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Long-range transport of soil dust and smoke pollution in the South Asian region

Bilkis A. Begum¹, Swapan K. Biswas¹, Gauri G. Pandit², I. Vijaya Saradhi², Shahida Waheed³, Naila Siddique³, M.C. Shirani Seneviratne⁴, David D. Cohen⁵, Andreas Markwitz⁶, Philip K. Hopke⁷

¹ Bangladesh Atomic Energy Commission (BAEC), Atomic Energy Centre, Dhaka (AECD), P.O. Box: 164, Dhaka, Bangladesh

² Bhaba Atomic Energy Centre, Trombay, Mumbai, 400085, India

³ Chemistry Division, Directorate of Science, Pakistan Institute of Nuclear Science and Technology (PINSTECH), P.O. Nilore, Islamabad, 45650 Pakistan

⁴ Atomic Energy Authority, 60/460, Baseline Road, Orugodawatta, Wellampitiya, Sri Lanka

⁵ Australian Nuclear Science and Technology Organisation (ANSTO) Locked Bag 2001, Kirrawee DC, NSW, Australia

⁶ GNS Science, 30 Gracefield Road, P.O. Box 31-312, Lower Hutt, New Zealand

⁷ Centre for Air Resources Engineering and Science, Department of Chemical and Biomolecular Engineering, Clarkson University, Potsdam, NY 136999-5708, United States

ABSTRACT

Transboundary transport of air pollution in the South Asian region has been an issue of increasing importance over the past several decades. Long–range transport of anthropogenic pollution is contrasted with that of pollution produced by natural processes such as dust storms or natural forest fires. Airborne particulate matter datasets covering the period from 2002 to 2007 from the neighboring countries like Bangladesh, India, Pakistan and Sri Lanka were used to find the source areas that are primarily responsible for long range transported pollutants. All four countries collected samples with the same type of sampler and follow the same technique for mass and BC measurements. It was found that high fine soil contributions were from dust storms. On the other hand, smoke in this region mainly comes from northern India where agricultural waste is often burned.

Keywords:

Airborne particulate matter Long-range transport Black carbon Soil

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Corresponding Author: Philip K. Hopke Tel: +1-315 268 3861 Fax: +1-315-268-4410 E-mail: hopkepk@clarkson.edu

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1. Introduction

The Asian Brown Cloud is a layer of air pollution that covers parts of South Asia, namely the northern Indian Ocean, India and Pakistan (Ramanathan et al., 2001; Srinivasan and Gadgil, 2002) and the cloud appears as a giant brown stain hanging in the air over much of South Asia and the Indian Ocean every year between January and March. The visible impact of air pollution is the haze, a layer of pollutants and particles from biomass burning and industrial emissions. This Asian Brown Cloud of pollutants has a brownish color and this brown cloud phenomenon is a common feature of industrial and rural regions around the world (UNEP and C^4 , 2002). Because of long-range transport of air pollutants, the mostly urban (fossil fuel related) and/or rural (biomass burning and brick kiln related) phenomenon is transformed into a regional haze (or cloud) that can span large areas including an entire continent. It is now becoming clear that the brown cloud may have huge impacts on agriculture, health, climate and the water budget of the planet (Ramanathan, 2008; Ramanathan and Carmichael, 2008).

The haze was really defined as a result of the INDOEX measurement campaign (Ramanathan et al., 2001; UNEP and C⁴, 2002). This haze consists of a combination of droplets and solid particles. The droplets in the haze are less than 1.0 μm in radius

(Pandve and Patil, 2008). There are two possible sources for the particles in haze. They are either generated naturally (e.g. sea salt, soil dust) or are man-made (e.g., sulfate and soot). From an aircraft, the haze appears brown when the fraction of soot or soil dust is large. The potent haze lying over the entire Indian subcontinent –from Sri Lanka to Afghanistan – has led to some erratic weather, sparking floods in Bangladesh, Nepal and northeastern India but drought in Pakistan and northwestern India (Ramanathan et al., 2007).

Haze is an atmospheric phenomenon where dust, smoke and other dry particles affect visibility and obscure the clarity of the sky. Sources for haze particles include farming, traffic, industry, and wildfires. Haze often occurs when dust and smoke particles accumulate in relatively dry air. Subsequent increases in humidity can result in hygroscopic growth and decreased visibility. When weather conditions block the dispersal of smoke and other pollutants they concentrate and form a usually low-hanging shroud that impairs visibility and may become a respiratory health threat. Industrial pollution can also result in dense haze.

