TRENDS IN PARTICULATE MATTER (PM) AND LEAD POLLUTION IN AMBIENT AIR OF DHAKA CITY IN BANGLADESH

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ABSTRACT

Airborne particulate matter (PM) samples collected at a semi-residential area in Dhaka during the period of 1997-2005 by Gent sample. The samples were analyzed by PIXE (Proton Induced X-Ray Emission). It was found that due to the banning of leaded gasoline together with other stepwise policy intervention, the air quality tends to become moderate. From the high standard deviation of Pb after banning of leaded gasoline, it can remarked that there is still emission of Pb which is not so frequent. As a result the yearly average Pb value has increased slowly. This increased value is not related with black carbon or bromine. It may come from paints, manufacturing products for Pb acid batteries and foundries etc. Therefore, suspect sources should be investigated and controlled for the management of pb level in the ambient air.

Key words: Proton Induced X-Ray Emission, Pb acid batteries, Particulate Matter

1. INTRODUCTION

Air pollution has become one of the serious environmental concerns in urban areas, especially in view of the adverse health effects that have been associated with ambient fine particles[1,2]. The rates of increase in pollutant concentrations in the cities of developing countries are higher than those of developed countries[3]. Up to 10% of respiratory infections and disease in Bangladesh may be attributable to urban air pollution[4]. While the problem is most severe in Dhaka, both because the air quality is poor and large number of people are exposed. The air pollution is also becoming a growing concern in other major cities of Bangladesh. Like other developing Southeast Asian countries, emissions from various kinds of diesel vehicle and badly maintained automobiles contribute most to air pollution problems. Ambient particulate matter (PM) with aerodynamic diameter <10 mm (PM_{10}) have both short-term and chronic adverse effects on health in both children and adults [5-7]. Fine particle pollution in the atmosphere primarily consists of micron and sub-micron particles from the manmade sources such as motor vehicles, industrial emissions, biomass burning, fossil fuel burning, and natural sources such as windblown soils and sea spray[8,9]. A high concentration of air pollutants such as black carbon in Dhaka City air has been reported [10]. Vehicular emissions, as well as burning of biomass for cooking and coal in the brick kilns around the city, are the main contributor to these emissions [11,12]. The characterization of these fine particles is very important for the regulators, and researchers due to their potential impact on human health [13], their ability to travel thousands of kilometers across countries, and their influence on climate forcing and global warming [14]. Several Government policy interventions such as banning of two-stroke baby taxi, promotion of compressed natural gas (CNG) as alternative fuel for vehicles, improved inspection and maintenance of vehicles, especially diesel vehicles, etc. have been implemented to reduce the PM pollution in Dhaka city.

Lead is an environmental toxicant that affects virtually every system in the human body[15]. In children, lead decreases intelligence, growth and hearing; cause anemia and can cause attention and behavior problems[16]. High levels of exposure to lead can cause severe brain damage or death. Leaded gasoline used to be the primary source of lead exposure in cities in most developing countries. It accounts for 80-90% of airborne lead pollution in large cities where it is still used[17]. The removal of lead from gasoline in the single most effective way to preventing lead poisoning in children and the key to rapidly eliminating childhood lead poisoning on a large scale. The successes achieved in many countries have demonstrated that lead elimination is feasible and can be carried out through cost-effective, relatively short-term programs[18].

The problem of lead pollution from automobile in the capital city Dhaka was identified in 90s [19-23]. A study on size fractionated ($PM_{2.5}$ and $PM_{2.5-10}$) airborne particulate matter was carried at a

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semi-residential site (AECD campus), Dhaka during 1993-95. The chemical analysis of the samples clearly identified the presence of high levels of lead and their origin from gasoline. Correlation between lead and bromine; and source apportionment analysis by receptor modeling technique clearly proved that most of the fine particle (PM_{25}) lead were of gasoline origin [24]. In view of reducing the Pb concentration in ambient air especially in Dhaka city, unleaded gasoline was introduced in 1999. Just after this policy intervention lead concentration dropped to one-third of its previous value[24]. Unfortunately, the story of environmental lead in Bangladesh is still an unfinished story and further decline has been very slow. There is a substantial amount of accumulated lead in dust from earlier depositions, which returns to ambient air as wind blown resuspended dust. Elevated lead concentrations in the soil dust collected from different roads of Dhaka city reflect this [25] phenomenon. Moreover, Pb is still being used in an uncontrolled manner in paints and there are also fugitive emissions from battery manufacturing and other industries. It is, therefore, essential to continue the fight for lowering lead level in air through the removal of lead in paints and through the control of emission from suspected industries especially lead based battery industries by regulatory measures.

