

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 heating² 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

Atmospheric Brown Clouds

Long-Range Transport and Climate Impacts

by V. Ramanathan and M.V. Ramana

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 Maldives⁶ 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.

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 (SO₂), 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. SO₂ 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 SO₂. 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,

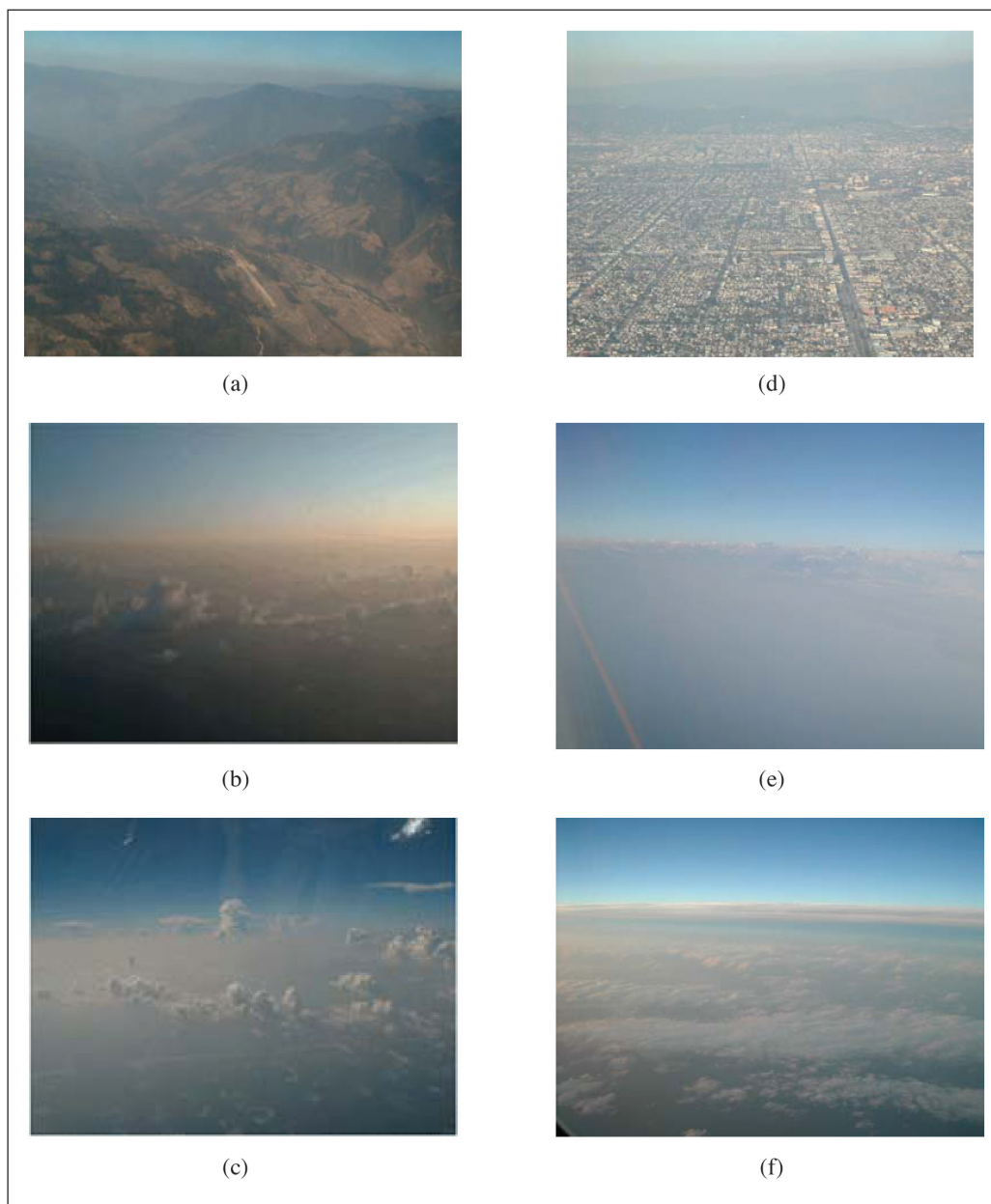


Figure 1. (a) Haze over the lower Himalayas, south of Mt. Everest²; (b) Haze over the Arabian Sea, March 25, 1999 (3.0°N, 74.5°E)⁴; (c) Haze over the Indian Ocean, February 24, 1999, just north of ITCZ (0.5°N, 73.3°E)⁴; (d) Haze over Los Angeles, December 27, 2002 (34°N, 118°E); (e) Haze over the Alps, 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).

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 values⁶). 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

