



Seasonal Characteristics of Water-Soluble Dicarboxylates Associated with PM₁₀ in the Urban Atmosphere of Durg City, India

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ABSTRACT

PM₁₀ samples were collected between July 2009 and June 2010 in the urban area of Durg City, India, and analyzed for water-soluble dicarboxylate species. Observed PM₁₀ concentrations varied from 94.0 to 432.1 µg/m³ with an annual average of 253.5 µg/m³. The annual average concentration of PM₁₀ was four times higher than the Indian Central Pollution Control Board (CPCB) National Ambient Air Quality Standard (Indian NAAQS) prescribed limit of 100 µg/m³. The high PM₁₀ mass concentration in Durg City are attributed to anthropogenic activities, including a high rate of construction activities, biomass combustion and mechanical disturbance of road dusts. The highest PM₁₀ value was recorded during the winter, a period characterized by extensive biomass burning, especially at night, while the lowest PM₁₀ concentration was recorded during the monsoon, when there was significant precipitation. The highest concentrations of dicarboxylates were found during winter and spring. On average, total water-soluble dicarboxylates (966 ng/m³) accounted for 0.39% of the PM₁₀ mass. Oxalate (C₂), followed by malonate (C₃) and succinate (C₄), dominated the total mass of dicarboxylates, the sum of these three species accounting for 77.5% of the total analyzed. The malonate to succinate concentration ratio calculated in this study was higher than those reported for vehicular emissions, suggesting that in addition to vehicular exhausts, secondary formation of particulate dicarboxylates via photo-oxidation also occurred. Principal component analysis (Varimax Rotated Component Matrix) revealed that secondary aerosol formation, coal and biomass combustion, and vehicular emissions were the major sources contributing to overall PM₁₀ mass in Durg City, India.

Keywords: PM₁₀; Water-soluble species; Mass concentration; Seasonal variation; Source identification.

INTRODUCTION

In recent years, many Asian countries have experienced significant economic growth leading to increased urbanization, motorization, and energy use (Colbeck *et al.*, 2010; Huang *et al.*, 2010; Pavuluri *et al.*, 2010; Shen *et al.*, 2011). As a result, poor air quality has emerged as a major environmental concern because of the lack of effective control measures. Particulate matter (PM) is a component of air pollution that adversely affects human health (Ravindra *et al.*, 2003; Tsai and Cheng, 2004; Begum *et al.*, 2008; Oh *et al.*, 2011), plays an important role in climate change (Gupta and Kumar, 2006; Duan *et al.*, 2007) and the chemistry of the atmosphere (Kouyoumdjian and Saliba, 2006; Bhaskar and

Mehta, 2010), alters biogeochemical cycles (Bari *et al.*, 2003; Shin *et al.*, 2009), affects visibility (Tsai, 2005; Tsai *et al.*, 2007; Bhaskar *et al.*, 2008), and contributes to the soiling of monuments (Tiwari *et al.*, 2009). Worldwide, urban air pollution is estimated to be responsible for 865,000 premature deaths every year, and about 60% of these deaths occur in Asia (World Health Organization, 2007). Of particular interest to human health are particles which have aerodynamic diameters less than 10 µm as these can enter into the lungs (Stone *et al.*, 2010; Ny and Lee, 2011). In 2008 World Health Organization (WHO) set 24 hr maximum thresholds for these particles (PM₁₀) at 50 µg/m³ (World Health Organization, 2008). The Clean Air Initiative for Asian Cities (CAI-Asia, 2008) has reported that in 64 cities, ambient PM₁₀ remains above WHO guidelines. Similarly, a study on urban air quality in Asia by Hopke *et al.* (2008) concluded that levels of particulate pollution in many cities were higher than standards in developed countries.

South Asia is one of the most important source regions on the globe for atmospheric particles and their precursors

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because of extensive economic development in India, similar to modern China (Pavuluri *et al.*, 2010). It was predicted in 2007 that South Asia's rate of growth in energy consumption would be the highest in the world by 2010 and that it would likely become the most important source region of anthropogenic emissions (Srivastava and Misra, 2007). Experiments over the Indian Ocean indicated a high PM loading influenced by growing levels of pollutants in India and Southeast Asia and their significant effect on radiative forcing (Lelieveld *et al.*, 2001).

The economy of India had an average gross domestic product growth of 6.1% and a high growth rate of energy consumption of 21% between 2001 and 2006 (World Bank, 2000). The mode of energy use and combustion technologies in India is very different from those of North America and Europe (Deshmukh *et al.*, 2010a). About 85% to 90% of cooking energy is from burning of biofuels like wood, agricultural waste, and dung cake, which account for 47% of the energy consumption (Tata Energy Research Institute, 2001). Even in urban areas, municipal solid wastes are mostly disposed into open landfills that are often ignited, resulting in uncontrolled field burning (Jha *et al.*, 2008). The emerging problem of air pollution as a consequence of the various developmental activities in industrial, transportation and other related endeavors in India is drawing the attention of policymakers, stakeholders, and the general public. Indeed, various stringent steps have already been taken in some megacities of India to tackle the air pollution problem, yet many small cities and towns are still to act (Gupta *et al.*, 2007).

Dicarboxylic acids (DCAs) are well recognized as ubiquitous components of water-soluble organic particles (Baboukas *et al.*, 2000; Kerminen *et al.*, 2000; Aggarwal and Kawamura, 2008; Hsieh *et al.*, 2009). They have strong impacts on radiative forcing of climate, human health, pollution transport, and visibility degradation (Wang *et al.*, 2002; Ho *et al.*, 2006). Water-soluble DCAs that are enriched in atmospheric particles have recently received greater attention because of their hygroscopic properties and the capability to act as cloud condensation nuclei (CCN) (Pavuluri *et al.*, 2010). Knowing the sources of water-soluble DCAs is important to understand the formation process of secondary organic aerosols (SOA) that act as CCN (Miyazaki *et al.*, 2009). Information regarding water-soluble DCAs is also useful for our understanding of atmospheric processes because they account for a substantial fraction of the water-soluble organic component of atmospheric aerosols, which may evolve from a series of chemical reactions occurring in the gas phase, on particle surfaces, and in cloud water (Yao *et al.*, 2004). This information is important for understanding regional air quality and global climate. DCAs are directly emitted from primary sources, such as fossil fuel combustion (Ho *et al.*, 2006; Huang and Yu, 2007), meat cooking operations (Robinson *et al.*, 2006), and biomass burning (Graham *et al.*, 2002; Falkovich *et al.*, 2005; Kumagai *et al.*, 2010; Tsai *et al.*, 2010).

However, it is widely believed, based on laboratory and field observations, that they are overwhelmingly produced

by the oxidation of various organic precursors via different photochemical pathways in the atmosphere, both in the gaseous and particulate phases, and in cloud water (Ho *et al.*, 2007; Legrand *et al.*, 2007; Miyazaki *et al.*, 2009; Kuo *et al.*, 2011). Oxalic acid (C_2) is produced from the oxidation of methylglyoxal via the formation of pyruvic and acetic acids as intermediates (Lim *et al.*, 2005). Photooxidation of 1, 3, 5-trimethylbenzene can also produce C_2 diacid (Aggarwal and Kawamura, 2008). Phthalic acid (Ph) is a photodegradation product of naphthalene (Ho *et al.*, 2007), and possibly other polycyclic aromatic hydrocarbons (Carlton *et al.*, 2006).