It is therefore; necessary to monitor the effectiveness of policy interventions and thus suggest future actions by the authority to further improve the air quality in Dhaka. In this backdrop, the aim of the present study is to look at the present situation of airborne PM and Pb pollution levels and its trends in Dhaka city.

2. METERIALS AND METHOD

2.1 Sampling and analysis: Sampling was performed from 1997 to 2005 at semi-residential site in Dhaka using a 'Gent' stacked filter sampler [26] capable of collecting air particulate samples in coarse (2.5-10 mm) and fine (2.5 mm,) size fractions. In the semi-residential area (SR), the sampler was placed on the flat roof of the Atomic Energy Centre, Dhaka (AECD) campus building. The sampler was placed so that the airflow around it was unobstructed. The samples in two fractions were collected twice in a week during the weekday.

The samples were collected on Nuclepore filters with 8 mm pores for the coarse fractions and 0.4 mm pores for the fine fractions. After sampling, the sample holder (Stack Filter Unit) was brought in the AECD (Atomic Energy Centre, Dhaka) Laboratory for retrieval of the filters and the samples were kept in airtight patrislides and stored in air-conditioned room (temperature 22°C and relative humidity of 50% approximately). The post weighing of the samples are usually done within one month of collection. Cross checking of the sampling method with Airmatrics MiniVol sampler shows that the GENT sampler has comparable sampling efficiency [27].

Twenty-four hour representative samples were collected at the sampling site twice a week in weekday only. About 100 samples (each sample comprises one fine and one coarse) were collected every year from each of the sampling station. The effective sampling time was varied between 6 and 20 h (depending on seasons) distributed uniformly over 24 h a day to avoid filter clogging and so that the flow rate remains within the prescribed limits of the sampler. This ensured proper size fractionation and collection efficiency. Intercomparison of GENT data with continuous 24 h Airmatrics MiniVol data by co-llocated sampling suggested [27] that the data generated using such time-sliced sampling procedure provides reasonably accurate average PM mass data.

2.2 PM mass and BC determination: Mass and BC for both Dhaka sites were measured in the AECD laboratory. The aerosol masses of both the coarse and fine fractions were determined by weighing the filters before and after exposure using a micro balance (METTLER Model MT5) and maintaining room temperature at 22°C and relative humidity at 50%. The aerosol samples both the coarse ($PM_{10-2.5}$) and fine ($PM_{2.5}$) fractions were equilibrated at constant humidity and temperature of the balance room before every weighing. A Po-210 (alpha emitter) electrostatic charge eliminator (STATICMASTER) was used to eliminate the static charge accumulated on the filters before each weighing. Quality control in filter weighing was ensured by using appropriate laboratory and field blanks.

The concentration of black carbon (BC) in the fine fraction of the samples was determined by reflectance measurement using an EEL type Smoke Stain Reflectometer [24]. Secondary standards of known black carbon concentrations are used to calibrate the reflectometer.

2.3 Meteorological condition: In Bangladesh, the climate is characterized by high temperatures, high humidity most of the year, and distinctly marked seasonal variations in precipitation. According to meteorological conditions, the year can be divided into four seasons, pre-monsoon (March-May), monsoon (June-September), post-monsoon (October-November) and winter (December-February) [(10)]. Winter season is characterized by dry soil conditions, low relative humidity, scanty rainfall and low northwesterly prevailing winds. The rainfall and wind speed become moderately strong and relative humidity increases in the pre-monsoon season when prevailing to southwesterly (marine). During monsoon season, the wind speed further increases and the air mass is purely marine in nature. In the post-monsoon season, the rainfall and relative humidity decreases, so also the wind speed. The direction starts shifting back to northeasterly. The meteorological data used in this study was obtained from a local meteorological station, located about 5 kilometers north of the semi-residential site.

