

MODELLING OF ACID DEPOSITION OVER THE SOUTH AFRICAN HIGHVELD

Yvonne Scorgie¹ and Gerrit Cornelius²

¹ ENVIRON Australia Pty Ltd, 100 Pacific Highway, Sydney, NSW 2060, Australia, yscorgie@environcorp.com

² Airshed Planning Professionals Pty Ltd, P O Box 5260, Halfway House 1685, South Africa, gerritk@airshed.co.za

Abstract

Trends in Nitrogen and Sulphur deposition were modelled over the South African Highveld using the integrated Gaussian puff modeling system CALPUFF. Modelled S deposition correlated with previous measurements for the central Highveld. Annual total S deposition rates were predicted to exceed 3 kg/ha-year over much of the Highveld, with a peak of over 35 kg/ha-year over the central Highveld and a lower peak (20 kg/ha-year) over the southern Vaal Triangle. Dry and wet deposition rates were found to vary independently resulting in significant spatial and temporal variations in ratios of dry to wet deposition. The contribution of dry S deposition was predicted to decrease from over 40% in the northwest parts of the Highveld to less than 10% in south eastern parts. Peak dry deposition contributions were simulated to occur near low level SO_x sources (>60%). Wet S deposition was projected to dominate in the vicinity of high stacks. Predicted N deposition rates were lower than measured rates due in part to gaseous ammonia deposition not having been accounted for in the modelling. Improved N deposition estimates could be achieved through the inventory and modelling of ammonia releases.

Keywords: Acid deposition, sulphur, nitrogen, Highveld, CALPUFF.

1. Introduction

Acid deposition over the South African Highveld has been investigated for over two decades, prompted by a growth in anthropogenic emissions and related concerns about potential degradation of soil and water resources. Long-term trends in acid deposition, and the likelihood and timing of critical load exceedances have been speculated about. The contribution of combustion processes to acid deposition, and the relative impact of such deposition on water quality compared to other pollutant sources are also of interest.

Regional acid deposition modelling was undertaken for the Highveld to inform a broader study into the effects of air pollutants on soils, water catchments and ecosystems. The modelling aimed to address the following information gaps identified from the review of past studies:

- Spatial and interannual trends in total sulphate (S) and nitrogen (N) deposition and ratios of wet to dry deposition.
- Trends in the contribution of sulphur dioxide (SO₂) and sulphate to S deposition
- Contributions of individual gaseous and particle constituents to total N deposition.
- Relative source contributions to S and N deposition.
- Variations in S and N deposition during high, low and average rainfall years.
- Historical and future trends in acid deposition.

Aspects of this regional study addressed in this paper include the modelling approach adopted, data inputs, and select study findings. Spatial and intra-

annual variations in S and N deposition are presented for a recent base case period (2000/1), and projected longer-term (1920 to 2020) trends in S and N deposition presented and discussed.

1.2. Previous Acid Deposition Studies

Wet deposition has been monitored on the Highveld since 1985, providing the foundation for a reasonable understanding of wet deposition rates (Wells *et al.*, 1987; Wells, 1989; Botha *et al.*, 1990; Turner, 1990; Bluff *et al.* 1991; Snyman *et al.*, 1991; Wells, 1993; Turner, 1993; Piketh and Annegarn, 1994; Turner *et al.* 1996; Zunckel *et al.*, 1994, 1996, 1999; Galpin and Turner, 1999a,b; Galpin and Held, 2002). Wet deposition measurement and analysis techniques are primarily based on methods used by the US-EPA and by the Warren Springs Laboratory in the United Kingdom. Although the process of wet deposition monitoring requires meticulous care, the procedure is relatively inexpensive and straightforward.

The possible significance of dry deposition, given the relative aridity of the Highveld climate, was raised in the 1980s and early 1990s (Wells, 1989; Turner, 1993; Wells, 1993). Dry deposition may be either measured directly or indirectly computed by inferential methods from the measurement of ambient air pollutant concentrations. Wells (1993) recommended the application of the inferential method for the estimation of dry deposition rates on the Highveld. Turner *et al.* (1995) demonstrated the effectiveness of the inferential technique for use on the Highveld, with estimated S deposition rates comparable with published data. Inferential

methods have remained a preferred approach for estimating dry deposition for the Highveld (Zunckel *et al.*, 1996, 1999; Mphepya and Held, 1999).

Atmospheric modelling of deposition has been undertaken for the Highveld since the late 1990s. Models used have included the US-EPA Lagrangian puff CALPUFF model, the MATCH Eulerian multi-layered 3D model developed by the Swedish Meteorological and Hydrological Institute, the Lagrangian-Eulerian Diffusion (LED) model, and the CAMx Eulerian photochemical dispersion model developed by ENVIRON (Zunckel *et al.* 2000a, 2000b; Scorgie *et al.*, 2002; Fourie *et al.*, 2005).

Total annual S deposition rates are documented in the above literatures as peaking in the range of 50 to 80 kg/ha-year over the main source area. Annual S deposition rates over the greater Highveld have been generally estimated to be over 8 kg/ha-year, with deposition rates of greater than 1 kg/ha-year predicted to occur over the entire north-eastern parts of the country.

The literature is more divergent with regard to the contribution of dry S deposition to total S deposition rates on the Highveld. Estimations range from dry S deposition constituting 30% to 80% of total deposition, with temporal and spatial variations in such ratios being poorly addressed by monitoring studies. Inaccuracies may arise due to ratios being calculated based on wet and dry deposition rates measured at different locations.

Nitrogen deposition rates for the Highveld are primarily published by Turner (1993), Galy-Lacaux *et al.* (2003, 2008) and Mphepya *et al.* (2004, 2005). Dry deposition rates at Amersfoort (situated on the Highveld to the southeast of the main source areas) and Louis Trichardt (remote site) have been calculated based on inferential modelling, using as input ambient NO₂ and NH₃ concentrations. Wet deposition was calculated from the measured NH₄⁺ and NO₃⁻ concentrations in precipitation and mean annual rainfall⁽¹⁾.

Total N deposition at Amersfoort has been quantified as 15 kg N/ha-year, comprising 63% wet deposition (9.5 kg N/ha-year) and 37% dry deposition (5.6 kg N/ha-year). The large difference in total N deposition between Amersfoort (15 kg N/ha-year) and Louis Trichardt (9 kg N/ha-year) illustrates the influence of industrial activities on the central Mpumalanga Highveld.

Based on measurements at Amersfoort, N deposition has been found to comprise: 42% dry deposition of gases (NO, NO₂, HNO₃, NH₃), 33% wet deposition of ammonium nitrate, 24% wet deposition of ammonium sulphate and nitric acid and <1% dry deposition of ammonium nitrate (Galy-Lacaux *et al.*, 2008).

