
An integrated perspective on assessing agricultural air quality

Viney P. Aneja*

Department of Marine, Earth and Atmospheric Sciences,
North Carolina State University, Raleigh, NC 27695 – 8208, USA
E-mail: viney_aneja@ncsu.edu
*Corresponding author

Dev Niyogi

Departments of Agronomy and Earth and Atmospheric Sciences,
Purdue University, West Lafayette, IN 47907 – 2054, USA
E-mail: dnyyogi@purdue.edu

Paul A. Roelle

CMR 420 Box 1971, APO AE 09063, USA
E-mail: roellep@gimail.af.mil

Abstract: The biogeochemical cycling of trace gases (e.g. nitrogen, sulphur, etc.), and contaminants on local, regional, and global scales is a complex system of emissions, transformations, transport, and deposition. To date, limited, if any, attempt has been made on quantifying and identifying direct emissions of gaseous sulphur compounds from agricultural operations. This represents a major regulatory need for sound and prudent environmental practice. In this paper, we summarise an integrated assessment framework for studying the agricultural air quality issues by discussing the various components of the research, education and outreach involved.

Keywords: agricultural air quality; air emissions; air pollution; ammonia; animal feeding operations; biogeochemical cycles; education; hydrogen sulphide; North Carolina; outreach.

Reference to this paper should be made as follows: Aneja, V.P., Niyogi, D. and Roelle, P.A. (2006) 'An integrated perspective on assessing agricultural air quality', *Int. J. Global Environmental Issues*, Vol. 6, Nos. 2/3, pp.137–148.

Biographical notes: Dr Viney P. Aneja is a Professor in Air Quality and Environmental Technology in the Department of Marine, Earth and Atmospheric Sciences at the North Carolina State University in Raleigh, North Carolina. He was recently selected to serve on the United States Environmental Protection Agency's Science Advisory Board, and is a member of the United States Department of Agriculture's Agricultural Air Quality Task Force.

Dr Dev Niyogi is an Assistant Professor with joint appointment in the Departments of Agronomy and Earth and Atmospheric Sciences at Purdue University. He is also the State Climatologist for Indiana and the Director of the Indiana State Climate Office. His research seeks to advance the understanding and representation of the effects of vegetation and land surface processes in environmental, weather forecasting, and climate analyses, using a combination of dynamical models and surface, satellite remote sensing and multiscale analysis products. Additional details regarding the projects and the activities of the land surface modelling group can be found at www.landsurface.org.

Dr Paul A. Roelle is a Major in the US Air Force and is the Commander of the weather unit providing meteorological support to the US Army's V Corps in Heidelberg, Germany. While in this position, Maj Roelle deployed to Iraq and served as the lead meteorologist in Iraq during Operation IRAQI FREEDOM and assisted with the reconstruction of the Iraqi Meteorological Service. Maj Roelle graduated from the Pennsylvania State University with a degree in Meteorology and then pursued a MS and PhD in Atmospheric Sciences from the North Carolina State University.

1 Introduction

Emissions of odour, gaseous nitrogen i.e. ammonia (NH_3), oxides of nitrogen ($\text{NO}_x = \text{NO} + \text{NO}_2$) and nitrous oxides (N_2O), and gaseous sulphur compounds (e.g. hydrogen sulphide) in eastern North Carolina have now become a significant problem, both politically and environmentally, owing to the enhanced intensively managed crop and animal agriculture. Intensive agriculture is a major industry accounting for revenues in billions of dollars in the United States. In North Carolina alone, the hog farming accounts for revenues in excess of \$2 billion. This has created environmental (air, water and soil) concerns that demand immediate attention (Aneja et al., 2006; NRC, 2003). Significant effort has been put into the understanding of gaseous nitrogen emissions from agricultural operations (e.g. Aneja et al., 2000, 2001a, b). Present research such as from the USDA Agricultural Air Quality initiative and large collaborative projects such as Project OPEN (Odour, Pathogen, and Emissions of Nitrogen) through the Animal and Poultry Waste Management Center (Aneja, 2001) has provided the initial framework and results related to ammonia and odour emissions from swine confined animal feeding operations (CAFOs) in North Carolina.

In this paper, we summarise an integrated assessment framework for studying the agricultural air quality issues by discussing the various components involved.

