ISSN: 0973-4929, Vol. 16, No. (Special Issue 1) 2021, Pg. 62-71

Current World Environment

www.cwejournal.org

Interlinkages Between Total Nitrogen and Doc Levels at an Urban Site of Saharsa District of Bihar (India)

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Abstract

Nitrogen is an element essential for building fundamental blocks of life. When present in excess amount in air, it causes adverse effects for the environment and human health. Different air pollutants when scavenged through rains are deposited on the surface. The chemical composition of rain water is an indicator of the levels of different air pollutants in the region. The present study reports the concentrations of Dissolved Organic Carbon (DOC) and the reactive nitrogen species (NO_3^- and NH_4^+) in rain water at an urban site located in the Saharsa district of Bihar. The sampling was carried out during July 2018 to October 2018. Total 18 samples were collected during this period. Results showed that the concentration of NO_3^- was considerably higher (2.91 mg/L) ranging from 0.26 to 11.84 mg/L than the NH_4^+ (0.84 mg/L) ranging from 0.00 to 3.2 mg/L. The pH value in the samples ranged from 5.50 to 7.68 with a mean value of 6.52. The DOC in rain water has been estimated by using the Shimadzu TOC analyzer. Apart from DOC, total carbon (TC), total nitrogen (TN) & inorganic carbon (IC) have also been also analyzed by Shimadzu TOC analyzer. The results showed that both anthropogenic and natural sources contributed to the dissolved organic carbon (DOC) in rain water. The linkages between the sources and the transformations of gaseous NH_3 with DOC have been established considering the fact that most of TN is represented by NH_3. Airmass back trajectory analysis showed that the site has trans-boundary source influence from Bay of Bengal and Bangladesh. The study suggests that there is need of continuous monitoring of these parameters in air in order to notice any impact on human health, soil and vegetation.

Article History
Received: 02 January 2021
Accepted: 20 May 2021

Keywords
Dissolved Organic Carbon;
Inorganic Carbon;
Reactive Nitrogen Species;
Total Nitrogen;
Wet Deposition.

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Doi: http://dx.doi.org/10.12944/CWE.16.Special-Issue1.07

Introduction

Air pollution is perceived as a major issue due to increased urbanization and industrialization. Nitrogen is a major atmospheric constituent. It makes up around 78% by volume of the Earth's atmosphere. The inert nitrogen is utilized by converting into different reactive compounds such as NH_3, NO_2, protein, urea etc. These nitrogen compounds are called reactive nitrogen species (Nr) which are chemically, biologically, photo chemically and radioactively active species.[1-2] NH_3 and NOX are the major reactive nitrogen species which play important role in the atmospheric chemistry and nutrient supply.[2] Major sources of atmospheric NH_3 include the nitrogen fixation, Haber-Bosch process and human excreta. NH_3 is a highly soluble in water and can be corrosive at high concentration. NH_3emitted in the air is either deposited directly through dry deposition or gets transformed into an NH_4^+ aerosols. These NH_4^+ aerosols play an important role in acid rain, nutrient supply and the radiative forcing.

Sources of atmospheric NO_x include transport, industries, lightning, combustion of fossil fuels and biomass burning.[3-5] Deposition of anthropogenic constituents via wet and dry removal processes from the atmosphere has adverse impacts on terrestrial and aquatic ecosystems.[6] The elevated concentrations of NO_x result in increased concentration of troposphere. High concentrations of nitrogenous compounds cause environmental problems such as eutrophication, soil acidification. Reaction of NO_2 with OH and forms nitric acid (HNO_3) which is deposited on the Earth's surface.[7] Nitrous oxide (N_2O) is mainly emitted by soils and paddy cultivation. N_2O is a potent greenhouse gas which plays an important role in the ozone destruction in the stratosphere.

