Atmospheric dry and wet deposition of sulphur and nitrogen species and assessment of critical loads of acidic deposition exceedance in South Africa

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Dates:
Received: 15 Oct. 2010
Accepted: 14 Feb. 2011
Published: 24 Mar. 2011

How to cite this article:
Josipovic M, Annegarn HJ, Kneen MA, Pienaar JJ, Piketh SJ. Atmospheric dry and wet deposition of sulphur and nitrogen species and assessment of critical loads of acidic deposition exceedance in South Africa. S Afr J Sci. 2011;107(3/4), Art. #478, 10 pages. DOI: 10.4102/sajs.v107i3/4.478

We tested the hypothesis that acidic atmospheric pollution deposition, originating from the South African central industrial area, poses an environmental threat across a larger region within the dispersal footprint. A network of 37 passive monitoring sites to measure SO_2 and NO_2 was operated from August 2005 to September 2007. The area extended over the entire northern and eastern interior of South Africa. Monitoring locations were chosen to avoid direct impacts from local sources such as towns, mines and highways. Dry deposition rates of SO_2 and NO_2 were calculated from the measured concentrations. Concentrations of sulphur and nitrogen species in wet deposition from a previous study were used in conjunction with measured rainfall for the years 2006 and 2007 to estimate the wet deposition over the region. The calculated total (non-organic) acidic deposition formed the basis for an assessment of exceedance of critical loads based on sensitivity of the regional soils. Regional soil sensitivity was determined by combining two major soil attributes available in the *World Inventory of Soil Emission Potentials* (International Soil Reference and Information Centre). Results indicate that certain parts of the central pollution source area on the South African Highveld have the potential for critical load exceedance, while limited areas downwind show lower levels of exceedance. Areas upwind and remote areas up and downwind, including forested areas of the Drakensberg escarpment, do not show any exceedance of the critical loads.

Introduction

Terrestrial acidification as a result of atmospheric pollution continues to be an environmental problem of concern for developing countries, in light of increasing emissions of acidic precursor trace gases.[1,2,3] South Africa has one of the largest industrialised economies in the Southern Hemisphere and is the only industrialised regional energy producer on the African continent.[4,5] A large proportion of this industrial infrastructure is concentrated on the Highveld plateau, which accounts for approximately 90% of South Africa's scheduled emissions of industrial dust, sulphur dioxide (SO_2) and nitrogen oxides (NO_x).[6] There are three pathways by which chemical species can be removed from the atmosphere: chemical transformation, wet deposition and dry deposition. The central task of this study was to derive combined dry and wet deposition of acidic trace gases, and to assess exceedances of critical sensitivity thresholds, taking into account soil properties and base cation deposition.

In 1988, the Convention on Long-range Transboundary Air Pollution adopted the critical-load concept:

> A quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge[7,8].

The adoption of this concept then constituted a basis for future developments concerning limitation of the emissions of air pollutants,[9] such as sensitivity mapping which gives a relative measure of the susceptibility of ecosystems to such changes.

The process of sensitivity mapping increases confidence in acidity evaluation, in the assessment of sensitivity to acidification in regions that could not be studied intensively. Various methods to calculate and map critical loads to inputs of acidity have been developed.[10] Global, regional and national maps showing ecosystem sensitivity to acidic deposition have been produced for a number of years. Global assessments include those of Troedsson and Nykvist[11] in 1973 and Rodhe et al.[12] in 1988, and led to the production of maps that give a broad overview of areas sensitive to acidic deposition. Kuylenstierna et al.[13] in 1995 produced a map using soil buffering, land cover and climatic variables that showed sensitivity of terrestrial ecosystems to acidic deposition. Prior regional-scale studies include a sensitivity distribution in Canada[14] in 1987; a surface water

alkalinity map for the USA (alkalinity of waters is highly correlated with soil and geological properties)[15] in 1982; a distribution in China[16] in 1994; and sensitivity maps for Japan[17] in 1983 and Australia[18] in 1996. In relation to other regional work, it is important to mention the critical loads map from the Regional Air Pollution Information and Simulation–Asia (RAINS-Asia) activity[19] in 1995.