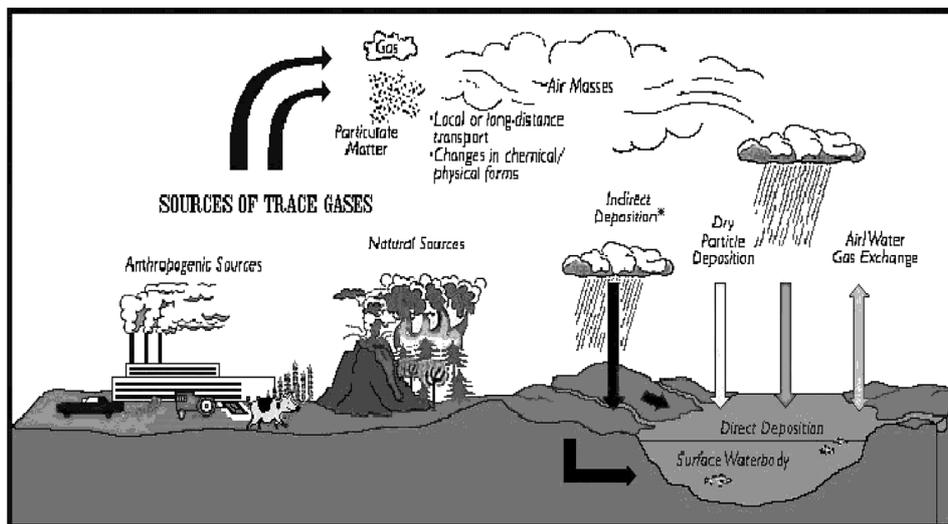
2 Components of integrated agricultural air quality assessment programme

2.1 Research needs

The biogeochemical cycling of trace gases (e.g. nitrogen, sulphur, etc.) the contaminants on local, regional and global scales is a complex system of emissions, transformations, transport and deposition (Aneja et al., 2001a, Figure 1). To date, limited, if any, attempts

have been made on quantifying and identifying direct emissions of gaseous sulphur compounds from agricultural operations. This represents a major regulatory need for sound and prudent environmental practice. Characterising simultaneous gaseous nitrogen and sulphur emissions from large-scale swine production facilities such as in North Carolina and the Midwestern United States requires an understanding of the multiple source terms and temporal patterns in animal production that exist for each type of operation. The emissions of gaseous nitrogen and sulphur compounds may pose serious problems, both from the point of odour (associated with reduced nitrogen and sulphur compounds) and fine particulate matter (PM_{fine}) formation (Baek and Aneja, 2004) that may affect recreation and human health. Atmospheric deposition of nitrogen compounds can contribute significantly to eutrophication and other nutrient loading effects.

Figure 1 Atmospheric emissions, transport, transformation and deposition of trace gases



*Indirect deposition is direct deposition to land followed by runoff or seepage through groundwater to a surface water body.

Source: Aneja et al. (2001a)

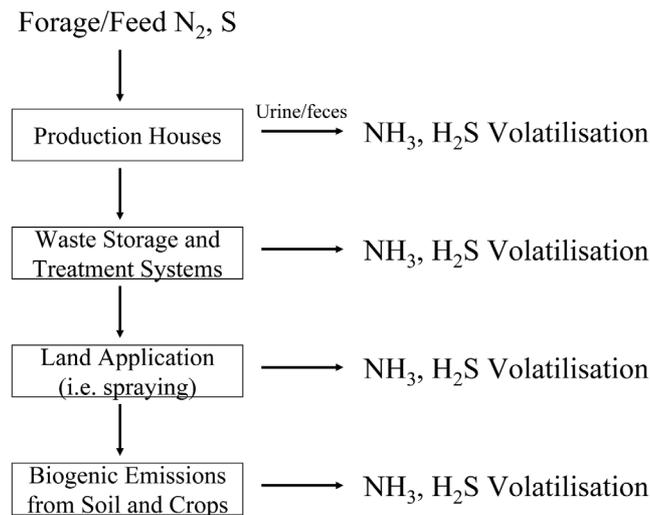
Most atmospheric nitrogen and sulphur compounds fall into two broad categories: oxides of nitrogen and sulphur, and reduced nitrogen and sulphur. The reduced form of nitrogen is primarily dominated by ammonia species ($\text{NH}_3 + \text{NH}_4^+$, denoted NH_x), and the reduced form of sulphur is primarily dominated by hydrogen sulphide (H_2S). Much attention has been devoted to studying the roles of oxides of nitrogen and sulphur in the atmosphere and towards reducing their emissions (NRC, 2003). However, there has been little focus on the cycling of NH_x compounds and H_2S in the atmosphere and their role in determining budgets of tropospheric acids and particulate matter.

