J. Ind. Geophys. Union (July 2017)
v.21, no.4, pp: 319-326

Atmospheric reactive nitrogen fluxes and scavenging through wet deposition Over Mathura (India)

Mudita Chaturvedi, Reema Tiwari and Umesh Kulshrestha*
School of Environmental Sciences, Jawaharlal University, New Delhi 110067, India
*Corresponding Author: umeshkulshrestha@gmail.com

ABSTRACT

In the era of excessive use of fertilizer and combustion of fossil fuel, the atmospheric deposition of reactive nitrogen species has led to the problem of acidification and eutrophication of the ecosystem. This study has been carried out for estimating the concentrations and wet deposition fluxes of N_r species in rain water along with their scavenging behaviour at a typical residential site under semiarid tropical region. For this purpose, sequential sampling of a rain event was performed for determining N_r levels during monsoon 2015 period. Samples were analysed for their cationic and anionic content using an ion chromatograph. The results showed a relative abundance of NH_4^+ (62.3 ± 2.3 μeql^{-1}) over NO_3^- (46.8 ± 40.1 μ eql^{-1}) in the rain water samples, subsequently creating a higher wet deposition flux of NH_4^+-N (3.6 kg ha^{-1}y^{-1}) in comparison to NO_3^- -N (2.1 kg ha^{-1}y^{-1}). This clearly indicated that the Nr deposition had very significant contribution over the study area. Scavenging patterns confirmed the presence of NH_4NO_3 showing co-variations along with the rainfall intensity. A strong correlation (0.92) of NH_4 and NO_3 also supported such observations, thereby, confirming the dominant forms in which these Nr species are being deposited over the study area.

Nr: It represents NH_4^+ and NO_3^- in this paper

Key words: Reactive nitrogen, wet scavenging, rainfall intensity, neutralization capacity.

INTRODUCTION

With the growing demand of food and energy production across the globe, the footprints of alteration in the N cycling are becoming significant in the ecosystem responses. This is evident from the tenfold rise in the transformation rates of inert N into reactive nitrogen compounds (Nr) creating cascade of environmental problems along their biogeochemical pathway (Galloway et al., 2004). Expanding sectors of industrial and agricultural activities along with the fossil fuel combustion processes has inadvertently increased their availability as NO_3^- and NH_4^+ in the environment (Galloway et al., 2008; Kulshrestha et al., 2014a). Their direct emission pathways have eventually resulted in these inorganic Nr species becoming the key driver of atmospheric chemistry and climate change (Dentener et al., 2006). However, with the changing dynamics of their atmosphere – biosphere interactions, there has been an exceedance in the Nr deposition fluxes beyond its critical threshold values leading to air pollution, acidification and eutrophication of the ecosystem (Reis et al., 2009; Vet et al., 2014).

Chemical characterization of deposition pathways, in this regard, has provided signatures for spatio temporal evolution of Nr emissions and their consequent interaction with the atmospheric transport dynamics and removal processes. Thus, depending on the Nr solubility and amount of precipitation received in the region, the wet as well as dry deposition processes are becoming detrimental to its fate by controlling the concentration of its gaseous and particulate species in the troposphere (Wallace and Hobbes, 2006). Amongst them, the mechanism of pollutant scavenging through precipitation has been more effective as an atmospheric purging process with more than 90% removal efficiency of their total atmospheric loading (Gromping et al., 1997). With the changing Nr emission scenarios over the past few decades, their precipitation weighted mean concentration has shown a rising trend over Africa (+19%) and Asia (+ 13.6%) but a declining trend over Europe (-2.7%) and North America (-4.3%) (Torseth et al., 2012; Singh et al., 2017). Introduction of NO_X emission reduction strategies has simultaneously resulted in the growing dominance of N reduced (NH_4^+) contribution to the total N wet deposition especially over the agricultural intensive areas. However, a few NO_x dominated emission areas in the eastern and western regions of U.S. has given more than 60% contribution of N oxidized (NO_3^-) to their total N wet deposition fluxes (Vet et al., 2014).

Hence, the chemical composition of rain varying through time and space could be used for deciphering the Nr scavenging patterns based on its source of influence and precipitation amount. A number of studies have already shown the alkaline nature of rain water in India due to the high dust loading of soil derived particles in its atmosphere (Parashar et al., 1996; Kulshrestha et al., 1996; Jain et al., 2000; Norman et al., 2001; Bhaskar and

