

Stable (C and N) isotopic composition of bulk aerosol particles over India and northern Indian Ocean

Rajesh Agnihotri¹, T.K. Mandal², S. G. Karapurkar¹, Manish Naja³, Ranu Gadi⁴, Y. Nazeer Ahammed⁵, Animesh Kumar⁶, T. Saud², M. Saxena²

¹National Institute of Oceanography (*Council for Scientific and Industrial Research*), Dona Paula, Goa-403 004, India

² National Physical Laboratory, Radio and Atmospheric Sciences Division, New Delhi 110012, India

³Aryabhata Research Institute of Observational Sciences (ARIES), Manora Peak, Nainital 263 129, Uttarakhand, India.

⁴ Indira Gandhi Institute of Technology, GGS Indraprastha University, Delhi-110006, India

⁵ Department of Physics, Yogi Vemana University, Kadapa, Andhra Pradesh, India

⁶National Environmental Engineering Research Institute, Nehru Marg, Nagpur, India

Abstract

Atmospheric carbonaceous aerosols of South Asian origin have received immense concerns in the Anthropocene owing to their plausible role in the observed regional to inter-continental scale climate anomalies. Tracking plausible sources and alterations during their transport (secondary processes) are keys to understanding their net influence on regional climate. Here, we report elemental concentrations of C and N (TC and TN), their isotopes ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$) and TC/TN ratios of bulk carbonaceous aerosol particles over northern Indian Ocean [Bay of Bengal (BOB) and Arabian Sea (AS)] collected as a part of integrated campaign of Aerosol Radiation Budget (ICARB) during March-May, 2006. In order to understand typical sources, we also measured same parameters of (i) aerosols emitted from typical bio-fuels conventionally burnt in north and northeastern India and (ii) aerosols particles present in ambient air over selected Indian cities during pre-monsoon season. Bulk aerosols over AS are characterized by significantly higher TC/TN ratios ($\sim 50 \pm 10$) compared to aerosol over Indian cities (5.6 ± 2.6) as well as over BOB (6.8 ± 12.5), most likely due to having significant inorganic carbon contributed by mineral dust. $\delta^{13}\text{C}$ of aerosols over AS and BOB do not show significant variation ($-25.6\text{‰} \pm 0.6$, $-26.5\text{‰} \pm 0.8$; $n=24$ and 21 respectively), however $\delta^{15}\text{N}$ values showed a conspicuous difference between the two branches of northern Indian Ocean ($10.6\text{‰} \pm 2.7$ over BOB and $1.4\text{‰} \pm 3.3$ over the AS). Depleted $\delta^{15}\text{N}$ of aerosols over AS can be interpreted in terms of significant mixing of isotopically depleted nitrogenous compounds (NH_3 and NO_{xs}) emitted from the underlying (denitrifying) waters.

Keywords: Aerosols, Arabian Sea, Bay of Bengal, pre-monsoon, Carbon isotopes, Nitrogen isotopes

1. Introduction

Atmospheric carbonaceous aerosols are micron size solid particles suspended in gaseous-liquid media present in lower tropospheric air. They are by-products of various natural (viz. mineral dust, sea salt spray, forest vegetation pollen etc.) and anthropogenic energy production activities (partial and complete combustion of live vegetation, biomass and fossil fuel). Aerosols, in addition to just scattering/ absorbing incoming solar radiation also provide active sites for the uptake of several chemical species and trace gases. These secondary uptakes on primary aerosols can result in their modified radiative as well as hydro-chemical properties. Due to lack of detailed knowledge of these effects, net role of carbonaceous aerosols remain highly uncertain in determining net climatic influence especially in South Asian region (Ramanathan et al., 2001; IPCC, 2007). Biomass and fossil fuel combustion are thought to be major anthropogenic sources of carbonaceous aerosols causing atmospheric smog and haze (Novakov et al., 2000), while industrial emissions and agricultural soils are also potential sources of anthropogenic aerosols. Atmospheric aerosols originating from thickly populated and rapidly developing South Asia have been assumed to affect regional as well as global climate potentially on intra-seasonal to inter-annual timescales (Goswami et al., 2006). Several recent researches have attempted to discern and quantify contributions from burning of live biomass and fossil fuel combustion proportion of anthropogenic sources of aerosols in South Asia in order to assess their net radiative and hydrological impact on regional climate using different approaches (Mayol-Bracero et al., 2002; Gustafsson et al., 2009; Kripa Ram & Sarin, 2010; Sudheer & Sarin, 2008). For instance, during the Indian Ocean Experiment (INDOEX), obtained data on aerosol samples collected over Indian Ocean showed that bulk of carbonaceous aerosols originated mainly from biomass/ bio-fuel burning (~75%) and had high concentrations of potassium (K) (Guozzatti et al., 2003; Kulshrestha et al., 2001). These studies suggested that biomass-biofuel burning in south Asia has large influences on concentration as well as composition of aerosols originating from these regions. During pre-monsoon period, low level northerly and north-easterly winds carry pollutants adhered to the ambient atmospheric aerosols from south-east Asia to oceanic regions. However, source characterization of carbonaceous aerosols over northern Indian Ocean AS and BOB especially during pre-monsoon period (February-May) is still not well studied using isotopic data. Marine processes occurring in both branches of northern Indian Ocean i.e. the AS and the BOB are capable of influencing overlying air-masses and thereby aerosol chemical composition. It is known that the AS acts as a source of several important greenhouse gases (e.g. CO₂, CH₄, N₂O etc.) (Naqvi et al., 2006; and references therein). In contrast, the BOB acts as a 'sink' for several greenhouse gases as it presents thickest fresh water lens produced by high riverine/ fresh water

discharge from major river-systems e.g. Ganga- Brahmaputra from northeastern and Krishna-Godavari from central India and Bangladesh (DileepKumar et al., 1996). These contrasting features of these marine regimes are thus expected to have an influence on composition of primary aerosols present in overlying air-mass.