2.1.1 Ammonia/ nitrogen from CAFOs

The role of ammonia as an alkaline component of the atmosphere has long been recognised (Junge, 1954). Ammonia is the most abundant gas-phase alkaline species in the troposphere, and can neutralise a great portion of acids produced through the oxidation of SO_2 and NO_x . Emissions of NH_3 are generally associated with intensively managed

agriculture, primarily intensive livestock agriculture (Figure 2). Current estimates of NH_3 budgets in the troposphere indicate that a significant fraction of NH_3 originates from animal manure (Battye et al., 2003; Dentener and Crutzen, 1994; Warneck, 1988). NH_3 differs from NO_x and SO_2 in that a large fraction of NH_3 may be deposited within a short distance of the source, because its emissions are almost entirely at ground level and its deposition velocity for most surfaces is relatively high (Hov and Hjøllo, 1994; Hov et al., 1994). NH_4^+ found in the atmosphere originates from the association of NH_3 with acid aerosols, and plays an important role in determining the aerosol composition. Knowledge of this composition and consequently deliquescence and optical properties of the aerosols is important to determine their potential radiative effects (Boucher and Anderson, 1995). Ammonium (NH_4^+) in aerosols can also be transported over large distances (Aneja et al., 2001a) and thus may play a significant role in nitrogen budgets in remote regions (Rhome et al., 2002). Quantification of the atmospheric deposition of both oxidized and reduced nitrogen compounds is of considerable interest because of the detrimental effects related to excess nutrient loading in sensitive ecosystems (Niyogi et al., 2003). Pearl (1997) and Pearl et al. (1999) suggest that reduced nitrogen species may be more biologically active compared to the oxidised forms of nitrogen in such environments, so that not only is it important to quantify the total nitrogen loading, but also the form in which it is delivered.

Figure 2 Major routes for NH_3 and H_2S emissions from conventional confined animal feeding operations in North Carolina, USA



Source: Aneja et al. (2001a)

2.1.2 Hydrogen sulphide/sulphur from CAFOs

Similar to the nitrogen compounds, sulphur compounds contribute substantial natural emissions into the troposphere. Anthropogenic emissions consist almost entirely of sulphur oxides (SO_2), whereas natural emissions are mainly in the form of reduced sulphur compounds (Aneja et al., 1979 a, b, 1980, 1984; Aneja, 1990; Finlayson-Pitts and Pitts, 1986). The major reduced sulphur compounds consist of hydrogen sulphide (H_2S), dimethyl sulphide (CH_3SCH_3), dimethyl disulphide (CH_3SSCH_3), carbonyl sulphide

(COS), carbon disulphide (CS₂), and methyl mercaptan (CH₃SH). Total reduced sulphur is the combined load of all its major reduced sulphur compounds naturally emitted into the atmosphere. Total reduced sulphur may, therefore, be defined as SH_x, where

$$\text{SH}_x = \text{H}_2\text{S} + \text{CH}_3\text{SCH}_3 + \text{CH}_3\text{SSCH}_3 + \text{COS} + \text{CS}_2 + \text{CH}_3\text{SH}.$$

Volatilisation from animal waste is an important source of gaseous reduced sulphur compounds, particularly in eastern NC, where the swine industry constitutes a major agricultural activity. Though estimates of natural sources of sulphur are available for volcanoes, oceans, biomass burning and soils and plants (Warneck, 1988), no such estimates are available for confined animal feeding operations. Preliminary measurements of hydrogen sulphide concentrations near these systems have found them to be significant.

Study of sulphur compounds is also important because of their contribution to the gaseous malodors (O'Neill and Phillips, 1992; Schiffman et al., 2001; Zahn et al., 2001).

Very little observational data on simultaneous ambient N and S compound level and deposition amounts are available to adequately quantify their budgets. For example, over the last decade, emissions of N and S compounds in the eastern US have increased significantly as a result of the intense growth in the livestock industry. Aneja et al. (1998), for instance, estimate that in North Carolina the nitrogen emitted as NH₃ is comparable to that emitted as NO_x. Such a budget for sulphur compounds does not exist. Current regulations under the Clean Air Act (CAA) directed towards the surface ozone control are likely to reduce the NO_x emissions while emissions of NH₃ could grow further in the future. These evolving emissions scenarios could potentially have a significant impact on tropospheric chemical composition over North America. Future studies should address these needs for intensively managed agriculture in general and on agricultural animal feeding operations in particular.

2.1.3 Deposition

Gaseous deposition from emissions associated with both crop and animal operations is of concern since it contributes to eutrophication and acidification of semi-natural ecosystems (Aneja et al., 1986, 2001a; Bull and Sutton, 1998; Erisman, 2001; Krupa, 2003). Deposition estimates have been traditionally obtained through analysis and systems such as the National Atmospheric Deposition Program (NADP) with wet deposition being the deposition pathway. However, as reviewed in Krupa (2003), for instance, reliance on wet deposition measurements alone can lead to considerable underestimates (by 40 to 60%, with higher value for agricultural regions) of the total (wet + dry) atmospheric N deposition.

