Research Journal of Recent Sciences ISSN 2277-2502
Vol. **2(ISC-2012),** 255-260 **(2013)** *Res.J.Recent.Sci.*

Characterization of Organic and Elemental Carbon in $PM_{2.5}$ Aerosols at Agra, India

Pachauri Tripti, Saraswat RK, Singla Vyoma, Lakhani Anita and Kumari Maharaj K
Dayalbagh Educational Institute, Dayalbagh, Agra 282 110, INDIA

Available online at: www.isca.in
Received 20th November 2012, revised 31st December 2012, accepted 21st January 2013

Absract

$PM_{2.5}$ samples were collected from May 2010 to April 2011 and were analyzed for OC (Organic carbon) and EC (Elemental carbon) using thermal optical transmittance (TOT) protocol. The results showed that the annual average concentration of $PM_{2.was}$ 79.7 ± 40.5 µg/m^3. In $PM_{2.5}$, OC and EC concentrations were 22.8 ± 17.1 and 3.4 ± 1.2 µg/m^3. Both OC and EC exhibited a clear seasonal pattern with highest concentration observed in winter followed by summer and monsoon which may be due to the combined effect of changes in emission rates and different meteorology in various seasons. TCA (Total carbonaceous aerosol) accounted for an averaged 50.3% of $PM_{2.5}$ mass. The annual average OC/EC ratio was 6.6 which is similar ratio for biomass burning emissions.

Keywords: Organic carbon, Elemental carbon, $PM_{2.5}$.

Introduction

Airborne carbonaceous aerosols are largest contributor to fine particles with an aerodynamic diameter smaller than 2.5 µm ($PM_{2.5}$) which have been found to be associated with human health problems causing serious respiratory and cardiovascular diseases and air quality problems such as visibility reduction. The carbonaceous matter consists of organic carbon (OC) and elemental carbon (EC). Elemental carbon is a primary pollutant emitted from incomplete combustion of fossil fuel and biomass while OC can be either released directly into the atmosphere from anthropogenic and biogenic sources (primary OC, POC) or formed within the atmosphere through gas – to – particle conversion of volatile organic compounds through photochemical reactions (secondary OC, SOC)[1,2]. The high loading of carbonaceous aerosols in fine particles has also been identified as the important factor in climate change, urban haze formation, crop production and atmospheric chemical reactions[3, 4,5]. Thus, in lieu of the importance in recent years, special attention has been drawn on carbonaceous species and studies have been carried out in a large variety of environments worldwide to understand the chemical composition and to control the mass concentration of fine particles Keeping the view in mind, the present study has been carried out to quantify the relative contribution of carbonaceous species in $PM_{2.5}$ mass, to identify the possible sources and factors affecting carbonaceous species.

Material and Methods

Description of sampling site: The study was carried out at Dayalbagh, a suburban site, which is 10 km away from the industrial sector of the Agra city where due to agricultural practices vegetation predominates. Agra (27°10′N, 78°05′E, and 169 m.s.l.) is located in the north central part of India. Two thirds of its peripheral boundaries (SE, W and NW) are bounded by the Thar Desert of Rajasthan and therefore is a semiarid area characterized by loose, sandy, and calcareous soil containing an excess of salts. Meteorologically the year is divisible into three distinct seasons; summer (March–June), monsoon (July–September) and winter (October – February). Agra is famous for Petha (famous Indian confectionary) and shoe industries which contribute to aerosol loading through their solid waste dumping and incineration. Apart from local sources, Mathura refinery, Firozabad glass industries and brick kiln factories are also situated within 40 km from Agra.

Sample collection: Sampling was carried out in Dayalbagh on the roof of Science Faculty building (12 m above the ground) in the Institute campus. All $PM_{2.5}$ were collected using Fine Particulate Sampler (Envirotech APM 550) operated at a constant flow rate of 16.6 Lmin^{-1} on pre-weighed 47 mm quartz fibre filters (Pallflex, Tissuquartz) for 24 hours from May 2010 to April 2011. Before exposure, the quartz fiber filters were pre-heated in a muffle furnace at 800°C for 3 h to remove organic impurities. The conditioned and weighed $PM_{2.5}$ filters were placed in cassettes and placed in polyethylene zip-lock bags and taken to the field for sampling to avoid contamination of the quartz filter on the way. After sampling, the $PM_{2.5}$ filter papers were removed with forceps and placed in the cassette and the cassette was wrapped with aluminum foil. The samples were wrapped in aluminum foil to prevent the degradation of organic compounds due to photo-oxidation. Before and after sampling, the filters were equilibrated in the desiccator for 24 h, and then weighed on an electronic microbalance (Mettler, Toledo) to determine the particulate matter mass.