Stable isotopes of C and N ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) have been well exploited for decoding and tracking biogeochemical processes in oceanography and limnology. In case of aerosols; Court et al. (1981) was probably the first one to suggest utility of C isotopes in identifying urban air particulate sources. $\delta^{13}\text{C}$ in conjunction with ratios of black carbon /total carbon (BC/TC) were utilized to deduce biomass burning emissions from different tropical ecosystems (Cashier et al., 1986). Widory et al. (2004) used combination of C and Pb isotopes to identify and semi-quantify aerosol sources such as road traffic versus industrial emissions in a typical urban environment of Paris city. $\delta^{13}\text{C}$ together with $\delta^{15}\text{N}$ of aerosols was used discerning C-3 versus C-4 vegetation type (Martinelli et al., 2002, Kelly et al., 2005). Likewise, Moore (1977) pioneered the use of $\delta^{15}\text{N}$ of NH_4^+ and NO_3^- in aerosols to investigate atmospheric N cycling. Thereafter, $\delta^{15}\text{N}$ has been widely used several studies (Heaton et al., 1997; Kawamura et al., 2004; Russell et al., 1998; Turekian et al., 1998; Widory, 2007; Yeatman et al., 2001a&b).

However, exploitation of C and N isotopes of aerosols for characterizing possible potential sources has been limited in Southeast Asia. Here, we report a detailed stable isotopic study of bulk carbonaceous aerosol particles over northern Indian Ocean (the BOB and AS) collected during pre-monsoon season (March-May) of year 2006, as a part of integrated campaign of Aerosol Radiation Budget (ICARB) programme. To characterize characteristic sources isotopically, we also collected total of 19 aerosols samples from different urban locations of India such as Bhubneshwar, Delhi, Nanital, Anantpur Nagpur and Goa including areas representing heavy road traffic (Delhi-Mathura highway) and industrial emission. In addition, we also measured $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of aerosols emitted from combustion of the typical biomasses and bio-fuel (coal) conventionally burnt for energy production in the north and northeast India. This study thus provides the first detailed stable isotopic benchmark data of atmospheric carbonaceous aerosols from India and nearby seas. Major outcome of this study is to find a clear difference between aerosols over AS and BOB plus aerosols over selected Indian cities in terms of significantly higher TC/TN ratios most likely owing to having significant inorganic carbon contributed by mineral dust and significantly lower $\delta^{15}\text{N}$, indicative of significant mixing of isotopically depleted N compounds (NH_3 , NO_{xs}) emitting from underlying denitrifying AS.

2. Sample collection and Methodology

Aerosol samples were collected using high volume sampler (HVS, Envirotech, APM -410) with quartz fiber filter as collection substrate from the oceanic environments of BOB and AS, *on-board* cruise *ORV Sagar kanya* (SK-223) during pre monsoon season (March 2006 to May 2006). The sampler was operated at an average rate of $\sim 1.12 \text{ m}^3 \cdot \text{min}^{-1}$, which was calibrated in laboratory before and confirmed after the cruise. The preconditioned (by first heating to 100°C and then desiccating) collection substrates were tare-weighed using a Sartorius five decimal balance (sensitivity of $\pm 0.01 \text{ mg}$) and kept in separate polythene bags, sealed and taken to ship, along with the sampler. The collection substrate was mounted carefully on the sampler installed at the front end of the ship deck (~ 10 meters above sea level), which enables the sampling of marine air from the front uncontaminated by the ship exhaust. The aerosol-laden substrates were sealed individually and brought to laboratory for analysis. From the quartz filter of size $20.3 \times 25.4 \text{ cm}$ containing aerosols, $5 \times 5 \text{ cm}$ portions were cut and kept for analyses of C and N isotopes. From these portions, actual isotopic analyses were made using two to three aliquots of 11 mm diameter circular (hole-punched) sub-portions. These sub-portions were placed in clean Tin cups (size $12 \times 4.5 \text{ mm}$) and made round pellets which were then combusted in a *Euro-Vector* Elemental Analyzer (EA) coupled with a *Delta V plus* stable isotope mass spectrometer (*Thermo*®) in a continuous flow mode. During combustion of packed cups (at 1050°C), flash combustion takes place in the presence of high purity O_2 . Evolved gases are passed through reduction furnace (at 670°C) filled with activated metallic copper that reduces the oxides of nitrogen (NO_x) to N_2 . Dry helium (5.5grade) is used as carrier gas for sample introduction. Gas streams are then passed through a moisture-trap filled with magnesium perchlorate (MgClO_4) ensuring removal of moisture and separated using gas chromatograph (at 50°C) before being introduced to open split of the *Delta* IRMS. $^{13}\text{C}/^{12}\text{C}$ and $^{15}\text{N}/^{14}\text{N}$ ratios of sample gases are then measured with respect to automated injections of standard (calibrated) CO_2 and N_2 gases. Final results are expressed as $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ relative to PDB and atmospheric N_2 standards respectively, and defined as-

$$\delta^{13}\text{C} \text{ and } \delta^{15}\text{N} = ((R_{\text{sample}} - R_{\text{standard}}) / R_{\text{standard}}) \times 1000 \text{ -----(1)}$$

where $R = ^{13}\text{C}/^{12}\text{C}$ and $^{15}\text{N}/^{14}\text{N}$

Primary calibration of the elemental and isotopic data was accomplished using a laboratory standard (ϵ -Amino-n-Caproic acid, $\text{C}_6\text{H}_{15}\text{NO}_2$; *ACA*) supplied by Prof. Mark A. Altabet (SMASST-UMASSD)

USA. Reported $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ values of ACA are 4.6‰ and -25.3‰ respectively. TC and TN contents in samples were also calculated from a calibration curve made of four ACA standards ranging from 1-4 μmole for N and 6-24 μmole for C. A laboratory standard COD was also used as control for analytical precision. We also ran several aliquots of aerosol filter in duplicates (total 5); their reproducibilities were within 0.5‰ for N and 0.3‰ for C isotopes. Similarly overall uncertainties for TC and TN concentrations were better than 2 and 3.5% respectively. We also estimated blanks from same size aliquots of pre-combusted quartz filters. N contents of blanks were almost negligible ($\leq 50\text{mv}$), while C contents of blank filter aliquots varied between 0.4-0.72 μmoles which is about 10-12 times smaller than the minimum analyte signal of C typically measured. TC concentrations have been corrected for mean blank filter value (i.e. 0.6 μmole); whereas we have not made blank corrections in the isotopic data of $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$.

