Chapter 48
Progress in Nitrogen Deposition Monitoring and Modelling

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Abstract The chapter reviews progress in monitoring and modelling of atmospheric nitrogen (N) deposition at regional and global scales. The Working Group expressed confidence in the inorganic N wet deposition estimates in U.S., eastern Canada, Europe and parts of East Asia. But, long-term wet or dry N deposition information in large parts of Asia, South America, parts of Africa, Australia/Oceania, and oceans and coastal areas is lacking. Presently, robust estimates are only available for inorganic N as existing monitoring generally does not measure the

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complete suite of N species, impeding the closing of the atmospheric N budget. The most important species not routinely measured are nitrogen dioxide (NO_2), ammonia (NH_3), organic N and nitric acid (HNO_3). Uncertainty is much higher in dry deposition than in wet deposition estimates. Inferential modelling (combining air concentrations with exchange rates) and direct flux measurements are good tools to estimate dry deposition; however, they are not widely applied. There is a lack of appropriate parameterizations for different land uses and compounds for input into inferential models. There is also a lack of direct dry deposition flux measurements to test inferential models and atmospheric model estimates.

Keywords Inorganic • Modelling • Monitoring • Organic • Wet and dry deposition

48.1 Introduction

This chapter reports the findings of a Working Group to address progress in monitoring and modelling of atmospheric nitrogen (N) deposition at regional and global scales. A background paper by Dentener et al. (2014, Chap. 2, this volume), presentations, and posters set the stage for Working Group discussions. The group focused on current knowledge in modelling and measurements of N deposition, identification of important gaps, and recommendation for future needs, including capacity-building at a regional level, assessing scientific uncertainties, understanding co-benefits beyond N deposition, and identifying links with other processes. The spatial and temporal trends of N emission and deposition around the world are described for inorganic and organic N.

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48.2 Status of Monitoring

The World Meteorological Organization's Scientific Advisory Group in Precipitation Chemistry (WMO SAG PC) is currently working on an assessment to review and synthesize the state of the science on the chemical composition of precipitation and deposition of major ions for the period 2000–2008. The regional presentations and posters further emphasized the different regional challenges regarding the understanding of key processes and sources as well as the quality assurance of the measurements. As an example, 90% of precipitation in India occurs from June to September, meaning that dry deposition is of much more importance in major parts of the year, in contrast to high precipitation regions in Northern Europe and parts of North America.

The observations show these general patterns:

- The highest levels of total nitrogen (N) deposition occur in China and India with much lower values in Africa. The Arctic and Antarctic represent the cleanest regions, depending on long-range transport for deposition inputs.
- Preliminary estimates of wet deposition show a range of 4–7 kg N ha^{-1} $year^{-1}$ in eastern North America, 5 to> 7 kg N ha^{-1} $year^{-1}$ in Europe, while sites in East Asia experience wet deposition between 2 and 60 kg N ha^{-1} $year^{-1}$. In Africa the estimated total deposition (dry plus wet) is typically between 7–10 kg N ha^{-1} $year^{-1}$.
- The emissions of NO_x and NH_3 have been increasing dramatically in Asia since the 1980s, and leading to increased N deposition which is of great concern both on a regional and global level. NO_x emissions have on the other hand decreased in North America and Europe.
- Organic N deposition is not routinely measured in all regions and the quality of measurements is often dubious; however the organic N contribution may be high (10–40%) in some areas (i.e. in parts of China).

Knowledge of the data quality of the measurements is certainly of great importance when comparing measurements within and across networks. In many regions, the recommended measurements guidelines (i.e., by WMO) are not followed, decreasing confidence in these measurements. Examples of measurement issues include improper sampling method and storage, and delay in chemical analysis. This may cause degradation of samples, which is especially important for N species. For example, ammonium in stored rainwater samples may oxidize to nitrate. Furthermore, many sites are not always representative for a large region (e.g., urban sites) and these have limited value especially for model validation (see also Dentener et al. 2014, Chap. 2, this volume).

48.3 Gaps and Recommendations for Monitoring

Several necessary improvements were identified during the workshop. Priorities vary by region and the intended use of data, whether it is for compliance monitoring to identify and quantify sources for better abatement strategies, or to better understand key atmospheric processes and/or to validate regional and global models. The most important gaps which are important for all these aspects include:

- Long-term monitoring sites for routine air concentration and wet N deposition measurement are lacking in several regions in the world; most evident are South America, large parts of Africa, and Central Asia.
- Several networks do not measure all species and it is recommended to increase the number of N species routinely measured to close the atmospheric N budget, most notably to include organic N, NH_3, HNO_3 and NO_2 in atmospheric concentration measurements, as the basis for estimating dry deposition more comprehensively.
- Greater attention must be paid to monitoring of organic N, which may be of significant importance in several regions. Furthermore, there is a need to develop a standard harmonized method for measuring organic N to improve the quality of the data and for comparability between regions/sites.
- In light of the relative importance of dry deposition to total deposition loads, it is highly recommended that more long-term direct flux measurements are established in major parts of the world, and to use recommended methods for partitioning of gaseous and particulate N (i.e., by using denuder sampling methods).
- The diurnal variation may vary considerably for several N species, and daily or weekly measurements will not capture these variations. It is therefore recommended that a few supersites in each region measure with higher temporal resolutions (i.e., hourly).
- The data quality is not always satisfactory, and there is a need to continue to increase capacity building for measuring wet and dry deposition in the developing world. The Working Group encouraged participation in WMO quality assurance programmes and further interaction and cooperation between regional networks and programmes.
- The Working Group recommended assessment of uncertainty in the deposition estimates to be used either for model validation or critical load assessment.