Carbonaceous analysis and quality control: A portion of filter samples (1.5 cm^2) was cut and analyzed for OC and EC by a thermal/optical Carbon Aerosol Analyzer (Sunset Laboratory, Forest Grove, OR) using NIOSH 5040 (National Institute of Occupational Safety and Health) protocol based on Thermal Optical Transmittance (TOT) which is described in detail elsewhere[6]. The analysis procedure includes two stages. In the first stage, OC is volatilized from the sample in a non-oxidizing atmosphere (helium) as the temperature is stepped to 900 ºC (340 ºC for 60 s, 500 ºC for 60 s, 615 ºC for 60 s, and 900 ºC for 90 s). In the second stage, after a helium cooling blow of 45 s, a 2% oxygen (by volume) in a balance of helium is introduced and then the oven temperature is stepped to 910 ºC (575 ºC for 45 s, 650 ºC for 60 s, 725 ºC for 45 s, 800 ºC for 45 s, and 910 ºC for 100 s). The split time between OC and EC is obtained by measuring the time required to return the filter to its initial transmittance value. At the end of every analysis, a fixed volume loop of methane is injected automatically as an internal standard to calculate the carbon results.

Water-soluble ions analysis and quality control: Water soluble K^+ was determined by using Dionex ICS 1100 Ion Chromatograph system (Dionex Corp, Sunnyvale, CA) equipped with guard column (CG12A), analytical column (CS12A), and cation self-regenerating suppressor (CSRS 300). To extract, half of each filter was sonicated for 45 min in 1% HNO_3 and K^+ concentrations was determined by using 20mM Methane Sulfonic Acid as an eluent.

Meteorological data analysis: Meteorological data such as ambient temperature, rainfall, relative humidity, wind speed and wind direction were collected through an automatic weather monitoring system (Envirotech's Wind Monitor WM271) mounted on the roof 8m above the ground level near the sampling site. It was programmed to collect data at 1 min. interval and store them in memory to be downloaded to a computer. The wind speed varied in a range of 0.4 to 12 m/s. The post monsoon and winter months showed very clam weather conditions (81.4 % calm) while during summer period the wind speed was very high reaching to its maximum value (10 – 12 m/s) especially during dust storms. The temperature varied from 3 to 48.7°C whereas relative humidity varied between 14.4 to 91.4%. The weather was found to be cold and calm during winter months especially during fog/haze events with lowest temperature (3°C), relative humidity (83.4%) and wind speed (0.45 m/s).

Results and Discussion

Concentration levels of $PM_{2.5}$: Mass concentration is the key criteria for the assessment of air quality. At this site, the mass concentrations ranged from 24.6 to 163.6 µg/m^3 while the annual average concentration was found to be 79.7 ± 40.5 µg/m^3. The average daily concentration of PM during the measurement period exceeded the 24 hour National Ambient Air Quality Standard[7] of India which are 60 µg/m^3 for $PM_{2.5}$. About 62% of the data exceeded this standard for $PM_{2.5}$ mass. Higher level of particulate matter at this sampling site may be attributed to the combined impact of climatic conditions and anthropogenic emissions by various local sources such as vehicular exhaust, waste incineration, coal and biomass combustion as well as resuspended dust.

The seasonal average concentrations was 72.2 ± 7.1 µg/m^3 in summer, 116.6 ± 39.1 µg/m^3 in winter and 39.2± 9.3 µg/m^3 in monsoon, respectively. In winter, $PM_{2.5}$ mass concentration was 1.6 and 2.9 times higher than in summer and monsoon seasons. The levels were found to be slightly elevated during winter season due to different emission sources (increased biomass, coal and fossil fuel combustion) and meteorological conditions, dominated by cold and calm atmosphere. A wind rose (WR) plot of winter months showed low and steady wind speed (figure-1) Thus, stable and cold conditions (i.e. less dispersion and low mixing heights) during most of the days in winter months favors the ambient particles to be remain for longer time in the atmosphere. Although the average summer values are lower than the winter values, some samples show very high concentration because in addition to local sources long range transport, especially on the days of dust storm also contribute to PM levels. In monsoon, heavy rainfall occurs (780 mm) and the levels of particulate matter can be efficiently removed by wet scavenging which leads to very low concentrations of PM.