Experimental setup adopted for the collection of carbonaceous aerosol particles being emitted from various bio-fuels (Table 3) is described in Parashar et al. (2005). Generally, ~ 50 gm of bio-fuel substance was burnt under controlled conditions typically for time range of 8-18 minutes. Emitted aerosols were collected on pre-combusted quartz fiber filters installed on high volume aerosol sampler operating at $1.12\text{ m}^3\cdot\text{min}^{-1}$ flow rate.

3. Results

Data of elemental concentrations (TC and TN) and isotopic ratios ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) of bulk aerosols collected at different locales of northern Indian Ocean (AS and BOB) during pre-monsoon period are presented in Table 1. Same parameters measured for aerosols collected over different continental locations over India during pre-monsoon season (February to March, 2009) are presented in Table 2. As aerosol sampling over Panaji (Goa) was accomplished using low volume filtration system (kept at NIO 7th floor roof, $\sim 21\text{m}$ above ground) to investigate isotopic signatures, isotopic data of these samples are reported in the Table 2. Table 3 contains data of aerosols emitted from combustion of some typical vegetation types, normally cultivated and burnt on tropical soil of India. Table 3 also contains data of aerosols emitted during coal combustion. Spatial variations of these parameters over land and marine realms (AS and BOB) are shown pictorially in Figs.-1 and 2 while Fig.-3 shows plots of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of aerosols over BOB and AS against their respective TC and TN concentrations. Spatial variability of the data is discussed region-wise in the following sections.

3.1 Variations in TC, TN concentrations, TC/TN wt. ratios and isotopes ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$) of bulk aerosol particles present over oceanic realms (BOB and AS)

TC and TN of aerosols over BOB vary in the ranges 8.4 ± 13.3 and $1.8\pm 0.9 \mu\text{g.m}^{-3}$ respectively, while $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of aerosols over BOB vary in a narrow range of -25.6 (Average) $\pm 0.6\%$ (SD) and $10.8\pm 2.5\%$, respectively. TC/TN wt. ratios of aerosols over BOB generally show lower values except southern BOB and overall vary in a range 6.8 ± 12.5 (Fig.-2c). TC of aerosols over AS show significantly higher values and vary in the range $35.2\pm 9.7 \mu\text{g.m}^{-3}$ compared to TN concentrations that vary in the range $0.72\pm 0.2 \mu\text{g.m}^{-3}$. Thereby, TC/TN wt. ratios of aerosols over AS show considerable higher values and vary in the range 50 ± 10 (Fig.-2c). $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of aerosol particles over AS vary in relatively narrow ranges i.e. $-26.5\pm 0.8\%$ and $1.4\pm 3.4\%$ respectively. $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of aerosols show weak positive correlations with respective TC and TN concentrations in case of aerosols over BOB (Fig. 3a,b), while poorer statistical relationships were observed in case of aerosols over AS (Fig. 3c,d). Likewise, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of aerosols over BOB show positive correlation while aerosols over AS do not any relationship between the two (Fig. 3e,f).

3.2 TC, TN, TC/TN wt. ratios and $\delta^{13}\text{C}$ & $\delta^{15}\text{N}$ of bulk aerosol particles present over Indian cities

$\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of aerosol particles present over continental stations (Table 2) vary in ranges $-25.4\pm 0.7\%$ and $8.1\pm 3\%$ respectively. $\delta^{15}\text{N}$ of aerosol over coastal city Panaji (Goa) station show much depleted values $4\pm 2\%$ compare to those over other cities. TC and TN over these cities (except Goa) vary in the ranges 26.4 ± 12 and $6\pm 5 \mu\text{g.m}^{-3}$. TC/TN wt. ratios of aerosols over these cities vary in a range 5.6 ± 2.6 . $\delta^{13}\text{C}$, $\delta^{15}\text{N}$ and TN of aerosol particles over heavy road traffic and industrial areas also fall in same range i.e. $\sim -25.2\%$, $\sim 9\%$ and $\sim 4\mu\text{g.m}^{-3}$, respectively. However, corresponding TC of aerosols from heavily polluted sources were considerably higher by a factor of 2-4, resulting in higher TC/TN wt. ratios (Table 2).

3.3 TC, TN, TC/TN wt. ratios, $\delta^{13}\text{C}$ & $\delta^{15}\text{N}$ of bulk aerosol particles emitted from typical bio-fuels

Aerosols emitted from combustion of typical vegetation types that are normally cultivated and burnt on tropical soil of India (Table 3) show narrow range for $\delta^{13}\text{C}$ i.e. $-27.9\pm 1.1\%$. However, $\delta^{15}\text{N}$ show larger variability with values ranging $13.2\pm 4.2\%$. Leguminous plants e.g. Arhar and

Mustard clearly have lower $\delta^{15}\text{N}$ values compared to typical C-3 type biomass due to their ability to fix atmospheric N (Table 3). TC/TN wt. ratios vary in a broad range $\sim 20 \pm 10$ (Table 3). Burning of two coal samples yielded mean $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values as -21.7‰ and $\sim 8\text{‰}$. Coal burning produced aerosols show significantly larger TC/TN wt. ratios (~ 50 ; Table 3).

4. Discussions

In general, $\delta^{13}\text{C}$ of aerosol particles over all continental stations are $-25.4 \pm 0.6\text{‰}$ appears to be an intermediate value of aerosols emitting from pre-dominant C-3 type vegetation burning (with mean as $\sim -28\text{‰}$) and coal combustion (average $\sim -22\text{‰}$) (Table 3). Emissions from increasing automobile vehicles and industries also could be significant sources of anthropogenic aerosols especially in urban/ sub-urban locales. $\delta^{13}\text{C}$ of aerosol particles emitted from heavy vehicular transport area (Delhi-Mathura highway) and industrial area in Nagpur (NEERI), however, showed similar ($\sim -25.2\text{‰}$) values (Table 2). However, corresponding TC concentrations show almost a factor of two differences, clearly showing diesel based road traffic is a potential agent for higher C emissions in air. $\delta^{13}\text{C}$ of aerosols from road traffic area i.e -25.2‰ , is however, $\sim 1\text{‰}$ higher than typical $\delta^{13}\text{C}$ of diesel fuel ($-26.5 \pm 0.5\text{‰}$; Widory et al., 2004) which indicates other sources of C in ambient urban air.