Durg City, the capital of the district of Durg in the state of Chhattisgarh, India, has been experiencing rapid urbanization and industrialization. Industries and vehicles are the two main sources of air pollution in Durg City (Deshmukh *et al.*, 2011a) and particulate pollution levels have increased at a very rapid rate. Data collected from the Indian Tata Energy Research Institute and Central Pollution Control Board showed that PM_{10} levels were relatively high in all Indian metropolitan cities and that they exceeded the Indian National Ambient Air Quality Standards (Indian NAAQS) and the WHO Ambient Air Quality Guidelines (Mitra and Sharma, 2002; Deshmukh *et al.*, 2010a). The existence of PM in the Indian environment is due not only to vehicular and industrial activities but also to a significant contribution from soil originated particles and re-suspended dust generated by strong winds and construction activities (Deshmukh *et al.*, 2010b, 2012a). The use of biofuels such as wood and cow dung cakes is also responsible for the high concentrations of PM in the cities of central India (Deshmukh *et al.*, 2011b).

Limited previous studies on particles associated with DCAs in India, primarily at several urban sites (Miyazaki *et al.*, 2009; Pavuluri *et al.*, 2010; Deshmukh *et al.*, 2012b), have identified the mass concentration and molecular distribution of water-soluble DCAs in the Indian urban atmosphere. In New Delhi, India, C_2 ($1.43 \mu\text{g}/\text{m}^3$) was found to be most abundant diacid species, followed by succinic (C_4 , $0.30 \mu\text{g}/\text{m}^3$) and malonic acids (C_3 , $0.05 \mu\text{g}/\text{m}^3$), and C_2 – C_4 diacids together represented an average 82% of total dicarboxylic acids (TDCAs) (Miyazaki *et al.*, 2009). C_2 was also the most abundant diacid species (58% of TDCAs) in Chennai aerosols, followed by terephthalic (t-Ph, 9% of TDCAs), C_3 (8% of TDCAs) and C_4 (7% of TDCAs) (Pavuluri *et al.*, 2010). Durg City is located within the Durg District in the eastern central region of India, and has large anthropogenic emissions, with a population of about 0.6 million (Deshmukh *et al.*, 2011a). Durg City is one of the most polluted cities in India today (Deshmukh *et al.*, 2011a) and yet, in spite of their importance in regional air quality and climate, to the best of our knowledge, no study has reported on the molecular characterization of DCAs in PM_{10} aerosols for Durg City. Reduction in ambient PM and improvement in air quality requires an understanding of the composition of ambient aerosol and its major sources. In this study, we conducted the first measurements of water-soluble dicarboxylates (DCs) in PM_{10} aerosol collected at an urban site in Durg City, India, from July 2009 to June 2010.

The mass concentrations and seasonal variability of water-soluble DCs in PM₁₀ in Durg City were studied. Varimax-rotated principal component analysis of water-soluble DCs was also performed to identify their possible sources.

EXPERIMENTAL

Site Description

The field work was conducted between July 2009 and June 2010 in Durg City (20°23'N to 22°02'N and 80°46'E to 81°58'E), India (Fig. 1) at an average altitude of ~317 m above sea level. Durg City is within the district of Durg in the state of Chhattisgarh, India, renowned for the presence of one of Asia's largest steel plants, Bhilai Steel Plant (BSP) in Bhilai. Durg is the second largest district in Chhattisgarh, covering an area of about 137 square kilometers. Durg City has a population of ~0.6 million (http://www.censusindia.gov.in/towns/chh_towns.pdf) and an average population and vehicular growth rate of 3.24% and 4.42% per year, respectively (Deshmukh et al., 2011a). The sampling site was located in front of the Government Science College in an area encompassed by educational and research institutes. The general area is crowded with a variety of industries such as steel plants, cement plants, sponge iron plants, and power plants, from which significant amounts of PM are

released into the atmosphere (Deshmukh et al., 2011a). Within a 30 km radius of the sampling site lie a large number of fertilizer plants, agro-forest plants, and electrical goods manufacturing plants that produce large quantities of fly ash and affect the local environment and surroundings via aerial dispersal. In addition, vehicular emissions from the national highways running through the college campus and the burning of wood, coal and cow dung as cooking fuel throughout the year in the adjacent village areas are also major sources of air pollution in Durg City. The closest street with significant road traffic, mainly trucks, buses, cars and 2-stroke automobiles, is about 200 m away from the sampling point. Durg City has more than 200,000 registered motor vehicles and, because of the high level of commercial activities and services within the city, the average daily traffic volume is about 2050–2120 per hour at a nearby highway (Deshmukh et al., 2011a). Additionally, about 180 trains per day pass through a major railway station located 500 m east of the sampling point. The existence of the national highway and railroads has favored industrial development in this region and the rapid population and vehicular growth rate has resulted in a significant rise in PM levels in Durg City. Emissions from small scale industries and workshops and soil derived particles are also considered as other important contributing sources.

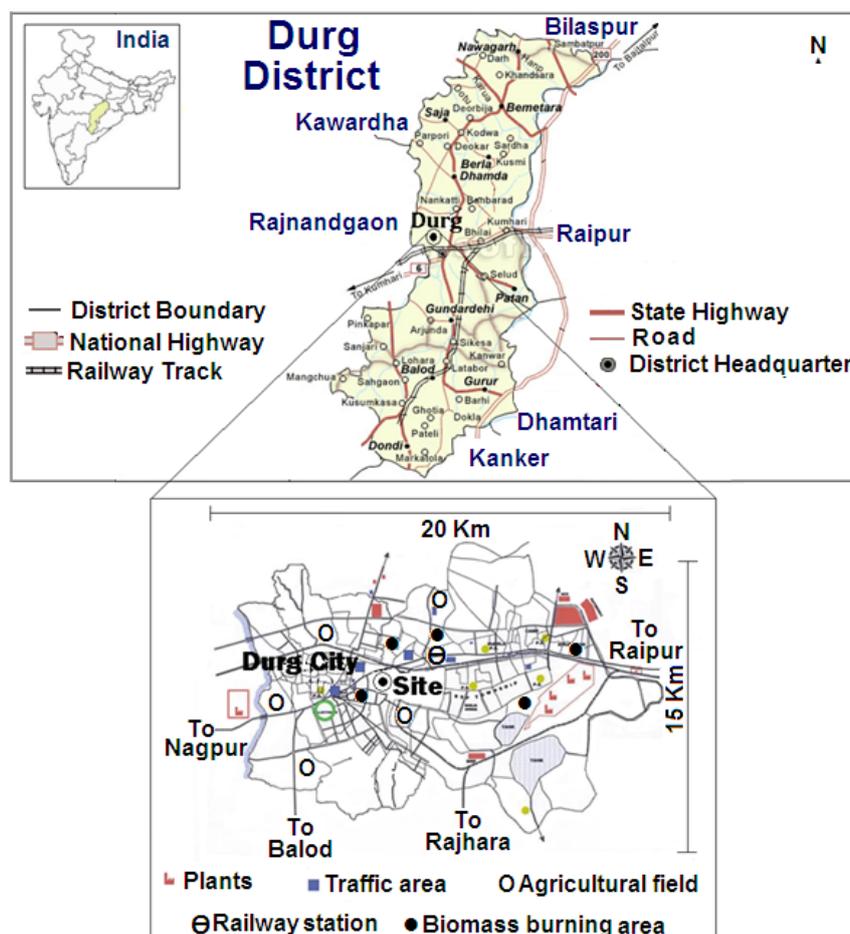


Fig. 1. Geographical location of the sampling site in Durg City, India.