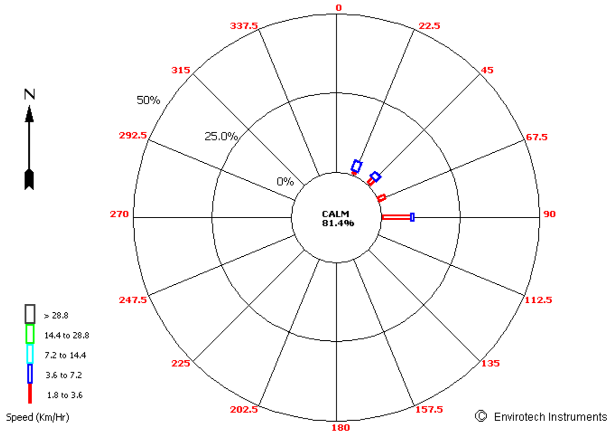

Figure-1
A wind rose plot for winter period

Concentration of carbonaceous species in $PM_{2.5}$: Seasonally averaged PM mass, TC (Total carbon), OC, EC, OC/EC ratio and TCA in $PM_{2.5}$ and TSP are summarized in table 1. Seasonally averaged $PM_{2.5}$ and TSP mass, TC, OC, EC and OC/EC ratio show strong seasonal variation with the highest concentration in winter followed by summer and monsoon. The average concentration of OC in $PM_{2.5}$ was 36.1 ± 19.1, 20.3 ± 7.1 and 8.0 ± 2.1 µg/m^3 during winter, summer and monsoon respectively. The OC mass concentration during winter season was about 1.7 and 4.5 times higher than summer and monsoon season respectively. However, the EC concentrations were found to be 5.0 ± 1.2, 2.9 ±1.1 and 1.7 ± 1.0 µg/m^3 during winter, summer and monsoon respectively. The results show that during winter, fine particles contribute more than half of OC and EC concentration to total suspended particulate matter. On applying t – test, seasonal variations in OC and EC concentrations was observed to be statistically significant. This may be attributed to different climatic conditions and emissions sources. The lower concentration of OC and EC during monsoon season can be due to wash out effect, as a significant fraction of OC is water soluble so it gets scavenged during precipitation while increased emissions from coal combustion heating and unfavorable atmospheric dispersion (e.g. low mixing height, frequent inversion etc.) leads to higher concentration during winter season.

Summarizes comparison of the carbonaceous levels with other cities of the world. In the present study, the OC concentration in $PM_{2.5}$ was found to be comparable with the two suburban sites of China, Guangzhou (22.6 µg/m^3, [8]) and Beijing (25.6 µg/m^3, [9]). The concentrations were also similar to those in urban (23.2 µg/m^3) site of Tianjin, China[5]. However, the OC concentrations were much higher than those in other areas such as Milan (9.6 µg/m^3, [10]) and Helsinki (3.0 µg/m^3, [11]). EC concentrations were found to be similar to those in Shanghai (3.1 µg/m^3, [9]) and Nanjing (2.9 µg/m^3, [12]), the two urban sites of China. However, the EC concentrations were found to be higher than those in Helsinki (1.1 µg/m^3), Milan (1.4 µg/m^3) and Yokohama (1.9 µg/m^3, [13]). The results show that the OC and EC concentrations were lower than the urban site of Kanpur. This observed variation may be due to the different source emissions.