¹ Nitrogen values are based only on gaseous and precipitation measurements of NO₂, NH₃, NO₃⁻ and NH₄⁺ and do not include species such as HNO₃, PAN and the dry deposition of N containing particles which could contribute significantly to the atmospheric burden of nitrogen.

2. Study Area

The study area extends across parts of the Mpumalanga, Gauteng, and Free State provinces. Situated at an altitude of about 1 500m, the region encompasses a mixture of commercial, industrial, mining, agricultural and residential activities. Major urban and industrial areas, situated primarily within the western part of the study area, include the Johannesburg, Tshwane (Pretoria) and Ekurhuleni metropolitan areas and the highly industrial 'Vaal Triangle' region.

The Mpumalanga Highveld, comprising the eastern half of the study region, is characterised by heavy industry within the cities of Witbank and Middelburg, coal-to-petroleum operations in Secunda, dispersed coal mining operations and several large coal-fired power stations situated across the region. Beyond the borders of the urban areas, land is semi-rural and primarily used for low intensity farming. The population of the region is over 10 million, with the greatest population densities occurring within the northwestern parts of the region, within the Johannesburg, Tshwane and Ekurhuleni metropolitan areas.

The study area was selected to include several water catchments identified as being of interest in the broader acid deposition study, namely the Sabi, Komati and Olifants catchments within Mpumalanga Province and the Sandspruit catchment situated at the border of the Mpumalanga, Free State and KwaZulu-Natal provinces.

3. Methodology and Data

3.1. Modelling Domain

The modelling domain extends 380 km east-west by 430 km north-south, with the south west corner at -28.8846° latitude and 27.44861° longitude.

Highveld emissions account for over 80% of the total S deposition occurring in the region (Zunckel *et al.*, 2000a). S and N deposition rates have been measured to be highest over the central Highveld in proximity to the major source areas. The modelling domain was therefore considered suitable in terms of encapsulating significant emission sources, maximum potential impact zones and the hydrological catchments identified for investigation.

The meteorology was modelled and the dispersion of pollutants simulated for the entire modelling domain, with ground-level concentrations and deposition rates being predicted for a Cartesian receptor grid comprising 76 grid cells (east-west) by 86 cells (north-south), with a grid resolution of 5 km.

Discrete receptor points were specified to coincide with monitoring stations and the headwaters of catchments to facilitate model verification and impact analysis respectively.

3.2. Source and Emissions Data

3.2.1. Emission Scenarios

Hydrological year, 1 October 2000 to 30 September 2001, was selected for model verification and base case acid deposition characterisation. This year was selected due to it being an average rainfall year. Source and emissions data were also more readily available for this period.

Several break point years, selected on the basis of projected historical and future emissions, were simulated using base case (2000/1) meteorology to predict long-term (~1920 to 2020) trends in atmospheric deposition.

3.2.2. Sources Inventoried

Sources inventoried are as follows:

- Power generation (primarily coal-fired power generation for the national grid);
- Industrial sources, including combustion and process emissions;
- Household fuel burning, including coal, wood, LPG and paraffin burning;
- Vehicle tailpipe emissions, including petrol- and diesel-driven vehicles;
- Biomass burning (agricultural and wild fires); and
- Institutional and commercial fuel burning (where available).

Anthropogenic fuel burning activities and biomass burning emissions account for the bulk of ambient SO₂ and NO₂ concentrations and associated atmospheric S and N deposition rates on the Highveld (Annegarn *et al.*, 2007). Sources of NO_x and/or SO_x emissions which were not quantified and included in the modelling include: spontaneous combustion within coal storage piles and coal discard dumps and natural sources such as biogenic emissions.

3.2.3. Emission Projections

Preliminary base case modelling indicated that large industrial and power generation sources dominate regional acid deposition predictions. More emphasis was therefore placed on the accurate projection of emissions for such sources.

Historical source and emissions data for Eskom power stations were made available for the 1927 to 2007 period. Future power station emissions were taken to comprise existing and recommissioned power stations operating at capacity, in addition to two new 4800 MW power stations (Kusile PS near existing Kendal PS; and Golf PS south of Sasolburg). Kusile PS, is under construction and expected to come on line between 2012 and 2014. Golf PS is proposed for construction and, pending its approval, will come on line prior to 2019. Both new power stations will implement wet fluidized gas desulphurisation (FGD) with a SO₂ control efficiency of 90-95%. Power generation forecasts for post 2025 are uncertain. It is conjectured that if South

Africa is still reliant on coal, new power stations are likely to be built in the Waterberg (situated to the north of the study domain).

The quantification of current and historical emissions from major industrial sources were based on: data supplied by companies, the emissions inventory compiled during the Vaal Triangle Air Quality Management Plan (AQMP) development project (Liebenberg-Enslin *et al.*, 2008), and the Department of Environmental Affairs and Tourism (DEAT) 1994 Emissions Inventory for Scheduled Processes.

Emission projections were not available for large industries. Initially emissions were projected based on economic growth predictions for petrochemical, metallurgical and pulp and paper sectors. Emissions are however unlikely to increase in line with industrial growth due to tightening regulations under the 2004 National Air Quality Act. Given the location of major industries within polluted airsheds which are being subject to air quality management, it is feasible that industrial emissions in 2020 will be equivalent to or lower than current emissions. Emission data for 2007 were therefore taken to be indicative of future emissions for existing major industries.

Data from previous studies (Scorgie *et al.*, 2004; Scorgie and Thomas, 2006) were used to quantify base case emissions for smaller industries, household fuel burning, vehicle tailpipe releases and vegetation burning. Past and future emission projections were based on published trends in manufacturing volumes, GDP, total household and electrified household numbers, and fuel sales data.

3.2.4. Base Case Emission Estimates

'Major sources' were defined as any facility emitting over 10 ktpa of either SO_x or NO_x, and included large operational coal-fired power stations and major industrial complexes.

Total annual SO₂ emissions were estimated to be 1,463 ktpa, comprising primarily major source emissions (96.9%), with minor contributions by vehicles (1.6%), other industry (0.8%), household fuel burning (0.3%) and biomass burning (0.04%).

Total annual NO_x emissions were estimated to be 685 ktpa, with significant contributions by major source emissions (76.9%) and vehicles (21.6%), and minor contributions by other industries (0.6%), residential fuel burning (0.5%) and biomass burning (0.5%).

Areas of maximum emissions are centralised over the eastern Mpumalanga Highveld and the Vaal Triangle. Elevated stack emissions (>100 m) account for 93% of the total SO₂ emissions estimated from all sources (with stacks >250 m accounting for 62% of total SO₂ emissions). In terms of NO_x emissions, elevated stack emissions (>100 m) account for 70% of the total emissions estimated from all sources (with stacks >250 m accounting for 50% of total NO_x emissions).