C-3 and C-4 types of plant generally burn with emission of aerosols of typically mean C isotopic ratio -26 and -12‰ (Cachier et al., 1986) with maximum fractionations of $\sim 0.5\text{‰}$ and $\sim 3.5\text{‰}$ respectively involved during combustion (Turekian et al., 1998). Table 3 does not include any typical C-4 type vegetation e.g. sugar cane especially considering its agricultural production is quite dominant in many parts of India. Though burning remnants of sugar cane crop is less common as they are mainly used as fodder and manure, aerosols produced by this vegetation type are expected to yield $\delta^{13}\text{C} \sim -13$ - 20‰ (Martinelli et al., 2002) and hence overall lowering of $\delta^{13}\text{C}$ of carbonaceous aerosols. In contrast, overall $\delta^{13}\text{C}$ of bulk aerosols over Indian cities during pre-monsoon season (i.e. $\sim -25\text{‰}$; Table 2) appear to be a proportionate mixture of burning of biomasses typically of C-3 type and coal (primarily used for energy production, cooking etc.). $\delta^{13}\text{C}$ of carbonaceous aerosols present over BOB almost show overlapping values with an average $-25.6 \pm 0.6\text{‰}$ (Table 1; Fig.-1), indicating their plausible continental origin during the pre-monsoon period. $\delta^{13}\text{C}$ of aerosols over AS are characterized by an average $-26.5 \pm 0.8\text{‰}$. Keeping in view of very different TC contents of aerosols over AS with respect to that over continent and over BOB, difference in $\delta^{13}\text{C}$ that is $\sim 1\text{‰}$ indicates inclusion of other C sources of similar isotopic composition.

Fig. 3(a,c) shows $\delta^{13}\text{C}$ - TC plots for aerosols over AS and BOB. In order to understand these statistical relationships better, data of four southern BOB aerosols (having higher TC/TN ratios >28 ; Table 1) have been excluded from Fig.-3a (BOB plot) and included in Fig.-3c (AS plot). In case of BOB region, there appears to be weak positive correlation ($r^2 = 0.27$; $p = 0.008$) between δ values and C contents, indicating that increasing contents of C from multiple sources are resulting in isotopic enrichment of C. However, poorer statistical relationship was observed between $\delta^{13}\text{C}$ and TC in case of aerosols over AS (Fig.-3c) and that can be mainly attributable to the presence of inorganic C in ambient air.

N in the atmospheric particulate matter mainly present as nitrate and ammonium (Seinfeld and Pandis, 1998). In anthropogenically active urban locations, N species arising from oxidation of gaseous NO_x (NO_2 and NO) produced by high temperature combustion processes associated with vehicle and industrial activities also could contribute N in aerosols. The major N compound in aerosol i.e. NH_4NO_3 can be produced either by atmospheric HNO_3 reacting with NH_3 generated from microbial degradation of agricultural stock waste, fertilizer production or/and gas to particle reactions of NH_3 with other acidic species, mainly occurring in fine mode fraction of aerosols. As continental polluted air is diluted, NH_4NO_3 can be disassociated and reformed HNO_3 can react with aerosol carbonate or sea salt in marine realm (Widory et al., 2007). Thus N incorporation in aerosols may involve complex chemical reactions resulting very broad range of $\delta^{15}\text{N}$ from -15 to $+30\text{‰}$ with considerable overlaps between data from different sources. Using $\delta^{15}\text{N}$ in conjunction with N contents can possibly cure this problem up to a certain extent (Widory et al., 2007). Using this framework, we attempt to interpret aerosol $\delta^{15}\text{N}$ measured in this study.

Contrary to $\delta^{13}\text{C}$, $\delta^{15}\text{N}$ of aerosol particles present over Indian cities show considerable variability especially aerosols over coastal city Panaji (Goa) are characterized by significantly lower values i.e. $\sim 4\text{‰}$ compare to that of other cities which have typical $\delta^{15}\text{N}$ values $\sim 8-10\text{‰}$. Except for two aerosols samples collected over a hill station Nanital on the same day, (yielding $\delta^{15}\text{N} \sim 5\text{‰}$), in general, $\delta^{15}\text{N}$ of aerosols over cities including a heavy traffic area and industrial emission show a narrow range with mean value of $\sim 10 \pm 1\text{‰}$ (Table 2), which is well within the range of $\delta^{15}\text{N}$ of aerosols, observed a typical urban locale of Paris (Widory et al. 2007). Among various bio-fuels, $\delta^{15}\text{N}$ of various C-3 type vegetation types show a typically high values $\sim 15\text{‰}$ (Table 3) except for leguminous vegetations e.g. Arhar and Mustard stem yielding $\sim 5-7\text{‰}$; Table 3) owing to nature these plants that fix atmospheric N_2 (of $\delta^{15}\text{N} \sim 0\text{‰}$). Fossil fuel combustion should also result in lowering

of $\delta^{15}\text{N}$ in aerosols (Kelly et al., 2005). Hence, aerosols from heavy road traffic and industrial areas showing $\delta^{15}\text{N}$ ($\sim 9\text{‰}$; Table 2) are probably indicating dominant N sources from biomass and bio-fuel burning.

$\delta^{15}\text{N}$ can have considerable variability during summer and winters in terms of sources as well as pathways of secondary N formation (Pavuluri et al., 2010; Widory et al., 2007), however, our study is based on sampling aerosols mainly during pre-monsoon season when pollutants originated from north, northeast of India find a way to disperse towards south due to prevailing winds from continent to southern marine regions (ITCZ withdrawal). Therefore, we anticipate $\delta^{15}\text{N}$ of aerosols over major cities studied and marine realms (AS and BOB) should be carrying winter biomass - bio-fuel burning signatures primarily. In that context, $\delta^{15}\text{N}$ of bulk aerosol particles over cities (except Panaji, Goa) are appearing also as a proportionate mixture of aerosols emitted from burning of major biomasses, coal burning and vehicular emissions. $\delta^{15}\text{N}$ of aerosols over BOB showing a weak positive correlation with TN concentrations ($r^2 = 0.28$; $p=0.0067$) (Fig.-3b) is consistent with observations made by Pavuluri et al. (2010) in south Indian metro city Chennai. Hence it appears that during the pre-monsoon season higher N concentrations are associated with heavier $\delta^{15}\text{N}$ in aerosols over BOB (Fig.-1a).