Contribution of carbonaceous species to $PM_{2.5}$: OC is the predominant contributor to total carbon (TC) accounting for about 86.6% in $PM_{2.5}$. Total carbonaceous aerosol was calculated by the sum of EC and organic matter (OM) which was estimated by multiplying the amount of OC by 1.6 (TCA = 1.6*OC + EC) [15, 16, 17]. Table-1 shows, the seasonally averaged concentration of TCA was 62.7, 34.1 and 14.5 µg/m^3 during winter, summer and monsoon season. However, the annual concentration of TCA was found to be 40.1 µg/m^3 accounting for 50.3% of $PM_{2.5}$ mass. This indicates that the carbonaceous fraction nearly accounted for about half of $PM_{2.5}$ mass which shows that fine particles are enriched with carbonaceous species. A significant correlation between water soluble K^+ and OC abundances ($R^2 = 0.63$) supports that the biomass burning emissions are the main source of carbonaceous species. K^+ is generally used as an index of biomass burning due to its release during combustion processes. The relatively higher value of K^+/EC ratios indicates biomass burning emissions which contribute carbonaceous particles in the atmosphere. Similarly, the K^+/ OC ratio exhibits a narrow range of 0.04- 0.13 for agricultural waste burning[18] and 0.19 - 0.21 during the wheat straw burning events. Similar results have been observed in our study where the average K^+/ OC ratio was found to be 0.02 during summer months which seems appropriate at the present site as it is surrounded by agricultural fields and harvest is over by this time. After the harvest agricultural waste are burnt which contribute to K^+/ OC ratio.

Relationship between OC and EC: The origin of OC and EC can be evaluated by the relationship between OC and EC[19, 9]. A good OC – EC correlation with correlation coefficient (R) of 0.88, 0.69 and 0.76 for $PM_{2.5}$ were obtained for summer, winter and monsoon respectively (figure 2). These results indicated the presence of common dominant sources for OC and EC (biomass burning, coal combustion and motor vehicular exhaust) because the relative rates of OC and EC would be proportional to each other. Atmospheric elemental carbon is from primary anthropogenic sources and not formed by atmospheric reactions whereas organic carbon may be emitted directly as primary particles or it can also be formed by atmospheric reactions as secondary organic carbon. The ratio of OC/EC ranged from 4.3 to 9.9, 3.1 to 14.7 and 2.3 to 8.5 respectively in $PM_{2.5}$ for summer, winter and monsoon. The highest OC/EC ratio during winter months could be attributed to several reasons namely: increased residential combustion of coal and wood contribute more to OC than EC, resulting in increased emission of volatile organic precursors, the stagnant and dry meteorological conditions resulted in more SOA formation in wintertime, more semi – volatile organic compounds condensed into aerosol in lower temperature. The annual average OC/EC ratio was found to be 6.6 ± 2.8 which is similar to that reported in the literature for biomass burning emissions. The OC/EC ratio is mostly greater than 2 for most of the samples indicating the dominance of OC derived from various burning sources (coal, biomass and biofuel) and secondary aerosol formation.

Estimation of secondary organic carbon (SOC): Due to lack of an analytical technique for directly quantifying primary organic carbon and secondary organic carbon (OC/EC) min ratio method received widespread application[20]. This approach suggests samples having low OC/EC ratio contain almost exclusively primary carbonaceous compounds. So concentration of SOC can be estimated from primary carbonaceous compounds and TOC (total organic carbon) using following equation: SOC = TOC – EC × (OC/EC) pri

Where (OC/EC) pri is the ratio for the primary sources contributing to the sample. The primary ratio of OC/EC is usually affected by many factors such as types of emission sources, temporal and spatial variation, ambient temperature etc. Thus, (OC/EC) pri was represented by the observed minimum ratio of (OC/EC) min[20]. The observed values of (OC/EC) min in this study were 4.7, 2.3 and 3.1 during summer, monsoon and winter respectively.

As shown in table 3, the seasonal average concentration of SOC in $PM_{2.5}$ samples were 20.5, 5.1 and 4.1 µg/m^3 in winter, summer and monsoon respectively. The results indicated that the SOC concentration in $PM_{2.5}$ samples was 4 times higher in winter than during summer. In the present study, the high SOC concentration might be due to great coal consumption for heating in winter which causes enhanced emissions of primary carbonaceous particles and organic gases which in addition to stagnant meteorological conditions (low mixing layer height) results in SOC precursors stagnation and SOC formation[21].

Table-1
Seasonally averaged PM mass, TC, OC, EC OC/EC & TCA for $PM_{2.5}$

Sampling season	Mass ($\mu g/m^3$)	TC ($\mu g/m^3$)	OC ($\mu g/m^3$)	EC ($\mu g/m^3$)	OC/EC	TCA ($\mu g/m^3$)	N
Winter	116.6	41.1	36.1	5.0	7.1	62.7	20
Summer	72.2	23.5	20.3	2.9	6.7	34.1	17
Monsoon	39.2	9.7	8.0	1.7	5.9	14.5	15
Annual	79.7	26.3	22.8	3.4	6.6	40.1	52