3.2.5. Breakpoint Year Emissions

Over the 1950 to 2007 period, quantifiable SO₂ emissions increased significantly from less than 30 ktpa to almost 1.9 Mtpa. Quantifiable NO_x (as NO) emissions are predicted to have increased over the same period from about 60 ktpa to over 900 ktpa. By 2020, SO₂ and NO_x emissions are estimated to be 2.26 Mtpa and 1.23 Mtpa respectively.

Suitable historical and future break point years were selected to assess the significance of changes in the magnitude and location of emissions in terms of spatial and interannual variations in acid deposition rates (Figure 1). Predicted SO₂ and NO_x emissions for breakpoint years selected are illustrated in Figure 2 and Figure 3 respectively.

3.3. Modelling

3.3.1. Meteorological Modelling

CALMET was used to simulate the meteorological field based on representative land use, topographical, upper air and surface meteorological data. Meteorological data was input for 19 surface stations, including the South African Weather Services' (SAWS) Johannesburg, Irene, Vereeniging, Witbank, Leandra, Ermelo, Standerton, Newcastle, Verkykkop and Bethal stations and Eskom's Elandsfontein, Majuba 1, Majuba 3, Kendal 2, Leandra, Makalu, Palmer, Verkykkop and Camden stations. Upper air data from SAWS ETA-model stations and two radiosonde stations, Irene and Bethlehem, were used. The three dimensional meteorological dataset generated included model projections at ground level, 20m, 200m, 500m, 1500m, and 3000m above ground thus parameterising the atmosphere within valley layers, transitional layers and atmospheric layers located above the terrain.

3.3.2. Dispersion Modelling

Gas phase reactions for SO_x and NO_x were computed internally by the CALPUFF model using the RIVAD/ARM3 Scheme. This scheme treats the NO and NO₂ conversion process in addition to the NO₂ and total NO₃ and SO₂ to SO₄ conversions, with equilibrium between gaseous HNO₃ and ammonium nitrate aerosol. Use was made of site-specific ozone measurement data (together with modelled radiation intensity) as surrogates for the OH concentration during the daytime when gas phase free radical chemistry is active. Hourly varying ozone data were input from Eskom's Verkykkop, Palmer, Elandsfontein and Kendal 2 stations.

Site-specific deposition velocities were input in the CALPUFF to facilitate dry deposition modelling. Such velocities were specified as seasonal-average 24-hour cycles of deposition velocities for SO₂, NO, NO₂ and HNO₃.

Wet deposition is determined by the scavenging coefficient which in turn is a function of the

characteristics of the pollutant (solubility, reactivity) as well as the nature of the precipitation. Default values of the scavenging coefficient for SO₂, sulphate, NO_x, HNO₃ and nitrate included in the CALPUFF model for liquid precipitation were used. Hourly precipitation data for 92 stations were input to improve the accuracy of wet deposition predictions.

Total S deposition modelled included wet and dry deposition of gaseous SO₂ and particulate SO₄. Total N deposition comprises dry deposition of NO, NO₂, HNO₃, NO₃ and ammonium sulphate in addition to wet deposition of HNO₃, NO₃ and ammonium sulphate.

To account for the contribution of ammonium sulphate and ammonium nitrate it was assumed that the predicted SO₄ and NO₃ are completely neutralized by NH₄ with the following factors being applied: 0.292 x SO₄ and 0.226 x NO₃. This method is frequently applied in the US to account for the ammonium associated with SO₄ and NO₃.

3.3.2. Study Limitations

SO₂ can be formed in the atmosphere through the reaction of hydrogen sulphide and ozone (thermal gas-phase photo-oxidation). Substantial H₂S emissions occur on the Highveld, with large industrial sources on the Mpumalanga Highveld alone accounting for over 70 ktpa. The modelling approach adopted did not allow for atmospheric SO₂ formation due to H₂S emissions.

Ammonia emissions were not quantified nor simulated during the study. Given that dry deposition of gaseous ammonia has been estimated to account for ~30% of total N deposition (Galy-Lacaux *et al.*, 2008), the omission of ammonia from the study is expected to affect the accuracy of total N deposition rates (including the magnitude and spatial patterns of such rates predicted) and the accuracy of projected wet to dry N deposition ratios.

3.4. Results

3.4.1. Model Verification

Predicted ambient SO₂, SO₄, NO, NO₂ and NO₃ concentrations correlated well with measured concentrations for the central Highveld. At Elandsfontein ratios of modelled to measured levels were within the range of 0.6 to 1.0 for annual averages and 0.7 to 1.1 for peak concentrations.

Predicted wet S deposition is of a similar order of magnitude as that measured by the acid rain monitoring network, with ratios of predicted to measured deposition rates being in the range of 0.6 to 1.6. Although predictions are higher than those published for Amersfoort (ratio of 1.4 to 1.6) it is notable that higher rainfall occurred in 2000/1 (720 mm) compared to the averaged 1996-8 (510 mm) and 1985-1992 (614 mm) periods for which measurements are published.

The magnitude and spatial variation in simulated wet S deposition were also found to be comparable to the rates modelled by Zunckel *et al.* (2000a), with wet S deposition rates of >3 kg/ha/annum occurring over the entire Highveld and rates of >10 kg/ha/annum over the main source areas.

The predicted dry S deposition compares closely with the deposition modelled using the inferential method published by Mphepya and Held (1999) and Zunckel 1999 for both Elandsfontein and Palmer. There is also a good correlation at the 'downwind' stations of Elandsfontein and Amersfoort with the Lagrangian-Eulerian Diffusion (LED) dispersion modelling undertaken by Fourie *et al.* (2005).

Concurrent dry and wet S deposition could only be found in the literature for two sites, Amersfoort (1996-8) and Suikerbosrand (November 1992 – March 1993). At Amersfoort, dry S deposition was measured to be 33% of total deposition. The percentage of dry S deposition predicted at Amersfoort for 2000/1 by this study is comparable (29%). At Suikerbosrand, dry S deposition was measured to be 44% of total deposition. The percentage dry S deposition predicted for 2000/1 at that site by this study is comparable (40%).

Deposition of SO₂ was predicted to account for 92% of the total S deposition at Elandsfontein, with the remainder (8%) due to SO₄ particle deposition. This is within the range of values published in the literature, with Turner *et al.* (1995) estimating that SO₄ contributed 5% of total S deposition, and Zunckel (1999) documenting that SO₄ accounts for 20% of S deposition at this station.

Predicted total wet N deposition is comparable to measured values published by Turner (1993) closer to the central Highveld area, with modelled values comprising 50% to 90% of measured values at Amersfoort and Ermelo respectively. Predicted wet N deposition rates however only represent 20% to 30% of the measured wet deposition rates at the more peripheral stations (Ladysmith, Vryheid, Piet Retief).