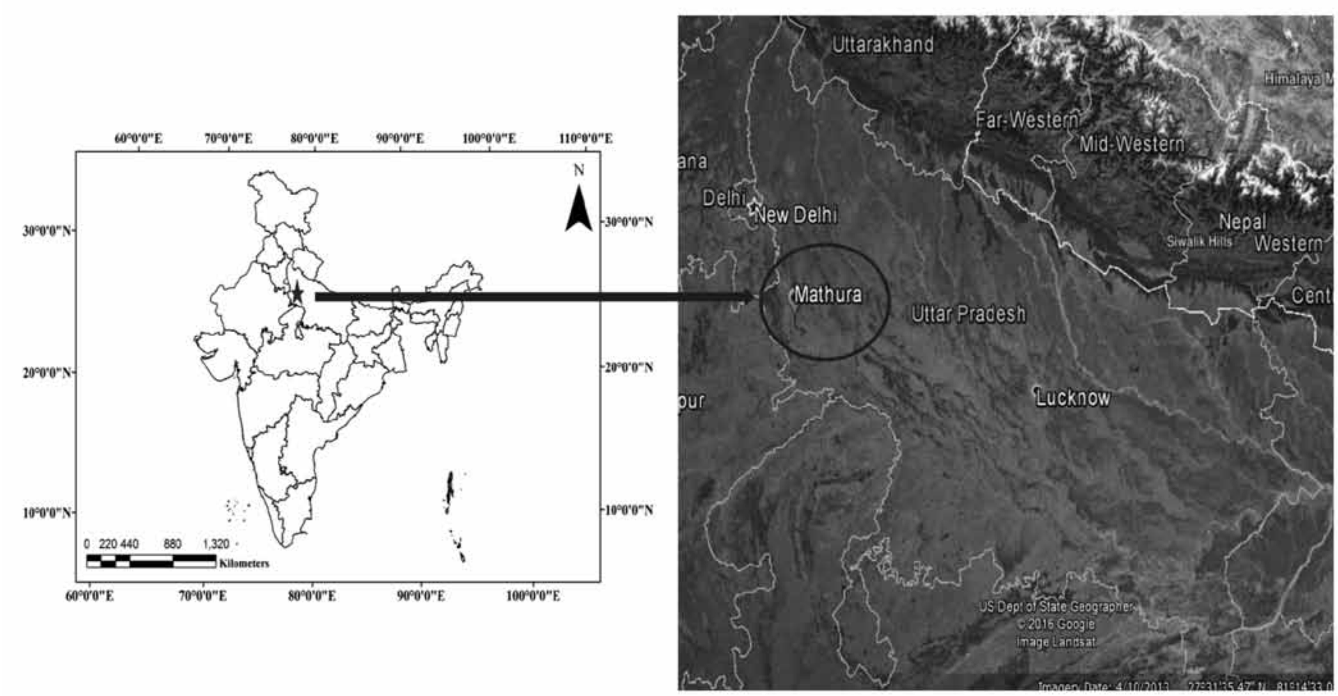

Figure 1. Map showing the sampling site.

Rao, 2016; Kulshrestha et al., 2005; Kulshrestha, 2013). Their below cloud scavenging during precipitation events can, therefore, result in the buffering of NO_3- acidity in rain water through following reaction

$CaCO_3$ (s) + $2HNO_3$ (g) —> $Ca(NO_3)_2$ (s) + H_2O (aq) + CO_2 (g) (below cloud scavenging)

Reduced Nr species like ammonia (NH_3), on the other hand, are involved in the removal of NO_3^- and SO_4^{2-} acidity from the troposphere through their secondary aerosol formation and in- cloud scavenging. As ammonia is the only common base present that is highly soluble in water, it is considered to be primary removal mechanisms of NO_y and SO_x from the troposphere by the way of its following neutralization reaction (Bauer et al., 2007).

NH_3 (g) + HNO_3 (g) <—> NH_4NO_3 (s)
NH_3 (g) + H_2SO_4 (g) —> $(NH_4)_2SO_4$(s)
(in cloud scavenging)

Considering the lack of comprehensive data for Nr deposition fluxes in the western Uttar Pradesh (U.P), restricted by the three month monsoon period (June – September) over the Indian region, the present study has been carried out to determine the levels of NH_4^+ and NO_3^- along with their wet deposition fluxes in Mathura, which is one of the highly polluted cities of U.P characterized by its rapid urbanization. Chemical composition of rain was analyzed for major water soluble ions. Nr scavenging pattern were established based on the rainfall intensity. Neutralization ratios were calculated for ascertaining the source of influence in the chemistry of wet scavenging. Such estimates could further be used for not only substantiating our current understanding of rain water chemistry but also in correlating the changes in the aquatic ecosystem arising from its deposition pathways.

METHOD

Sampling location

Mathura is located at 27.45°N 77.67°E in U.P state of India. Lying on the western side of river Yamuna at 140Km south east of Delhi and 60km North West of Agra, it is a suitable representation of typical urban site characterized by rapid population growth and unplanned urban sprawl. Being a major tourist hub, it is also known for hosting a number of tourist activities resulting in frequent traffic congestions from the vehicular movement throughout the year. Presence of Mathura oil refineries near the city premises has further added to its worsening air quality. The temperature averages at 25.6°C, which becomes highest during June (34.7°C) and lowest during January (14.7°C). The city receives an average precipitation of 707 mm, with a minimum during April (2 mm) and maximum during August (271 mm).

Sample collection:

Rain water samples were collected using a manual set up of polypropylene funnel with a 14cm diameter and bottle of 1 litre capacity. Prewashed assemble of funnel and bottle was fixed with paraffin and were kept at a 13m height for rain

Table 1. Duration of collection of each rain event along with one set of sequential sampling during 7th event.