Comparison of carbonaceous species with other sites

Table-2
Comparison of $PM_{2.5}$, OC and EC concentrations with other cities in the world

Location	Site	Period	OC	EC	OC/EC	References
Beijing, China	AES (suburban)	July, Nov 2002	25.6	5.6	4.6	8
Guangzhou	LG (suburban)	Dec 2002, July 2003	22.6	6.5	3.5	8
Shanghai, China	JD (suburban)	Oct 2005 – Aug 2006	17.4	3.1	5.6	9
Tianjin, China	Urban	Jan, April, July 2007	23.2	5.1	4.4	9
Milan, Italy	Urban	2002 - 2003	9.6	1.4	6.9	10
Helsinki, Finland	Urban	July 2001 - 2001	3.0	1.1	2.6	11
Nanjing, China	PMO (suburban)	Feb 2001	14.2	2.9	4.9	12
Yokohama, Japan	Urban	Sept 2007 – Aug 2008	3.75	1.94	1.9	13
Kanpur, India	Urban	Oct 2008	47.0	7.7		14
Agra, India	Sub urban	May 2010 – April 2011	22.8	3.4	6.6	Present study

Table-3
Levels of SOC estimated from OC/EC ratios

Sampling season	(OC/EC)min	POC ($\mu g/m^3$)	SOC ($\mu g/m^3$)	SOC/OC %
Winter	3.1	15.5 ± 3.8	20.5 ± 17.4	47.5 ± 23.6
Summer	4.7	15.1 ± 6.6	5.1 ± 3.3	24.5 ± 15.1
Monsoon	2.3	3.9 ± 2.5	4.1 ± 1.8	43.9 ± 21.6
Annual	3.3 ± 1.2	12.0 ± 6.9	10.8 ± 13.3	42.2 ± 23.6

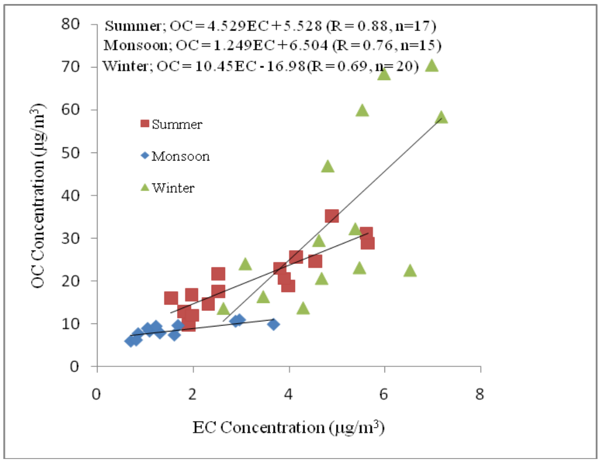

Figure-2
Seasonal correlation between OC and EC concentration in $PM_{2.5}$

Back Trajectory Analysis: In the present study, the air mass backward trajectory analysis was carried out using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single – Particle Lagrangian Integrated Trajectory (HYSPLIT) model based on the GDAS global wind field developed by NOAA/ ARL[22]. The back trajectory analysis indicate that during summer most of the air masses come from the west with a rapid moving speed carrying aeolian dust from the Thar Desert of Rajasthan (figure 3a). These strong air moving episodes during summer season results in a significant increase of aerosol particles. A five day air mass back trajectories are drawn for the 1500m height for monsoon season. The back trajectory results show the origin of southwest monsoon. Figure 3b shows the moisture laden winds originating from Arabian Sea towards north central region of India. These moisture rich air masses travelling from Indian Ocean to Indian Subcontinent are responsible for heavy rains during monsoon. However, the backward trajectory during winter season show that there is no long range transport of aerosols and the aerosol particles were anthropogenic in origin (figure 3c).