Depleted $\delta^{15}\text{N}$ of aerosols over Panaji (Goa) (a coastal city) could be due to (i) much lesser population and thereby less anthropogenic activity compared to other Indian cities and (ii) dominance of marine aerosols owing to its close proximity to the adjacent marine region of AS. Between the two, later reason appears to be more likely as $\delta^{15}\text{N}$ of aerosols over AS show a clear contrast compared to that over Indian cities and over BOB with significantly lower average values $1.4\pm 3.4\text{‰}$ (Table 1; Fig.-1a). Corresponding average TN concentrations over both the marine realms are not as significantly different (0.72 ± 0.2 , $1.8\pm 0.8 \mu\text{g}\cdot\text{m}^{-3}$ for AS and BOB respectively). Hence, TN contents have to be seen in light of TC and TC/TN ratios which are significantly different between the two marine realms (Fig.-2) mainly attributable to the presence of significant proportion of inorganic carbon (TIC) originated from a variety of mineral dust sources in the AS during the pre-monsoon season (Kumar et al., 2008). On average, the proportion of particulate organic matter (POM) is just estimated as $\sim 2\%$ of the total, which is considerably less compare to mineral dust proportion which is $\sim 44\%$ (Kumar et al., 2008). Considering such a low proportion of POM in aerosols over AS marine boundary layer owing to dilution by mineral dust, TN concentrations are not proportionately as low as it should have been if major portion of TN would have been contributed by anthropogenic organic sources from the sub-continent. Thus, amount of inorganic N

(NH₃, NO_{xs}) in the TN concentrations are suspected to be dominant in AS and major reason for the lowering of overall $\delta^{15}\text{N}$ of bulk aerosol particles could be incorporation of N species (NH₃ and NO_{xs}) with lower isotopic values, emitting as a result of perennial and seasonal intense denitrification in the water column. It is known that gaseous products like N₂ and N₂O are isotopically lighter compare to substrate NO₃⁻ (Naqvi et al., 2006). In addition, AS is also known as a source of several greenhouse gases, thereby secondary processes of gas to particle conversions on ambient aerosols could be significant, both these processes appear to result in a net depletion of $\delta^{15}\text{N}$ of aerosols as observed (Fig.-1a). This observation is in good agreement with Jickells et al. (2003) where they reported isotopically light ammonium associated with aerosols from marine source compared to aerosols containing ammonium from terrestrial origin in the north-east Atlantic Ocean. The observed relatively poor statistical relationship between $\delta^{15}\text{N}$ and TN for AS (Fig.-3d) also appears to corroborate the presence of other forms of (inorganic) N.

The observed depletion of $\delta^{15}\text{N}$ of bulk aerosol particles over AS indicating mixing of inorganic N species emitted from underlying (denitrifying) waters, can result in alterations of surface properties of aerosols significantly. As residence time of aerosols is ~7-15 days, they can be efficiently removed to the ocean surface layer and supply new nitrogen. Productivity blooms, observed in the eastern AS during spring-monsoon season could thus be partly supported by this atmospheric new N inventory.

Though not much data of C and N isotopes of aerosols from south Asia are available for comparison, very recently Pavuluri et al. (2010) reported highly elevated $\delta^{15}\text{N}$ (+15.7 to +31.2‰; 23.9±3.3 average) in aerosols over south Indian metro-city Chennai, which they interpret in terms of sources from local animal excreta and bio-fuel/biomass burning from South and Southeast Asia. We did not encounter such elevated $\delta^{15}\text{N}$ values in bulk aerosol particles over any of cities studied (Table 2), however, lower ends reported by Pavuluri et al. (2010) in the early winter (18.0‰) and summer (15.7‰) in Chennai aerosols are comparable to bulk aerosols collected from burning of some biomass types used as fuel (Table 3). We suspect that in addition to fractionations involved during combustion of biomass-bio-fuels, local scale secondary processing of atmospheric aerosols in heavily populated metro-cities could also be responsible for higher $\delta^{15}\text{N}$.

5. Conclusions

This is the first $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ data of atmospheric carbonaceous aerosols over AS and BOB along with aerosols over some of the major cities of India. We also present end-member values measured

on aerosols produced by burning of some typical bio-fuels used in north and northeast of India. Major conclusions of this study are-

1- Aerosols over urban locations are typically characterized by $\delta^{13}\text{C} \sim -25.4 \pm 0.6\text{‰}$, that could be an intermediate value of proportionate mixing of aerosols emitted by biomass and coal burning predominant during winter in north and northeast of India and characterized by $\delta^{13}\text{C}$ values of $\sim -28\text{‰}$ and -22‰ respectively.

2- Though, various biomasses studied here yield typical $\delta^{13}\text{C} \sim -28\text{‰}$, their $\delta^{15}\text{N}$ show sufficient variability, indicating that coupled measurements of $\delta^{15}\text{N}$ with $\delta^{13}\text{C}$ data could be very useful for source apportionment studies of aerosols ($\delta^{13}\text{C}$ for primary sources and $\delta^{15}\text{N}$ for in-situ N cycling in atmosphere).

3- Aerosols over the eastern and western branches of the northern Indian Ocean (AS and BOB) show almost overlapping $\delta^{13}\text{C}$ values (former being depleted by $\sim 1\text{‰}$), despite significant differences in TC concentration and TC/TN ratios. This indicates mixing of other sources of C in atmospheric carbonaceous aerosols over AS, having similar isotopic composition.

4- Aerosols over AS are also characterized by significantly lower $\delta^{15}\text{N}$ compared to that over major continental stations as well as over BOB. We interpret this in terms of significant mixing of inorganic N species emitting from the underlying denitrifying waters which are known to emit isotopically depleted N species as products. Gas to particle conversions could also be significant in the AS. Phase-specific N isotope analyses could provide vital clues to complex aerosol chemistry (in-situ N cycling) in the lower atmosphere in this biogeochemically and climatically sensitive region.

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Table 1: Elemental composition and isotopes of C and N with C/N ratios of bulk aerosols collected over northern Indian Ocean during pre-monsoon period (March to May, 2006).