Event no.	Date of sampling	Time in	Time out	Duration	pH	Volume
1.	04/07	16:05	17:00	55min	6.57	250
2.	09/07	16:25	17:24	60min	6.47	450
3.	02/08	11:50	13:48	118min	6.18	125
4.	09/08	11:30	14:39	189min	6.38	340
5.	10/08	15:15	15:45	30min	5.68	125
6.	14/08	19:27	21:50	143min	5.9	60
7.(I)	15/08	13:55	14:10	15 min	6.12	220
(II)	15/08	14:10	14:25	15 min	6.13	260
(III)	15/08	14:25	14:40	15 min	6.27	45
(IV)	15/08	14:40	18:36	236 min	6.15	36

water collection. Sample collector was withdrawn after each single rain event and the amount collected was determined using measuring cylinder before storing the sample in 125ml polypropylene bottles with thymol. A total of 7 rain events based sampling has been carried out, of which the last event involved collection of 4 sequential samples with varying time period according to the intensity and amount of precipitation. This was done for the purpose of obtaining representative samples during different rain events, to demonstrate the wet scavenging process. The total volume of samples collected was 1911ml during monsoon period of June, July and August.

Sample analysis:

Physical parameters viz. volume, pH and conductance were measured immediately after collection of samples. Chemical composition of rain water, on the other hand, were analyzed for its major cations (K^+, NH_4^+, Na^+, Ca^{2+}, Mg^{2+}) and anions (F^-,Cl^-,NO_3^-, SO_4^{2-}) with the help of ion chromatography (Metrohm–883 basic plus model). Metrosep A SUPP 4, 250/4.0 column and an eluent of 1.8 mmol/L Na_2CO_3 and 1.7 mmol/L $NaHCO_3$ at a flow rate of 1.0ml/min with Metrohm suppressor technique were used for determination of anions. The cation separation was achieved with the metrosep C4-100/4.0 and an eluent of 1.7 mmol/L Nitric acid and 0.7 mmol/L dipicolinic acid at a flow rate of 0.9 ml/min without suppressor. Calibration of the method and quantification of components were carried out using MERCK reference standards (CertiPUR) of 1, 2, 5 ppm for anions and 2, 5, 10 ppm for cations.

Data quality assessment:

The quality of data was checked with the help of ion balance assuming the accountability of major cations (K^+, NH_4^+, Na^+, Ca^{2+}, Mg^{2+}) and anions (F^-,Cl^-,NO_3^-, SO_4^{2-}) for most of the ions in the sample. Based on the principle of electro- neutrality, the cationic equivalents of the rain water sample should be equal to its anionic equivalents (Ayers, 1995; WMO, 1994). The average sum of equivalent cations ($\sum C$) and anions ($\sum A$) was observed to be $\sum C = 270.8\ \mu eq/l$ and $\sum A = 180.5\ \mu eq/l$. The relative ion difference was calculated to be 0.2 using the following formula:

$$\text{Relative ion balance} = (\sum C - \sum A) / (\sum C + \sum A)$$

Total percent ion difference was found to be well within the acceptable range with slight deviation arising from the unanalyzed HCO_3^- (Kulshrestha et al., 2003).

RESULTS AND DISCUSSION

Ionic composition of rain water

Equivalent concentration of all the major cations and anions in the rain events is given in Table 2. The average values showed the dominance of Ca^{2+} in the rain water composition due to its strong crustal source of influence over the Indian atmosphere (Kulshrestha et al., 1998, 2003). Strong neutralization affinity of Ca^{2+} with SO_4^{2-}, on the other hand, has inadvertently resulted in the dominance of SO_4^{2-} amongst the major anions (Kulshrestha et al., 2003; 2014b). This has also been evident from our sequential sampling of 15th August single rain event where a decreasing trend in the SO_4^{2-} concentrations clearly indicated scavenging by Ca^{2+} at every interval. The order of its average ionic concentration in the rain water sample was, therefore, observed to be as: $\mathbf{Ca^{2+} > SO_4^{2-} > Cl^- > NH_4^+ > NO_3^- > Na^+ > Mg^{2+} > K^+ > F^-}$

Changing dynamics of the ionic contribution with the sequential progression of rainfall events were analysed for its cationic and anionic fractions (Figure 2). The observations showed a decline in the contribution of anionic fraction for the samples that were collected during the consecutive period of rainfall events. Absence of active sources of NO_3^- and Cl^- has furthermore provided a sharp decline in their percent contribution to the total ionic composition as

Table 2. Concentration of major ions in rain samples (μeq l^{-1}).