Predicted total N deposition at Amersfoort was lower than the total 'measured' N deposition rates published for Amersfoort by Galy-Lacaux *et al.* (2003), with the ratio of predicted to measured deposition being 0.3. The underprediction is due in part to dry deposition of ammonia gas not having been accounted for in the predictions.

Predicted and measured N deposition constituents (excluding ammonia gas dry deposition) were comparable. Total N deposition was predicted and measured to be dominated by wet deposition of nitrate and ammonium nitrate (~50%), with total wet deposition accounting for over 80% of the total deposition.

It is notable that if gaseous NH₃ deposition was accounted for in the modelling, the contribution of nitrate and ammonium nitrate would be reduced from ~50% to ~30% with dry gaseous NO, NO₂, HNO₃ deposition being more significant (~40%).

Accounting for dry NH₃ deposition, total wet deposition would account for less than 60% of the total N deposition at Amersfoort.

3.4.2. Predicted Base Case Sulphur Deposition

The total annual S deposition maximum occurs over the central Highveld (between Witbank, Secunda and Bethal). Deposition rate peaks of >35 kg/ha/year are predicted to occur in the vicinity of large point sources including coal-fired power stations, petrochemical plants and large steelworks. A second maximum, smaller in magnitude and spatial extent, occurs over the southern Vaal Triangle, with peak deposition rates of >20 kg/ha/year predicted to occur to the east of Sasolburg (Figure 4).

Wet and dry removal of gaseous SO₂ contributes significantly to predicted total annual S deposition maximums. Dry removal of SO₂ was predicted to contribute most significantly to peak deposition rates in the vicinity of low level major industrial sources. Wet deposition of gaseous SO₂ was however projected to be a more effective removal mechanism for sulphur emitted from high stacks such as those of the large-scale power stations.

Significant spatial and temporal variations in the ratio of wet to dry S deposition were predicted to occur, and partially explain the range of ratios presented in the literature. The spatial variations are due to a range of factors including: spatial variations in rainfall (magnitude, frequency), and the location and height of significant SO_x sources.

The contribution of dry S deposition to total S deposition was predicted generally to decrease from over 40% in the northwest parts of the study area to less than 10% in the south-east. The highest contribution of dry deposition occurred in the vicinity of significant low level SO_x sources, such as to the west of Witbank (>60% dry) and at Vanderbijlpark (>50% dry). In the vicinity of large-scale power stations and petrochemical plants with high stacks, wet S deposition was predicted to account for over 70% of total S deposition.

Higher dry S deposition rates occur in the summer and winter over the central Highveld. Averaged inferred deposition velocities for SO₂ are estimated to be larger during summer and smallest during winter. This is attributable to increased solar radiation, leaf area index (LAI), the photosynthetic activity of vegetation and variations in meteorological variables such as temperature and surface wetness which occurs during summer. The higher deposition velocities during summer are however offset by the lower ambient SO₂ concentrations which occur. In contrast, the effect of the lower deposition velocities during winter is offset by increased ambient SO₂ concentrations.

No distinct diurnal pattern is evident in wet S deposition, as is to be expected. Neither are diurnal trends in total S deposition apparent due to the contribution of wet S deposition to total S deposition

rates. A diurnal trend is however evident in dry S deposition, with the morning peak coinciding with convective mixing of elevated plumes to ground.

The bulk of the dry deposition is due to daytime deposition of SO₂ over the Highveld associated with the mixing down of plumes emitted from tall power plant stacks within the daytime convective boundary layer. At night vertical down mixing of plumes is inhibited due to stable boundary layer conditions.

3.4.3. Predicted Base Case Nitrogen Deposition

The omission of ammonia emissions from the modelling is expected to result in an underprediction of dry N deposition (and consequently an underprediction of total N deposition and exaggeration of wet/dry deposition ratios). Study findings presented should therefore be taken as indicative of NO_x emission related deposition only.

The total N deposition maximum was predicted to occur at a more easterly location compared to the S deposition maximum with the peak centred between Bethal and Ermelo, reflecting the projected wet N deposition pattern (Figure 5). Wet N deposition was estimated to account for over 80% of total deposition over the entire modelling domain.

Total N deposition was predicted to be dominated by wet deposition of nitrate and ammonium nitrate, with time taken for formation of these products and spatial variations in rainfall accounting for the deposition maximum being located more remotely from significant NO_x source areas. Higher rainfalls generally occurred over the eastern parts of the study domain.

Dry N deposition, which is primarily due to dry deposition of gaseous NO, NO₂ and HNO₃, was predicted to peak over the central Highveld, coincident with the widespread elevated NO₂ concentrations projected. Over the entire modelling domain contributions of specific species to dry N deposition were predicted as follows: NO (~20%) NO₂ (~40%) and HNO₃ (~40%).

As indicated above, model results are expected to under predict total N deposition due to gaseous ammonia deposition not having been accounted for. The percentage contribution of wet N deposition to total N deposition is likely to be significantly overstated due to the omission of dry deposition of gaseous ammonia (which has been estimated to contribute over 30% of total N deposition).

3.4.4. Long-term Trends in Acid Deposition

Predictions for breakpoint years were based on the use of meteorology for 2000/1 (average rainfall year). Trends in Highveld SO₂ and NO_x emissions over breakpoint years and resultant total S and N deposition at specific discrete receptor points are depicted in Figure 6 and Figure 7. Predicted spatial variations in total S deposition for breakpoint years are illustrated in Figure 8.

The relationship between emissions of acid deposition precursors and resultant deposition

fluxes is complex and, in many cases, non linear. This non-linearity is expressed as differences in the rate of change of emissions and total deposition, differences in the response of wet and dry deposition rates to emission trends, and spatial variations in deposition trends.

In the USA, strong near-linear correlations were found between large scale SO₂ emission reductions and large reductions in sulphate concentrations in precipitation in the Northeast, one of the areas most affected by acid deposition (NADP, 2007). In the UK however, significant non-linearities have been noted in the relationship between sulphur emissions and deposition over the past two decades (Fowler *et al.*, 2007). In the case of the US, the locations and emission heights of sources remained relatively unchanged despite reductions in emissions from such sources. Whereas in the UK, there have been significant changes in the location and configuration of sources with urban emissions becoming more prominent as industrial emissions have increasingly been reduced.

No clear linear relationship is projected between SO₂ emissions and catchment receptor S deposition during the 1948 to 1979 period (Figure 6). This is to be expected since the location of major SO₂ sources varied significantly over this period, with peak source areas switching from Witbank (pre 1950) to Witbank, Vereeniging and Germiston (1950-5), and subsequently including Sasolburg (1956-1962). Thereafter, Komati PS situated near Bethal became the dominant power generation source (1963-1967).