Generally, sensitivity mapping in South Africa is consistent with previous global approaches. The method used by Kuylenstierna et al.[20,21] followed the pattern of sensitivity mapping in the United Kingdom, Scandinavia and other European countries.[22,23,24,25] In South Africa, two critical loads assessments have been undertaken previously.[26,27] The first assessment investigated the potential of the critical loads approach as a key component of an acid deposition risk advisory system (ADRAS) for South Africa. A region, constituting most of Mpumalanga Province, was selected for a test study and a first attempt was made to define actual deposition and critical loads for soils and surface waters. Terrestrial critical loads prediction had high uncertainty in a part of the region because of the lack of monitoring stations. A soil critical load map could not be constructed as a result of the lack of information on mineral weathering rates expected under the climatic conditions within the studied region.[26] Instead, a map of relative sensitivity was constructed, based on bedrock lithology, annual rainfall, soil chemistry and land use. This map showed the most to least sensitive areas. The areas ranked as very sensitive corresponded to the areas along the Mpumalanga Escarpment, the areas ranked as sensitive corresponded to the north-east of Mpumalanga and the area categorised as moderately sensitive corresponded to the eastern part of the region. Portions of the Kruger National Park were ranked as resistant. The need for a refined classification of land use and soil types was highlighted.[26]

The second assessment used the rain chemistry data and mean annual rainfall data to infer wet deposition results; dry deposition was not taken into account.[27] These estimates for soil critical loads were derived by producing a sensitivity map (using soil parameters such as pH, cation exchange capacity and base saturation). Then critical loads were assigned to the final sensitivity classes. The report concluded that there was little threat of acidification through wet deposition and it was suggested that future investigations focus on more accurate quantification of the wet component of regional acidification.[27]

This study combined a dry acidic deposition distribution and estimates of the wet deposition as its first task. Dry deposition values were based on measured ambient concentrations.[28] The second task was to assess any current exceedance of the critical sensitivity thresholds for soils where the soil buffering capacity to acidity is becoming saturated. The assessment was performed over a wider area, extending a few hundred kilometres upwind and downwind of the major point sources. This was the first study of this scale undertaken in southern Africa, although similar studies have been conducted worldwide, particularly in the developed countries of North America and Europe.[29,30,31,32]

Methods

We used inferential methods to estimate both dry and wet deposition. A database of ambient concentrations of acidic trace gas species SO_2 and NO_2, measured at 37 remote sites across the region, was used.[28] Deposition velocities were obtained from an empirical set of dry deposition results measured in this region.[33] The wet deposition computation used prior long-term measurements of precipitation chemistry for the same region,[34] which were combined with precipitation measurements concurrent with the trace gas monitoring campaign.

The approach of critical loads exceedance assessment in this study followed the procedure developed for critical loads assessment in developing countries.[20,21] However, unlike the modelled atmospheric concentrations used in the original study, the procedure adopted here used measured concentrations (gaseous and wet acidic anions).[28,34] Total (dry and wet) acid deposition loads were adjusted to account for mitigation as a result of base cation deposition. Soil sensitivity maps were generated, based on an empirical world soil database inventory.[35] Two soil attributes, cation exchange capacity and base saturation, were extracted to determine sensitivity categories (buffering capacity). Finally, exceedance maps were generated by overlaying the total acidic deposition layer onto the sensitivity map.

Monitoring network design

The passive sampler monitoring network for this research study was established in a 600 km x 600 km array centred around Witbank on the South African Highveld. Gaseous pollutants of interest for this study included SO_2 and NO_2. The main sources of these two components are the nine coal-fired power plants located on the industrial Highveld. Other sources of air pollution in this region include major ore smelters, multiple surface coal mines, and a major coal liquefaction and petrochemical complex. Although prevailing winds are from the north-west, seasonal stagnation and recirculation necessitated a network design covering the entire inland area of South Africa.[36] For logistical reasons, the network was constrained to the political borders of South Africa. Elandsfontein (Site 17) monitoring station was chosen as a reference site for the network. This site was intentionally chosen as it is in the zone of highest pollution, at the centre of the industrial Highveld and is ideally located to provide maximum ground-level concentrations within the network.