In contrast to many other pollutant gases, which are consistently deposited, the exchange of NH₃ between the atmosphere and the land surface is controlled by both atmospheric and land surface processes and can be bi-directional (Aneja et al., 1998, Niyogi et al., 2003; Phillips et al., 2004). The occurrence of either emission or deposition depends on the nitrogen (N) status of the ecosystem. For intensively managed crop agricultural ecosystems, the N balance is mainly driven by agricultural fertilisation (Harper et al., 1996; Schjoerring, 1991; Sutton et al., 1993). However, in unfertilised semi-natural and natural ecosystems, deposition of atmospheric NH₃ is typical (Duyzer, 1994; Sutton et al., 1995; Wyers and Erisman, 1998), and other forms of fixed nitrogen (e.g. wet deposition of NH₄⁺ and NO₃⁻, dry deposition of HNO₃ and NO₂) can influence biodiversity. There are important feedback effects of ecosystems on the atmosphere:

intense long-term N and S inputs may lead to changes in the ecosystem functioning, related to the increased nutrient status of the vegetation and litter, that decrease net N and S input to the ecosystem (Sutton et al., 1995). In addition, intensively managed agricultural systems have the potential to produce significant NH_3 and H_2S emissions, and thus influence the atmospheric N and S balance. Field measurements and modelling analyses are needed which estimate deposition (velocity) for N (primarily ammonia) and S compounds in the vicinity of the CAFOs where emission flux measurements should be undertaken. The regional wet deposition also needs to be systematically analysed using regional networks such as NADP (Aneja et al., 2003; NADP, 2003; Walker et al., 2000) and Clean Air Status Trends Network (CASTNet, 2002).

2.1.4 Source – receptor analysis: back trajectories and stable isotope measurements

Understanding the source (emission) – receptor (deposition) relationships will provide information that is important for regulators and policy makers. This can be investigated by:

- the combination clustering of back trajectory analyses from Hybrid Single Particle Lagrangian Integrated Trajectories – HY-SPLIT Model results (Aneja et al., 2003; Dorling et al., 1992; Draxler, 1997; Rhome et al., 2002) and emission source categorisation based on EPA emission inventory; along with
- stable isotope measurements of N and S deposition samples.

The potential of the stable isotopes of nitrogen and sulphur to investigate the sources – receptor relationships for atmospheric sulphur and nitrogen has long been recognised (Freyer, 1978; Grey and Jensen, 1972; Heaton, 1986; Moore, 1977; Nriagu et al., 1991; Xiao and Liu, 2001). Using stable isotopes as a source tracer requires that isotopic composition of one source be significantly different from other potential sources (Macko and Ostrom, 1994). By combining the isotopic composition of the major acidic components of atmospheric deposition (N and S) with information on air mass source region, it is possible to determine where, geographically, the nitrogen and sulphur compounds originate (Xiao and Lui, 2001). However, there are limited studies of this type published in the literature and this analysis involving regional meteorology/air mass analysis and regional isotopic measurement has a knowledge gap at this time, which needs to be addressed in agricultural air quality studies.

2.1.5 Air quality modelling

Attempts at detailed 3D modelling of the nitrogen and sulphur compounds for assessing the effects associated with agricultural air quality have also been minimal. Although some modelling efforts have characterised the atmospheric distributions of NH_x compounds over North Carolina (Mathur and Dennis, 2003), Europe (EMEP, 2002; EUROTRAC, 1997, Fowler et al., 1998), and on global scales (Adams et al., 1981; Dentener and Crutzen, 1994), similar studies for characterising their regional and local distributions over the continental US are lacking. The primary confounding factors limiting such investigations have been the lack of understanding, and consequently the model treatment, of the sources, sinks, and chemical interactions of NH_x and reduced sulphur compounds in the atmosphere. At present, the magnitude and temporal variability of anthropogenic and natural emissions of NH_3 are still poorly quantified (Bouwman et al., 1997). The

atmospheric chemical and thermodynamic coupling of reduced nitrogen compounds with the oxides of sulphur and nitrogen, is now beginning to be understood (e.g. Baek and Aneja, 2004; Seinfeld and Pandis, 1998). It dictates that the modelling framework be capable of accurately representing the oxidant chemistry related to the formation of ozone and acidic substances. Further, the local to regional nature of NH_3 emission sources, coupled with its atmospheric lifetime (of the order of a few hours), suggest that the modelling framework should have sufficient spatial and temporal resolution. A model for atmospheric behaviour of NH_x and H_2S must also be able to describe both the atmospheric transport of NH_3 and H_2S near the source, as well as the transport of NH_4^+ and sulphates (SO_4^{2-}) over long distances.