Increasingly power generation shifted to the Mpumalanga Highveld and large industrial sources were commissioned in the region. By the late 1970s / early 1980s, power generation emissions primarily occurred over the Mpumalanga Highveld and other major sources (e.g. Highveld Steel and Vanadium) were commissioned. Throughout the 1980s emissions intensified over this region with Duvha and Matla power stations being commissioned and their operating capacity stepped up, and Sasol 2 and 3 becoming operational at Secunda.

The significance of the Mpumalanga Highveld region as the dominant source of SO₂ emissions persisted throughout the 1990s and 2000s. This region is projected to become even more significant by 2020 as existing power stations increase their output and the planned Kusile PS near Witbank is commissioned.

Given continued SO₂ emissions from elevated stacks situated on the Mpumalanga Highveld, the projected linear relationship between SO₂ emissions and predicted S deposition during the 1980 to 2020 period is conceivable (Figure 6). The closest relationship is found with deposition trends projected for the Olifants and Komati catchments which are situated, along with the Elandsfontein receptor, within the major source region.

The increasing intensity of emissions on the Mpumalanga Highveld over the past three decades is also apparent in projected spatial N deposition patterns and deposition rates predicted for catchment receptors (Figure 7).

4. Conclusions

Regional acid deposition modelling was undertaken for the Highveld to address information gaps regarding intra-annual trends and spatial variations in S and N deposition, and to project longer-term trends in deposition rates.

Dispersion model results correlated reasonably well with measured SO₂, SO₄, NO, NO₂ and NO₃ concentrations and S deposition rates for the central Highveld. This correlation reinforced the earlier finding by Zunckel *et al.* (2000a) that Highveld emissions account for well over 80% of the total S deposition occurring in the region. The modelling domain and methodological approach was therefore considered suitable for simulating S deposition trends within the hydrological catchments on the central Highveld. Model results generally underpredicted ambient concentrations and deposition rates at peripheral sites situated at the outer extents of the modelling domain.

Annual total S deposition rates exceed 3 kg/ha-year over much of the Highveld, with a peak of over 35 kg/ha/year over the central Highveld (between Witbank, Secunda and Bethal) and a lower peak (20 kg/ha-year) over the southern Vaal Triangle.

Spatial and temporal trends in dry and wet deposition vary independently, thus accounting for significant spatial and temporal trends in ratios of dry to wet deposition. The contribution of dry S deposition was predicted to decrease from over 40% in the northwest parts of the study domain to less than 10% in the south-eastern corner. The highest contribution of dry deposition occurred near low level SO_x sources (>60%), whereas wet S deposition was predicted to account for over 70% of total S deposition in the vicinity of high stacks. According to global modelling by Dentener *et al.* (2006), wet deposition was found to contribute between 50% and 70% of the total global SO_x deposition.

Predicted dry and total N deposition rates were lower than measured rates due in part to gaseous ammonia deposition not having been accounted for in the modelling. The percentage contribution of wet N deposition to total N deposition is likely to be significantly overstated due to the omission of dry deposition of gaseous ammonia, which has previously been estimated to contribute over 30% of total N deposition. Study findings presented are therefore indicative of NO_x emission related N deposition only. Improved N deposition estimates could be achieved through the inventory and modelling of ammonia releases and biogenic NO_x sources.

Highveld SO₂ and NO_x emissions have grown significantly over the past eighty years from <100 ktpa to over 900 ktpa for NO_x and 1.9 Mtpa for SO₂. By 2020, SO₂ and NO_x emissions are estimated to reach 2.26 Mtpa and 1.23 Mtpa respectively. No clear linear relationship was found between SO₂ emissions and predicted S deposition prior to the mid 1970s due to significant shifts in the location of major SO₂ sources. Following this date power generation and large industries were progressively established on the Mpumalanga Highveld. The region persisted as a dominant source of SO₂ and NO_x emissions throughout the 1990s and 2000s, and is projected to remain prominent in the foreseeable future. A near linear relationship between trends in SO₂ emissions and trends in S deposition predicted for central Highveld catchments is consequently projected for the 1980 to 2020 period. The increasing intensity of NO_x emissions over the past three decades is similarly predicted to result in increments in N deposition within catchments on the central Highveld.

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Figures

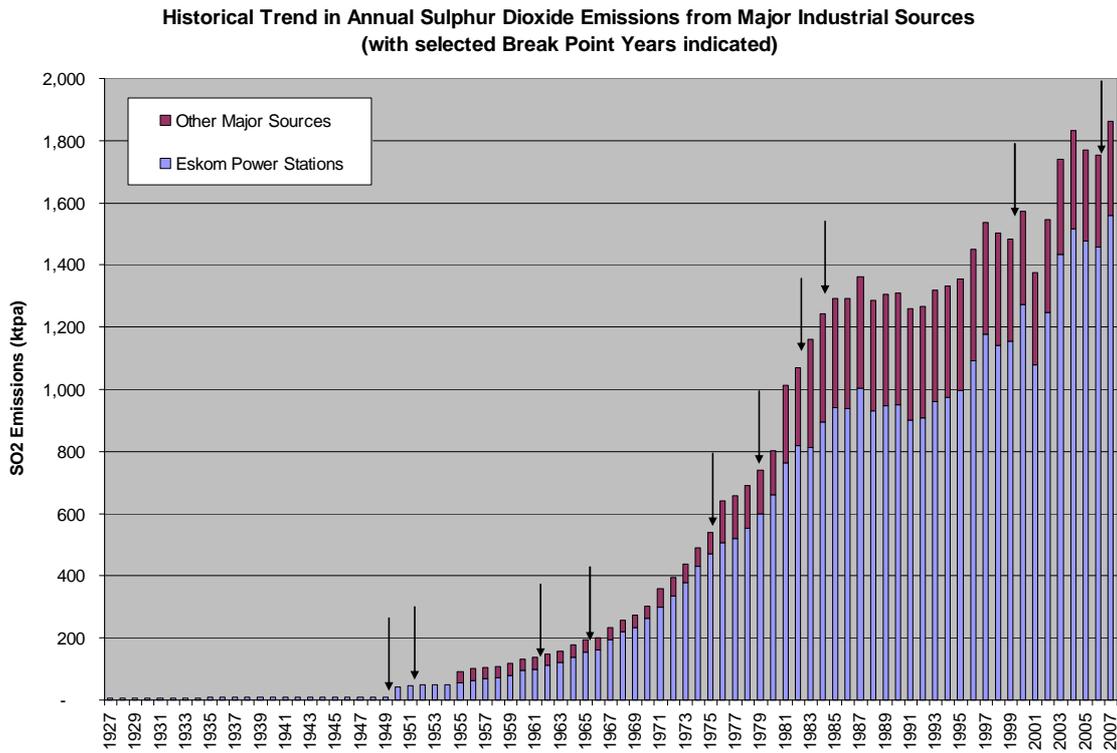


Figure 1. Historical trends in annual SO₂ emissions from major sources with selected break point years indicated