In spite of continuous emissions of Nr species, atmosphere keeps on cleaning itself and the constituents are scavenged in due course of time through wet deposition and dry deposition.[8] Wet deposition occurs in the form of rain, snow or fog. Wet deposition plays an important role in removing the organic carbon from the atmosphere.[9] A fraction of such carbon is soluble in rain water called dissolved organic carbon (DOC) which passes easily through the filter. The natural source of organic compounds includes VOCs, bio aerosols, from vegetation, forest fires, sea and volcanoes. The anthropogenic source includes combustion of coal, petroleum, fossil fuels, biomass burning, fugitive emission and Haber Bosch nitrogen fixation10. According to reports, half of the primary organic carbon is transformed into secondary organic aerosols (SOA). About 60% of OC is removed from the atmosphere through wet atmospheric deposition while 40% through dry deposition process. It is reported that atmospheric DOC can influence cloud albedo and cloud condensation nuclei, rainwater pH, photochemical processes and visibility.[11] Coinciding the importance of DOC, the present study was carried out to determine the DOC content in rainwater and to correlate its associated nitrogenous compounds in order to identify their co-variation and common source linkages. Apart from this, the study also aimed to determine the concentration of NO_3^- and NH_4^+ at the site as well as to find out the dominance of local vs transported sources of carbon and Nr species in the wet deposition at the study area.

Methodology

Sampling Site

As shown in Fig. 1, the site Saharsa town is a district headquarter (25.88°N, 86.6°E) located in the eastern Bihar and falls in the northern plains of India. The location helps in estimating the air quality of the region as well. The district occupies an area of 1,687km^2 at an altitude of 44 mamsl. The area has a population of 19,00,661. The district is surrounded by the river Kosi on the western side. Agriculture is the main occupation of people in the district. The main crops grown in the district are paddy, maize, jute, wheat, barley and sugarcane. Major sources of pollution at the site include vehicular emission, domestic cooking, biomass burning, chimneys and road dust. As the site lies close to the Indo-Nepal and Indo-Bangladesh borders, it would help in the study of trans-boundary pollution.

Sample Collection

The sample collection was carried out at the terrace of a building at a height of 20m. An assembly of polythene funnel of 20cm diameter fitted onto 2L capacity polythene bottle was used for the collection of rain water. The samples were collected on event basis. After each rain event, the volume of the sample was measured using a measuring cylinder which was properly washed with de-ionized water.

After the collection, samples were kept in prewashed small polythene bottles of 125ml volume. A small amount of thymol was added to the samples for preservation. A total of 18 samples were collected from July to October 2018. Further, the samples were brought to the laboratory and kept at 4°C in a refrigerator before analysis.

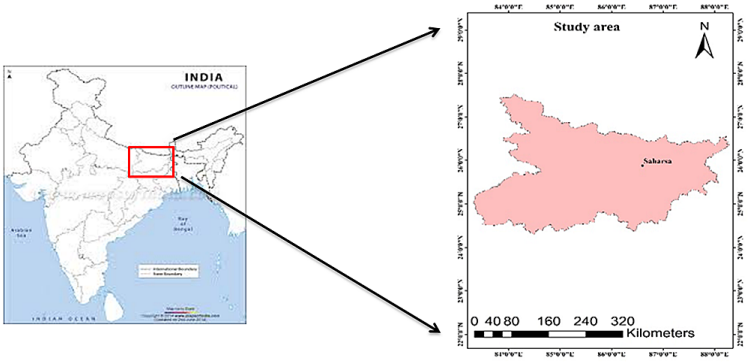

Fig.1:Study Area Map showing Saharsa town

Chemical Analysis

After the collection of samples, pH and electrical conductivity of the samples were measured immediately. The NO_3^- and NH_4^+ were analyzed by spectrophotometric method.[12] NH_4^+ was measured by colorimetrically using the indophenol blue method with the help of UV-vis spectrophotometer (Perkin Elmer, USA). The NO_3^- was determined by using the TRI solution method using UV-vis spectrophotometer.[13] The bicarbonate ion (HCO_3^-) and alkalinity of samples were measured using titration method.[14]

The DOC in rainwater was determined by high temperature combustion with a normal sensitivity catalyst technique using Shimadzu TOC analyzer. The DOC determination involves two steps – i) quantification of total carbon (TC)and ii). determination of inorganic carbon (IC). The standard for TC was prepared by dissolving potassium hydrogen phthalate in ultrapure water while the standard for IC measurement was prepared from a mixture of anhydrous sodium carbonate and sodium hydrogen carbonate. For the TC measurements, the sample was injected into the combustion tube. TC is the sum of IC and DOC (in filtered sample). For IC measurements, sample was acidified with H_3PO_4 and purged with a CO_2 free carrier gas in order to remove and measure CO_2 produced by the inorganic species.