3. RESULTS AND DISCUSSION

3.1 PM mass variation: The annual average of the air particulate matter (PM) masses, BC and Pb are shown in box and wisher plots (Figure 1) during the collection periods of 1997-2005. Here the dotted lines in the box indicate the mean value and the solid lines indicate the median values of the annual concentrations of PM_{10}, $PM_{2.5}$, BC and Pb. A line in the box represents the median of the particle mass concentration. The range of the box depicts the bounds of the 25^{th} and 75^{th} percentile of the data. The outliers in each data set are also shown as dots. It appears that there was a change in $PM_{2.5}$ concentration after 2000. From 2001 to 2002, the Government took different steps to reduce the vehicular emissions, which according to available information is the single largest source of air pollution in Dhaka city. As in Bangladesh, the vehicular emission contributes more in $PM_{2.5.}$ It may also be seen that the annual average of $PM_{2.5}$ and PM_{10} mass over the periods are much higher than the Bangladesh National Ambient Air Quality Standard and the USEPA recommended standard, which are set at 15 and 60 µg m^{-3} respectively on yearly average basis before the year 2001. From 2001 both the values are close to the standards although not meeting the standard. As the Government took different steps to reduce the PM mainly on the vehicular emissions, it would be found that (Figure 1) $PM_{2.5}$ emissions decreased significantly. But there are a few outliers, which are caused by meteorological conditions. It would be found that in case of PM mass and BC, the mean and median are almost same. But in case of Pb (Figure 1), there is a big difference between the mean and median values. This is because Pb does not come from the fixed source continuously. It comes from various sources in discrete way.

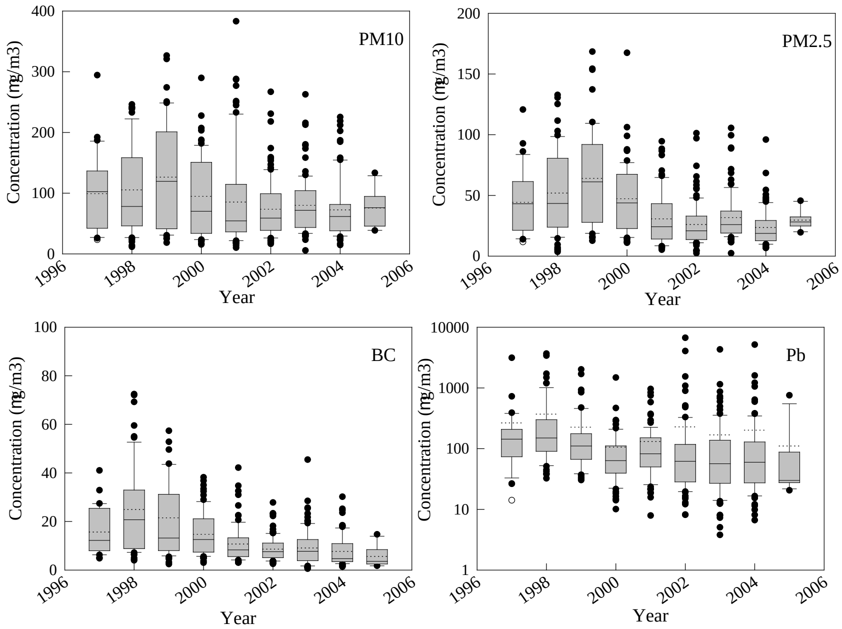

Figure 1: Box and whisker plot for annual PM masses, BC and Pb concentrations for fine during the study period.

Emissions from motor vehicles are estimated to be the single, largest source of air pollution, including fine particles and BC in Dhaka. Two major contributors to high ambient concentrations of PM_{10} in the transport sector were two-stroke engine gasoline vehicles and heavy-duty diesel vehicles. The daily air pollution levels are determined by the combined effect of emission and dispersion of pollutants and the latter, of course, depends on the weather conditions. It would be found that the fine PM is slowly decreasing from the successive year but the PM_{10} concentrations remain almost unchanged. It may be that increasing the total traffic has resulted in an increase in resuspended road dust.