The main objective of this study is to locate the sources of significant events of fine soil dust and smoke particles in the atmospheric aerosol that are major causes of haze in the South



Asian region. Regional fine air particle data dated from January 2002 to December 2007 that includes mass, black carbon and elemental concentrations in Bangladesh, India, Pakistan, and Sri Lanka measured using appropriate nuclear analytical techniques were analyzed to understand the haze problem and long-range transport of fine particles.

2. Methodology

2.1. Sample description and analysis

All the four countries collected samples use the Gent stacked filter sampler (Hopke et al., 1997), which is capable of collecting air particulate samples in coarse (2.5–10 μ m) and fine (<2.5 μ m) size fractions. The sampling locations are shown in Figure 1 and the sampling sites description and analytical techniques used for sample analysis for individual countries are given in Table 1. In Dhaka, Bangladesh, the site is at a semi–residential area located at the Atomic Energy Centre, Dhaka (AECD) Campus (23.73°N, 90.40°E). The sampler was placed on the flat roof of the building at a height of 5 m above the ground. The intake nozzle of the sampler is located 1.8 m above the roof. The intake is about 80 m away from the roadside. The sample was placed so that the airflow around it was unobstructed.



Figure 1. Map locating the sampling sites.

The sampler in Mumbai, India was placed at a height of 15 m above the ground on the terrace of a building in Vashi that is situated in Navi Mumbai. Navi Mumbai is the largest planned new city near Mumbai (19.07°N, 72.97°E). The city is very close to Thane Belapur area that was known as largest industrial belt in India. The industrial estate is mainly comprised of chemical, bulk drugs and intermediates, dye and dye intermediates, pharmaceutical, pesticide, petrochemical, engineering goods and textile manufacturing industries. A national highway passes 2 km from the sampling site.

Table 1. Information of the sampling sites

Country	Site description	Latitude	Longitude	Sampling period	Analytical Technique
Bangladesh	Residential	23.73°N	90.40°E	2002 2007	IBA
India	Suburban	19.05°N	73.02°E		XRF, IBA
Pakistan	Suburban	33.72°N	73.05°E	2002-2007	IBA
Sri Lanka	Residential	6.82°N	79.85°E		IBA

The sampler in Colombo, Sri Lanka was located at the Atomic Energy Agency (AEA) facility (6.933°N, 79.833°E) and the sampler was placed on the flat area on the first floor of the AEA building. This building is set in a residential area on the northwestern side of Colombo. There are moderately busy roads within 300 m.

The sampler in Nilore, Islamabad, Pakistan $(33.37^{\circ}N)$ and $73.06^{\circ}E$) is located on the roof of a building at PINSTECH at a height of approximately 5–6 m above ground. The inlet of the sampler is at a height of ~1 m above roof level. The nearest road is 1 km away. The air flow is unobstructed as the buildings are well separated. The surrounding area consists of some residences and farms. The Nilore site is located on the outskirts of the city of Islamabad. This area was mostly farmland until 10 years ago, but now many housing units are being developed in this area. The road leading to PINSTECH from the main city is being widened that will further increase this urbanization process.

The samples were collected on Nuclepore filters with 8 μ m pores for the coarse fraction samples and 0.4 μ m pores for the fine fraction samples. In general, samples were collected for representative 24–hour periods at least once a week. In some locations, it was not possible to collect the sample over a full 24 hours because of filter clogging. Then the sampler was operated with alternating time on and time off (e.g. one hour on followed by one hour off) over the course of the 24–hour period to provide a representative sample during that day. In all cases, the total sampling time was never less than 8–hours equally distributed over 24–hours.

The methods used for the gravimetric mass measurement and the black carbon measurements are same for the four countries (Hopke et al., 2008). The collected samples were analyzed using nuclear analytical methods including Ion Beam Analysis (IBA) (Cohen et al., 1996), X–ray Fluorescence (XRF) and Instrumental Neutron Activation Analysis (INAA). These methods are described in detail elsewhere (Landsberger and Creatchmam, 1999).

2.2. Source fingerprints

The IBA or XRF analyses of PM samples provided opportunities of detecting sufficient number of elemental concentrations to develop fingerprints for a number of particle sources. It is useful to combine some of these elements and estimate the concentrations of the major compounds such as ammonium sulfate from the measured sulfur concentration. It is also possible to derive other combinations of the elements that represent signatures for interesting aerosol components. These combinations are called pseudo–elements such as "soil" (Malm et al., 1994). Thus, these composite variables and pseudo–elements provide a better understanding of the composition of the fine particles and lead to better estimates of possible sources and their contributions to the average ambient aerosol (Malm et al., 1994).