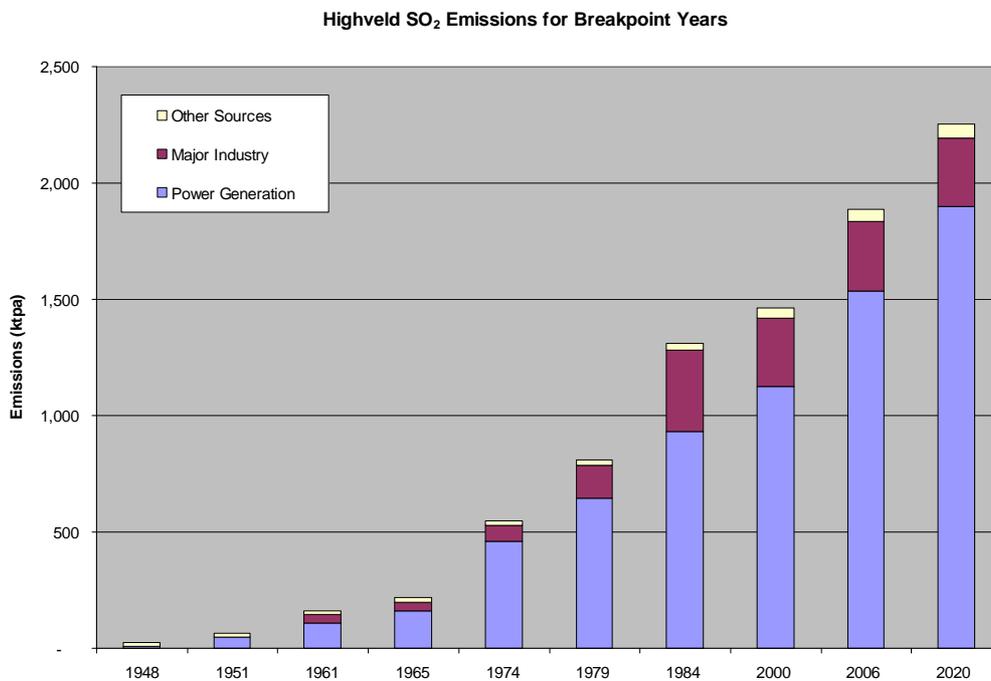


Figure 2. Annual SO₂ emissions for breakpoint years selected

Highveld NO_x (as NO) Emissions for Breakpoint Years

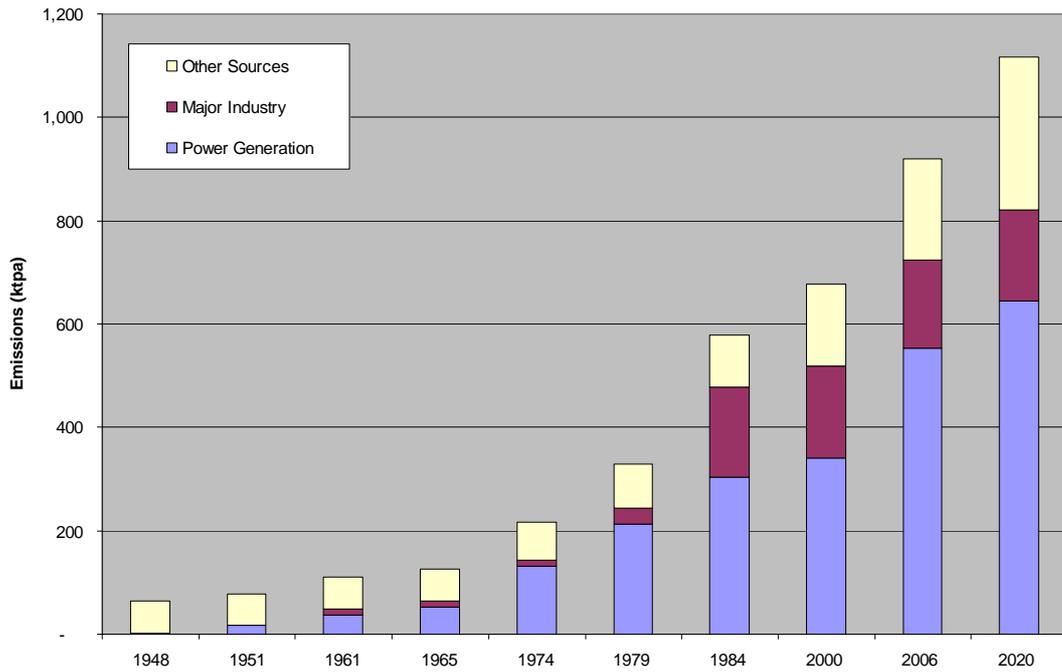


Figure 3. Annual NO_x emissions for breakpoint years selected

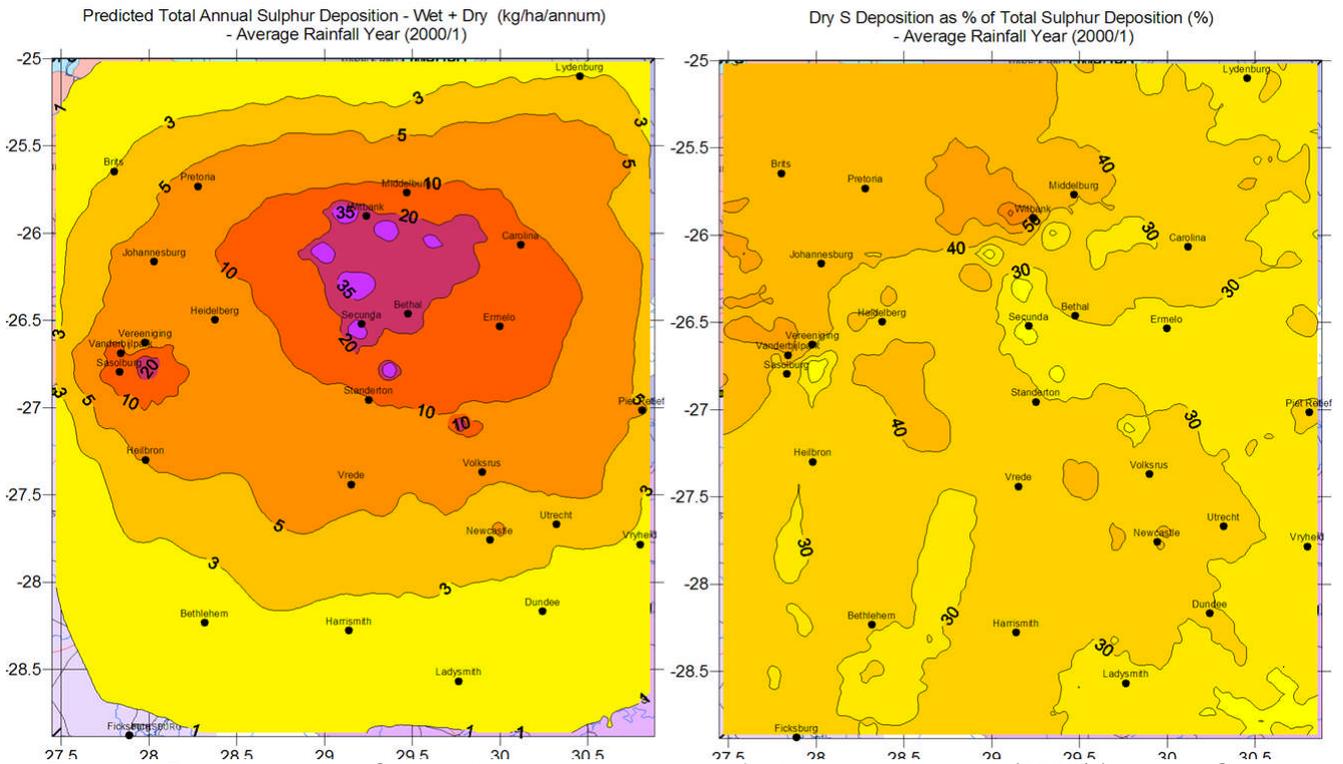


Figure 4. Predicted annual S deposition over the Highveld for the base case year (2000/1) and