

## Gaps in Nitrogen Deposition Measurements in South Asia

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The growing demand of energy and food has resulted in increased consumption of fossil fuels giving rise to huge emissions of NO<sub>x</sub> and NH<sub>3</sub> gases. Both these gases are important reactive nitrogen species. Coal, petrol and diesel are the major sources of NO<sub>x</sub> while urea fertilizer is the major source of NH<sub>3</sub><sup>1,2</sup>. Urea is produced through Haber-Bosch process during which inert nitrogen (N<sub>2</sub>) is converted into urea. When we apply urea in the agricultural fields, its thermal dissociation gives rise to NH<sub>3</sub> and CO<sub>2</sub> in air. For every one molecule of NH<sub>3</sub>, one molecule of CO<sub>2</sub> is emitted.<sup>3</sup> Globally, urea fertilizer consumption has increased from 50 million tons in 1961 to 215.37 million tons in 2019. Similarly, in India, the urea consumption is increased from 1 million ton in 1960s to around 33.5 million tons in 2019-20 (<https://factly.in/data-chemical-fertilizer-consumption-increased-by-about-16-in-the-last-six-years/>). Global consumption of fossil fuels such as petroleum oil is increased from 17790 TWh in 1965 to 51170 TWh in 2021, coal is increased from 16140 TWh in 1965 to 44473 TWh in 2021 and gas from 6304 TWh in 1965 to 40375 TWh in 2021 (<https://ourworldindata.org/fertilizers>). these increasing trends of energy consumption which are reflected in the study reporting increasing atmospheric NO<sub>x</sub> trends in different regions in India.<sup>4</sup> Most of south Asian countries have geared up their fossil fuel usage for energy production after 1990.

Both NH<sub>3</sub> and NO<sub>x</sub> are deposited from the atmosphere through wet and dry deposition processes and are generally estimated as NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> respectively. Chemical composition of atmospheric deposition is an indicator of air pollution of the region. In the process of wet deposition, the pollutants are deposited through rain, snow and hails. In the dry deposition process, the pollutants are removed through dry deposition process during dry weather conditions. Dustfall is also a kind of dry deposition of coarse particles taking place during dry weather conditions. Dustfall is very common phenomenon in the south Asian region.<sup>5</sup>

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The atmospheric dust being alkaline in nature, controls acid rain occurrence through buffering action.<sup>6</sup> However, high loadings of particulate matter due to atmospheric dust are considered responsible for poor air quality.

According to the reports, the Himalayan snow is highly affected by the impact of long range transport of reactive nitrogen species.<sup>7</sup> In another study, Singh and co-workers<sup>8</sup> have reported that  $\text{NO}_3^-$  content in rain water was increased by 11.7 times in 2011 as compared to 1994 values. Significantly large increase in ambient  $\text{NO}_x$  in air and  $\text{NO}_3^-$  concentrations in rain water in Delhi during past two decades indicates an increase in fossil fuel consumption. Among  $\text{NH}_3$ -N,  $\text{NH}_4^+$ -N and  $\text{NO}_3^-$ -N, gaseous  $\text{NH}_3$  contributes maximum fraction of Nr in the atmosphere in the region. The Indo-Gangetic region experiences the highest wet deposition of  $\text{NH}_4^+$  due to very high population density and the related anthropogenic activities. At rural sites, the high  $\text{NH}_3$  levels can be attributed to the fertilizers and biomass burning while at urban sites, major sources of gaseous  $\text{NH}_3$  include human excreta, municipality waste and vehicular traffic etc.<sup>9</sup> In addition, the high temperature of the tropics and the alkaline nature of aerosols also favour the building up of  $\text{NH}_3$  in the atmosphere in this region.<sup>10</sup> Abundance and phase distribution studies show that particulate  $\text{NH}_4^+$  is noticed lower than gaseous  $\text{NH}_3$  in Delhi during all the seasons.

A total of 1.97 Tg of wet deposition and 1.67 Tg of dry deposition of Nr species takes place in India.<sup>11</sup> Emissions vs deposition budget still needs more measurements of atmospheric deposition through systematic dense network of sites. In general, the number of reactive nitrogen studies of wet and dry deposition from South Asia is very limited. Also, there are very few measurements about the throughfall deposition in the south Asian region. However, there has been an appreciable development about the measurements of nitrogen assessment from individual groups in the region. At present, UKRI-GCRF-South Asian Nitrogen Hub, WMO-Global Atmospheric Watch (GAW) and DRS Net-India programs are carrying out measurements of atmospheric deposition of reactive nitrogen species in south Asia. Earlier atmospheric deposition studies in the region have been carried out under Composition of Asian Deposition (CAD), Composition of Aerosol and Precipitation in India and Nepal (CAAP) programs of Swedish Development Authority (SIDA).<sup>12-14</sup>