2.3. Soil

Windblown soil is composed mainly of the oxides of Mg, Al, Si, Ca, Ti and Fe with many other trace elements. The average composition of sandstone and sedimentary rocks and the summation of the 5 major oxides of Al, Si, Ca, Ti and Fe account for more that 85% of the total composition (Weast and Astle, 1982). So the equation for soil is:

$$Soil = 2.20 Al + 2.49 Si + 1.63 Ca + 1.94 Ti + 2.42 Fe$$
(1)

Equation (1) assumes that the two common oxides of iron Fe_2O_3 and FeO occur in equal proportions. The factor of 2.42 for iron also includes the estimate for K_2O in soil through the (K/Fe)=0.6 ratio for sedimentary soils.

2.4. Smoke

Smoke is the collection of airborne solid and liquid particulates and gases (Mulholland, 1995) emitted when a material undergoes combustion or pyrolysis, together with the quantity of air that is entrained or otherwise mixed into the mass. It is an unwanted byproduct of fires (including stoves, candles, oil lamps and fireplaces). Smoke is also a component of internal combustion engine exhaust gas, particularly diesel exhaust. The composition of smoke depends on the nature of burning fuel and conditions of combustion. Fine potassium is an accepted indicator for smoke from biomass burning/brick kiln (Baxla et al., 2009; Begum et al., 2004; Begum et al., 2009). In order to obtain a reliable smoke indicator from the fine potassium, it is necessary to subtract the fine potassium associated with soil and sea salt component from total K (Weast and Astle, 1982). Hence, smoke is obtained by using the following equation:

$$Smoke = (K_{tot} - 0.036 Na - 0.6 Fe)$$
(2)

The haze is seen from the southern edge of the Himalaya Mountains southward over the Bay of Bengal every year. The duration of this haze varies from country to country but will often be observed from early December to early March. The haze, mostly a mixture of urban and industrial pollution, often collects at the base of the mountains in the wintertime. This cloud of haze that frequently lingers over parts of Asia from Pakistan to China and even the Indian and Pacific Oceans has been called the "Asian Brown Cloud". The brownish haze consists of a three kilometers thick mixture of anthropogenic sulfate, nitrate, organics, black carbon, dust and fly ash particles and natural aerosols such as sea salt and mineral dust. The brownish color is due to the absorption and scattering of solar radiation by anthropogenic black carbon, fly ash, part of the soil dust and NO₂ (Ramanathan and Ramana, 2003).

The brown haze often exists over large South Asian cities in part due to particulate emissions within the urban area, dust storm, and distant fires. Emitted soil dust and gases have the capacity to be transported over long distances, sometimes many hundreds of kilometers and may give rise to deposition in another country. In addition to the respiratory problems the persistent haze can cause, it also appears to hinder crops by blocking sunlight and could be altering regional weather. Haze can have impacts on agricultural productivity through direct and indirect effects. The direct effects include:

(1) Reduction of total solar radiation (sum of direct and diffused) in the photo–synthetically active part of the spectrum (0.4 to 0.7 micron) reduces photosynthesis.

(2) Settling of particles (e.g. fly ash, black carbon, and dust) on the plants can shield the leaves from solar radiation.

(3) In addition, particle deposition can increase acidity and cause plant damage.

The indirect effects include:

(1) Changes in surface temperature that can directly affect the growing season. In the tropics, surface cooling (such as expected from particles) can extend the growing season (while a greenhouse warming can decrease it).

(2) Changes in rainfall or surface evaporation can have a large impact (UNEP and C^4 , 2002).

Atmospheric black carbon (BC) can act in two ways. First as a direct absorber of visible light and that provides direct warming in the lower atmosphere (Ramanathan and Carmichael, 2008). Secondly, the deposition of black carbon on ice or snow such as in the Himalayan glaciers is part of what is causing them to melt quickly (Kehrwald et al., 2008). Thus, there are good reasons to understand the extent and sources of BC.

Biomass burning in low quality cook stoves is an important source of methane and up to 6% of the methane emissions are due to biomass burning (Islam, 2002). BC may also originate from vehicular combustion sources.

2.5. Meteorology

Bangladesh has a climate of tropical monsoon, mild winter (October to March), hot, humid summer (March to June) and humid warm rainy monsoon (June to October). January is the coolest month with temperature averaging 26°C and April is the warmest month with temperature ranging from 33°C to 36°C. It rains mostly during June to October.