DATE	Sample ID	LOCATION	LAT	LONG	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\mu\text{gC.m}^{-3}$	$\mu\text{gN.m}^{-3}$	TC/TN ratio
19/03	NPL_ICARB_21082008_2	BOB	17.45	84.18	13.7	-24.1	3.98	2.84	1.40
20/03	NPL_ICARB_21082008_3	BOB	19.66	88.11	15.2	-24.9	5.38	2.65	2.03
22/03	NPL_ICARB_21082008_4	BOB	18.97	89.45	14.3	-24.9	3.90	2.62	1.49
23/03	NPL_ICARB_21082008_5	BOB	17.96	85.52	12.2	-24.6	4.62	2.36	1.96
24/03	NPL_ICARB_21082008_6	BOB	17.00	89.20	13.8	-24.8	4.41	2.94	1.50
25/03	NPL_ICARB_21082008_7	BOB	16.00	91.94	12.5	-24.9	4.15	3.32	1.25
26/03	NPL_ICARB_21082008_8	BOB	15.00	90.63	12.7	-25.1	3.44	3.01	1.14
27/03	NPL_ICARB_21082008_9	BOB	14.99	86.44	11.9	-25.6	2.17	1.45	1.49
28/03	NPL_ICARB_21082008_10	BOB	13.99	86.63	11.7	-25.6	1.96	1.48	1.33
29/03	NPL_ICARB_21082008_11	BOB	13.00	85.75	11.7	-25.8	2.50	2.21	1.13
30/03	NPL_ICARB_21082008_12	BOB	12.11	86.44	11.1	-25.8	1.53	1.23	1.24
31/03	NPL_ICARB_21082008_13	BOB	12.04	82.92	12.4	-25.8	1.27	1.61	0.79
01/04	NPL_ICARB_21082008_14	BOB	12.53	80.63	6.4	-25.9	8.62	2.31	3.74
02/04	NPL_ICARB_21082008_15	BOB	11.22	81.07	10.3	-26.3	1.31	0.98	1.34
03/04	NPL_ICARB_21082008_16	BOB	9.91	83.66	11.0	-25.8	1.18	0.94	1.24
04/04	NPL_ICARB_21082008_17	BOB	9.99	87.52	7.8	-26.3	1.06	1.03	1.03
05/04	NPL_ICARB_21082008_18	BOB	11.04	90.65	8.1	-25.8	0.99	1.03	0.96
06/04	NPL_ICARB_21082008_19	BOB	10.20	91.82	7.7	-26.4	1.39	0.93	1.49
07/04	NPL_ICARB_21082008_20	BOB	7.34	91.08	7.5	-26.5	1.12	0.74	1.52
08/04	NPL_ICARB_21082008_21	BOB	8.09	87.94	8.5	-26.6	2.33	3.17	0.74
09/04	NPL_ICARB_21082008_22	BOB	8.27	84.77	7.4	-25.3	23.14	0.62	37.61
10/04	NPL_ICARB_21082008_23	BOB	5.95	81.60	10.6	-25.6	35.01	1.24	28.20
11/04	NPL_ICARB_21082008_24	BOB	7.14	78.17	10.6	-25.9	41.64	1.27	32.91
12/04	NPL_ICARB_21082008_25	BOB	8.19	77.00	5.6	-25.7	45.14	1.21	37.44
18/04	NPL_ICARB_21082008_26	AS	9.48	75.16	1.7	-26.7	39.45	1.01	39.11
19/04	NPL_ICARB_21082008_27	AS	9.00	67.60	-1.9	-25.4	33.11	0.87	38.10
20/04	NPL_ICARB_21082008_28	AS	9.00	60.19	-1.2	-27.1	31.86	0.63	50.68
21/04	NPL_ICARB_21082008_29	AS	10.00	59.30	3.0	-27.3	33.88	0.67	50.53
22/04	NPL_ICARB_21082008_30	AS	11.00	63.20	5.3	-27.0	24.07	0.64	37.56
23/04	NPL_ICARB_21082008_31	AS	11.00	68.03	2.7	-27.1	71.12	1.21	58.59
24/04	NPL_ICARB_21082008_32	AS	11.00	72.58	0.2	-26.3	30.72	0.85	36.30
25/04	NPL_ICARB_21082008_33	AS	12.57	74.43	-2.3	-24.3	37.93	0.99	38.23
26/04	NPL_ICARB_21082008_34	AS	15.26	72.39	-0.5	-26.3	28.40	0.64	44.33
27/04	NPL_ICARB_21082008_35	AS	17.34	70.12	-0.9	-26.3	28.71	0.56	51.64
28/04	NPL_ICARB_21082008_36	AS	16.98	68.36	1.4	-27.2	44.76	0.79	56.78
29/04	NPL_ICARB_21082008_37	AS	14.46	66.67	3.2	-27.3	28.77	0.48	60.08
30/04	NPL_ICARB_21082008_38	AS	14.00	62.66	12.7	-26.3	31.69	0.94	33.61
01/05	NPL_ICARB_21082008_39	AS	15.16	59.04	2.7	-26.7	35.05	0.60	58.30
02/05	NPL_ICARB_21082008_40	AS	18.58	59.71	0.1	-27.5	33.65	0.52	64.77
03/05	NPL_ICARB_21082008_41	AS	21.12	62.15	1.1	-26.5	31.08	0.63	49.10
04/05	NPL_ICARB_21082008_42	AS	17.00	30.97	2.7	-27.4	34.60	0.57	60.64
05/05	NPL_ICARB_21082008_43	AS	16.52	63.32	2.8	-26.6	29.71	0.48	61.73
06/05	NPL_ICARB_21082008_44	AS	18.18	65.59	-0.8	-25.7	33.70	0.56	60.42
07/05	NPL_ICARB_21082008_45	AS	20.93	68.08	-1.5	-26.1	31.34	0.69	45.68
08/05	NPL_ICARB_21082008_46	AS	19.48	71.27	-0.5	-26.1	45.29	0.87	51.83

Table 2: Elemental concentrations of total C & N, their isotopes $\delta^{13}\text{C}$, $\delta^{15}\text{N}$, and C/N ratios of bulk aerosols collected over different continental locations over India during pre-monsoon period (February to March, 2009).