Table 1: Descriptive Statistics of Various Parameters

N=18	pH	Conductivity (µS/cm)	NH4+ (mg/L)	NO3- (mg/L)	TOC (mg/L)	IC (mg/L)	TC (mg/L)	TN (mg/L)	ON (mg/L)
Mean	6.52	1.91	0.84	2.91	1.91	2.16	4.34	1.88	0.74
Max	7.68	4.91	3.2	11.84	4.31	6.12	9.08	4.37	3.75
Min	5.5	0.77	0	0.26	0.77	0.38	1.93	0.48	0.01

Results and Discussion

Ph Variation and Ionic Composition of Rainwater

Table 1 gives the descriptive statistics of different parameters measured in rainwater in mg/L. The pH values in the samples ranged from 5.5 to 7.68 with a mean value of 6.52 (Fig. 2) The alkaline nature of the observed sample is mainly because of high loadings of suspended $CaCO_3$ rich particulate matter in the atmosphere which is a common feature of Indian region.[6,15] However, one out of 18 samples was having acidic (pH below 5.6) which was due to prolonged and continuous rain. The continuous

rains result in settling down of dust particles from atmosphere due to which the buffering capacity of rain water against sulphate and nitrate becomes low. Generally, as mentioned earlier the particulate matter in India consists of $CaCO_3$ that results in high pH of rainwater in the region.[16] It was further confirmed by high values of bicarbonate ion (HCO_3^-) and alkalinity of samples. The concentration of HCO_3 varied from 19.99 to 48 mg/L with an average value of 27.90 mg/L. The concentration of alkalinity varied from 16.39 to 39.36 mg/L with an average value of 22.90 mg/L. The conductivity of the samples varied from 1.91 to 4.91 µS/cm with an average of 0.77.

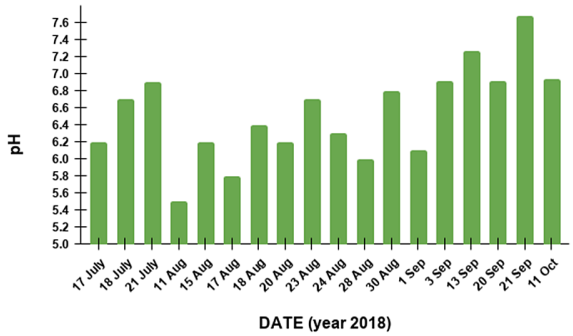

Fig. 2: pH of rain water samples collectedat the site

Variation of NO_3^- and NH_4^+

In rainwater, N is mainly present as NO_3^- and NH_4^+ ions.[17] In order to consider their equivalents for chemical transformation, NO_3^- and NH_4^+ were calculated inµeq/L for further discussion in this section. The concentration of NO_3^- in the samples varied from 4.2 to 191µeq/Lwith an average of 52µeq/L. The sources of NO_3^-mainly include combustion of fossil fuel, biomass burning, brick kilns and vehicular emission at the site.[18] The Fig. 3 shows the variation in the pattern of NO_3^- and NH_4^+ with time in the region. The sudden increase in the level of NO_3^- on 30th August 2018 might be due to heavy precipitation (13mm) occurred through strong lightning. Additionally, NO_3^- associated with crustal particulate matter might have been washed out. The lightning breaks N_2 forming NO and NO_2 which are oxidized to NO_3. In the reaction mentioned below, nitric oxide (NO) reacts with molecular oxygen to form nitrogen dioxide (NO_2) which further reacts with ozone (O_3) and forms nitrate (NO_3).