PM Collection and Mass Measurement

PM₁₀ samples were collected with a Tisch Ambient Eight-Stage Cascade Impactor (Model TE 20–800, Tisch Environmental, City of Cleves, Ohio) between July 2009 and June 2010. The impactor separated particles into eight-size fractions between 0.4 µm and 10.0 µm, according to the following equivalent aerodynamic cutoff diameter, 10.0–9.0 µm (stage 0), 9.0–5.8 µm (stage 1), 5.8–4.4 µm (stage 2), 4.4–2.5 µm (stage 3), 2.5–2.1 µm (stage 4), 2.1–1.0 µm (stage 5); 1.0–0.7 µm (stage 6) and 0.7–0.4 µm (stage 7). Particulate matter of diameter less than 0.4 µm was collected in backup filter installed in the final stage of cascade impactor. Analysis was done for the eight size ranges of particles and PM₁₀ was calculated by summing the mass concentrations of eight size fractions.

The sampler was operated on the terrace of a double-storied building at a height of 15 meter above ground in a location free from obstruction. Samples were collected on pre-weighed Whatman 41 filters, conditioned at 20°C and 40% relative humidity for ~24 hrs, at a flow rate of 28.3 liters per minute. Each sampling started at 0600 hrs (local time) and ran for 24 hrs, during which the flow rate was monitored with a DryCal DCLite flowmeter (BIOS International, Butler, New Jersey). Generally, samples were collected on every third day throughout the year covering all four seasons, i.e., monsoon (July to September), winter (October to January), spring (February to March) and summer (April to June). During the study period 120 samples of PM₁₀ aerosols were collected. Field blanks ($n = 24$), i.e., filters taken to the field and installed in the sampler but through which no air was pumped, were also collected to quantify and subtract artifacts due to adsorption of gas-phase organic components onto the filters.

After the collection of the sample, the filter holder, still loaded with filters and protected in a polyethylene bag, was taken from the sampling site to the laboratory. The filters were unloaded in a dust-free room using forceps that were cleaned with ethanol and then Milli-Q water for each filter and subsequently stored in Petri dishes prior to gravimetric analysis, when filters were again conditioned at a temperature of 20°C and a relative humidity of 40% for ~24 hrs and then weighed three times using a Sartorius electronic balance (Model CP225D) with a precision of 10 µg. PM₁₀ aerosol mass (µg) was measured as the mass difference of filters before and after sampling and the concentration (µg/m³) of PM₁₀ aerosols was determined by dividing the particulate mass by total volume of air sampled (40.8 m³/day). After gravimetric analysis, the loaded filters and field blanks were stored in petri dishes placed inside an unlit freezer below 4°C to prevent loss of volatile or semi-volatile species from the sample filters.

Chemical Analysis and Quality Assurance

PM₁₀ samples were analyzed for water-soluble DCs using a method reported by Hsieh *et al.* (2009) and Tsai *et al.* (2008). To measure water-soluble DCs, a filter was placed in a polyethylene bottle, 10.0 mL of deionized water (resistivity: 18.0 MΩ-cm at 25°C, Barnstead) was added to it and the bottles were shaken (Yihder TS–500 Shaker) in

an unlit refrigerator at 4°C for 90 minutes to prevent the decomposition of extracted dicarboxylates. The liquid was then filtered through a 0.2 µm ester acetate filter and the aqueous filtrate was characterized using IC. The IC (DX–600, Dionex) was equipped with a gradient pump (Model GP50), an ASRS–Ultra anion self-regenerating suppressor, a conductivity detector (CD25), a Spectrasystem automated sampler (AS1000) with 2 mL vials, and a Teflon injection valve using a 1000 µL sample loop, in combination with an AS11 analytical column (250 mm × 4 mm I.D.) with an AG11 guard column (50 mm × 4 mm I.D.), an anion trap column (ATC–3), 5–100 mM NaOH (gradient) and 100% methanol (eluent). The flow rate was maintained at 2.0 mL/min during the water-soluble DCs analysis.

Using 0.1 ppm laboratory prepared calibration standards, the method detection limits (MDLs) were calculated as three times the SD (where SD is the standard deviation of the replicate analyses for dicarboxylate species) to determine the lowest concentration level that could be detected and be statistically different from a blank. The obtained MDLs were found to be in the range of 1.49 to 7.06 µg/L for dicarboxylate species. Table 1 shows the method detection limits, % spike recovery, relative standard deviation and field blank concentrations of individual dicarboxylate species for the quality control and assurance of the analyzed aerosol parameters. To assess spike recovery of DCs, a free mixed standard (C₂–C₆, M, F, hC4, TA, Ph) was prepared and 0.5 ppm laboratory prepared DC was added separately to blank samples. Thereafter, a complete ion chromatographic extraction procedure was followed and water-soluble DCs analyzed. Recoveries were found to be in the range of 92.8% to 106.1% while the precision, estimated from the standard deviation of repeated measurements of standards and samples, was found to be in the range of 0.86% to 2.35% (Table 1), demonstrating good repeatability of the analytical method (Tsai *et al.*, 2008). Blank filters were subjected to the same procedures followed for the extraction and preparation of sample-containing filters and it was found that, over the one year experimental period, mean concentrations of DCs were in the range of 0.59 to 12.1 ng/m³. All the reported PM mass and water-soluble DCs concentrations were adjusted to account for the field blank concentrations.

Prevailing Meteorology in Study Area

The meteorological parameters including temperature (°C), rainfall (mm), relative humidity (%), water vapor pressure (kPa), wind speed (m/s), and wind direction (degree) were collected from the Department of Agrometeorology, Indira Gandhi Agricultural University, Raipur, India. The climate is hot and humid during the monsoon, hot and dry in the spring and summer, and dry in the winter. Thus, the study region receives heavy rainfall during the monsoon but very little precipitation during summer and winter. The annual mean (± standard deviation) and ranges of the above meteorological parameters are given in Table 2. Highlights include the highest daily temperature; 42.4°C on May 24, 2010, and the lowest as 5.8°C on January 3, 2010. The week beginning April 11, 2010 was the driest (relative humidity 11%) and the week beginning August 24, 2009 was the

Table 1. Method detection limits, recovery ratio, relative standard deviation and field blank concentrations of water-soluble dicarboxylate species analyzed by ion chromatography.

Dicarboxylate species	Method detection limit (MDL, $\mu\text{g/L}$)	Recovery ratio (% , $n = 3$)	Relative standard deviation (RSD, %)	No. of field blanks below detection limit ($n = 24$)
Oxalate (C_2)	7.06	99.8	2.35	2
Malonate (C_3)	1.49	106.1	0.86	3
Succinate (C_4)	4.20	103.7	1.48	6
Glutarate (C_5)	2.43	101.1	1.27	8
Adipate (C_6)	2.28	92.8	1.68	8
Maleate (M)	1.58	99.2	0.93	4
Fumarate (F)	2.45	104.3	1.34	6
Phthalate (Ph)	5.28	103.2	1.83	18
Malate (hC_4)	2.41	97.0	1.39	4
Tartrate (TA)	4.89	92.8	1.68	2

^aAverage value.

Table 2. Descriptive statistics of meteorological variables and PM_{10} concentration between July 2009 and June 2010 in Durg City, India.