In the context of these needs, research is required which will provide the development of a comprehensive, multidisciplinary integrated study that combines observational and model development and application that is capable of representing the transport, transformation and deposition of NH_3 , NH_4^+ , PM_{fine} , H_2S and SO_4^{2-} derived from it. Improvements in the quantification of atmospheric sources and sinks of these compounds, suggested by the isotopic and back trajectory analysis conducted during the field measurements should be synthesised in a comprehensive air quality model:

- to assess the fate of these emissions on a regional basis
- to provide quantitative estimates of the atmospheric budgets of these compounds
- to assess the potential impact on airborne PM levels arising from changes in emissions associated with changes in future agricultural practices.

Studies should also present in detail the applications of the air quality models to the different regions of the US and the development of seasonal and annual estimates of both ambient concentrations and deposition amounts for nitrogen and sulphur species and evaluate the results against systematic measurements from specialised observations and regional networks.

2.2 Outreach needs

Air quality affects everyone. However, the air quality impacts, regulations and technological developments have different connotations for different sectors. For agricultural air quality the stakeholders include the community in the vicinity of the agriculturally intensive area, ecologists, government regulators, industries, technology providers, academic professionals, concerned citizens, the news media and elected officials. Each of these groups may perceive problems and opportunities differently, but all need access to information as it is developed. The diversity and complexity of air quality problems contribute to the challenges faced by extension and outreach professionals who must communicate with stakeholders having diverse backgrounds.

To address these challenges, while communicating air quality research results and concepts to stakeholders, three areas of information needs are evident.

- A basic understanding of the fundamental concepts regarding air pollutants and their measurement and control is needed by all stakeholders.
- Each problem or set of problems must be reasonably well defined since a comprehensive solution of all problems simultaneously may not be feasible; for instance, the solution of an odour problem associated with animal production may not address atmospheric effects due to ammonia emissions.
- The integrity of the communication process must be preserved by avoiding prejudice and protectionism; although stakeholders may seek to modify information to enhance their interests, extension and outreach professionals must be willing to present unwelcome information or admit to a lack of information.

A solid grounding in fundamental concepts, clearly defined statement of the air quality issue(s), and a resolute commitment to integrity and credibility will enable effective communication of air quality information to and among diverse stakeholders and should be the focus of the outreach component.

The objective of the outreach programme should be to increase the scientific understanding of citizens, regulators, and farm operators on issues related to air quality and public health. The outreach programme should incorporate a variety of extension teaching methods, including publications, workshops, presentations, field tours, news articles and websites targeted at specific audiences. The success of the outreach programme can be evaluated based on the implementation of best farm management practices for controlling emissions and improvements in regulations and enforcement programmes. For this task, the Cooperative Extension Service (CES) are typically well-positioned to conduct a comprehensive outreach programme on managing air emissions from animal feeding operations.

2.3 Educational needs

The effect of intensively managed agriculture in general and large scale confined animal feeding operations (CAFOs) in particular, is a critical issue being hotly debated in Europe. The US Research on agricultural air quality should be integrated into both the undergraduate and graduate education programmes. Opportunities exist to develop field-research orientated courses in the area of current environmental concerns. Students in the introductory courses can visit the research sites, help take observations and incorporate data from the ongoing research into reports on different aspects of the air quality issues. They can present the results of their research to other students and colleagues. Graduate education is an integral part of the research programme; all research efforts can involve graduate students as part of their thesis work. In addition to these courses, short courses on emerging agricultural air quality issues such as sulphur compounds, nitrogen compounds and particulate matter in the atmosphere should also be developed.

3 Conclusions

The current scientific knowledge base for emissions of nitrogen, carbon, hydrocarbon and sulphur compounds from intensively managed agriculture, both crop and animal, and the ultimate fate of the compounds is directly comparable to the situation in the 1980s with regard to the contribution of the agricultural nonpoint sources to nutrient contamination of water. There is just enough scientific information to recognise that there is a potential for serious problems, but the information is not sufficiently detailed or complete to understand the extent of such problems or to make scientifically credible recommendations on their potential solutions. At present, there are insufficient facts for objective regulators to make important decisions that may have a great influence on both air and water quality, human health and the economy of agricultural regions.

Both the lack of, and need for, information on gaseous emissions from animal operations and their effects were well illustrated by a feature article in the July 20, 2003 edition of a Raleigh, N.C. newspaper (News and Observer). The paper reported that a company was considering building a large CAFO about ten miles from the state's largest wildlife refuge. The article detailed immediate opposition to the installation because of the potential for air quality problems in the refuge. Others pointed out the very significant economic boost for an economically depressed region. The problem that the decision makers will have is that their decision cannot be based on sound scientific data, because the data base is simply insufficient. The ultimate permit decision will be based on incomplete scientific knowledge. Although this is a regional decision, the scientific information gap would exist anywhere in the US where a new CAFO might be proposed. This problem is not unique to N.C. but exists in several areas of the US and Europe.