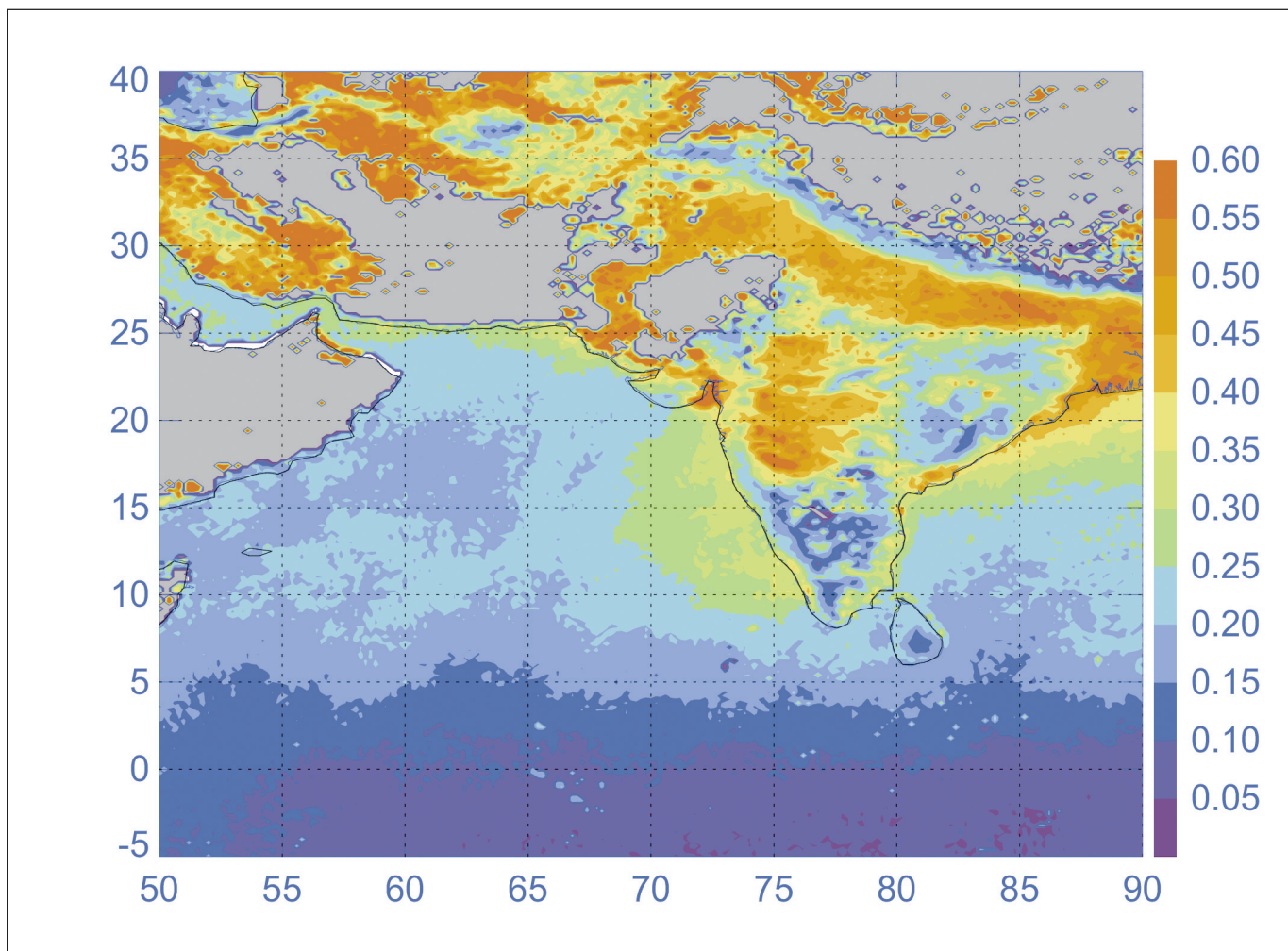


Figure 2. Mean aerosol optical depth (AOD; 550 nm land and ocean) at visible wavelength from December 2001 to May 2002. The data were obtained from the MODIS instrument onboard NASA's Terra Satellite.⁹

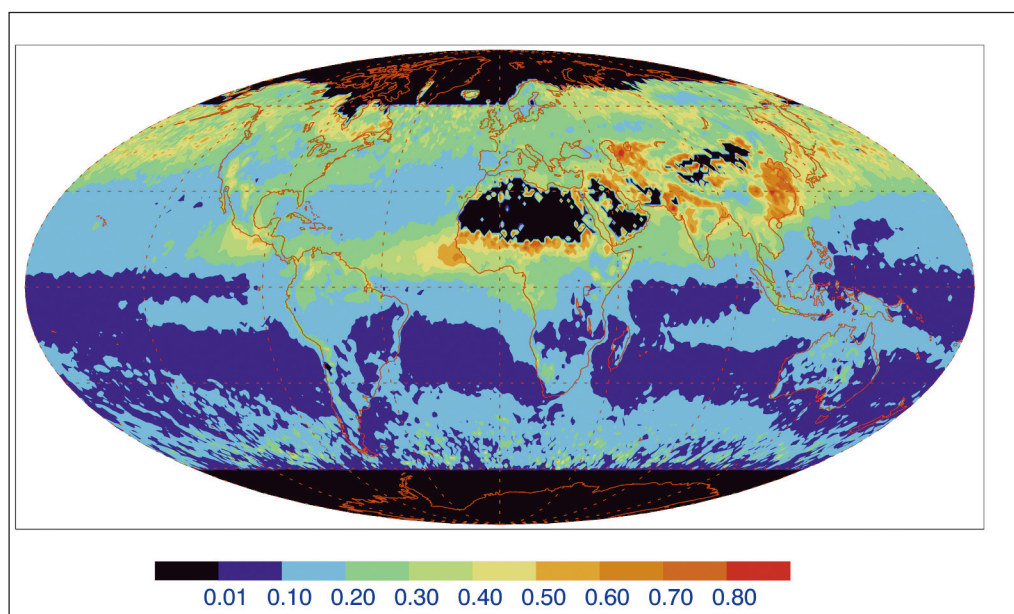


Figure 3. Global distribution of mean AOD (550 nm) for April and May, 2001 and 2002. The data were obtained from the MODIS instrument onboard NASA's Terra Satellite.⁹ Black area represents no data.

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 measurements⁷ 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