Mumbai (India) has a tropical wet and dry climate. Mumbai's climate can be best described as moderate temperatures with high level of humidity. Its coastal nature and tropical location ensures moderate temperatures throughout the year, average of 27.2 °C and average precipitation of 2 422 mm. The temperature on average is about 30 °C in summer and 18 °C in winter. Mumbai's experiences four distinct seasons winter: (December–February); summer: (March–May); monsoon (June–September) and Post Monsoon (October–December).

Pakistan has four distinct seasons: a cool, dry winter from December through February; a hot, dry spring from March through May; the summer rainy season, or southwest monsoon period, from June through September; and the retreating monsoon period of October and November. The onset and duration of these seasons vary somewhat according to location. The climate in the capital city of Islamabad varies from an average daily low of 2°C in January to an average daily high of 40 °C in June. Half of the annual rainfall occurs in July and August, averaging about 255 millimeters in each of those two months. The remainder of the year has significantly less rain, amounting to about 50 mm per month. Hailstorms are common in the spring.

Sri Lanka's climate is similar to that experienced in southern India, and involves two monsoon seasons and a dry season. Colombo's weather, and much of the weather of the southwestern section of the island, is impacted by the Yala monsoon during the period from May to August. The dry season runs from December to April. In the northern and eastern sections of the island, the weather patterns are different: the monsoon season is from October to January and the dry season lasts from May to September. During the months of October and November all parts of Sri Lanka experience occasional heavy downpours and thunderstorms.

Therefore, it was observed that from early December to early March, the winter season exist within this region and the wind direction is mainly northwesterly.

2.6. Back trajectory calculation

Using models of atmospheric transport, a trajectory model calculates the position of the air being sampled backward in time from the receptor site from various starting times throughout the sampling interval. The trajectories are presented as a sequence of latitude and longitude values for the endpoints of each segment representing each specific time interval being modeled. The vertical motion of air parcels is considered during this model. The NOAA Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT–4) (Draxler and Rolph, 2003) model was used to calculate the air mass backward trajectories for those days when fine particles were sampled. Archived REANALYSIS meteorological data were used as input. The latitude/longitude was used depending on the site of location of four countries and trajectories were computed backward in time up to 240 hours (10 days). Tick marks on the trajectory plots indicate 6–hour movement locations.

Back trajectories were calculated for periods of 10 days. Although the error in positions of the individual trajectory endpoints increase with increasing trajectory duration (Kahl, 1996), 10–day back trajectories have proven useful in assessing the likely source regions for particulate pollution (Polissar et al., 2001).

3. Results and Discussion

Although airborne particles are generally associated with global cooling effects, recent studies have shown that they can actually have a positive radiative forcing effect particularly in certain regions such as the Himalayas (Ramanathan et al., 2007). In South Asia, many countries recognize air pollution as a major public health concern and have undertaken projects or regulatory programs to control air pollution, but data provided by some of these countries indicate that in many cities, air quality still falls below world standards for acceptable air quality (Hopke et al., 2008). These studies showed that major sources of pollutants are soil dust, smoke and traffic, which form haze in this continent (Pandve, 2008). Thus, the data for these sources are discussed.

3.1. Long-range transport of soil

In order to exclude low local source contributions, only daily events with concentrations that are two standard deviations above the mean value for a measured species were considered. Table 2 shows the mean, standard deviation, and the threshold values for soil, smoke and black carbon concentrations. The meteorological condition in this region during winter leads to prevailing northerly and northwesterly winds. There is then the possibility of transboundary events (Adhikary et al., 2007) affecting local air quality. Figure 2 shows the time series plot for the fine soil concentrations for all four countries during the study period. It was found that there was a large peak on 5 March 2003 in case of Bangladesh and the value was 38 $\mu g/m^3.$ On the other hand, for India, the large peak was on 20 February 2003 and the value was $24 \ \mu g/m^3$. In case of Pakistan, there is no large peak in the time interval from February 2003 to March 2003. For Sri Lanka, there are no data available during this period. From the meteorological data, it was observed that winter season predominates during the month of December to February in this region and the direction of wind is usually northwesterly. Beginning in March, the premonsoon starts and the average wind speed becomes higher than in the wintertime.

