observations of Arctic Haze were not made until the 1950s,[7] its existence most likely dates back to the advent of widespread coal use in Europe. Ice core studies