The black carbon concentration is determined only in the fine fraction of the collected PM. Black carbon is produced during incomplete burning. The $PM_{2.5}$ fine particles are also mostly produced in high temperature processes and they behave in a similar way in respect of suspension time and dispersion. Hence, the effect of some weather dependent variations in $PM_{2.5}$ components can be filtered out by calculating the ratio of concentration of the component with that of elemental carbon. It can be seen that there is a change in BC concentration in the ambient air over the period. This trend is similar to that observed in the APM mass concentration variations. The production of black carbon (BC) is uniquely linked to the combustion of fossil fuel, vehicular exhausts and biomass burning.

3.2 Pb in the PM samples: Lead in PM in Dhaka was mainly due to the significant emissions of the toxic matter from motor vehicles before unleaded gasoline was introduced. These particles were agglomerates of auto exhaust emissions(24). Before 2000, there was substantial amount of Pb in air of Dhaka city(28,29). It was observed in particles containing Pb-halides, most typically Pb-bromide(24). They indicated the primary emissions of auto exhausts from vehicles using leaded petrol. However, Pb was often associated in the particles with high sulphur content, which indicated Pb-sulfates(24). There is a good relationship between Pb and Br before banning of Leaded gasoline. There is also a good correlation between BC and Pb which also support the emission of Pb from motor vehicles (Figure 2 & 3). As the association of Pb in fine fraction is high, it observed that these relationships show much better in fine samples.

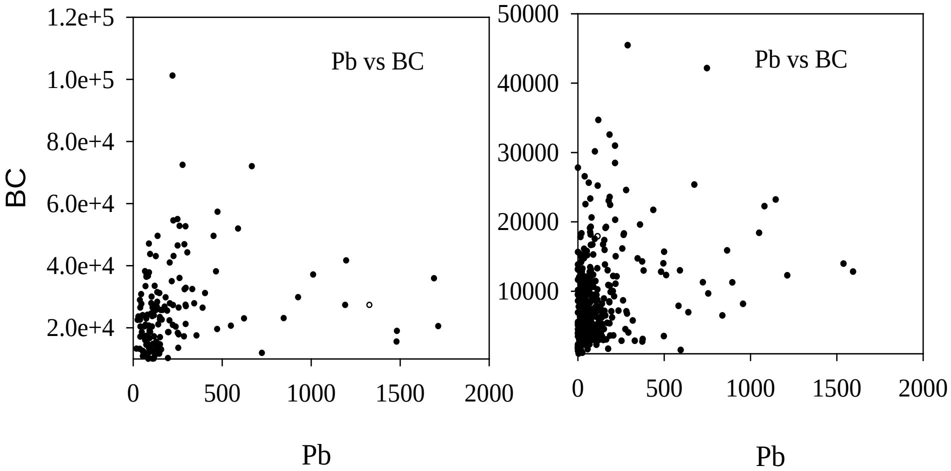

Figure 2: The relation between BC with Pb before and after banning of Pb-gasoline

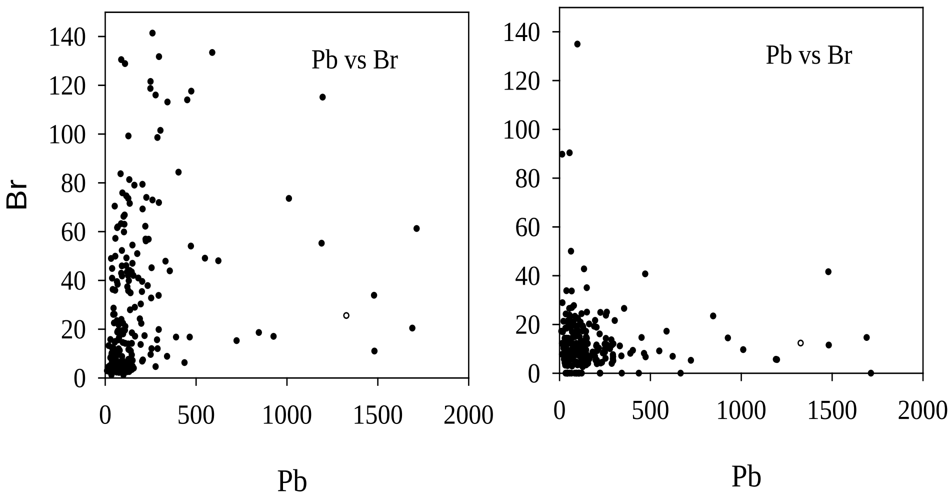

Figure 3: The relation between Br with Pb before and after banning of Pb-gasoline