Table 2. The mean, standard deviation and peak values of soil, smoke and black carbon concentrations $(\mu g/m^3)$ during the studying period

Parameter	Statistics	Bangladesh	India	Pakistan	Sri Lanka
Soil	Mean	3.51	6.82	3.17	2.00
	Median	2.69	5.03	1.98	1.83
	STD	3.05	6.53	3.63	1.48
	Threshold Value	9.60	19.9	10.4	4.95
Smoke	Mean	0.31	0.33	0.15	0.06
	Median	0.24	0.12	0.10	0.05
	STD	0.24	0.81	0.22	0.12
	Threshold Value	0.78	1.96	0.59	0.31
BC	Mean	8.97	7.86	2.62	10.8
	Median	7.54	6.70	2.34	10.3
	STD	6.34	4.63	1.61	4.15
	Threshold Value	21.7	17.1	5.84	19.1



Figure 2. Time series plots of fine soil concentrations in Bangladesh, India, Pakistan, and Sri Lanka.

For the observed potential transboundary episode resulting in the highest soil contribution (Figure 2), the elemental data were examined and it was seen that the concentration of silicon, a signature for soil was also very high for those samples. To explore the long-range transport of soil, back trajectories were calculated on these days. Thus, back trajectories of air parcels at different heights starting from 300 m, 500 m, 1000 m and 1500 m on 5 March 2003 (10 days) for Dhaka, Bangladesh and 20 February 2003 (7 days) for Mumbai, India were tried. Figure 3 shows representative trajectories for 500 m and 1000 m height. They show that the air parcels come over Iran and Pakistan before reaching the sampling site at Dhaka and possibly over North Africa before reaching the sampling site at Mumbai. In case of Dhaka Bangladesh, it was found that for 1 500 m starting height, the air parcels came from northwest direction. There was a thick dust plume (light brown) that blew westward and then routed northward by strong southerly winds (NASA web site).



Figure 3. Air parcel back trajectories showing long range transport of soil dust in Bangladesh and India in February 2003.

Every year starting from February to March due to the change of season several dust storms occur and the dust particles travel thousands of miles. The trajectories calculated at lower heights (300 m and 500 m) show areas of Iran and Oman. It is also seen in the satellite image that there were two thick plumes of desert dust blew over Oman and across the Gulf of Oman toward Iran and Pakistan on 18 February 2003 (see the Supporting Material–SM, Figure S1). Therefore, it can be concluded that the high fine soil contribution is most likely due to long–range transport of the desert dust from these regions. For India, it has been found that the trajectory plots at starting heights of 1 000 m and 500 m overlap and show the same origins of desert dust. From the NASA web site it was found that there were several dust storms in early February 2003 that blew in a northeasterly direction. It has been found from calculated trajectories that the dust particles travel about 7 000 km to reach both of the receptor sites (Dhaka and Mumbai).

High concentrations of fine soil were found on 21 and 26 February 2004 in Colombo, Sri Lanka and Islamabad, Pakistan respectively. The backward trajectories of air parcels at different starting heights of 300 m, 500 m, 1 000 m on 21 and 26 February 2004 in Colombo, Sri Lanka (10 days back) and Islamabad, Pakistan (5 days back) respectively were calculated (Figure 4). It was found that the trajectory plots for both receptor sites at 300 m and 500 m were similar and overlap. Therefore, the trajectories with a 300 m starting height were discarded for clarity. The trajectory plots (500 m and 1 000 m) showed that the air parcels came from a northwesterly direction and traveled about 4 000 km and 12 000 km to reach Islamabad and Colombo, respectively. Therefore, the origin of dust source was not same. From NASA web site (see the SM, Figure S2) it was found that there were several dust storms in Sahara, Jordan and Iran in February 2004 which blew over northeastern part of Southern Asia.



Figure 4. Air parcel back trajectories showing long range transport of soil dust in Pakistan and Sri Lanka in February 2004.

3.2. Long-range transport of smoke

The scatter plot for BC and smoke concentrations (Figure 5) shows that both come from the same source. Figures 6 and 7 show the time series of BC and smoke concentrations and the smoke concentrations calculated from Equation (2) for the four countries. In order to remove the local contributions to smoke, similar criterion values as described previously were calculated to identify the likely long-range transport episodes. From the Dhaka time series, the peak concentration above threshold value, which was found on 10 November 2003, to be 1.13 µg/m³, and the BC concentration on that day was 25.4 μ g/m³. In case of India, on 5 November 2003, the high concentrations for smoke and BC were $2.66 \ \mu\text{g}/\text{m}^3$ and $13.6 \ \mu\text{g}/\text{m}^3,$ respectively. For Pakistan, on 15 November 2003, the high concentrations for smoke and BC were 1.15 µg/m³ and 8.93 µg/m³, respectively. For Sri Lanka, on 12 December 2003, the high concentrations of smoke and BC were 0.48 μ g/m³ and 16.2 μ g/m³, respectively.