Meteorological variables	Annual ($n = 120$)	Monsoon ($n = 30$)	Winter ($n = 40$)	Spring ($n = 20$)	Summer ($n = 30$)
	Mean \pm Stdev ^g (Range)				
T^a ($^{\circ}\text{C}$)	26.4 ± 5.5 (5.8–42.4)	25.8 ± 0.8 (21.6–30.8)	19.0 ± 3.4 (5.8–30.5)	24.6 ± 3.8 (10.4–38.2)	31.5 ± 2.5 (19.7–42.4)
RH^b (%)	64.0 ± 19.0 (11.0–96.0)	83.0 ± 6.0 (58.0–96.0)	67.0 ± 8.0 (22.0–95.0)	47.0 ± 10.0 (15.0–89.0)	42.0 ± 18.0 (11.0–86.0)
RF^c (mm)	$1272.2^i \pm 54.7$ (0.0–335.2)	$998.3^i \pm 95.2$ (5.2–335.2)	$96.5^i \pm 12.5$ (0.0–52.0)	$11.4^i \pm 2.2$ (0.0–6.8)	$166.0^i \pm 19.1$ (0.0–62.6)
VP^d (kPa)	1.9 ± 0.8 (0.7–3.4)	2.9 ± 0.2 (2.7–3.4)	1.5 ± 0.6 (0.7–3.0)	1.3 ± 0.2 (1.0–1.7)	1.8 ± 0.6 (1.0–2.8)
WS^e (m/s)	4.8 ± 3.1 (1.0–2.7)	6.2 ± 3.2 (3.0–12.7)	2.1 ± 1.3 (1.0–6.2)	3.4 ± 1.8 (1.4–6.6)	7.5 ± 1.8 (4.1–10.2)
PWD^f	SW & W	SW & W	NW & NE	NW & SW	SW & W
Particulate matter	Annual ($n = 120$)	Monsoon ($n = 30$)	Winter ($n = 40$)	Spring ($n = 20$)	Summer ($n = 30$)
	Mean \pm Stdev ^g (Range)				
PM_{10} concentration ^h	253.5 ± 99.4 (94.0–432.1)	158.0 ± 79.2 (95.1–350.5)	356.1 ± 37.7 (237.2–432.1)	273.9 ± 79.6 (192.5–432.2)	198.5 ± 31.6 (94.0–230.4)

^aTemperature; ^bRelative humidity; ^cRainfall; ^dWater vapor pressure; ^eWind speed; ^fPrevalent wind direction; ^gStandard deviation; ^hUnit in $\mu\text{g}/\text{m}^3$; ⁱTotal rainfall.

most humid (relative humidity 96%). The daily highest wind speed was recorded as 12.7 m/s on July 19, 2009 and the lowest as 1.0 m/s on January 17, 2010, while the daily highest rainfall was 335 mm on July 11, 2009. The daily highest water vapor pressure was 3.4 kPa on July 24, 2009 and lowest was 0.7 kPa on January 21, 2010.

During the mid-monsoon month of August, the winds are stronger, moist, and from the western regions surrounding India. From October, the wind patterns start shifting in direction from southwest to northeast, while from April to June the winds are from the southwest and northwest. Wind rose plots generated using a WRPLOT View (Lakes Environmental, Canada) show that winter and spring are relatively calm (a condition of no wind or a wind with a speed less than 0.5 m/s) seasons (71.4% and 66.3%) compared with summer (17.9%) and monsoon (16.8%) (Fig. 2).

RESULTS AND DISCUSSION

Mass Concentration of PM_{10} Aerosols

The annual mean (\pm standard deviation) and ranges of PM_{10} values collected between July 2009 and June 2010 in Durg City, India, are shown in Table 2. The PM_{10} concentration ranged from 94 to 432 $\mu\text{g}/\text{m}^3$ with an average and standard deviation of $253.5 \pm 99.4 \mu\text{g}/\text{m}^3$. The observed 24 hrs PM_{10} mass concentrations in Durg City were significantly higher than the prescribed limits of 100 $\mu\text{g}/\text{m}^3$ (NAAQS) and 50 $\mu\text{g}/\text{m}^3$ (WHO) and also slightly higher than reported for other urban sites in India (Tiwari et al., 2009; Bhaskar and Mehta, 2010; Deshmukh et al., 2010a; Singh et al., 2010).

The seasonal concentrations of PM_{10} aerosols in Durg City are given in Table 2. The table shows that there is a

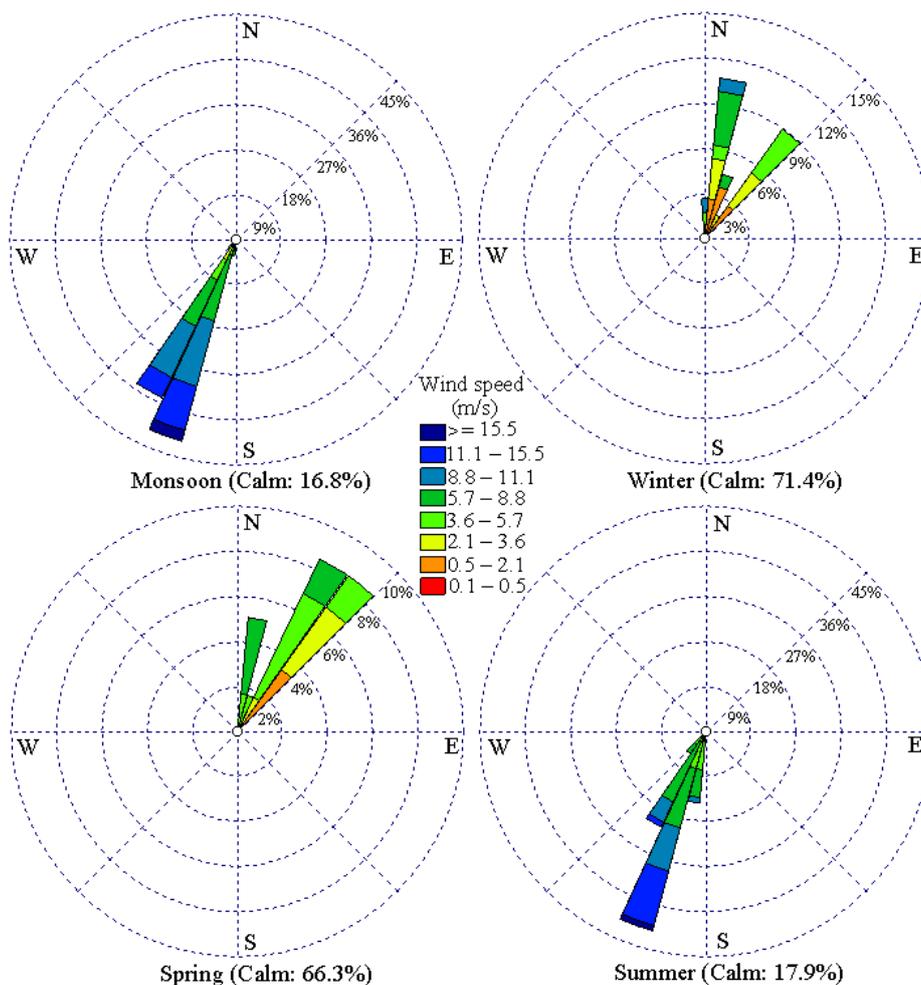


Fig. 2. Wind rose plots for all four seasons between July 2009 and June 2010.

higher mass concentration of PM_{10} during the winter and spring than the summer and monsoon seasons. This seasonal variation is influenced by both anthropogenic activities and meteorological conditions. Chhattisgarh is known as the rice bowl of Central India and significant agricultural activity in and around Durg City in winter. Biomass burning contributes to the higher level of PM_{10} . The main crops are rice and wheat and agricultural or crop waste, in the form of cereal straws and woody stalks, are generated during winter and find use as thatching for rural homes, as the major fuel for cooking, and for heating. These materials combined with the use of other combustibles such as wooden blocks, fire wood, and cow dung cake, lead to the emission of significant quantities of ash into the atmosphere. Also, some fractions of the unutilized waste are burned in the field.