A very large percentage of the vast scientific database which currently exists for nonpoint source water pollution was obtained in research projects supported, in part, by the United States Department of Agriculture programme. Sound scientific information is the only way in which good decisions can be made with regard to regulation or lack of a need for the regulation of emissions of these gases.

The scientific information gathered from this much needed research can be shared with the scientific community via education, outreach and extension. The feedback from these activities will ensure that the data and interpretations made from them are scientifically credible. The results ought to be presented to State and Federal Regulatory and Legislative bodies as appropriate for their understanding of processes involved and the probable results of any rules or regulatory actions. The information is also valuable for developing the best management practices as recommendations for producers.

Acknowledgments

It is our pleasure to acknowledge support from the US Department of Agriculture National Research Initiative Competitive Grants Program Contract # 2003-05360 (Dr R. Knighton, Project Officer). We also sincerely appreciate the discussions and support provided by the graduate students at the Air Quality Research Program at North Carolina State University.

References

- Adams, D.F., Farwell, S.O., Robinson, E., Pack, M.R. and Bamesberger, W.L. (1981) 'Biogenic sulphur source strengths', *Environ. Sci. Technol.*, Vol. 15, pp.1493–1498.
- Aneja, V.P. (1990) 'Natural sulphur emissions into the atmospheric', *Journal of Air and Waste Management Association*, Vol. 40, pp.469–476.
- Aneja, V.P. (2001) 'An integrated study of the emissions of ammonia, odor and odorants, and pathogens and related contaminants from potential environmentally superior technologies for swine facilities', Project OPEN, Proposal funded by Animal and Poultry Waste Management Center/Smithfield Foods, Raleigh, NC, December 2001.
- Aneja, V.P., Adams, D.F. and Pratt, C.D. (1984) 'Environmental impact of natural emissions', *Journal of the Air Pollution Control Association*, Vol. 34, No. 8, pp.799–803.
- Aneja, V.P., Chauhan, J.P. and Walker, J.T. (2000) 'Characterization of atmospheric ammonia emissions from swine waste storage and treatment lagoons', *Journal of Geophysical Research*, Vol. 105, pp.11535–11545.
- Aneja, V.P., Malik, B.P., Tong, Q., Kang, D. and Overton, J.H. (2001b) 'Measurement and modelling of ammonia emissions at waste treatment lagoon-atmospheric interface', *Journal of Water, Air and Soil Pollution-Focus*, Vol. 1, pp.177–178.
- Aneja, V.P., Murray, G.C. and Southerland, J. (1998) 'Atmospheric nitrogen compounds: emissions, transport, deposition, and assessment', *EM*, April, pp.22–25.
- Aneja, V.P., Nelson, D., Roelle, P.A., Walker, J. and Battye, W. (2003) 'Agricultural ammonia emissions and ammonium concentration associated with aerosols and precipitations in the Southeast United States', *Journal of Geophysical Research-Atmosphere*, Vol. 108, pp.ACH 12-1–12-11.
- Aneja, V.P., Overton, J.H., Cupitt, L.T., Durham, J.L. and Wilson, W.E. (1979a) 'Direct measurements of emission rates of some atmospheric biogenic sulphur compounds', *Tellus*, Vol. 31, No. 2, pp.174–178.
- Aneja, V.P., Overton, J.H., Cupitt, L.T., Durham, J.L. and Wilson, W.E. (1979b) 'Carbon disulphide and carbonyl sulphide from biogenic sources and their contribution to the global sulphur cycle', *Nature*, Vol. 282, No. 5738, pp.493–496.
- Aneja, V.P., Overton, J.H., Cupitt, L.T., Durham, J.L. and Wilson, W.E. (1980) 'Measurements of emission rates of carbon disulphide from biogenic sources and its possible importance to the stratospheric aerosol layer', *Chemical Engineering Communication*, Vol. 4, No. 6, pp.721–727.
- Aneja, V.P., Roelle, P.A., Murray, G.C., Southerland, J., Erisman, J.W., Fowler, D., Asman, W.A.H. and Patni, N. (2001a) 'Atmospheric nitrogen compounds II: emissions, transport, transformation, deposition and assessment', *Atmos. Environ.*, Vol. 35, pp.1903–1911.
- Aneja, V.P., Rogers, H.H. and Stahel, E.P. (1986) 'Dry deposition of ammonia at environmental concentrations on selected plant species', *Journal of the Air Pollution Control Association*, Vol. 36, No. 12, pp.1338–1341.
- Back, B.H. and Aneja, V.P. (2004) 'Measurement and analysis of the relationship between ammonia, acid gases, and fine particles in Eastern North Carolina', *Journal of Air and Waste Management Association*, Vol. 54, pp.623–633.
- Battye, W., Aneja, V.P. and Roelle, P.A. (2003) 'Evaluation and improvement of ammonia emissions inventories', *Atmospheric Environment*, Vol. 37, pp.3873–3883.
- Boucher, O. and Anderson, T.L. (1995) 'General circulation model assessment of the sensitivity of direct climate forcing by anthropogenic sulfate aerosols to aerosol size and chemistry', *J. Geophys. Res.*, Vol. 100, No. 26, pp.117–26, 134.
- Bouwman, A.F., Lee, D.S., Asman, W.A.H., Dentener, F.J., Van Der Hoek, K.W. and Oliver, J.G.J. (1997) 'A global high resolution emission inventory for ammonia', *Global Biogeochem. Cycl.*, Vol. 11, pp.561–587.