Sample	Cl^-	NO_3^-	SO_4^{2-}	F^-	Na^+	NH_4^+	K^+	Ca^{2+}	Mg^{2+}
04-Jul	180.9	78.5	160.6	4.0	43.1	60.1	60.2	2.2	64.4
09-Jul	22.9	24.8	35.6	1.7	48.8	63.2	7.3	73.6	13.3
02-Aug	305.9	145.2	170.8	1.1	38.2	56.7	91.6	119.4	117.0
09-Aug	70.1	66.7	78.6	2.0	46.2	61.0	25.2	129.2	19.2
10-Aug	74.0	0.9	73.0	4.0	47.2	62.5	26.8	61.4	75.2
14-Aug	27.5	28.2	16.0	0.8	48.6	63.1	9.4	96.4	22.6
15-Aug (I)	48.5	40.6	132.9	0.6	46.3	62.9	19.8	131.8	37.0
15-Aug(II)	20.4	19.2	52.4	1.7	47.6	63.3	9.3	86.8	18.1
15 – Aug (III)	13.9	18.3	36.1	1.7	51.8	65.8	4.7	99.2	15.9
15 Aug (IV)	15.1	45.6	25.2	1.7	48.0	62.5	3.4	116.5	23.0
Average	**77.9**	**46.8**	**78.1**	**1.9**	**46.6**	**62.3**	**25.8**	**91.6**	**40.6**
Min.	**13.9**	**0.9**	**16.0**	**0.6**	**38.2**	**56.7**	**3.4**	**61.4**	**13.3**
Max.	**305.9**	**145.2**	**170.8**	**4.0**	**51.8**	**65.8**	**91.6**	**131.8**	**117.0**
S.D.	**87.33**	**40.14**	**49.15**	**0.92**	**3.47**	**2.32**	**25.96**	**23.26**	**33.22**

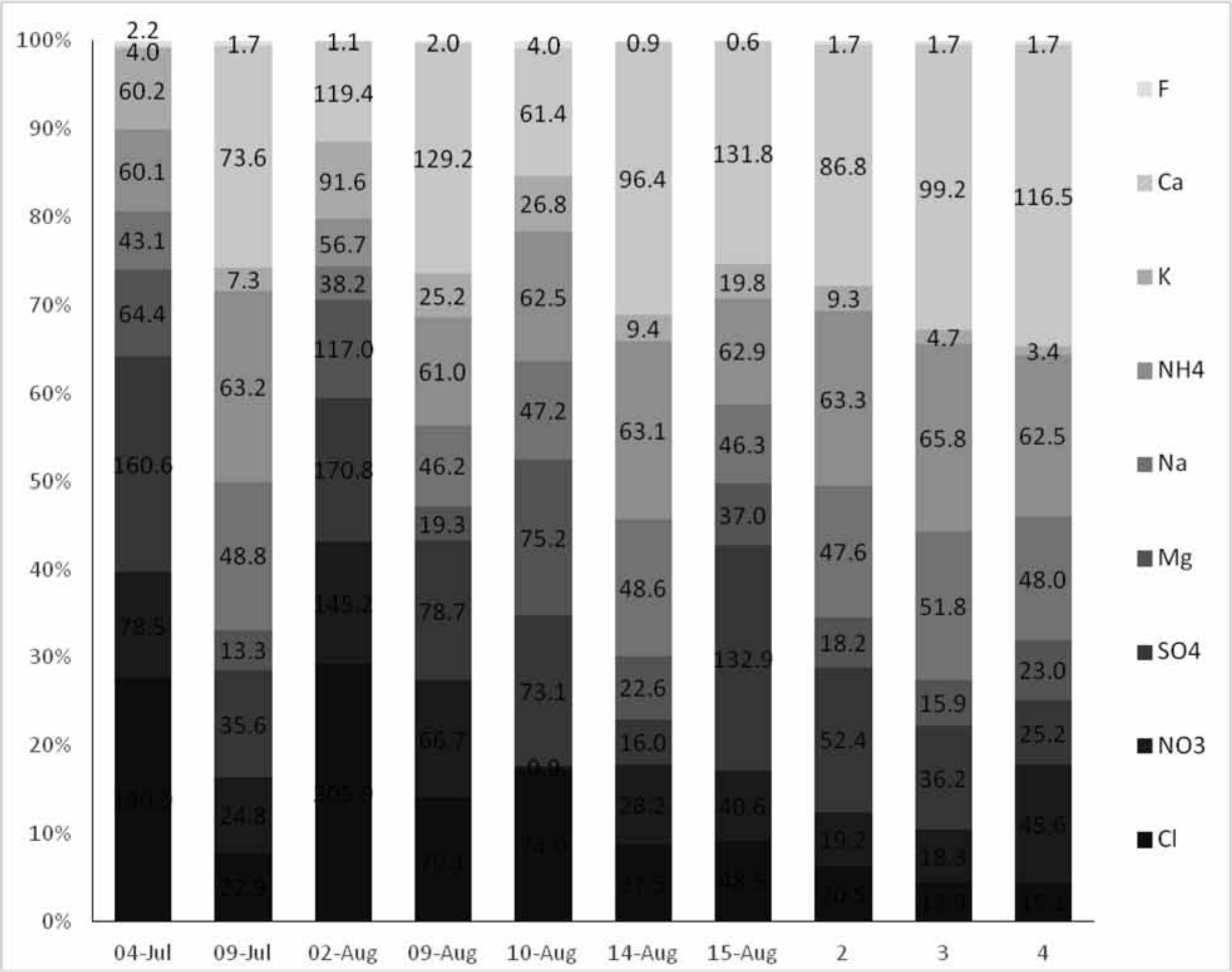

Figure 2. Percentage contribution of major cations and anions for rain event samples.