dry S deposition as a percentage of total S deposition

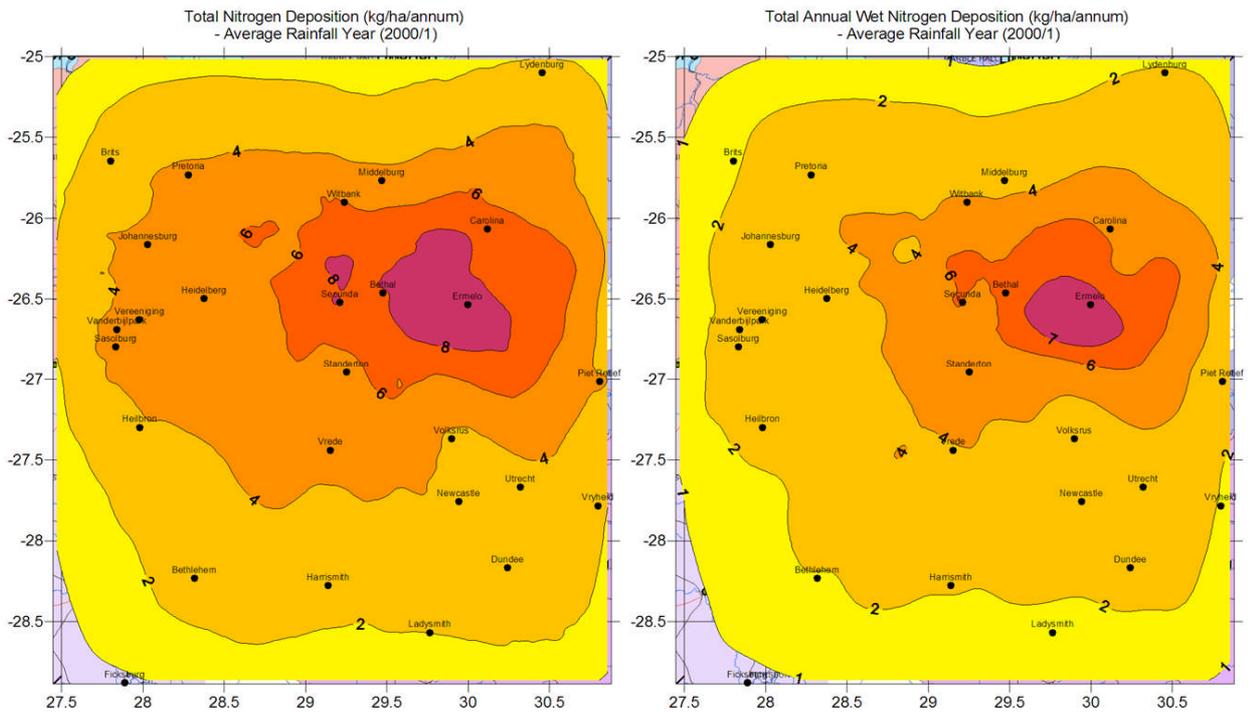


Figure 5. Predicted annual wet and total N deposition over the Highveld for the base case year (2000/1)

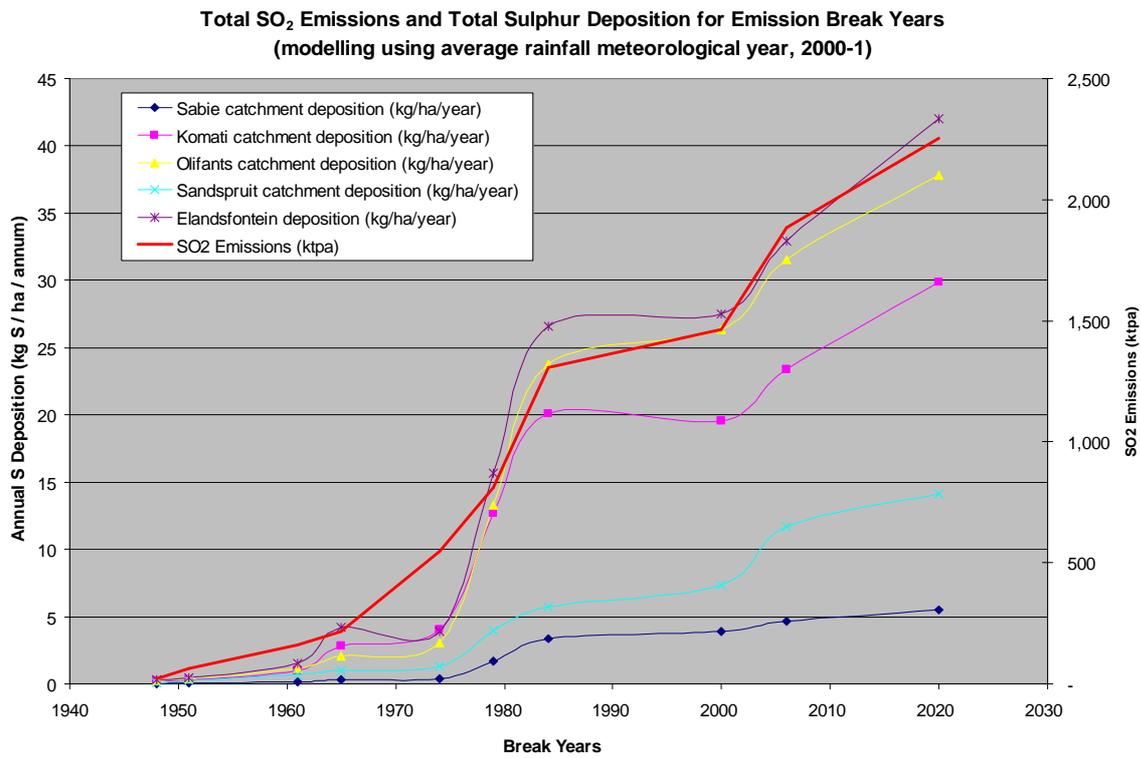


Figure 6. Trends in Highveld SO₂ emissions and predicted total sulphur deposition rates at selected discrete receptors during breakpoint years