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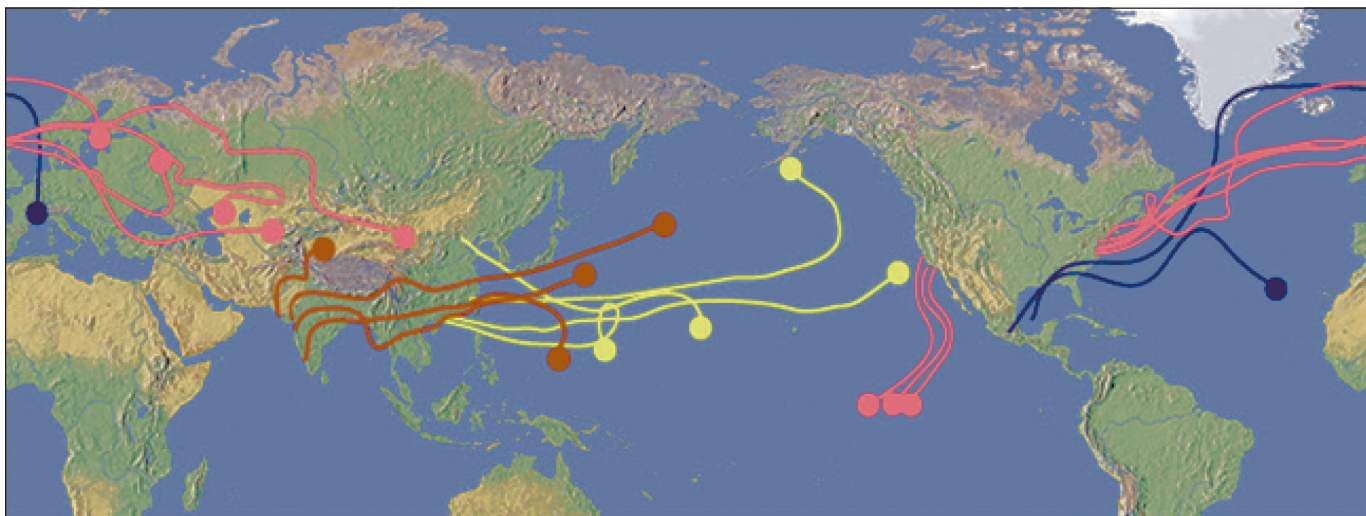


Figure 4. Seven-day forward particle trajectories at 3-km altitude during April originating from India, China, Mexico, the East and West Coasts of the United States, London, Paris, and Berlin, showing potential transcontinental nature of the "brown haze" (courtesy of T.N. Krishnamurti).

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 ($\geq 30^\circ\text{N}$ 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 Xu²² 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.

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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 SO₂. 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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REFERENCES