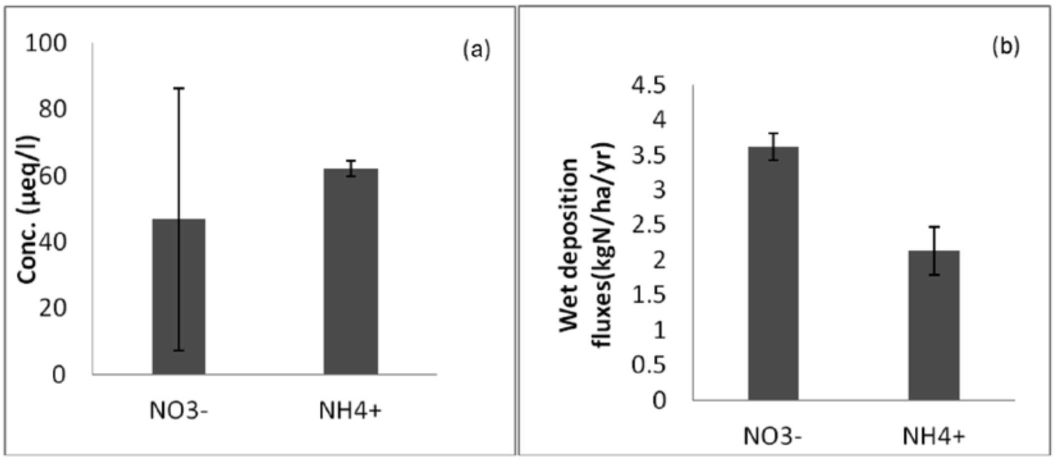

Figure 3. Mean values of NH_4^+ and NO_3^- showing (a) relative abundances in rain water samples (μ eq l^{-1}) and (b) wet deposition fluxes (kgN ha^{-1} y^{-1}) over Mathura.

evident from their 9th and 10th August sequential samples. Similar pattern of ionic contribution has also been observed for the intermittently collected samples from the single rain event of 15th Aug due to its continuous scavenging process for the acidic anionic fractions.

Wet deposition fluxes of Nr:

Relative abundance of reduced (NH_4^+) as well as oxidized (NO_3^-) species of Nr in rain water is shown in Figure 3 (a). While NH_4^+ exhibited minimum variability in its equivalent concentration values ranging from 56.7 to 65.8 μeq l^{-1} with an average of 62.3 ± 2.32 μeq l^{-1}, NO_3- showed a range of 0.9 - 145.2 μeq l^{-1} at an average concentration of 46.8 ± 40.14 μeq l^{-1}. Such variability could be attributed to the progression of monsoon providing active source of NH_4^+ through organic matter decomposition on one hand and continuous scavenging of acidic NO_3^- during consecutive rainfall events on the other.

The resulting wet deposition fluxes of Nr were calculated from their abundances in the rain water sample using the following formula (Du and Liu, 2014).

$$R_i = \Sigma\, C_i \times P_i \times 0.01$$

Where R_i (kg $ha^{-1}y^{-1}$) is annual wet deposition flux, C_i (mg/l) is the concentration of Nr for each precipitation event and P_i is the depth of precipitation (mm).

In synchronization with their relative abundances, the wet deposition fluxes of NH_4^+-N (3.62 kg $ha^{-1}y^{-1}$) were also observed to be higher than NO_3^- - N (2.13 kg $ha^{-1}y^{-1}$) as shown in Figure 3 (b). Such a pattern clearly reflected the growing influence of ammonium source and their active emissions from agriculture and sewage contributing towards their increased scavenged values during monsoon. Lack of active sources of NO_3^- resulting from the low solubility of their precursor gases (NO_X) in the rain water samples and its continuous wet scavenging through buffering reaction has consequently lowered the wet deposition fluxes of NO_3^- - N over the study area. These values might be underestimated values for entire year due to non - inclusion of pre and post monsoonal rain events in the sampling schedule.

Scavenging pattern of Nr:

Variability in the concentration of NH_4^+ and NO_3^- in rain water samples was evaluated with the changing rainfall intensity as shown in Figure 4. NO_3^- being acidic in nature showed higher concentrations during initial period of sampling, thereby, indicating towards their dominant removal through below cloud scavenging reactions. However, with the gradual monsoon progression, a declining trend in NO_3^- concentration from the high rainfall intensities was observed especially for consecutive rain fall events. Such a trend was further established in the sequential sampling of 15th Aug single rain event showing a decline in the NO_3^- concentrations after each intermittent sampling due to its continuous scavenging. NH_4^+ being basic in nature showed variability irrespective of the rain fall intensities. This indicates the dominant removal through in-cloud scavenging process.