The prevailing winter meteorological conditions also affect seasonal variation in mass concentrations of PM_{10} . The lower temperature ($5.8^{\circ}C$) and wind speed (1.0 m/s) during winter causes the mixing height to fall and the PM concentration increases to a maximum. The wind rose plot for winter (Fig. 2) shows that the atmosphere was very stable due to low and steady wind speed with 71.4% calm conditions. Thus, stable, cold and dry meteorology on most

winter days favored the prolonged life of PM in the ambient air.

Lower concentrations of PM_{10} were found during summer, which can be attributed not only to the lower volume of biomass burning but also to a very high mixing height due to the high temperature ($42.4^{\circ}C$) and wind speed (10.2 m/s). The monsoon winds of July, August and September are characterized by heavy rain; the lower concentrations of PM_{10} observed during the monsoon were due to the cleaning of streets and dust control offered by heavy precipitation (998.3 mm). Similar trends have also been observed at other locations in India (Ravindra *et al.*, 2003; Gupta and Kumar, 2006; Deshmukh *et al.*, 2012a).

Molecular Composition of Dicarboxylates (DCs)

The mean (\pm standard deviation) and ranges of DCs analyzed in PM_{10} samples in Durg City, India is given in Table 3. The concentrations of total dicarboxylates (TDCs) varied between 353 ng/m^3 and 1862 ng/m^3 with a mean concentration of 966 ng/m^3 . The relative abundance of individual DCs to the PM_{10} mass and TDCs analyzed in PM_{10} samples was found to be in the range of 0.21 to 0.69% with an average of 0.39% (Fig. 3). The annual mean concentrations of TDCs in Durg were found to be

Table 3. Descriptive statistics of concentration (ng/m^3) of water-soluble dicarboxylate species in PM_{10} between July 2009 and June 2010 in Durg City, India.

Dicarboxylate species	Annual ($n = 120$)	Monsoon ($n = 30$)	Winter ($n = 40$)	Spring ($n = 20$)	Summer ($n = 30$)
	Mean \pm Stdev ^b (Range)				
Oxalate (C_2)	642.7 \pm 264.5 (299.2–1171.3)	455.9 \pm 139.2 (299.2–775.9)	911.6 \pm 221.5 (496.6–1171.5)	628.8 \pm 215.6 (327.1–1126.3)	480.3 \pm 99.1 (343.7–733.4)
Malonate (C_3)	44.1 \pm 32.5 (4.8–118.6)	7.6 \pm 3.6 (4.8–14.5)	54.2 \pm 33.5 (21.5–118.6)	79.8 \pm 17.1 (54.2–100.7)	43.5 \pm 15.2 (18.9–75.8)
Succinate (C_4)	42.4 \pm 31.5 (5.1–112.1)	8.0 \pm 3.8 (5.1–15.4)	76.1 \pm 22.0 (38.2–112.1)	52.8 \pm 14.7 (31.2–86.7)	25.1 \pm 9.2 (13.5–39.8)
Glutarate (C_5)	16.4 \pm 12.3 (1.6–77.9)	2.4 \pm 1.1 (1.6–4.8)	28.0 \pm 22.8 (3.5–62.7)	25.4 \pm 22.7 (5.5–77.9)	8.8 \pm 3.0 (4.5–12.9)
Adipate (C_6)	15.9 \pm 9.2 (1.4–76.9)	2.3 \pm 1.0 (1.4–4.4)	27.4 \pm 22.7 (3.1–62.1)	24.8 \pm 22.6 (5.1–76.9)	8.1 \pm 2.7 (4.1–11.8)
Maleate (M)	41.7 \pm 22.2 (5.8–150.0)	9.1 \pm 4.2 (5.8–17.3)	63.3 \pm 28.4 (27.6–114.6)	55.5 \pm 38.6 (19.5–150.0)	36.5 \pm 21.8 (9.6–72.1)
Fumarate (F)	39.2 \pm 25.5 (8.3–147.4)	11.3 \pm 2.6 (8.3–16.0)	60.3 \pm 44.1 (20.2–147.4)	27.9 \pm 14.3 (12.7–65.7)	46.5 \pm 30.2 (10.0–88.4)
Phthalate (Ph)	39.9 \pm 22.4 (7.0–196.2)	10.5 \pm 4.1 (7.0–18.4)	53.4 \pm 24.9 (21.9–98.0)	62.3 \pm 51.9 (21.7–196.2)	28.2 \pm 13.1 (11.4–48.7)
Malate (hC_4)	38.2 \pm 29.4 (7.4–131.7)	11.8 \pm 5.7 (7.4–22.8)	61.7 \pm 45.3 (13.8–131.7)	73.0 \pm 30.2 (29.2–113.7)	50.0 \pm 31.6 (12.4–99.0)
Tartarate (TA)	37.6 \pm 11.6 (6.4–123.7)	10.0 \pm 4.5 (6.4–18.7)	59.3 \pm 35.1 (20.9–123.7)	54.6 \pm 20.2 (31.7–107.4)	25.1 \pm 19.7 (12.4–92.4)
TDCs ^a	966.1 \pm 415.1 (352.9–1862.1)	528.7 \pm 167.8 (352.9–905.1)	1395.3 \pm 263.7 (898.2–1862.1)	1085.1 \pm 269.3 (705.5–1591.1)	752.1 \pm 172.6 (500.4–1187.1)
Concentration ratios of dicarboxylate species	Annual ($n = 120$)	Monsoon ($n = 30$)	Winter ($n = 40$)	Spring ($n = 20$)	Summer ($n = 30$)
	Mean \pm Stdev ^b (Range)				
C_2/C_4	27.2 \pm 21.5 (4.5–76.7)	60.9 \pm 11.8 (43.5–76.7)	13.6 \pm 6.7 (4.5–29.8)	13.2 \pm 8.4 (7.4–36.1)	21.0 \pm 6.6 (10.8–30.4)
C_3/C_4	1.2 \pm 0.8 (0.2–4.3)	1.0 \pm 0.1 (0.9–1.4)	0.7 \pm 0.5 (0.2–1.5)	1.6 \pm 0.3 (1.0–2.0)	2.0 \pm 1.2 (0.8–4.3)
hC_4/C_4	1.4 \pm 1.0 (0.2–6.5)	1.5 \pm 0.1 (1.4–1.6)	0.8 \pm 0.5 (0.2–1.6)	1.4 \pm 0.5 (0.5–1.8)	2.1 \pm 1.6 (0.9–6.5)
F/M	1.2 \pm 1.0 (0.3–7.2)	1.3 \pm 0.2 (0.9–1.5)	1.0 \pm 0.6 (0.3–1.9)	0.6 \pm 0.2 (0.4–0.8)	1.6 \pm 1.7 (0.4–7.2)
Ph/ C_6	4.6 \pm 4.2 (0.6–424.2)	4.7 \pm 0.3 (4.2–5.1)	4.5 \pm 1.6 (0.8–13.7)	6.5 \pm 7.9 (0.6–24.2)	3.4 \pm 0.8 (1.3–4.3)

^aTotal dicarboxylates; ^bStandard deviation.

comparable to those reported in Chinese cities (904 ng/m^3 , Ho *et al.*, 2007), and the East China Sea (850 ng/m^3 , Mochida *et al.*, 2003a) but lower than those reported in the Sea of Japan (1200 ng/m^3 , Mochida *et al.*, 2003b) and higher than those reported in Gosan, Jeju Island (660 ng/m^3 , Kawamura *et al.*, 2004), Tokyo Japan (355 ng/m^3 , Kawamura and Yasui, 2005), Chichi-jima Island (170 ng/m^3 , Mochida *et al.*, 2003b), Western North Pacific (200 ng/m^3 , Mochida *et al.*, 2003a) and Sapporo, Japan (406 ng/m^3 , Aggarwal and Kawamura, 2008).