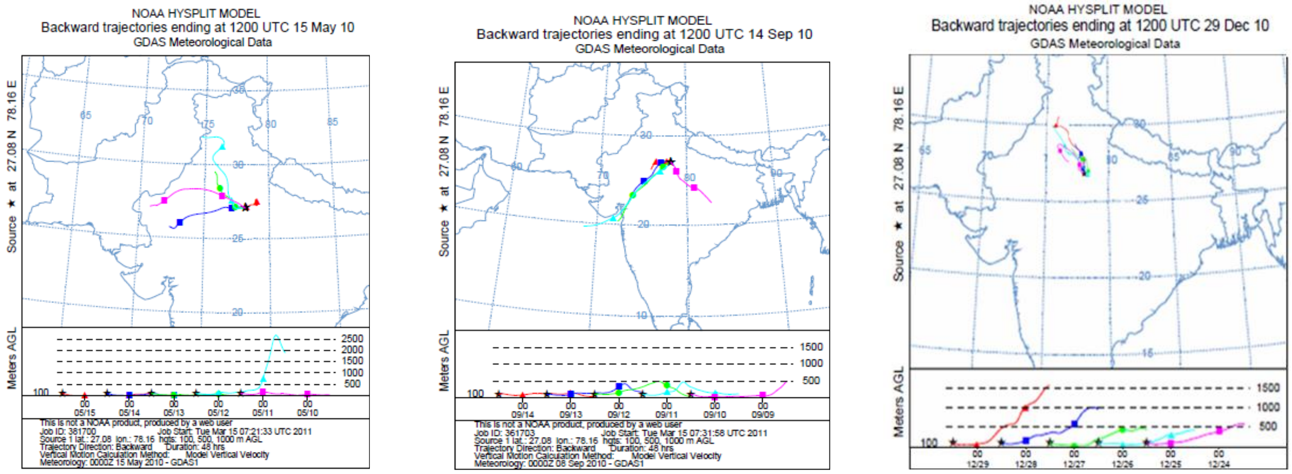

Figure-3

The typical air mass trajectories arriving at the sampling site: (a) Backward trajectory pointing to the dust source from northwest Asian desert region in summer. (b) Backward trajectory air masses originated from Indian Ocean and travelling through the Indian sub continent. (c) Backward trajectory during winter month indicating anthropogenic origin of aerosols

Conclusion

Abundance and seasonal characteristics of $PM_{2.5}$ was investigated at suburban site of Agra, India. The carbonaceous aerosol samples were collected from May 2010–April 2011. In $PM_{2.5}$, OC mass concentration during winter season was about 1.7 and 4.5 times higher than summer and monsoon season respectively. The results show that during winter, fine particles contribute more than half of OC and EC concentration to total suspended particulate matter. The average OC and EC concentrations were 22.8 ± 17.1 and 3.4 ± 1.2 $\mu g/m^3$. Both OC and EC exhibited a clear seasonal pattern with highest concentration observed in winter followed by summer and monsoon. This may be attributed to the combined effect of changes in emission rates and different meteorology in various seasons. TCA (Total carbonaceous aerosol) accounted for an averaged 50.3% of total $PM_{2.5}$ mass. The annual average OC/EC ratio was 6.6 which is similar ratio for biomass burning emissions. The SOC concentrations were found to be higher during winter season. The back trajectory analysis shows that the origin of these particles is mainly anthropogenic in nature.

Acknowledgements

The authors are thankful to Director, Dayalbagh Educational Institute, Agra; Head, Department of Chemistry for facilities provided and Department of Science and Technology, DST project No: SR/S4/AS:273/07, New Delhi for financial assistance.

References

1. Seinfeld J.H., Pandis S.N., Atmospheric chemistry and physics: from air pollution to climate change, *John Wiley and Sons, New York* **(1998)**

2. Mkoma S.L., Chi X. and Maenhaut W., Characteristics of carbonaceous aerosols in ambient PM_{10} and $PM_{2.5}$ particles in Dar es Salaam, Tanzania, *Sci. of Total Environ.*, **408**, 1308–1314 **(2010)**

3. Chameides W.L., Yu H., Liu S.C., Bergin M., Zhou X. and Mearns L., Case study of the effects of atmospheric aerosols and regional haze on agriculture: An opportunity to enhance crop yields in China through emission controls, *Proceedings of the National Academy of Science of the United States of America*, **96**, 13626–13633 **(1999)**

4. Ramanathan V., Crutzen P.J., Lelieveld J., Mitra A.P., Althausen D. and Anderson J., Indian ocean experiment: An integrated analysis of the climate forcing and effects of the great Indo-Asian haze, *J. Geophys. Res.*, **106**, 28371–28398 **(2001)**

5. Li W. and Bai Z., Characteristics of organic and elemental carbon in atmospheric fine particles in Tianjin, China, *Particuology*, **7**, 432–437 **(2009)**