Date (dd/mo/yr)	LOCATION	Sample ID	LAT	LONG	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\mu\text{gC.m}^{-3}$	$\mu\text{gN.m}^{-3}$	TC/TN ratio
14/2/09	Bhubneswar	NPLAerosol_B1	20.30	85.83	13.0	-25.0	28.9	6.8	4.3
15/2/09	Bhubneswar	NPLAerosol_B2	20.30	85.83	11.7	-25.5	33.2	7.5	4.4
17/2/09	Delhi	NPLAerosol_D1	28.63	77.17	9.5	-25.8	32.6	10.1	3.2
18/2/09	Delhi	NPLAerosol_D2	28.63	77.17	10.7	-25.2	51.6	10.5	4.9
12/2/09	Nainital	NPLAerosol_N1	29.40	79.44	5.3	-26.7	17.1	2.1	8.3
13/2/09	Nainital	NPLAerosol_N2	29.39	79.46	5.2	-26.4	15.8	2.3	7.0
14/2/09	Nainital	NPLAerosol_N3	29.39	79.45	8.9	-26.5	19.7	2.4	8.3
13/3/09	Anantpur	Anantpur-7_1	20.04	86.33	9.1	-25.0	31.8	19.0	1.7
14/3/09	Anantpur	Anantpur-8_1	20.04	86.33	9.1	-24.6	35.9	4.2	8.5
15/3/09	Anantpur	Anantpur-9_1	20.04	86.33	10.3	-24.8	28.1	4.7	5.9
24/3/09	Nagpur	Nagpur-14_1	21.15	79.09	9.0	-25.4	9.9	5.1	1.9
25/3/09	Nagpur	Nagpur-15_1	21.15	79.09	11.9	-26.0	12.1	1.4	8.8
20/02/2009	Goa	Aero_NIO09_1	15.46	73.80	5.79	-25.2	-ND-	-ND-	-ND-
25/02/2009	Goa	Aero_NIO09_2	15.46	73.80	2.34	-25.08	-ND-	-ND-	-ND-
17/03/2009	Goa	Aero_NIO09_3	15.46	73.80	6.1	-25.2	-ND-	-ND-	-ND-
03/04/2009	Goa	Aero_NIO09_4	15.46	73.80	1.9	-24.5	-ND-	-ND-	-ND-
13/04/2010	Road Traffic_CRRI	NPL_Aero_CRRI road	28.63	77.17	9.6	-25.3	98.4	4.5	21.7
18/04/2010	Industrial area NEERI	NPL_Aero_NERRI	21.15	79.09	8.9	-25.1	48.5	3.7	13.3

Table 3: Elemental C and N concentrations, their isotopic values ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) and C/N ratios of bulk aerosols collected from burning of different types of biomass used as fuels and typical end member values of road traffic and industrial areas in India.

Type of Fuel	Sample ID	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	$\mu\text{gC.m}^{-3}$	$\mu\text{gN.m}^{-3}$	TC/TN ratio
Eukeliptus	NPLAerosol_UP2	14.3	-28.3	9013.9	294.4	31.0
Neem	NPLAerosol_UP5	13.5	-28.4	11562.2	474.5	24.8
Arhar	NPLAerosol_UP8	5.2	-28.3	13523.5	614.4	22.5
Mustard stem	NPLAerosol_UP11	7.6	-28.4	13891.6	450.3	31.6
Cowdung cake*	NPLAerosol_UP13	8.1	-34.87*	78979.5	3695.1	21.4
Babool	NPLAerosol_UP22	14.3	-28.8	21643.1	2758.5	7.9
Chilly stem	NPLAerosol_UP25	10.4	-25.9	39662.6	4989.9	8.0
Desi Keekar	NPLAerosol_UP28	14.4	-26.9	77491.0	3939.4	19.7
Sheesham	NPLAerosol_UP40	20.5	-29.4	11947.1	1043.7	11.7
Arandi	NPLAerosol_UP43	18.3	-26.9	59645.6	3222.3	18.6
Coal -1	Bi219-coal	8.7	-21.8	7483.5	181.7	42.0
Coal -2	Bi220-coal	7.1	-21.7	9487.5	173.2	55.6

* Lower confidence value and has not been used for considering average.

Figures Captions

Figure 1: $\delta^{15}N$ and $\delta^{13}C$ of bulk aerosol particles collected over AS and BOB during pre-monsoon season of 2006. Black squares over India depict isotopic values observed on aerosols emitted from selected sites and typical road traffic area of Delhi-Mathura highway and industrial area of Nagpur (NEERI) (Table 2).

Figure 2: TC and TN ($\mu\text{g} \cdot \text{m}^{-3}$) and TC/TN wt. ratios of bulk aerosol particles collected over AS and BOB during pre-monsoon season of 2006. Black squares over India depict values observed on aerosols emitted from selected sites and typical road traffic area of Delhi-Mathura highway and industrial area of Nagpur (NEERI) (Table 2).

Figure 3: $\delta^{13}C$ and $\delta^{15}N$ of aerosols over AS and BOB plotted against corresponding concentrations of TC and TN ($\mu\text{g} \cdot \text{m}^{-3}$) (a-d), and with respect to each other (e,f).