Oxalate (C_2) was the most abundant dicarboxylate, with a concentration in the range 299 to 1171 ng/m^3 and an average concentration of 643 ng/m^3 . The relative abundance of C_2 ranged from 0.10 to 0.44% as a percentage of the

PM_{10} mass and 38.6 to 89.0% as a percentage of TDCs, with an average of 0.27% and 69.3%, respectively. The predominance of C_2 has also been observed in samples collected from different regions including urban Tokyo (Sempere and Kawamura, 2003), Chinese megacities (Wang *et al.*, 2002), Hong Kong (Yao *et al.*, 2004), Southern India (Pavuluri *et al.*, 2010), New Zealand (Wang and Shooter, 2004), Amazonia (Kundu *et al.*, 2010), rural northeastern United States (Sundar Raman *et al.*, 2008) and Europe (Legrand *et al.*, 2007) and is emitted as the final product of photooxidation of aromatic hydrocarbons, isoprene, ethylene, and acetylene (Lim *et al.*, 2005) as well as from fossil fuel combustion (Sorooshian *et al.*, 2007) and biomass burning activities (Huang and Yu, 2007). The second and third

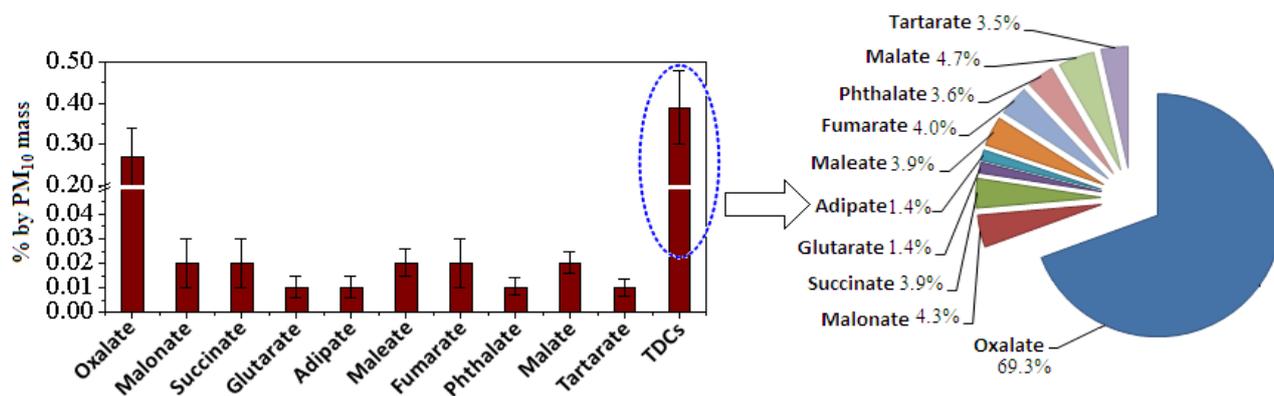


Fig. 3. Percentage contribution of dicarboxylate species in overall PM₁₀ mass and total dicarboxylates.

most abundant DCs were malonate (C₃) and succinate (C₄) followed by maleate (M) and phthalate (Ph). Concentrations of C₃ and C₄ ranged from 4.8 to 119 ng/m³ (average 44 ng/m³) and 5.1 to 112 ng/m³ (average 42 ng/m³), respectively, while relative abundance to the PM₁₀ mass and TDCs analyzed was 0.02% and 4.3%, respectively for C₃, and 0.02% and 3.9%, respectively for C₄. The high abundance of C₃ and C₄ in TDCs was also reported in urban PM in Sapporo, Japan (Aggarwal and Kawamura, 2008), Chinese megacities (Ho *et al.*, 2007) and Southern India (Pavuluri *et al.*, 2010). Maleate (42 ng/m³) and phthalate (40 ng/m³) contributed 3.9% and 3.6% of the TDCs concentration in PM₁₀. The predominance of maleate over fumarate (39 ng/m³) suggested that the PM₁₀ in Durg City contains relatively fresh oxidation products of aromatic hydrocarbons emitted from local pollution sources (Aggarwal and Kawamura, 2008). Phthalic acid (Ph), directly emitted from combustion sources and/or generated by atmospheric degradation of aromatic hydrocarbons such as naphthalene (Kawamura *et al.*, 2004), was found at concentrations in the range 7.6 to 196 ng/m³ with an average concentration of 40 ng/m³. Phthalate was between 1.0 and 12.3% (average 3.6%) of the TDCs analyzed in PM₁₀ samples. The higher molecular weight DCs, i.e., glutarate (C₅) and adipate (C₆), in PM₁₀ were less abundant. Concentrations of C₅ and C₆ ranged from 1.6 to 78 ng/m³ (average 16 ng/m³) and 1.4 to 77 ng/m³ (average 16 ng/m³), respectively, and in sum they accounted for 2.8% of the TDCs analyzed.

Seasonal Distributions of Dicarboxylates (DCs)

The seasonal average concentrations and ranges of DCs analyzed in PM₁₀ are given in Table 3. Concentrations were highest in winter and spring and then summer and were lowest in the monsoon season. The formation of inversion layers due to the low mixing height and low wet deposition are believed to be the major reasons for the high concentrations of dicarboxylates in winter (Pathak *et al.*, 2003). Conversely, high temperature and wind speed, the washout effect from rainfall and the partition between gas and particle phase are the major reasons for low concentrations of dicarboxylates in summer and monsoon seasons. The TDCs concentrations varied between 898 and 1862 ng/m³ in winter, 706 and 1591 ng/m³ in spring, 500 and 1187 ng/m³

in summer, and 353 and 905 ng/m³ in the monsoon season, with an average of 1395 ng/m³, 1085 ng/m³, 752 ng/m³ and 529 ng/m³ in the respective seasons. Winter concentrations were higher than those in Chinese cities (904 ng/m³, Ho *et al.*, 2007), Gosan, Jeju Island (499 ng/m³, Kundu *et al.*, 2010) and Chennai, India (695 ng/m³, Pavuluri *et al.*, 2010), as were spring concentrations compared with those reported at Gosan, Jeju Island (735 ng/m³, Kundu *et al.*, 2010).

Summer concentrations were also higher than those reported for Chennai, India (503 ng/m³, Pavuluri *et al.*, 2010) but were lower than those reported in Chinese cities (892 ng/m³, Ho *et al.*, 2007) and Gosan, Jeju Island (784 ng/m³, Kundu *et al.*, 2010). The most abundant dicarboxylate, C₂, was present in higher concentrations in winter and spring followed by summer and monsoon. It is reported that C₂ may be produced by the photo-oxidation by ozone of cyclo-olefins and aliphatic diolefins emitted from motor vehicles (Nolte *et al.*, 2001). Since traffic emissions remained relatively constant all year round, this seasonal cycle might be more related to other emission sources such as coal combustion and the prevailing meteorological conditions during winter. Coal, the combustion efficiency of which is very low, is commonly used in central India for heating and cooking purposes and the high concentration of C₂ in winter may be due to increased emissions from coal-fueled heating sources. As noted, summer and monsoon are favorable to atmospheric diffusion and pollutant dispersion due to high wind speed and precipitation. Hence, the low level of C₂ in these seasons may be explained either by less burning activity or a rainfall that allows efficient scavenging of water-soluble particulate species (Wang *et al.*, 2008).