After the banning of use of Pb-gasoline, the ambient air quality has improved. There is a sharp decrease of Pb/BC ratio [24] during that period which proved that the primary source of Pb was Pb-gasoline. It s been found that there is no relation between Pb vs Br after the banning of Pb-gasoline (Figure 3).

Table 1 shows the annual average of Br, Pb with standard deviation. It also contains the ratio of Br/Pb, Pb/BC and $PM_{2.5}/PM_{10}$. $PM_{2.5}$ has a decreasing trend although PM_{10} does not decrease much. The main reason for this is the resuspended road dust contribution to PM_{10} due to construction and road maintenances. It was observed that there is a decreasing trend of $PM_{2.5}$ (Figure 4) because of several policy interventions within Dhaka city[30]. It would also be found that after banning of leaded gasoline in July 1999, the concentration of Pb has decreased and it continued up to 2000. After that this concentration has not decreased. This might be due to use of Pb in the manufacturing industries specially paints, lead-acid batteries and foundries [25].

It would found that Pb has a tendency to be associated more with the fine fraction than the coarse fraction [31]. This is because the Pb particles are formed at high temperature. However, handling of lead batteries in preparation for the reclamation of the lead produces coarse particle lead [32]. It would be found that after the banning of Pb-gasoline (Figure 2), there are two different sources from where Pb may originate. From the ratio of Pb/BC, it is confirmed that Pb comes from the discrete sources. There is a slight correlation of Pb with the BC (Figure 3) after banning of leaded gasoline. As BC also originates from the Brick kiln, it may also come from there. Although, Pb was eliminated from the gasoline in Bangladesh beginning in July 1999, there may be substantial accumulated lead in the dust near roadways because lead has a very long residence time in surface soil [31]. There are also Pb-battery recycling plants in old town of Dhaka. In addition, there are Pb weights used on tires to balance the wheels[33]. They get thrown off and crushed in traffic, which is also a significant source of coarse Pb.

From the time series (Figure 4), it would be found that the concentration of PM, BC and Br have seasonal trend due to meteorological condition. Pb has also seasonal trend before banning of leaded gasoline. After banning of leaded gasoline, there are discrete peaks of Pb through the year.