$$2NO + O_2 \rightarrow 2NO_2 \quad ...(1)$$

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \quad ...(2)$$

$$NO_3 + NO \rightarrow 2NO_2 \quad ...(3)$$

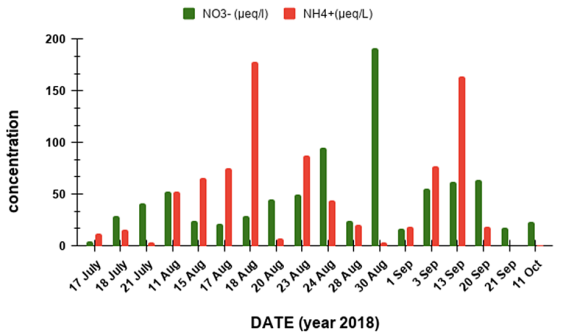

Fig.3: Variation of NO_3^- and NH_4^+ in rainwater samples

The concentrations of NH_4^+ varied from 0 to 177.8µeq/L with an average of 51.1µeq/L. The Fig. 5 shows the variation in the pattern of NH_4^+ in the region. The higher levels of NH_4^+ in rain water are mainly due to the release of NH_3 from various sources such as use of ammonia containing

fertilizers in agriculture, emissions from livestock, excreta of animal and human being as well as biomass burning in the area.[5,13] NH_3 readily reacts with H_2SO_4 and HNO_3 forming respective salts.[19] Generally, due to abundance of $CaCO_3$ in air, free H_2SO_4 is not present in this region. When rain is continued for a longer period, the crustal material is washed out resulting in H_2SO_4 and $(NH_4)_2SO_4$ formation.[8,15,20]

$$NH_3 + H_2SO_4 \rightarrow NH_4HSO_4$$

$$NH_3 + NH_4HSO_4 \rightarrow (NH_4)_2SO_4$$

The present concentrations of NH_4^+ and NO_3^- are not so high and hence, are not a threat to the soil fertility at present, due to very high critical load values in this region.[21]

Total nitrogen (TN) is the sum total of all the organic and inorganic nitrogen compounds present in the sample. NH_3 is the major contributor in TN. The concentration of total nitrogen varied from 34.6 to 312.6µeq/L with an average of 138.2µeq/L (Fig.4). The high level of TN on 21 July 2018 indicates organic nitrogen to be one of the sources, apart from inorganic nitrogen. This needs further investigation.

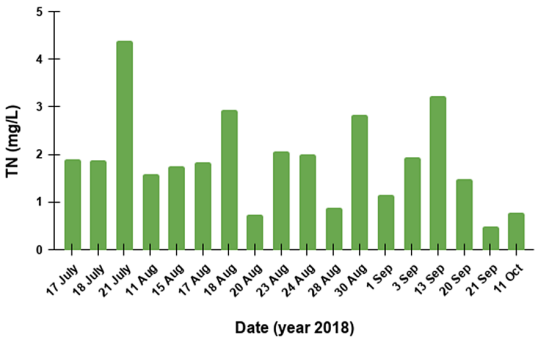

Fig.4: Variation of total nitrogen (TN) in rainwater samples

Water soluble organic Nitrogen is the deposition of organic nitrogen as gases or particles in precipitation. In order to evaluate the concentration of organic nitrogen in rain water, we have included NH_4^+ and NO_3^-. The NO_2^- is excluded as it is an unstable ion and is considered as a minor species in the rain water.[10] Organic nitrogen in atmosphere consists of a large number of reactive compounds, from small molecules to complex biological polymers. Some Organic nitrogen compounds have phytotoxicity effect and possess serious impact on human health. Organic nitrogen is derived as the difference in the measured concentration of dissolved inorganic nitrogen (DIN) and total nitrogen in the sample.