The second most abundant dicarboxylate, C₃, was also present in higher concentrations in winter and spring followed by summer and monsoon. On average, C₂ was the most abundant of the detected DCs in winter, being 12 and 16 times more abundant than C₄ and C₃, respectively, while in summer and spring C₂ was followed by C₃ and then C₄ diacids. C₄ diacid can serve as a precursor of C₃ diacid during photochemical reactions in summer (Kawamura *et al.*, 1996; Ho *et al.*, 2007) and a significant fraction of C₃ diacid may be secondarily produced in the atmosphere by photochemical degradation of C₄ diacid during summer (Kundu *et al.*, 2010). This relationship between C₄ and C₃

may explain the molecular composition of diacids reported in winter aerosols from the temperate zone, including in Tokyo (Kawamura and Ikushima, 1993) and on Jeju Island (Kawamura *et al.*, 2004), where C_4 is more abundant than C_3 in winter but vice versa in summer.

Concentrations of phthalate, a tracer of anthropogenic emissions (Kawamura and Yasui, 2005), were higher in spring and winter, and lower in summer and monsoon. High values for this tracer in spring and winter indicate that primary emission from significant coal burning during these seasons is the possible source of phthalate in Durg City. The least abundant DCs i.e., C_5 and C_6 , were also present in higher concentrations in spring and winter, and made a minor contribution to the TDCs detected in PM_{10} particles.

Concentration Ratios of Selected Dicarboxylate Species

The annual and seasonal concentration ratios of selected dicarboxylate species analyzed in PM_{10} in Durg City, India, are shown in Table 3. Previous studies suggested that water-soluble DCs are produced in the atmosphere via photochemical reactions involving unsaturated hydrocarbons of fatty acids as well as their oxidation products. Some saturated dicarboxylic acids such as C_6 diacid may be generated from cyclic olefin via O_3 and OH radical reactions in the gas phase (Atkinson and Arey, 2003). In contrast, unsaturated dicarboxylic acids are considered to be generated via photochemical oxidation of other precursors such as aromatic hydrocarbons and methylcycloalkenes (Legrand *et al.*, 2007).

In many previous studies, C_2/C_4 mass ratios were used to evaluate photochemical aging and track the possible sources of diacids (Ho *et al.*, 2007; Legrand *et al.*, 2007; Kundu *et al.*, 2010). Kundu *et al.* (2010) reported a C_2/C_4 ratio of 3.5 in Brazil and suggested that this low number indicated that dicarboxylates were likely emitted not only from biomass burning but also from incomplete combustion of fossil fuels. In the present study, the C_2/C_4 ratio ranged between 4.5 and 76.7 (average 32.1) and the higher ratios in the range are attributed to photochemical production of C_2 from C_4 within biomass burning plumes. The C_2/C_4 ratios in monsoon, winter, spring and summer seasons were 69.0, 13.6, 13.2 and 21.0, respectively.

Because C_3 diacid is produced by photochemical oxidation of C_4 diacid in the atmosphere, the C_3/C_4 mass ratio can be an informative indicator in understanding the production of dicarboxylic acids and the source strength of anthropogenic and biogenic precursors in the atmosphere (Yao *et al.*, 2004; Ho *et al.*, 2007; Hsieh *et al.*, 2008). A C_3/C_4 mass ratio of 0.3 to 0.5 indicates an overwhelming contribution from traffic emissions for these diacids (Wang *et al.*, 2002; Yao *et al.*, 2004) whereas a ratio greater than 1.0 indicates an origin from photochemical reactions associated with elevated concentrations of oxidants and therefore a sourcing from secondary reactions (Kawamura and Ikushima, 1993; Kawamura and Sakaguchi, 1999). In the current research, the C_3/C_4 ratio ranged from 0.2 to 4.3 with an average of 1.2, indicating significant photochemical modification of the aerosol over Durg City. The annual mean C_3/C_4 mass ratio found in this study was higher than

that reported for Beijing, China (0.53, Ho *et al.*, 2010) and in a suburban area of southern Taiwan during non-episodic and PM episodic pollution (0.95, Hsieh *et al.*, 2008). In the latter study, nighttime ratios during non-episodic pollution were lower (0.92) than daytime (0.98), suggesting that primary vehicle exhaust is the dominant source of particulate DCs (Hsieh *et al.*, 2008). The difference in nighttime and daytime ratios (0.76 and 1.15) was greater during PM episodic pollution, suggesting a greater contribution of secondary sources to particulate DCs formation during daytime (Hsieh *et al.*, 2008). Hsieh *et al.* (2007) also reported an average C_3/C_4 mass ratio of 0.60 and 0.79 during nighttime and daytime, respectively, in the suburban area of southern Taiwan and suggested a greater input from secondary sources to particulate DCs formation.

The C_3/C_4 ratios in monsoon, winter, spring and summer seasons in the current study were 1.0, 0.7, 1.6 and 2.0, respectively. This result suggests that in addition to primary exhaust, secondary formation of particulate DCs by photo-oxidation reactions may have also occurred. The low C_3/C_4 mass ratio during winter indicates origin from vehicle exhaust (Kerminen *et al.*, 2000) during this season whereas the high ratio in summer indicates an origin from secondary reactions (Yao *et al.*, 2004). The lower chain length DCs are more water-soluble and wash out more effectively during the monsoon season and results may suggest that a significant fraction of C_3 dicarboxylate is formed as secondary product in the atmosphere by photochemical degradation of C_4 dicarboxylate. The C_3/C_4 mass ratio found in this study during winter is higher than that reported for northern and southern Chinese cities (0.59 and 0.63, respectively) (Ho *et al.*, 2007) but lower than that reported for Beijing, China (1.14, Huang *et al.*, 2005). The C_3/C_4 mass ratio found in this study during summer is also higher than that reported for northern and southern Chinese cities (0.61 and 1.12, respectively) (Ho *et al.*, 2007) and for Beijing (1.15) (Huang *et al.*, 2005). In northern and southern Chinese cities, but especially in southern China, higher C_3/C_4 mass ratios were found during summer than during winter, indicating the importance of secondary formation of dicarboxylic acids (Ho *et al.*, 2007).

The ratio of hC_4 (malate) to C_4 (succinate) varied between 0.5 and 6.5 with an average of 1.5 during the entire study period and was 1.5, 1.1, 1.4, and 2.1 during monsoon, winter, spring and summer respectively. The higher hC_4/C_4 ratio in summer shows that hC_4 can be photochemically generated from C_4 via hydroxylation reaction (Ho *et al.*, 2007). Maleic acid (M) can be isomerized to fumaric acid (F) in the presence of sunlight. Therefore, this isomerization reaction is enhanced in summer (Pavuluri *et al.*, 2010). Kundu *et al.* (2010) reported a higher F/M ratio in summer and attributed it to the enhanced isomerization reaction under seasonally intensified solar radiation. Adipic (C_6) and phthalic (Ph) acids may be produced by the oxidation of anthropogenic cyclohexene and aromatic hydrocarbons such as naphthalene, respectively (Schauer *et al.*, 2002). A Ph/ C_6 ratio of 4.74 obtained in this study is lower than those reported for other urban areas in India (Miyazaki *et al.*, 2009).

Influence of Meteorology in Ambient Pollutants Concentration

The residence time of pollutants in the atmosphere and the formation of secondary pollutants is controlled by the rate of emission of those pollutants from their source into the air, the presence of oxidants, and by meteorological factors. Meteorological factors such as temperature, relative humidity, rainfall, mixing height and wind speed all play an important role in determining the air pollution level in the atmosphere (Bhaskar and Mehta, 2010).