The long term data limited to rain water chemistry are available from the WMO GAW sites.<sup>15</sup> Other important reports in this regard include the studies reported by Dentener and co-workers<sup>16</sup> and other groups.<sup>17-18</sup> Deposition fluxes of coarse mode  $\text{NH}_4^+$  and  $\text{NO}_3^-$  particles on the indoor plants have been reported highlighting their impacts on the biochemical properties of the plants.<sup>19</sup> The Indian Nitrogen Assessment' is a very recent compilation of various studies about different aspects of reactive nitrogen in India.<sup>20</sup> However, forest N is not much covered in this report. In Nepal, the precipitation is found to have significant influence of pollution even at a remote site in the Khumbu region of the Himalayas where relatively high value of scavenging ratio of  $\text{NO}_3^-$  was noticed as compared to other ions.<sup>21</sup>  $\text{NO}_3^-$  concentrations in aerosols were one third of  $\text{NH}_4^+$  but in precipitation, the  $\text{NO}_3^-$  concentrations were greater than  $\text{NH}_4^+$  due to the presence of gaseous  $\text{HNO}_3$  in the air. This feature was more dominant when air was blowing from more polluted areas.

Model vs measurement comparison studies are very important for developing deposition prediction capabilities. In a pioneering effort, the measurements of the reactive nitrogen species such  $\text{NH}_4^+$  and  $\text{NO}_3^-$  in rain water in India have been compared with the modeling output by Kulshrestha and co-workers<sup>14</sup> by using MATCH model.<sup>22</sup> The output of the model was helpful in providing an interpretation of the observational data of  $\text{NH}_4^+$  and  $\text{NO}_3^-$ . However, such exercises are further needed to reduce the uncertainties of the model outputs.

This is to mention that there have been some issues of QA/QC of the data in south Asiaregion mainly due to the problems associated with sampling and chemical analysis of the reactive nitrogen species such as  $\text{NH}_4^+$  and  $\text{NO}_3^-$ . Kulshrestha and co-workers have reviewed the precipitation chemistry data in the region.<sup>14</sup> These workers found that in most of the studies,  $\text{NH}_4^+$  was under estimated due to analytical problems such as delay in analysis, no addition of preservative, improper storage of the samples etc. while  $\text{NO}_3^-$  was over estimated due to its additional contribution from local soil. Ion balance and conductivity balance

approaches are supposed to be reported to support the analytical assurance. But most of the earlier studies lack such estimates of errors. Most of the measurements included  $F^-$ ,  $Cl^-$ ,  $NO_3^-$ ,  $SO_4^{2-}$ ,  $Na^+$ ,  $K^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $NH_4^+$  in ionic and conductivity balance calculations. The pH of rain water in India is relatively high having high concentrations of  $HCO_3^-$  ion due to the influence of calcareous soils of this region. Hence, ion balance and conductivity balance checks without  $HCO_3^-$  do not really assure the data quality. Sometimes the results are influenced by the shape and material type (steel, plastic, glass) of the collection assembly. Improper transport, preservation and delay in analysis are also very important factors affecting the quality of the data.<sup>23-24</sup> In order to have good quality measurements, it is necessary to select sites which can represent larger areas. Overall, an adherence to QA/QC of data is mandatory for ensuring reliable and robust data sets in the long run.

### Gaps and Recommendations

The south Asia region needs the following actions to fill the existing knowledge gaps related to N cycle-

- i) Need to develop an integrated assessment plan for various Nr species including their emissions, abundance, transport, transformations, scavenging, impacts, forest pool etc.
- ii) Good quality data availability is limited for Nr depositions in Indian region which needs a dedicated data and parameter specific protocol, inter-lab comparison exercises and a reference standards development initiative.
- iii) A long term measurement network covering a number of sites of different characteristics with the aim to use its research findings in the policy making.
- iv) In order to understand local, trans-boundary and long range transport of Nr species, the deposition studies need to be coupled with the trajectory analysis. It will be a good idea to constitute a program to monitor an import and export of pollution within south Asian countries under The South Asian Association for Regional Cooperation (SAARC).
- v) We need to focus on the impact studies of various Nr species.
- vi) Budgets of dry deposition of gases and aerosols especially  $NH_3$  and  $NH_4^-$  in indoor environment will be highly useful.
- vii) In order to understand the process involving in gas-aerosol interactions, scavenging, transport, evapo-transpiration, deposition and uptake etc., we need to develop a separate task force. Appropriate emphasis needs to be given on dry deposition studies of Nr in order to reduce uncertainties in Nr budget of this region.
- viii) Also, there is a need to develop a common modeling group involving active scientist/groups. It would be more appropriate if we include some socio-economic expert(s) which will help in translating the scientific findings into an impactful report needed for the policy makers.

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