- Bull, K.R. and Sutton, M.A. (1998) 'Critical loads and the relevance of ammonia to an effects based Nitrogen Protocol', *Atmos. Environ.*, Vol. 32, pp.565–573.
- Clean Air Status Trends Network (CASTNet) (2002) World wide web site, <http://www.epa.gov/castnet>, accessed December 2002.
- Dentener, F.J. and Crutzen, P.J. (1994) 'A three-dimensional model for global ammonia cycle', *J. Atmos. Chem.*, Vol. 19, pp.331–369.
- Dorling, S.T., David, T.D. and Pierce, C.E. (1992) 'Cluster analysis: a technique for estimating the synoptic meteorological controls on air and precipitation chemistry – method and applications', *Atmos. Environ.*, Vol. 26A, pp.2575–2581.
- Draxler, R.R. (1997) 'Description of the HYSPLIT-4 modelling system', National Oceanic and Atmospheric Administration, Techn. Mem. ERL ARL – 224.
- Duyzer, J.H. (1994) 'Dry deposition of ammonia and ammonium aerosols over heathland', *J. Geophys. Res.*, Vol. 99, pp.18757–18763.
- EMEP Programme (Cooperative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe) web page, <http://www.emep.int>, 2002.
- Erisman, J.W. (2001) *De Vliegende Geest. Ammoniak uit Delandbouw en de Gevolgen voor de Natuur*, Bergen: BetaText, p.271.
- EUROTRAC (1997) *Biosphere-Atmosphere Exchange of Pollutants and Trace Substances*, S. Slanina (Ed), Heidelberg, Germany: Springer.
- Finlayson-Pitts, B.J. and Pitts, J.N. (1986) *Atmospheric Chemistry*, New York: John Wiley & Sons, pp.993–999.
- Fowler, D., Pitcairn, C.E.R., Sutton, M.A., Fléchar, C., Loubet, B., Coyle, M. and Munro, R.C. (1998) 'The mass budget of atmospheric ammonia within 1 km of livestock buildings and deposition patterns within adjacent woodland', *Environ. Pollut.*, Vol. 102 (S1), pp.343–348.
- Freyer, H.D. (1978) 'Seasonal trends of NH₄⁺ and NO₃⁻ nitrogen isotope composition in rain collected at Jillich, 3Germany', *Tellus* Vol. 30, pp.83–92.
- Grey, D.C. and Jensen, M.L. (1972) 'Bacteriogenic sulphur in air pollution', *Science*, Vol. 177, pp.1099–1100.
- Harper, L.A., Bussink, D.W., van der Meer, H.G. and Corre, W. (1996) 'Ammonia transport in a temperate grass system: I. Seasonal transport in relation to soil fertility and crop management', *Agron. J.*, Vol. 88, pp.614–621.
- Heaton, T.H.E. (1986) 'Isotopic studies of nitrogen pollution in the hydrosphere and atmosphere: a review', *Chemical Geology*, Vol. 59, pp.87–102.
- Hov, H. and Hjøllø, B.A. (1994) 'Transport distance of ammonium in Northern Europe 2. Its relation to emissions of SO₂ and NO_x', *J. Geophys. Res.*, Vol. 99 (D9), pp.18,749–18,755.
- Hov, H., Hjøllø, B.A. and Eliassen, A. (1994) 'Transport distance of ammonia and ammonium in Northern Europe 1. Model description', *J. Geophys. Res.*, Vol. 99 (D9), pp.18,735–18,748.
- Junge, C.E. (1954) 'The chemical composition at atmospheric aerosols I. Measurements at Round Hill field station', *J. Meteorol.*, Vol. 11, pp.223–333.
- Krupa, S.V. (2003) 'Effects of atmospheric ammonia (NH₃) on terrestrial vegetation: review', *Environmental Pollution*, Vol. 124, pp.179–221.
- Mathur, R. and Dennis, R.L. (2003) 'Seasonal and annual modelling of reduced nitrogen compounds over the Eastern United States: emissions, ambient levels, and deposition amounts', *J. Geophys. Res.*, Vol. 108 (D15), pp.ACH 22–1 ACH 22–19.
- Macko, S.A. and Ostrom, N.E. (1994) 'Pollution studies using stable isotopes', in K. Lajtha and R.M. Michener, (Eds) *Stable Isotopes in Ecology and Environmental Science*, London: Blackwell Scientific, pp.45–62.
- Moore, H. (1977) 'The isotopic composition of ammonia, nitrogen dioxide, and nitrate in the atmosphere', *Atmospheric Environment*, Vol. 11, pp.1239–1243.