In order to find out the scavenging neutralization reactions of Nr, regressions of NO_3^- were plotted with NH_4^+ and Ca^{2+} for ascertaining their wet scavenging mechanism (Figure 5). The results showed the ionic concentrations of NO_3^- and NH_4^+ following a similar pattern with a strong correlation of 0.92 possibly arising from their coexistence in the atmosphere. This, in turn is indicative of their dominant removal through in cloud scavenging process via NH_4NO_3 formation. Also with NH_4^+ and NO_3^- showing systematic variations with rain intensities, the removal of

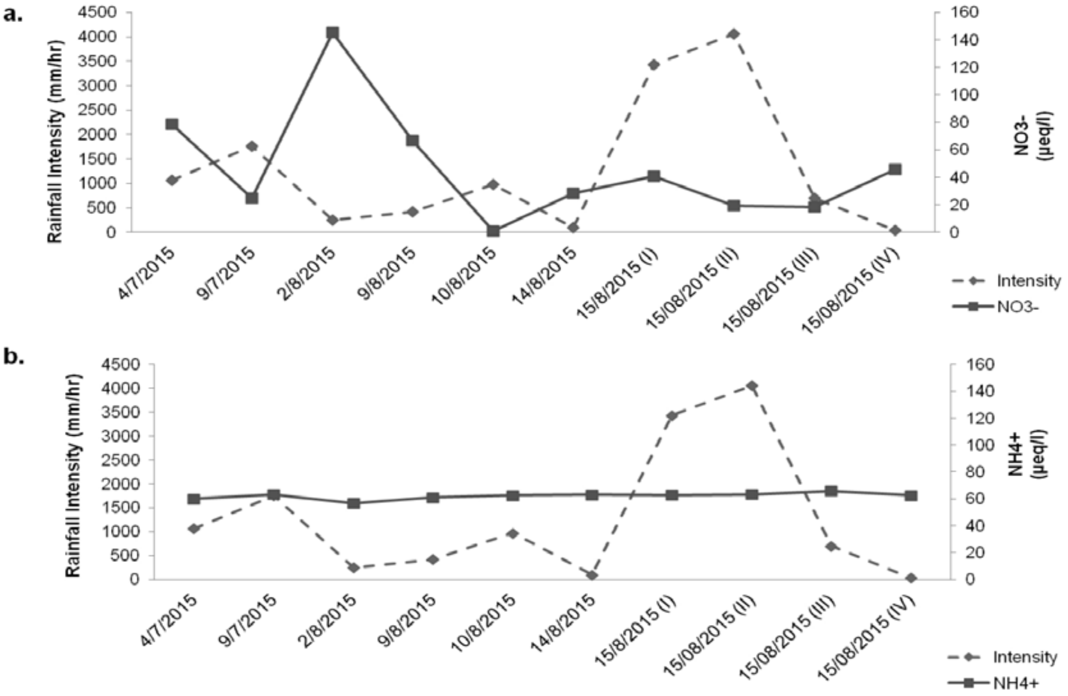

Figure 4. Nr variability with rainfall intensity for a) NH_4^+ and b) NO_3^-.

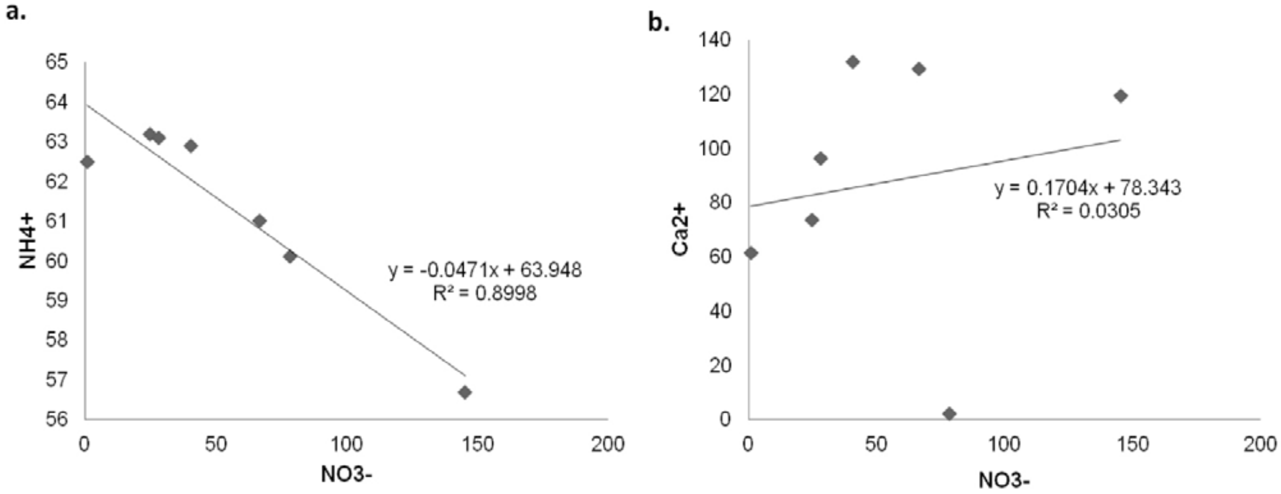

Figure 5. Regression plots of NO_3^- with a) NH_4^+ and b) Ca^{2+} in μeq l^{-1}.

NH_4NO_3 could, therefore, be rendered more effective under low intensity rain through small droplets.