Figure 1

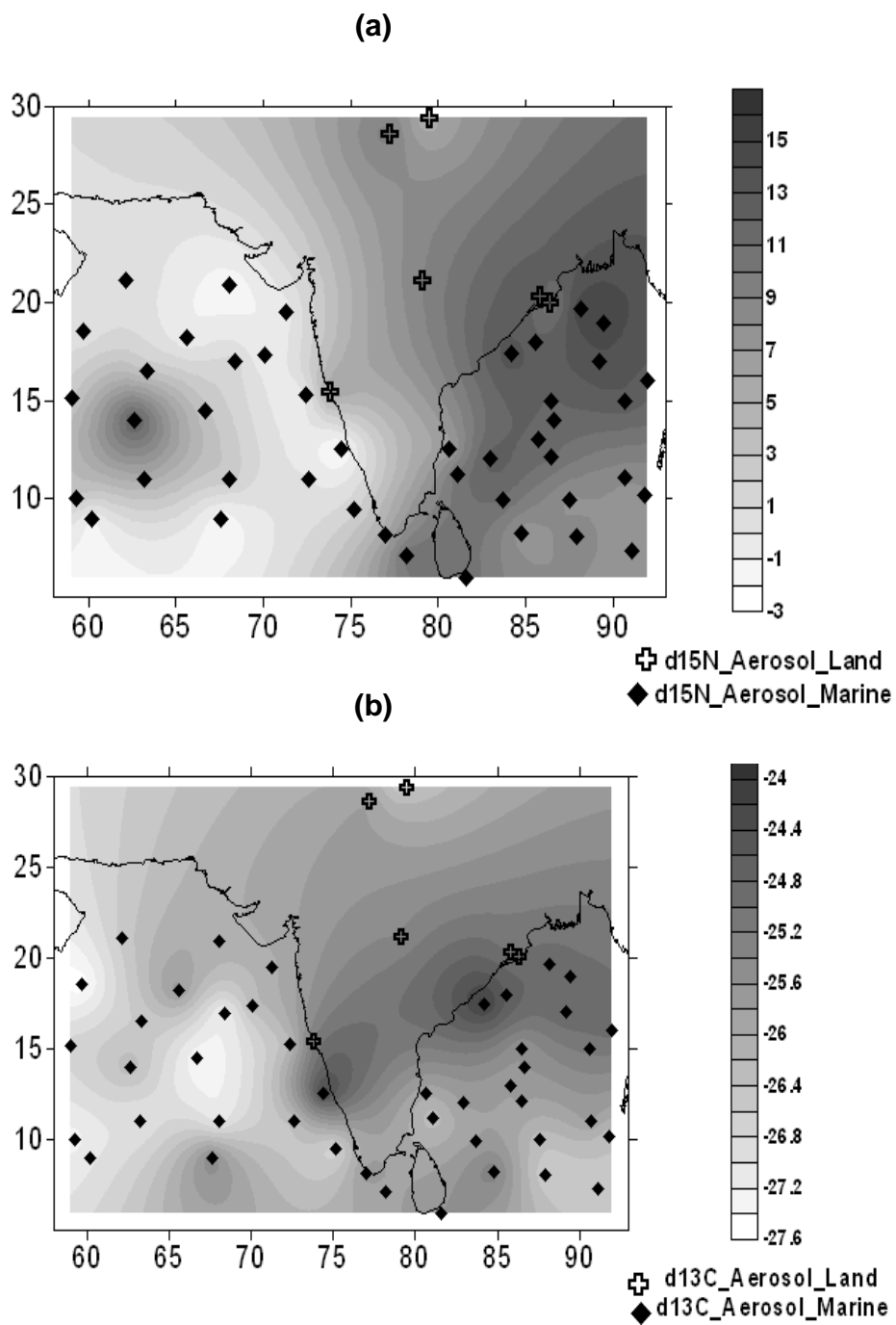


Figure 2

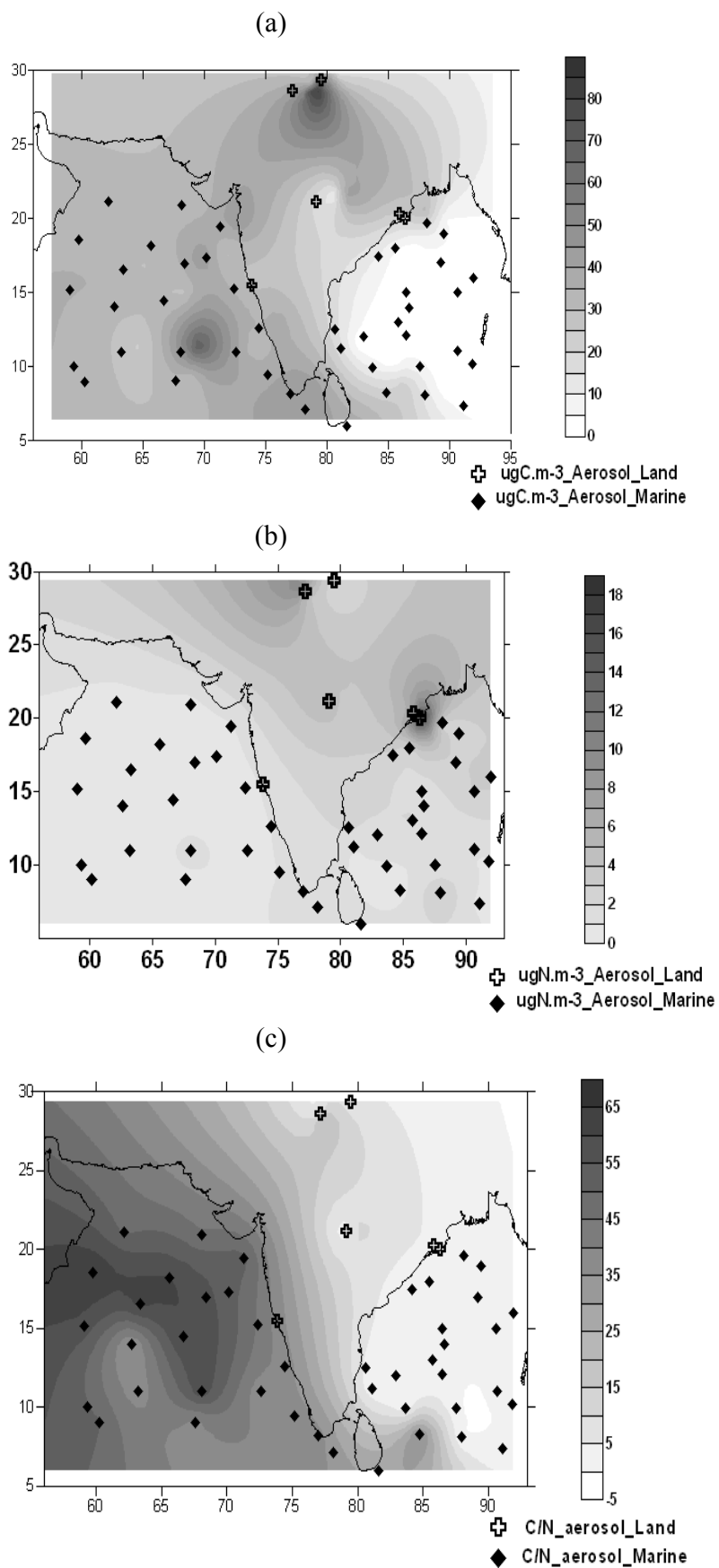


Figure 3-