- NADP (2003) National Atmospheric Deposition Program/National Trends Network, <http://nadp.sws.uiuc.edu>, accessed May 2003.
- National Research Council (NRC) (2003) *Air Emissions from Animal Feeding Operations*, The National Academies Press, Washington, DC, p.263.
- Niyogi, D., Alapaty, K. and Raman, S. (2003) 'A photosynthesis based dry deposition modelling approach', *Water Air Soil Pollution*, Vol. 144, pp.171–194.
- Nriagu, J.O., Coker, R.D. and Barrie, L.A. (1991) 'Origin of sulphur in Canadian arctic haze from isotope measurements', *Nature* Vol. 349, pp.142–245.
- O'Neill, D.H. and Phillips, V.R. (1992) 'A review of the control of odour nuisance from livestock building: part 3, Properties of the odorous substance which have been identified in livestock wastes or in the air around them', *Journal of Agricultural Engineering Research*, Vol. 53, pp.23–50.
- Pearl, H.W. (1997) 'Coastal eutrophication and harmful algal blooms: importance of atmospheric deposition and groundwater as "new" nitrogen and other nutrient sources', *Limnol. Oceanography*, Vol. 42, pp.1154–1165.
- Pearl, H.W., Willey, J.D., Go, M., Peierls, L., Pinckney, J.L. and Fogel, M.L. (1999) 'Rainfall stimulation of primary production I western Atlantic Ocean waters: roles of different nitrogen sources and co-limiting nutrients', *Marine Ecology Series*, Vol. 176, pp.205–214.
- Phillips, S.B., Arya, S.P. and Aneja, V.P. (2004) 'Ammonia flux and dry deposition velocity from near-surface concentration gradient measurements over a grass surface in North Carolina', *Atmospheric Environment*, Vol. 38, pp.3469–3480.
- Rhome, J., Niyogi, D. and Raman, S. (2002) 'Assessing seasonal transport and deposition of agricultural emissions in eastern North Carolina, USA', *Pure Appl. Geophys.*, Vol. 160, pp.117–141.
- Schiffman, S.S., Bennett, J.L. and Rayner, J.H. (2001) 'Quantification of odors and odorants from swine operations in North Carolina', *Agricultural and Forest Meteorology*, Vol. 108, pp.213–240.
- Schjoerring, J.K. (1991) 'Ammonia emission from the foliage of growing plants', in T.D. Sharkey, H.A. Mooney, and E.A. Holland, (Eds) *Trace Gas Emissions by Plants*, New York: Academic Press, pp.267–292.
- Seinfeld, J.H. and Pandis, S.N. (1998) *Atmospheric Chemistry and Physics*, New York: John Wiley.
- Sutton, M.A., Pitcairn, C.E.R. and Fowler, D. (1993) 'The exchange of ammonia between the atmosphere and plant communities', *Adv. Ecol. Research.*, Vol. 24, pp.301–393.
- Sutton, M.A., Schjørring, J.K. and Wyers, G.P. (1995) 'Plant-atmosphere exchange of ammonia', *Phil. Trans. R. Soc. Lond. A*, Vol. 351, pp.261–278.
- Walker, J.T., Aneja, V.P. and Dickey, D.A. (2000) 'Atmospheric transport and wet deposition of ammonium in North Carolina', *Atmos. Environ.*, Vol. 34, pp.3407–3418.
- Warneck, P. (1988) *Chemistry of the Natural Atmosphere*, San Diego, CA: Academic, pp.426–441.
- Wyers, G.P. and Erisman, J.W. (1998) 'Ammonia exchange over coniferous forest', *Atmos. Environ.*, Vol. 32, pp.441–451.
- Xiao, H.Y. and Liu, C.Q. (2001) 'Sources of nitrogen and sulphur in wet deposition at Guiyang, southwest China', *Atm Environment*, Vol. 36, pp.5120–5130.
- Zahn, J.A., Hatfield, J.L., Laird, D.A., Hart, T.T., Do, Y.S. and DiSpirito, A.A. (2001) 'Functional classification of swine manure management systems based on effluent and gas emission characteristics', *J. Environ. Qual.*, Vol. 30, pp.635–647.