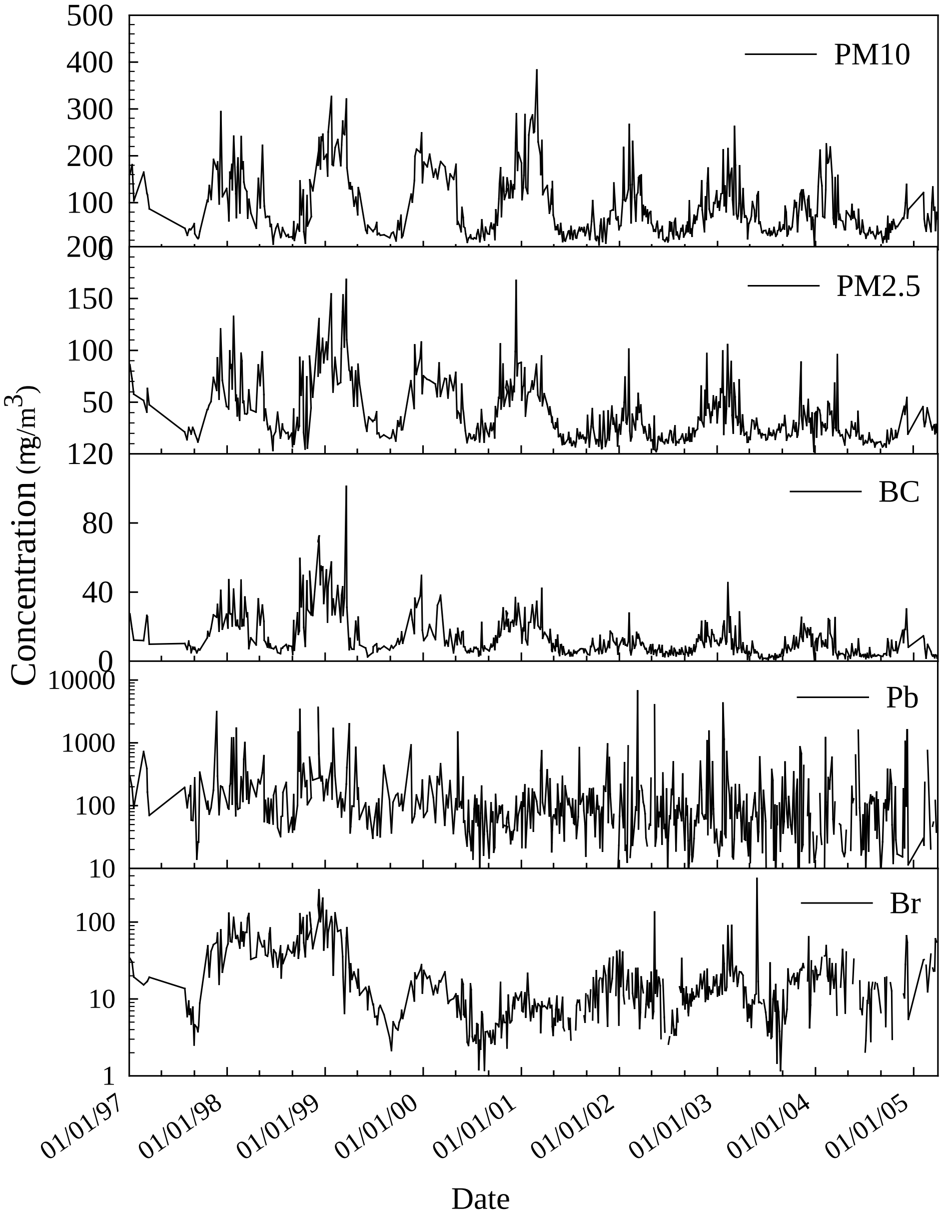

Figure 4: Time series of PM_{10}, $PM_{2.5}$, Br, BC and Pb during the study period.

Table 1.
The yearly mean with standard deviation (ng/m^3) of Br and Pb and the ratio of Br/Pb, Pb/BC and $PM_{2.5}/PM_{10}$

Year	*Br* **ng/m^3**	*Pb* **ng/m^3**	*Br/Pb* **%**	*Pb/BC* **%**	*$PM_{2.5}/PM_{10}$* **%**
1997	23.5±21.1	265±549	17.3	1.69	47.5
1998	71.3±44.9	370±644	47.1	1.44	52.0
1999	28.7±34.5	225±370	21.4	1.64	56.2
2000	7.76±5.39	106±179	13.1	0.80	56.1
2001	9.33±7.97	130±163	19.6	1.64	41.6
2002	12.8±14.6	227±784	24.5	2.81	38.2
2003	20.0±39.0	166±467	48.9	2.26	41.5
2004	13.8±14.1	198±611	45.0	2.57	35.6
2005	25.4±19.4	102±207	90.7	2.16	44.7

Unfortunately, the story of environmental lead in Bangladesh is still an unfinished story and further decline is likely to be very slow. There is a substantial amount of accumulated lead in dust from earlier depositions. Elevated lead concentrations in the soil reflect this [25]. On the other hand, Pb is still being used in an uncontrolled manner in paints and there are also fugitive emissions from battery and other industries. Therefore it could be found that after 2000 year, the Pb level has shown an increasing tendency (Figure 3) and needs to be controlled. It is, therefore, essential to continue the fight for lowering lead level in air through the removal of lead in paints and through the control of emission from suspected industries especially lead based battery industries by regulatory measures.