Neutralization capacity of the wet deposition:

Neutralization capacity of the rain water samples was calculated for further substantiating the observed pattern of Nr scavenging (Table 3). It is defined by the relative abundance of sulphate plus nitrate in comparison with calcium and ammonium in rain water samples using the following formula (Alastuey et al., 2001):

$$\text{Neutralizing capacity} = \frac{SO_4^{2-} \quad + \quad NO_3^-}{Ca^{2+} \quad + \quad NH_4^+}$$

Neutralization capacity of the rain water samples was observed to be lowest for the first few initial period of sampling due to the presence of heavy loading of neutralizing agent (Ca^{2+} and NH_4^+) predominantly from their crustal sources in the atmosphere. Subsequent progression of

Table 3. Neutralization capacity of rain water on event basis.

Event	Neutralization capacity
04-Jul	0.38
09-Jul	0.44
02-Aug	1.79
09-Aug	0.76
10-Aug	0.6
14-Aug	0.28
15-Aug (I)	0.89
15 – Aug (II)	0.48
15 – Aug (III)	0.33
15 – Aug (IV)	0.40

the monsoon, on the other hand, provided increasing neutralization capacity from their continuous scavenging of acidic components (NO_3^- and SO_4^{2-}). Nevertheless, their values were observed to be <1 for most of the samples. This suggests the exceedance of major cations against the major anions, especially during consecutive rain events. Sequential sampling of the single rain event of 15th Aug showed similar pattern of neutralization capacity having lowest values during first 15 minutes of rain. With the results showing a 45% decline for the initial 15 minutes followed by 31% decline during the next 15 minute change in its neutralization capacities, a faster scavenging of coarse mode particle of Ca^{2+} associated with SO_4^{2-} and NO_3^- in comparison to their fine mode fraction could be established.

CONCLUSION

The expanding industrial and agricultural sectors significantly contribute to the atmospheric Nr, which are eventually removed by rainfall. The observations showed an abundance of NH_4^+ (62.3 ± 2.3 μeql^{-1}) over NO_3^- (46.8 ± 40.1 μeql^{-1}) in the rain water samples eventually creating a significant contribution of wet deposition fluxes of NH_4^+ - N (3.6 kg ha^{-1} y^{-1}) in comparison to the NO_3^- - N (2.1 kg ha^{-1} y^{-1}) at Mathura. Scavenging patterns confirmed the presence of NH_4NO_3, showing variations along with the rainfall intensity. Correlation of 0.92 between NH_4 and NO_3 also supported these graphical observations, thereby, confirming the dominant forms in which these Nr species are being deposited over the study area. Their scavenging pattern confirmed initial period of rain events are dominated by the below cloud scavenging process. The subsequent monsoonal advancement reveals that the below cloud scavenging remains less effective, especially during consecutive rain events. Neutralization capacities of the rain water samples further substantiated such patterns in the Nr scavenging, thereby, confirming its effective removal through NH_4NO_3 formation by small droplets of rain during low intensity rainfall.

ACKNOWLEDGMENTS

We sincerely thank the financial support received from DST PURSE and UGC to conduct this research work. Thanks are due to Dr. Saumya Singh for proper evaluation of the manuscript and useful suggestions. We sincerely thank Chief Editor for continued support and editing of the manuscript.

Compliance with Ethical Standards

The authors declare that they have no conflict of interest and adhere to copyright norms.

REFERENCES

Alastuey, A., Querol, X., Chaves, A., Lopez - Soler, A., and Ruiz, C.R., 2001. Wet only sequential deposition in a rural area in north eastern Spain. Tellus B, v.53, no.1, pp: 40-52.

Ayers, G.P., 1995. (November). Some practical aspects of acid deposition measurement. In Presentation to the Third Expert Meeting on Acid Deposition Monitoring Network in East Asia Japan: Niigata Prefecture. pp: 14-16.

Bauer, S.E., Koch, D., Unger, N., Metzger, S.M., Shindell, D.T., and Streets, D.G., 2007. Nitrate aerosols today and in 2030: a global simulation including aerosols and tropospheric ozone. Atmospheric Chemistry and Physics, v.7, no.19, pp: 5043-5059.

Bhaskar, V.V., and Rao, P.S.P., 2016. Annual and decadal variation in chemical composition of rain water at all the ten GAW stations in India. Journal of Atmospheric Chemistry, pp: 1-31.

Dentener, F., Drevet, J., Lamarque, J.F., Bey, I., Eickhout, B., Fiore, A.M., Hauglustaine, D., Horowitz, L.W., Krol, M., Kulshrestha, U.C., and Lawrence, M., 2006. Nitrogen and sulfur deposition on regional and global scales: a multimodel evaluation. Global biogeochemical cycles, v.20, no.4, pp: 1-21.

Du, E., and Liu, X., 2014. High rates of wet nitrogen deposition in China: A synthesis. In Nitrogen Deposition, Critical Loads and Biodiversity Springer Netherlands., pp: 49-56.