$$ON = TN-(NO_{3-} + NH_4^+)$$

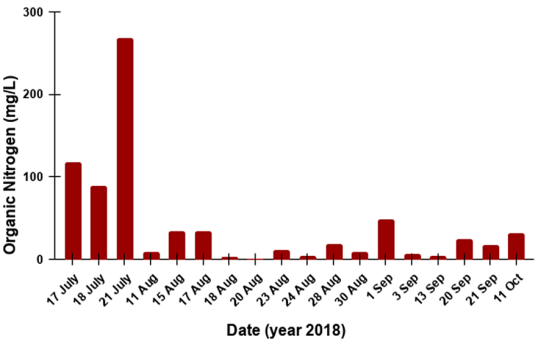

Fig.5: Variation of Organic Nitrogen in rainwater samples

Emission of organic nitrogen in the atmosphere is either as particle phase or gas phase that further reacts to form the secondary particles. It is quite difficult to identify the exact source because through

many continuous chain reactions in the atmosphere, formation of the secondary pollutants occurs. Source of precursors (e.g. formation of organic nitrates through photochemical reactions of hydrocarbons and nitrogen oxides may be recognized, but no single source is identifiable.[22] Some of the prominent sources of organic N compound areamines, amino acids, nitrophenols, urea, alkyl amides etc. There are different studies that show the identification of organic N based on the correlations with other species (NO_3^-, NH_4^+) whose source is known. DON has shown correlation with NH_4^+ and with ammonium and amino acids.[23]

The concentration of dissolved organic nitrogen varied from 267.8 to 0.9µeq/L with an average of 49.9µeq/L at the site (Fig 5). The concentration of DON is very much dependent on the volume of the amount of rain collected. DON is quite high in the first three samples because of the low volume of the precipitation collected i.e. 0.67mm (first day), 0.48mm (second day) and 0.26mm (third day). Sources of organic N compounds is from the use of fertilizers in the agricultural land (Urea), livestock & animal husbandry (Amines and Urea), landfills (amines), and biomass burning (nitrophenols).[24]