48.4 Status of Modelling

As a general statement, it is necessary to emphasize that models are essential additions to measurements as they provide evidence of source attribution and chemistry and transport pathways, and can be used for emissions scenario analysis. Models can also help to fill gaps in areas where monitoring is limited (chemically and geographically). However, it is important to note that the models are no better than the inputs included. Thus, high quality input data (i.e., spatial and temporal emission data) and good validation tools (measurement networks) are a necessity for having confidence in the results. Therefore, the confidence in the model results is very much reflected in the measurement gaps described above. Generally, it can be said that:

- Regional and global model representation of wet deposition is reasonable in Europe and the US, but still very uncertain in other parts of the world.
- Preliminary global model estimates show the highest levels of total N deposition (wet + dry, oxidized N + reduced N) occur in China (15–45 kg N ha^{-1} $year^{-1}$) and India (15–35 kg N ha^{-1} $year^{-1}$), followed by Europe (15–25 kg N ha^{-1} $year^{-1}$) and eastern North America (10–20 kg N ha^{-1} $year^{-1}$), with the lowest values in Africa (5–15 kg N ha^{-1} $year^{-1}$), South America (5–15 kg N ha^{-1} $year^{-1}$) and Oceania (< 5 kg N ha^{-1} $year^{-1}$); typical ranges are given.
- The relative contribution of oxidized N to total is dominant in North America, Europe, northern Asia and northern Africa. Reduced N is dominant in South America, Asia and Africa.
- Dry deposition dominates in California, northern Africa, parts of South America, Africa and Oceania.
- Modelled dry deposition fluxes are very uncertain, and can seldom be compared to measurements, due to many factors including incompleteness of observations, uncertainties in chemistry schemes, and sparseness of measurements.
- The confidence level in model outputs increases with accurate emissions data and available data for validation.

48.5 Gaps and Recommendations for Modelling

In addition to problems with high quality measurement data to validate the model output and good emission data, more specific challenges in the model parameterizations exist and necessary improvements were identified by the Working Group:

- Chemical transport modellers should closely follow improvements in meteorological parameterization coming from the meteorological community. Currently, the global and regional chemical transport models have problems characterizing the hydrological cycle (precipitation amounts), especially in situations with complex orography. Further, the night time boundary layer representation must be improved and cloud/fog deposition approaches should be developed.
- Land use data included in models vary between groups, and the Working Group recommended harmonizing land-use between atmospheric and ecosystem models and further development of the sub-grid scale representations.
- The dry deposition processes described in the models need to be developed; bi-directional exchange of NH_3 should be incorporated, and unidirectional deposition of oxidized N and sulphur (S) species should be updated further.
- Since there is a serious lack of wet and dry deposition measurements, models need to be verified and improved utilizing alternative datasets as much as possible, such as ambient concentrations, dedicated field study data and model intercomparisons. Promising datasets are becoming available from satellites, but more work must be done to consistently combine the model and measurement frameworks.

- New emission datasets are becoming available, and should be checked using knowledge in countries and regions i.e. 'ground-truthing'. For instance, NO_x emission estimates from lightning and ships are important sources for some regions and must be addressed in the models.
- In order to obtain accurate spatial and temporal emission estimates, especially for NH_3, meteorology driven modules should be applied.
- Capacity for modelling wet and dry deposition should be increased in the developing world.
- Understanding of the sources of bias should be developed further. Areas that must be addressed include missing sources, such as lightning NO_x and NH_3 emissions from soil and organic N; uncertain processes like dry deposition/air surface exchange; and occult deposition (cloud droplet deposition), wintertime photochemistry/free troposphere chemistry.
- There are uncertainties in the split of coarse and fine nitrate aerosol that have implications for dry deposition processes. The Working Group recommended using intensive campaign data to further improve the parameterization of gas/particle phase of N, as well as the size distribution of aerosols species.

48.6 Conclusions

- The Working Group expressed confidence in the inorganic N wet deposition estimates in U.S., eastern Canada, Europe and parts of East Asia. We are lacking long-term wet or dry N deposition information in large parts of Asia, South America, parts of Africa, Australia/Oceania, and oceans and coastal areas.
- Presently, robust estimates are only available for inorganic N as existing monitoring generally does not measure the complete suite of N species, impeding the closing of the atmospheric N budget. The most important species not routinely measured are nitrogen dioxide (NO_2), ammonia (NH_3), organic N and nitric acid (HNO_3).
- Uncertainty is much higher in dry deposition than in wet deposition estimates.
- Inferential modelling (combining air concentrations with exchange rates) and direct flux measurements are good tools to estimate dry deposition; however, they are not widely applied. There is a lack of appropriate parameterizations for different land uses and compounds for input into inferential models. There is also a lack of direct dry deposition flux measurements to test inferential models and atmospheric model estimates.

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References

Dentener, F., Vet, R., Dennis, R. L., Du, E., Kulshrestha, U. C., & Galy-Lacaux, C. (2014). Progress in monitoring and modelling estimates of nitrogen deposition at local, regional and global scales. In M. A. Sutton, K. E. Mason, L. J. Sheppard, H. Sverdrup, R. Haeuber, & W. K. Hicks (Eds.), Nitrogen deposition, critical loads and biodiversity (Proceedings of the International Nitrogen Initiative workshop, linking experts of the Convention on Long-range Transboundary Air Pollution and the Convention on Biological Diversity). (Chapter 2; this volume). Springer.