6. Birch M.E. and Cary R.A., Elemental carbon-based method for monitoring occupational exposures to particulate diesel exhaust: methodology and exposure issues, *The Analyst*, **121**, 1183–1190 **(1996)**

7. Central Pollution Control Board, *National Ambient Air Quality Standards*, New Delhi, India **(1994)**

8. Feng J., Chan C.K., Fang M., Hu M., He L. and Tang X., Characteristics of organic matter in $PM_{2.5}$ in Shanghai, *Chemos.*, **64**, 1393–1400 **(2006)**

9. Feng Y., Chen Y., Guo H., Zhi G., Xiong S., Li J., Sheng G. and Fu J., Characteristics of organic and elemental carbon in $PM_{2.5}$ samples in Shanghai, China, *Atmos. Res.*, **92**, 434 –442 **(2009)**

10. Lonati G., Ozgen S. and Giugliano M., Primary and secondary carbonaceous species in $PM_{2.5}$ samples in Milan (Italy), *Atmos. Environ.*, **41**, 4599–4610 **(2007)**

11. Viidanoja J., Sillanpaa M., Laakia J., Kerminen V.M., Hillamo R., Aarnio P. and Koskentalo T., Organic and black carbon in $PM_{2.5}$ and PM_{10}: 1 year of data from an urban site in Helsinki, Finland, *Atmos. Environ.*, **36**, 3183–3193 **(2002)**
12. Yang H., Yu J.Z., Ho S.S.H., Xu J., Wu W-S., Wan C.H., Wang X. and Wang L., The chemical composition of inorganic and carbonaceous materials in $PM_{2.5}$ in Nanjing, China, *Atmos. Environ.*, **39**: 3735–3749 **(2005)**
13. Khan M. F., Shirasuna Y., Hirano K. and Masunaga S., Characterization of PM $_{2.5}$, $PM_{2.5-10}$, and $PM_{>10}$ in ambient air, Yokohama, Japan, *Atmos. Res.*, doi: 10.1016/j. atmosres.2009.12.009 **(2010)**
14. Ram K. and Sarin M.M., Day – night variability of EC, OC, WSOC and inorganic ions in urban environment of Indo – Gangetic Plain: Implications to secondary aerosol formation, *Atmos. Environ.*, doi: 10. 1016 / j. atmosenv. 2010.09.055 **(2010)**
15. Turpin B.J. and Huntzicker J.J., Identification of secondary organic aerosol episodes and quantification of primary and secondary organic aerosol concentrations during SCAQS, *Atmos. Environ.*, **29**, 3527–3544 **(1995)**
16. Cao J J, Lee S C, Ho K F, Zhang X Y, Zou S C, Fung K, Chow J C, Watson J G, Characteristics of carbonaceous aerosol in Pearl River Delta Region, China during 2001 winter period, *Atmos. Environ.*, **37,** 1451–1460 **(2003)**
17. Gu J., Bai Z., Liu A., Wu L., Xie Y., Li W., Dong H. and Zhang X., Characterization of atmospheric organic carbon of $PM_{2.5}$ and PM_{10} at Tianjin, China, *Aer. Air Quality Res.*, **10**, 167–176 **(2010)**
18. Andreae M.O. and Merlet P., Emission of trace gases and aerosols from biomass burning, *Global Biogeo. Cycles*, **15**, 955–966 **(2001)**
19. Chow J.C., Watson J.G., Lu Z., Lowenthal D.H., Frazier C.A., Solomon P.A., Thuillier R.H. and Magliano K., Descriptive analysis of $PM_{2.5}$ and PM_{10} at regionally representative locations during SJVAQS/AUSPEX, *Atmos. Environ.*, **30**, 2079–2112 **(1996)**
20. Castro L.M., Pio C.A., Harrison R.M. and Smith D.J.T., Carbonaceous aerosol in urban and rural European atmospheres: Estimation of secondary organic carbon concentrations, *Atmos. Environ.*, **33**, 2771–2781 **(1999)**
21. Park S.S., Kim Y.J. and Fung K., Characteristics of $PM_{2.5}$ carbonaceous aerosol in the Sihwa industrial area, Korea, *Atmos. Environ.*, **35**, 657–665 **(2001)**
22. Draxler R.R., Rolph G.D., HYSPLIT Model Acess via NOAA ARL READY). NOAA Air Resources Laboratory, Silver Spring, MD http://www.arl.noaa.gov/ ready/hysplit4.html 2003 **(2003)**