CONCLUSION

The results of this study show that significant reductions of fine PM were achieved because of different policy interventions. Additional policy interventions, such as paving of the roadways, removal of surface road dust by street sweeping, and other measures to reduce resuspension are necessary to reduce the PM_{10}. Although Pb was eliminated from the gasoline in Bangladesh in July 1999, from the present study, it would be found that still there is substantial amount of Pb in air. There are two reasons for this. One is, there may be accumulated Pb in the dust near roadways. The other reason is there could also be fugitive Pb emission from paints, battery and other industries especially foundries.

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REFERENCES

1. D. W. Dockery, F. E. Speizer, D. O. Stram, J. H. Ware, J. D. Spengler, and B. G. Ferris. Effects of inhalable particles on respiratory health of Chieldren. Am. Rev. Diseases *139*, 134,1989.
2. D. J. Dockery, and C. A. Pope. Acute respiratory effects of particulate air pollution. Ann Rev Public Health *15*, 107,1994.
3. N. Kato, Y. Ogawa, T. Koike, T. Sakamoto, S. Sakamoto, and R. Group. Analysis of the structure of energy consumption and the dynamics of emissions of atmospheric species related to the global change (SO_x, NO_x and CO_2) in Asia. NISTEP Report no 21, 4th Policy-oriented Research Group, National Institute of Science and Technology Policy, Tokyo.,1991.
4. Bangladesh. Country Environmental Analysis, Bangladesh Development Series Paper No: 12, www.worldbank.org.bd/bds.2006.
5. J. Heinrich, M. Pitz, W. Bischof, N. Krug, and P. J. A. Borm. Endotoxin in fine (PM2.5) and coarse (PM2.5-10) particle mass of abmient aerosols. A temporo-spatial analysis. Atmos. Environ. *37*, 3659,2003.
6. C. A. Pope, and D. W. Dockery (1999). Epidemiology of particle effects, Academic Press).
7. US-EPA. Air qualilty Criteria for Particulate Matter. 2002.
8. D. D. Cohen, M. Graham, M. Bailey, and R. Kondepudi. Elemental analysis by PIXE and other IBA techniques and their application to source fingerprinting of atmospheric fine particle pollution. Nuclear Instrumental Method (B) *109*, 218,1996.

9. D. D. Cohen. Characterization of atmospheric fine particle using IBA techniques,. Nuclear Instrumental Methods (B) *136*, 14,1998.

10. A. Salam, H. Bauer, K. Kassin, S. M. Ullah, and H. Puxbaum. Aerosol chemical characteristics of a mega-city in Southeast Asia (Dhaka, Bangladesh). Atmos. Environ. *37*, 2517,2003.

11. A. Chaloulakou, D. Assimacopoulos, and T. Lekkas. Forecasting daily maximum O3 concentrations in the Athens Basin. Environmental Monitary and Assessment *56*, 2559,1999.

12. P. Kassomenos, V. Kotroni, and G. Kallos. Analysis of climatological and air quality observations from greater Athens area. Atmos. Environ. *29B*, 3671,1995.

13. D. W. Dockery, C. A. Pope, X.Xu, J. D. Spengler, J. H. Ware, M. E. Fay, B. G. Ferris, and F. E. Speizer. An Association between Air Pollution and Mortality in six US Cities. New England Journal of Medicine *329*, 1753,1993.

14. IPCC. The third assessment report of working group I of the intergovernmental panel on climate change, technical summary, lead authors, D.L. Albritton (USA), L.G. Meira Filho (Brazil), Shanghai. 2001.

15. CDC. Preventing lead poisoning in young children: a statment by the Centers for Disease Control. Atlanta, GA: US Department of Health and Human Services, Public Health Service.1991.

16. ATSDR. Toxicological profile for lead, Atlanta, GA: US Department of Health and Human Services, Agency for Toxic Substances and Disease Registry.1999.

17. M. Lovei. Eliminating a silent threat:World Bank support for the global phaseout of lead from gasoline. Lead poisoning Prevention and Treatment: Implementing a National Program in Developing Countries. Proceedings of the International Conference on Lead Poisoning Prevention and Treatment, February 8-10, 1999. Bangalore, India: The George Foundation, 1999, p.169-180.1999.