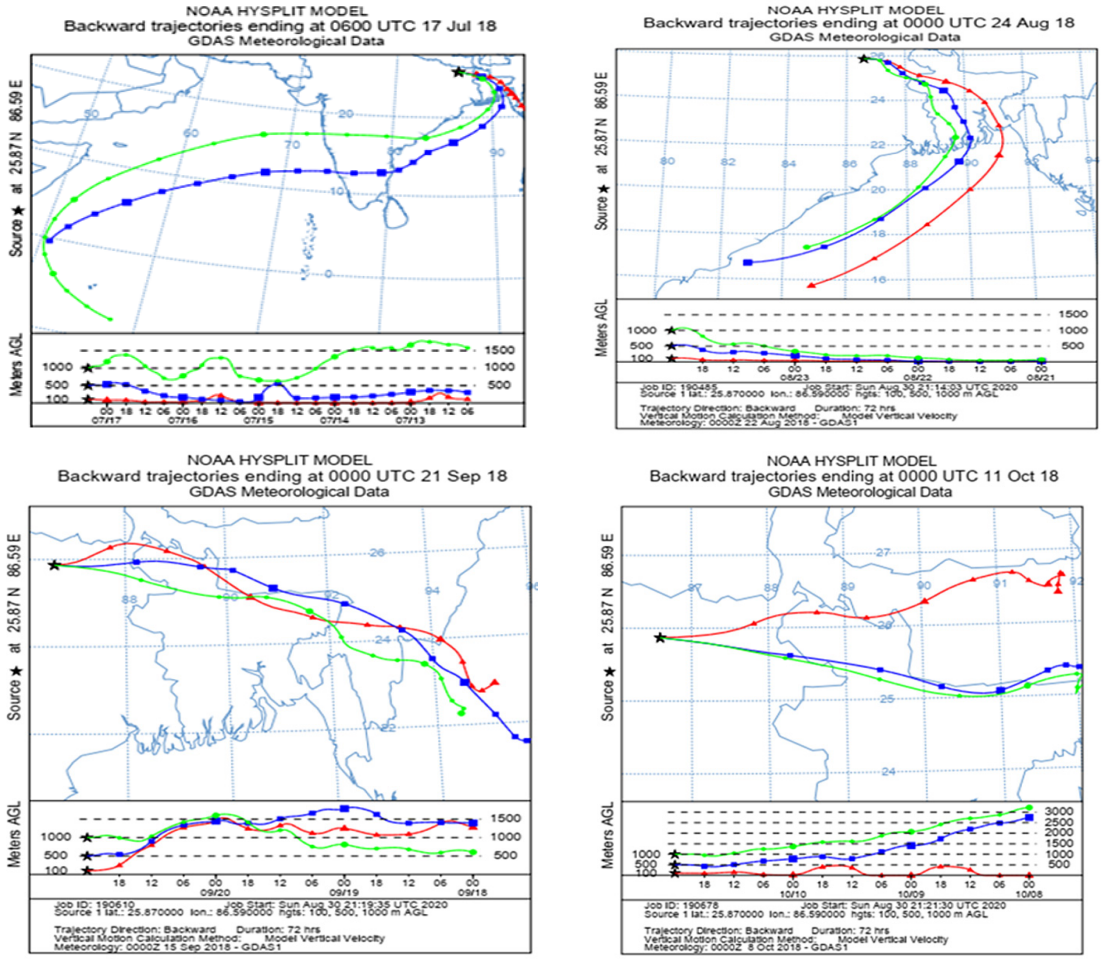

Fig.6: Airmass Back Trajectory at the sampling site of four different months respectively

Backward Trajectory Analysis

Back-trajectories were generated using the HYSPLIT 4 model of the Air Resources Laboratory of NOAA. The back trajectory helps in demonstrating trans-boundary and long range transport of air pollution. In this study, back trajectory of single event from every month (July 2018 to October 2018) has been shown in Fig.6. Back-trajectories were calculated up to 1000 m above ground level (AGL) for 72-h at 00:00 UTC for each of the rain event. In July, during

monsoon, the air masses were coming from the horn of Africa. In August, these were coming from the central part of Indian Ocean and Bay of Bengal that contributed these pollutants. But all these have passed through Bangladesh. In September and October also, these were coming from Bangladesh area. The possibility of trans-boundary pollution from nearby countries cannot be ruled out.

The air mass trajectories are used to figure out the source region as well as different factors relating to atmosphere and climate such as transport of pollutants, air quality, climatology, source apportionment, meteorology, residence time analysis, precipitation chemistry.[25-27] Several studies have been reported on long range and transboundary transport of pollution using air mass trajectories.

Variation of TOC, TC, IC & TN

At this site, the concentration of inorganic carbon (IC) varied from 0.38 to 6.12mg/L with an average of 2.16mg/L(Fig. 7a). Inorganic carbon includes carbonate and bicarbonate present in rain water.[28] Sources of carbon at the site included the biomass burning by the local people, fossil fuel burning mainly coal in the brick kilns, domestic cooking, by Barauni Oil Refinery and vehicular emissions. However, carbon can be transported from the nearby countries also as mentioned in the backward trajectory section.