1. Ramanathan, V.; et al. INDOEX White Paper: Indian Ocean Experiment; available at http://www.indoex.ucsd.edu/publications/white_paper/.
2. Ramanathan, V.; et al. The Indian Ocean Experiment: An Integrated Analysis of the Climate Forcing and Effects of the Great Indo-Asian Haze; *J. Geophys. Res.* **2001**, *106*, 28371.
3. Ramanathan, V.; Crutzen, P.J.; Kiehl, J.T.; Rosenfeld, D. Aerosols, Climate, and the Hydrological Cycle; *Science* **2001**, *294*, 2119.
4. Lelieveld, J.; et al. The Indian Ocean Experiment: Widespread Air Pollution from South and Southeast Asia; *Science* **2001**, *291*, 1031.
5. Atmospheric Brown Clouds (ABC), under the sponsorship of the U.N. Environmental Programme (UNEP) and National Oceanic and Atmospheric Administration (NOAA). See <http://www.asianbrowncloud.ucsd.edu/>.
6. Satheesh, S.K.; Ramanathan, V. Large Differences in Tropical Aerosol Forcing at the Top of the Atmosphere and Earth's Surface; *Nature* **2001**, *405*, 60.
7. Heymsfield, A.J.; McFarquhar, G.M. Parameterizations of INDOEX Microphysical Measurements and Calculations of Cloud Susceptibility: Applications for Climate Studies; *J. Geophys. Res.* **2001**, *106*, 28653.
8. Kinne, S.; Pueschel, R. Aerosol Radiative Forcing for Asian Continental Outflow; *Atmos. Environ.* **2001**, *35*, 5019.
9. Bush, B.C.; Valero, F.P.J. Surface Aerosol Radiative Forcing at Gosan during ACE-Asia Campaign; *J. Geophys. Res.* **2003**, *108*, 8660.
10. Markowicz, K.M.; Flatau, P.J.; Ramana, M.V.; Crutzen, P.J.; Ramanathan, V. Absorbing Mediterranean Aerosols Lead to a Large Reduction in the Solar Radiation at the Surface; *Geophys. Res. Lett.* **2002**, *29* (20), 1968.
11. Kaufman, Y.J.; Tanre, D.; Boucher, O. A Satellite View of Aerosols in the Climate System; *Nature* **2002**, *419*, 215.
12. Hignett, P.; Taylor, J.P.; Francis, P.N.; Glew, M.D. Comparison of Observed and Modeled Direct Aerosol Forcing during TARFOX; *J. Geophys. Res.* **1999**, *104*, 2279.
13. Ichoku, C.; Remer, L.A.; Kaufman, Y.J.; Levy, R.; Chu, D.A.; Tanre, D.; Holben, B.N. MODIS Observations of Aerosols and Estimation of Aerosol Radiative Forcing over Southern Africa during SAFARI 2000; *J. Geophys. Res.* **2003**, *108*, 8499.
14. Chung, C.E.; Ramanathan, V.; Kiehl, J.T. Effects of the South Asian Absorbing Haze on the Northeast Monsoon and Surface-Air Heat Exchange; *J. Climate* **2002**, *15*, 2462.
15. Menon, S.; Hansen, J.; Nazarenko, L.; Luo, Y. Climate Effects of Black Carbon Aerosols in China and India; *Science* **2002**, *297*, 2250.
16. Krishnan, R.; Ramanathan, V. Evidence of Surface Cooling from Absorbing Aerosols; *Geophys. Res. Lett.* **2001**, *29* (10), 1029.
17. Levitus, S.; Antonov, J.I.; Wang, J.; Delworth, T.L.; Dixon, K.W.; Broccoli, A.J. Anthropogenic Warming of Earth's Climate System; *Science* **2001**, *292*, 267.
18. Rosenfeld, D. Suppression of Rain and Snow by Urban and Industrial Air Pollution; *Science* **2000**, *287*, 1793.
19. Hulme, M.; Osborn, T.J.; John, T.C. Precipitation Sensitivity to Global Warming: Comparison of Observations with HadCM2 Simulations; *Geophys. Res. Lett.* **1998**, *25*, 3379.
20. Ramaswamy V.; et al. Radiative Forcing of Climate Change. In *Climate Change 2001: The Scientific Basis, Synthesis Report, Intergovernmental Panel on Climate Change*; Cambridge University Press: Cambridge, 2001.
21. Rotstayn, L.; Lohman, V. Tropical Rainfall Trends and the Indirect Aerosol Effects; *J. Climate* **2002**, *15*, 2103.
22. Xu, Q. Abrupt Change of the Mid-Summer Climate in Central East China by the Influence of Atmospheric Pollution; *Atmos. Environ.* **2001**, *35*, 5029.
23. Kiehl, J.T.; Briegleb, B. The Relative Roles of Sulfate Aerosols and Greenhouse Gases in Climate Forcing; *Science* **1993**, *260*, 311.
24. Ramanathan, V.; Kiehl, J.T.; Bettge, T.; Chung, C.; Washington, W.M. The South-Asian Brown Cloud: A Coupled Ocean-Atmosphere Model Study of Its Impact on the Monsoon, El-Nino, and the Hydrological Cycle. Presented at the 14th Symposium on Global Change and Climate Variations, American Meteorological Society, Longbeach, CA, January 2003, and at the 14th Conference IUGG/MI02 Symposium on Cloud, Aerosol and Climate, Sapporo, Japan, June 30–July 3, 2003.
25. Stone, R. Counting the Cost of London's Killer Smog; *Science* **2002**, *298*, 2106.

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