18. M. Kojima, and M. Lovei. Environmental Concerns and Priorities in Urban Air Quality Management. Urban Air Quality Management: Coordinating Transport, Environment and Energy Policies in Developing Countries. Washington, DC; The World Bank, p. 1.2001.

19. M. Khaliquzzaman, S. K. Biswas, S. A. Tarafdar, A. Islam, and A. H. Khan. Trace Element Composition of Airborne Particulate Matter in Urban and Rural Areas of Bangladesh, AECD/AFD-CH/3-44. 1995.

20. M. Khaliquzzaman, S. K. Biswas, S. A. Tarafdar, A. Islam, and A. H. Khan (1996). Trace Element Composition of Airborne Particulate Matter at a Rural Site in Bangladesh (1993-95). Paper presented at: Presented at the RCM for the coordinated Research Programme on Applied Research on Air Pollution using Nuclear Related Analytical Techniques (Kingston, Jamaica).

21. M. Khaliquzzaman, S. A. Tarafdar, S. K. Biswas, A. Islam, and A. H. Khan. Background Ait Pollution Studies in Urban and Rural Areas in Bangladesh, IAEA-report, NAHRES-26, Vienna, International Atomic Energy Agency. 1995.

22. M. Khaliquzzaman, S. A. Tarafdar, S. K. Biswas, A. Islam, and A. H. Khan. Nature and the extent of airborne particulate matter pollution in Urban and Rural areas of Bangladesh during 1993-98. Technical report *AECD/AFD-CH/9-50,*1999.

23. M. Khaliquzzaman, S. A. Tarafdar, S. K. Biswas, A. Islam, and A. H. Khan. Trace Element Composition of Airborne Particulate Mater in a Rural Site in Bangladesh, AECD/AFD-CH/5-47, Dhaka. 1996.

24. S. K. Biswas, S. A. Tarafdar, A. Islam, M. Khaliquzzaman, H. Tervahattu, and K. Kupiainen. Impact of unleaded gasoline introduction on the concentration of lead in the air of Dhaka, Bangladesh. J. Air and Waste Management Association *53*, 1355,2003.

25. F. Ahmed, and H. Ishiga. Trace metal concentrations in street dusts of Dhaka city, Bangladesh. Atmos. Environ. *40*, 3835,2006.

26. P. K. Hopke, Y. Xie, T. Raunemaa, S. Biegalski, S. Landsberger, W. Maenhaut, P. Artaxo, and D. Cohen. Characterization of Gent stacked filter unit PM10 sampler. Aerosol Science and Technology *27*, 726,1997.

27. B. A. Begum, and S. K. Biswas. Comparison of PM collection efficiency of Gent and Airmatrics MiniVol Portable Air Sampler. Nuclear Science and Application *I14*, 79,2005.

28. M. Khaliquzzaman, and S. K. Biswas. Monitoring of Heavy Metal Pollutants in Air Particulates in Bangladesh., 15 1992.

29. M. Khaliquzzaman, S. A. Tarafdar, S. K. Biswas, and A. H. Khan (1997). Analytical Quality Assurance in Trace Element Analysis using Different Techniques in the Bangladesh Atomic Energy Commission. Paper presented at: Int. Symp. on Harmonization of Health Relader Environmental Measurements using Nuclear and Isotopic Techniques (Hydrabad, India, IAEA).

30. B. A. Begum, S. K. Biswas, and P. K. Hopke. Impact of banning of Two Stroke Engines on airborne particulate matter concentrations in Dhaka, Bangladesh. J. Air and Waste Management Association *56*, 85,2006.

31. T. M. Young, D. A. Heeraman, G. Sirin, and L. L. Ashbaugh. Resuspension of soil as a source of airborne lead near industrial facilities and highways. Environmental Science & Technology *36,* 2484,2002.

32. G. E. Glover, P. K. Hopke, S. J. Vermette, S. Landsberger, and D. R. D'Auben. Source apportionment with site specific source profiles. J Air Waste Management Association *41*, 294,1991.

33. R. A. Root. Lead loading of urban streets by motor vehicles wheel weights. Environmental Health Perspective *108*, 937,2000.