The vertically mixed model starting at different heights from 300 m, 500 m and 1 000 m above the ground level was used to calculate the seven, ten, ten and seven days backward trajectories arriving at the receptor sites in Dhaka, Mumbai, Islamabad and

Colombo on 10 November, 03 November, 15 November and 12 December 2003, respectively (Figure 8). The trajectory plots show the area of northwest part of South Asia.



Figure 5. Scatter plot between BC and smoke concentrations.



Figure 6. Time series plots of black carbon (BC) concentrations in Bangladesh, India, Pakistan, and Sri Lanka.

The smoke signature reaches at Dhaka by traveling about 2 200 km to 5 000 km from the northwest direction and shows the area of India, Iran and Iraq. In case of Mumbai, the source region is northern India. For Sri Lanka, the source regions are Bangladesh, Afghanistan, Pakistan, and Vietnam and travels about 2 500 km to 8 000 km to reach Colombo. For Pakistan, the source regions of smoke signature are west part of India and Kagakhantan and travels about 800 km to 2 200 km to reach at Islamabad.

There were two satellite images (see the SM, Figures S3 and S4) that show the potential source areas in northwestern India. These two satellite images show the evidence of haze that occurred due to smoke from agricultural fires in northern India starting from 3 November 2003 and backing up against the Himalayan Mountains until 6 November 2003. Haze is created by a range of airborne particles and pollutants released from incomplete burning of biomass, agricultural waste, or traffic related fuel. The cloud is associated with the winter monsoon (November/December to April) during which there is no rain to wash pollutants from the air. The increasing aerosol loading in

Asian countries associated with industrialization during last two decades has caused major health–related problems associated with worsening air quality and has also had impact on aviation safety (Lau et al., 2008). Studies over South Asian region have found that anthropogenic aerosols may significantly change the energy balance of the atmosphere and earth's surface (Ramanathan et al., 2001; UNEP and C⁴, 2002). These anthropogenic aerosol forcing could influence the seasonal rainfall distribution in the monsoon regions over South Asia (Ramanathan et al., 2001; Chung et al., 2005; Ramanathan et al., 2005). The observed weakening Indian monsoon and in China, drought in the north and flooding in the south are influenced by the haze (Lau et al., 2008). Asian glacial melting could lead to water shortages and floods for the hundreds of millions of people who live downstream.



Figure 7. Time series plots of fine smoke concentrations in Bangladesh, India, Pakistan, and Sri Lanka.



Figure 8. Air parcel back trajectories showing the likely source areas for smoke in India, Bangladesh, Pakistan, and Sri Lanka.

Every year during the winter, starting from December to early March, there are high air pollution episodes in this region and it often collects at the base of the mountains. The high air pollution is due to anthropogenic activities and it has been found that BC concentrations are high during this season (Nair et al., 2007; Begum et al., 2008). For South Asia, general circulation climate models (GCMs) simulations suggest that a two-to three-fold increase in soot loading is sufficient to substantially weaken the monsoon circulation, decrease rainfall by more than 25% and increase drought frequency significantly (Menon et al., 2002).

4. Conclusion

Haze consists of a combination of suspended water droplets and minute particles in the atmosphere. From this study, it was observed that the air quality problem is largest during the winter monsoon (the dry season) in meteorologically stable and cloud free atmospheric conditions. It was also seen that the peak concentrations of smoke observed in Bangladesh during November 2003 is likely due to agricultural fires in northern India during that period and the high fine soil contribution is likely due to long–range transport of the desert dust which was blew over Oman and across the Gulf of Oman towards Iran and Pakistan on 18 February 2003.

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Supporting Material Available

Thick plumes of desert dust blew over Oman and across the Gulf of Oman toward Iran and Pakistan on February 18 2003 (Figure S1), An intense dust storm blew through the skies over Jordan on February 22, 2004 (Figure S2), Smoke from agricultural fires in northern India, continues to back up against the Himalayan Mountains on November 6, 2003, it started from November 3 2003 (Figure S3), To the southeast of a dense cluster of agricultural fires (red dots), a pall of smoke hangs over northwestern India (Agricultural fires in northern India, October 23, 2003) (Figure S4). This information is available free of charge via the Internet at http://www.atmospolres.com.

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