Galloway, J.N., Dentener, F.J., Capone, D.G., Boyer, E.W., Howarth, R.W., Seitzinger, S.P., Asner, G.P., Cleveland, C.C., Green, P.A., Holland, E.A., and Karl, D.M., 2004. Nitrogen cycles: past, present, and future. Biogeochemistry, v.70, no.2, pp: 153-226.

Galloway, J.N., Townsend, A.R., Erisman, J.W., Bekunda, M., Cai, Z., Freney, J.R., Martinelli, L.A., Seitzinger, S.P., and Sutton, M.A., 2008. Transformation of the nitrogen cycle: recent trends, questions, and potential solutions. Science, v.320, no.5878, pp: 889-892.

Grömping, A.H.J., Ostapczuk, P., and Emons, H., 1997. Wet deposition in Germany: long-term trends and the contribution of heavy metals. Chemosphere, v.34, no.9-10, pp: 2227-2236.

Jain, M., Kulshrestha, U.C., Sarkar, A.K., and Parashar, D.C., 2000. Influence of crustal aerosols on wet deposition at urban and rural sites in India. Atmospheric Environment, v.34, no.29, pp: 5129-5137.

Kulshrestha, U.C., 2013. Encyclopedia of Environmental Management. v.6, no.7, pp: 8.

Kulshrestha, U.C., Sarkar, A.K., Srivastava, S.S., and Parashar, D.C., 1996. Investigation into atmospheric deposition through precipitation studies at New Delhi (India). Atmospheric Environment, v.30, no.24, pp: 4149-4154.

Kulshrestha, U.C., Saxena, A., Kumar, N., Kumari, K.M., and Srivastava, S.S., 1998. Chemical composition and association of size-differentiated aerosols at a suburban site in a semi-arid tract of India. Journal of Atmospheric Chemistry, v.29, no.2, pp: 109-118.

Kulshrestha, U.C., Kulshrestha, M.J., Sekar, R., Sastry, G.S.R., and Vairamani, M., 2003. Chemical characteristics of rainwater at an urban site of south-central India. Atmospheric Environment, v.37, no.21, pp: 3019-3026.

Kulshrestha, U.C., Granat, L., Engardt, M., and Rodhe, H., 2005. Review of precipitation monitoring studies in India—a search for regional patterns. Atmospheric Environment, v.39, no.38, pp: 7403-7419.

Kulshrestha, U.C., Kulshrestha, M.J., Satyanarayana, J., and Reddy, L.A.K., 2014a. Atmospheric deposition of reactive nitrogen in India. In Nitrogen Deposition, Critical Loads and Biodiversity. Springer Netherlands, pp: 75-82.

Kulshrestha, M.J., Singh, R., Duarah, R., and Rao, P.G., 2014b. Influence of crustal aerosols on wet deposition at a rural site of North-East India. International Journal of Environmental Studies, v.71, no.4, pp: 510-525.

Norman, M., Das, S.N., Pillai, A.G., Granat, L., and Rodhe, H., 2001. Influence of air mass trajectories on the chemical composition of precipitation in India. Atmospheric Environment, v.35, no.25, pp: 4223-4235.

Parashar, D.C., Granat, L., Kulshreshta, U.C., Pillai, A.G., Naik, M.S., Momin, G.A., Rao, P.P., Safai, P.D., Khemani, L.T., Naqvi, S.W.A., and Narvekar, P.V., 1996. Chemical composition of precipitation in India and Nepal: a preliminary report on an Indo-Swedish project on atmospheric chemistry. Department of Meteorology; Stockholm University; Stockholm; Sweden.

Reis, S., Pinder, R.W., Zhang, M., Lijie, G., and Sutton, M.A., 2009. Reactive nitrogen in atmospheric emission inventories. Atmospheric Chemistry and Physics, v.9, no.19, pp: 7657-7677.

Singh S., Kumar B., Sharma A., and Kulshrestha U.C., 2017. Wet deposition fluxes of atmospheric inorganic reactive nitrogen at an urban and rural site in the IndoGangetic Plain. Atmospheric Pollution Research, http://dx.doi.org/10.1016/j.apr.2016.12.021.

Tørseth, K., Aas, W., Breivik, K., Fjæraa, A.M., Fiebig, M., Hjellbrekke, A.G., Lund Myhre, C., Solberg, S. and Yttri, K.E., 2012. Introduction to the European Monitoring and Evaluation Programme (EMEP) and observed atmospheric composition change during 1972–2009. Atmospheric Chemistry and Physics, v.12, no.12, pp: 5447-5481.

Vet, R., Artz, R.S., Carou, S., Shaw, M., Ro, C.U., Aas, W., Baker, A., Bowersox, V.C., Dentener, F., Galy-Lacaux, C., and Hou, A., 2014. A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus. Atmospheric Environment, v.93, pp: 3-100.

Wallace, J.M., and Hobbs, P.V., 2006. Atmospheric science: an introductory survey. Academic press, v.92.

WMO (World Meteorological Organization) 1994. Report of the Workshop on Precipitation Chemistry Laboratory Techniques, WMO TD., v.658, pp: 14-15.

Received on: 6.3.17; Revised on: 16.4.17; Accepted on: 18.4.17