**Total Nox (as NO) Emissions and Total Nitrogen Deposition for Emission Break Years
(modelling using average rainfall meteorological year, 2000-1)**

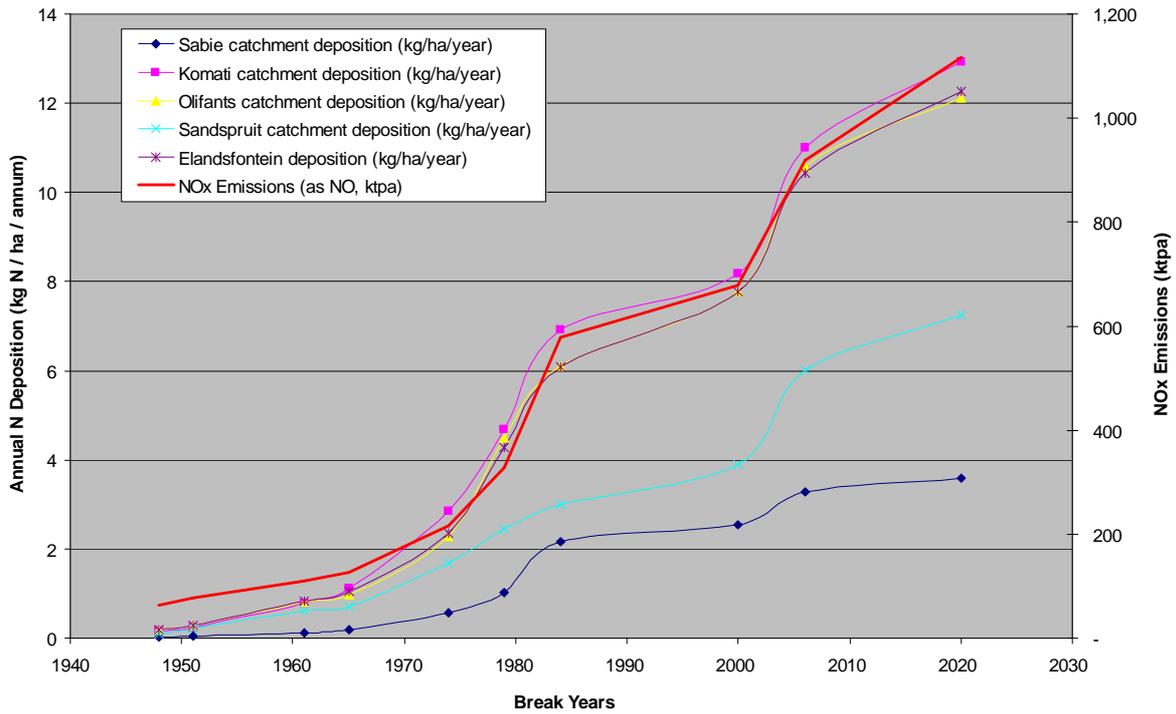


Figure 7. Trends in Highveld NO_x (as NO) emissions and predicted total nitrogen deposition rates at selected discrete receptors during breakpoint years

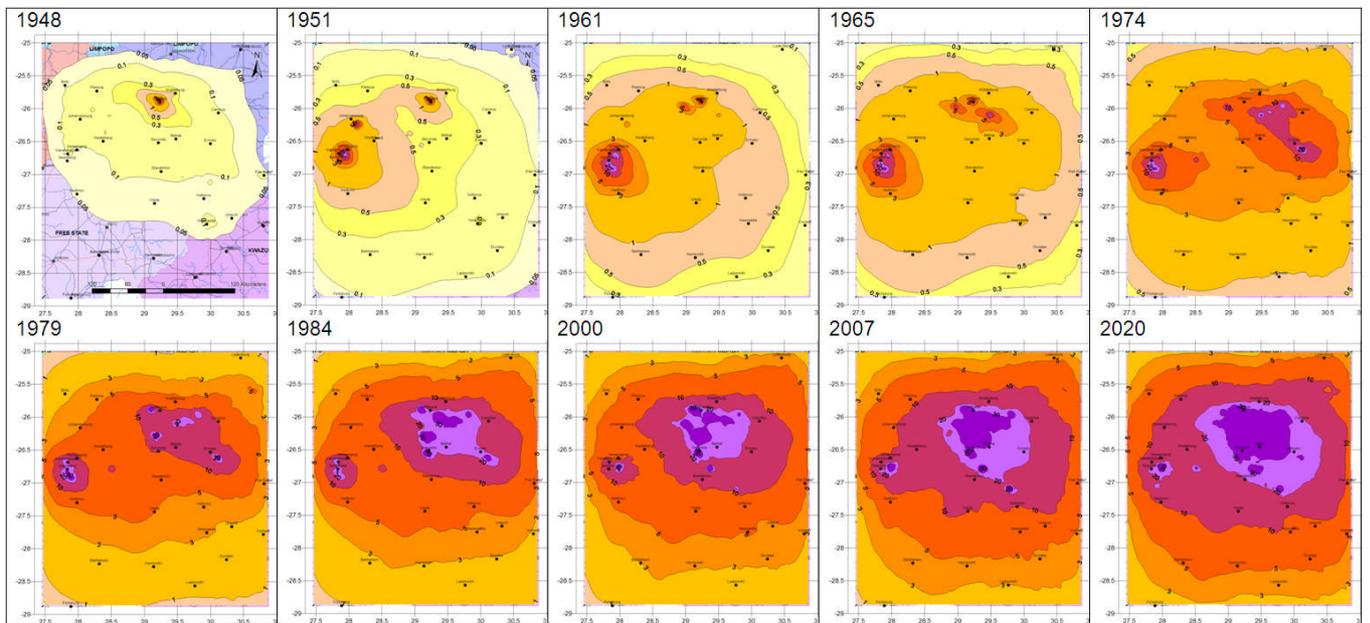


Figure 8. Predicted spatial variations in total S deposition for breakpoint years (kg/ha/year). Predictions for breakpoint years were based on the use of meteorology for 2000/1 (average rainfall year).