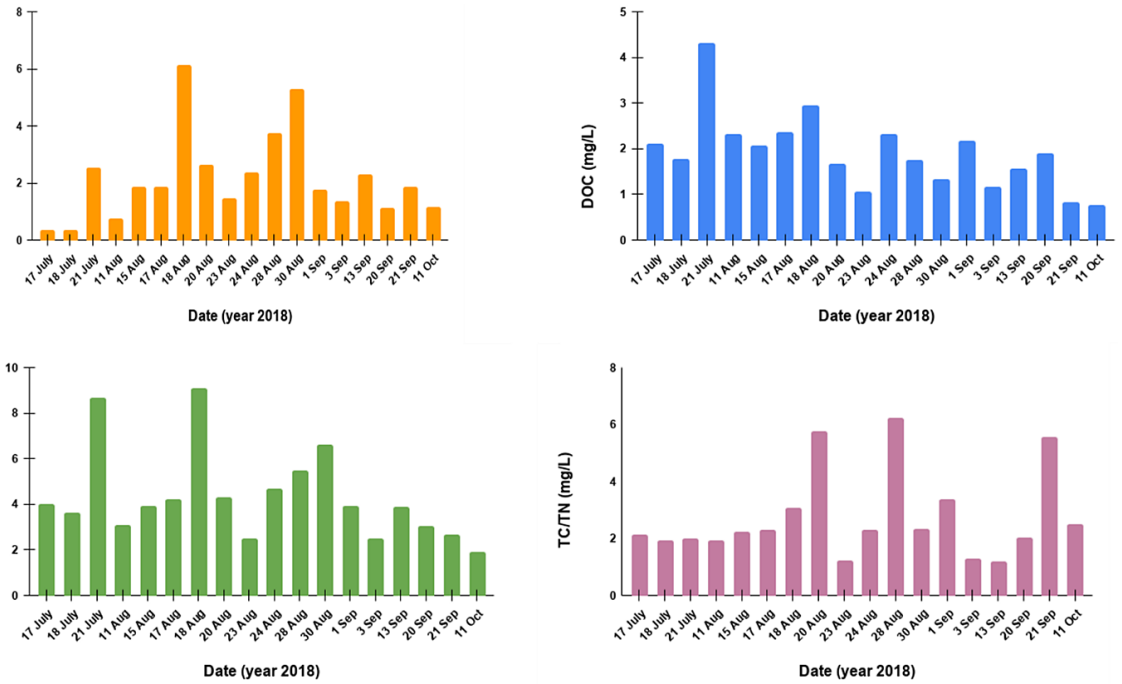

Fig.7: Variationof Inorganic Carbon (a), Total Carbon (b), DissolvedOrganic Carbon (c) and Total C/Total N (d) ratios in the rainwater samples

The total carbon (TC) in the precipitation is the sum of total organic carbon and inorganic carbon. The concentration of TC at the site varied from 1.93 to 9.08 mg/L with an average of 4.34 mg/L as shown in Fig. 7(b).The sources of TCare mainly the biomass burning, domestic cooking, brick kilns etc.

The dissolved organic carbon (DOC) in this study represents TOC as the samples were filtered.[29] Figure 7(c) shows the concentration of dissolved organic carbon that varied from 0.77 to 4.31 mg/L with an average value 1.91 mg/L. In the wet deposition process, the organic particles present get scavenged from the atmosphere and dissolve in the water, thereby contributing in the concentration of total organic carbon. During the early days of rain events, the organic compounds are scavenged more from the atmosphere than the later stages of rainfall.[30-31] Therefore, the concentration of DOC is more in such samples. Another factor that

determines more efficient scavenging of organic compounds from the atmosphere is the lower rate of precipitation in each event of rain.

Figure. 7(d) shows the ratio between total carbon (TC) and total nitrogen (TN). The high value of TC/TN ratio gives the dominant carbon source in the samples. Anthropogenic activities near site generally include burning of biomass, household waste and plastics which releases carbon into the atmosphere. Domestic cooking plays an important role in the carbon emission which might be a factor for high C/N ratios.

Conclusions

The average value of pH of the rain water samples at the study site in Saharsa district of Bihar was recorded as 6.67 having the range from 5.50 to 7.68. This range of pH is similar to other sites reported in earlier studies in north India. The high loading of $CaCO_3$ rich suspended particulate matter in the atmosphere results in the higher pH of rain water. Further, due to presence of carbonate or bicarbonate ion, the inorganic carbon concentration in the samples ranged from 0.36 to 6.12 mg/L. The average concentration of total carbon was 4.34 mg/L and total organic carbon was 1.91 mg/L.In this study, the NH_4^+ had an average value of 0.84 mg/L while NO_3^- as 2.91 mg/L in rain water. The probable sources of NH_4^+ contributions include fertilizer application in agriculture fields, human excreta and rearing of domestic animal could be the reason while that of NO_3^- include biomass burning, auto mobilee missions, coal burningand presence of brick kilns around the area. However, the back trajectory analysis showed an influence of trans-boundary pollution transported from Bay of Bengal and Bangladesh side too. The study suggests to carry out long term measurements of rain chemistry in order to quantify the local vs trans-boundary pollution at this site.

Acknowledgement

We sincerely acknowledge the financial support received from DST PURSE programme. The author, Akanksha Roy acknowledges the award of JRF received from UGC, India which helped in the completion of this work. This study is a part of our group activities viz. DRS-NET-India group and UKRI GCRF South Asian Hub project.

Funding

The author(s) received no financial support for the research, authorship, and/or publication of this article.

Conflict of Interest

The authors do not have any conflict of interest.

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