The annual and seasonal means and ranges of the above meteorological variables were summarized earlier and shown in Table 2. The relationship of PM₁₀ and water-soluble DCs with meteorological variables was investigated by correlation analysis and results are given in Table 4. PM₁₀ mass showed significant negative correlation with temperature ($p < 0.01$). As shown in Table 2, PM₁₀ concentration was higher at low temperatures during winter and lower at high temperature during summer. Higher temperature increases the boundary layer height, which increases the ventilation effect and further leads to faster dispersion of PM, thus resulting in a decrease in PM concentrations. The converse is true with low temperature. Similar results were found in previous studies conducted in other Indian cities (Bhaskar et al., 2008; Tiwari et al., 2009; Deshmukh et al., 2011b). Photochemical processes play an important role in controlling the atmospheric concentrations of DCs (Ho et al., 2006). As shown in Table 4, DCs (C₂, C₃, C₄, F, hC₄) were well correlated with temperature during the study period, indicating that dicarboxylates are produced in the atmosphere by photochemical chain reactions involving unsaturated hydrocarbons or fatty acids as well as their oxidation products (Ho et al., 2006). High relative humidity acts to lower the inversion layer and mixing height and thereby limits dispersion of PM in the atmosphere, leading to a higher PM mass. Therefore, most DCs (C₂, C₃, C₄, C₅, F, and hC₅) were well correlated with relative humidity ($p < 0.01$, $p < 0.05$) during the study period. The effect of rainfall on PM₁₀ and water-soluble DCs was also investigated.

As Table 2 shows, PM₁₀ and water-soluble DCs were negatively correlated with rainfall, showing that the scavenging of PM from the atmosphere due to precipitation

is an important removal mechanism (Sahu et al., 2004). Strong winds also act to flush pollutants out of the system while low winds allow pollution levels to increase (Chaloulakou et al., 2003). In the present study, maximum levels of PM₁₀ concentration were observed at low wind speed during the winter and spring seasons, and minimum levels were seen at high wind speed during summer and monsoon (Table 2). The increase in wind speed causes an increase in ventilation effects, thereby dispersing the PM₁₀ in the ambient air and causing a decrease in the observed PM₁₀ mass concentrations. PM₁₀ and water-soluble DCs showed significant negative correlation ($p < 0.01$) with wind speed and indicated the predominance of local sources.

Source Identification by Principal Component Analysis

Principal component analysis (PCA) was used to identify possible sources of water-soluble DCs in PM₁₀ particles in Durg City, India. We conducted varimax rotation to determine principal components (PCs) having an eigenvalue larger than 1. Each PC shows correlation of each variable as loadings. A factor loading greater than 0.70 was considered to be important in this study. Varimax-rotated PCA was performed using the statistical software, SPSS (Statistical Package for the Social Sciences, version 16.02) using the data set ($n = 120$) over the entire study period, and results are shown in Table 5. Three PCs (PC 1, PC 2 and PC 3) were extracted. PC 1 explains 48% of the variance and shows the heavy loading of PM₁₀ mass and five DCs (C₂, C₃, C₄, F and hC₄). These species in PC 1 correspond to the various photochemical sources along with the formation of secondary aerosol in the airborne particulates. The sources identified in this study are similar to those reported in the literature (Hsieh et al., 2007, 2008; Pavuluri et al., 2010). PC 2 explains 22% of the variance and shows the heavy loading of C₆ and Ph DCs, indicating that these are mainly derived by photooxidation of anthropogenic hydrocarbons. The coal which is used in domestic and industrial activities is the major source of these species in the atmosphere. PC 3 explains 18% of the variance and shows significant loadings of tartarate and maleate. Traffic emission related VOCs are converted to tartaric and maleic acid and these two species can be regarded as the species to index traffic pollution (Hsieh et al., 2009). These three factors explained almost

Table 4. Correlation coefficients of PM₁₀ and dicarboxylate species with meteorological variables in Durg City, India.

	Temperature	Relative humidity	Rainfall	Wind speed
PM ₁₀	-0.66^a	-0.10	-0.41	-0.62^a
Oxalate (C ₂)	0.59^a	0.86^a	0.10	-0.59^a
Malonate (C ₃)	0.52^b	0.92^a	-0.15	-0.51^b
Succinate (C ₄)	0.78^a	0.69^a	0.26	-0.50^b
Glutarate (C ₅)	0.23	0.53^b	0.02	-0.16
Adipate (C ₆)	0.08	0.08	0.18	-0.10
Maleate (M)	0.10	0.17	-0.02	-0.49^b
Fumarate (F)	0.49^b	0.94^a	-0.09	-0.41^b
Phthalate (Ph)	0.38	0.01	0.12	-0.60^a
Malate (hC ₄)	0.69^a	0.50^b	0.09	-0.48^b
Tartarate (TA)	0.07	0.29	0.02	-0.45^b

^aCorrelation is significant at 0.01 level (two tailed); ^bCorrelation is significant at 0.05 level (two tailed).

Table 5. Varimax-rotated PCA analysis of water-soluble dicarboxylate species in PM₁₀ in Durg City, India.

Variables	PC 1	PC 2	PC 3
PM ₁₀	0.94	0.10	0.09
Oxalate (C ₂)	0.94	0.19	−0.11
Malonate (C ₃)	0.98	0.16	−0.24
Succinate (C ₄)	0.80	0.29	0.20
Glutarate (C ₅)	0.11	0.76	−0.06
Adipate (C ₆)	−0.38	0.72	−0.22
Maleate (M)	−0.22	−0.23	0.86
Fumarate (F)	0.76	0.18	−0.08
Phthalate (Ph)	0.05	0.91	0.28
Malate (hC ₄)	0.68	−0.28	0.11
Tartarate (TA)	0.30	0.19	0.92
Eigenvalue	10.2	5.3	2.2
Cumulative eigenvalue	10.2	15.5	17.7
% Total variance	48.4%	22.2%	18.2%
Cumulative % variance	48.4%	70.6%	88.8%

Extraction method: Principal Component Analysis.

Rotation method: Varimax with Kaiser Normalization.

88.8% of the variance of the total data set (Table 5), demonstrating that the PCA analyses worked well for the obtained data set by extracting three factors or sources that were the strong correlation with overall PM₁₀ mass concentrations in the urban area of central India.

SUMMARY AND CONCLUSIONS

PM₁₀ samples were collected in an urban area of Durg City, India between July 2009 and June 2010 and analyzed for water-soluble dicarboxylate species. The annual mean concentration of PM₁₀ was found to be 253.5 µg/m³, significantly higher than the NAAQS of India and WHO prescribed limits. The concentration of PM₁₀ was higher in winter followed by spring and summer and lower in monsoon. The higher concentration observed during winter can be attributed to increased biomass burning coupled with low temperature and wind speed which lead to lower mixing height and poor dispersion conditions. The concentration of total dicarboxylates was between 353 ng/m³ and 1862 ng/m³ (0.2 and 0.7% of the PM₁₀ mass) with an average of 966 ng/m³ (0.4% of the PM₁₀ mass). Oxalate was the most abundant dicarboxylate (69.3% of TDCs) followed by malonate and succinate. Concentrations of DCs were higher in winter and spring due to biomass burning activities combined with the effects of low wind velocity and low mixing layer height and lower in summer and monsoon. The higher C₂/C₄ mass ratios found in this study indicated photochemical production of C₂ from C₄ within biomass burning plumes while the higher C₃/C₄ mass ratios suggest that in addition to primary exhaust, secondary formation of particulate dicarboxylates via photo-oxidation reactions occurred. Source identification via PCA identified photochemical and secondary aerosol, anthropogenic emissions, and vehicular emissions and explained 88.8% of the variance. It is anticipated that increases in population, traffic, industrialization and energy consumption in the future may further increase the PM emission load and

adversely affect the already poor air quality in Durg City, with adverse effects on the public health and social wellbeing of this city. Therefore, it is recommended that effective control measures should be implemented in order to reduce at least the local atmospheric pollution in Durg City and to protect human health.

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