Passive sampling of acidic trace gases

Trace gases were monitored using the passive sampling technology as applied in the International Geosphere Atmosphere Chemistry–Deposition of Biogeochemically Important Trace Species (IGAC–DEBITS–Africa/IDAF) network.[37] These measurements are based on the molecular

diffusion of gases onto species-specific impregnated filters.[38,39] Passive samplers have been tested and validated since the beginning of 1998 in the IDAF network at six African stations.[40,41] This technique has also been tested in different tropical and subtropical regions.[42] Precision of the passive samplers, expressed as mean percentage difference between duplicates, has been found to be in the range 5% – 10% for NO_2 and 10% – 25% for SO_2.[43,44]

Monitoring sites were selected at integral one-degree grid intersections, with some additional sites positioned at half-degree grid intervals. To avoid the impacts from local air pollution sources, care was taken to position the sites away from urban centres, industrial point sources or other local disturbances. Samplers were also located away from major roads. Sites were mostly in rural farmland, forestry and natural reserves, sparsely inhabited and often non-electrified locations. In instances where the grid point was close to potential interferences, sites were shifted from the original fixed grid point by distances of not more than 15 km. Passive samplers were exposed in the field for 30-day intervals, corresponding to calendar months, for 2 years (two annual cycles running from September 2005 to August 2007). The monitoring sites were jointly managed with field volunteers who serviced them on a monthly basis. Samples were prepared in the laboratory and couriered or mailed to the site operators.

Quality assurance and quality control

Quality assurance and control for passive sampling has been described previously[28] and so only a short overview is given here. An accredited atmospheric chemistry laboratory with its own quality assurance and control measures was used. Throughout the sampling campaign, measures were undertaken for field quality assurance. These ranged from duplicate sampling and seasonal site visits to monitors and procedural inspections of the monitoring campaign. For internal quality control purposes, comparisons were made between the two annual cycles of the monthly, seasonal and annual means. In addition, data analysis included checking for extreme concentrations that prompted the scientific flagging of outliers.

Quality assurance and control for the wet chemistry was undertaken by a licensed industrial laboratory. Ion chromatography chemical analyses performed in the laboratory were annually evaluated by the World Meteorological Organization (WMO) and the results compared to reference values.[33]

Dry gaseous deposition calculation

Dry deposition rates were calculated from the measured ambient concentration and dry deposition velocity for the species derived from an inferential model.[45,46] The dry deposition rate, F is determined as follows:

$$F = -V_d \times C \qquad \text{[Eqn 1]}$$

where C is the atmospheric concentration and V_d is the deposition velocity (downward flux). For an inferential model a number of meteorological parameters are required. Those parameters were measured and included in an inferential model in a deposition calculation presented by Mphepya.[33] His work, based on a set of 3 years of monitored data (1996 – 1998), was used to calculate the deposition velocities for two sites – Elandsfontein and Palmer. A mix of 66% grass and 34% maize was considered to be representative of the Elandsfontein site, while a mix of 85% grass and 15% forest was considered for Palmer.[33] Such surface vegetation mixes were found to be representative for the sites of this study and thus both sets of data could be used for this study.

Differentiated deposition velocities for day and night were given for each season[33] in each year of observation as well as for the 3-year means. The 3-year means (per day/night and season) for both Elandsfontein and Palmer were then averaged and the Elandsfontein–Palmer averages applied to each site in the dry deposition calculations of this study. The V_d differed more in terms of day–night and for different seasons for different sites than for different sites under the same daytime and seasons, indicating that surface cover is not the determining factor of V_d (Table 1). Nevertheless, gaseous deposition velocities remain a major uncertainty as no direct deposition velocities for SO_2 and NO_2 were measured locally. However, the values shown in Table 1 were considered to be the most suitable ones for this study.

Another important step taken in this calculation was to record periods of light and dark for each monitoring site for each month of sampling, based on the geoposition of each monitoring site.[47]

Wet acidic deposition

In most cases, the long-term spatial variation in wet deposition is determined more by variations in the precipitation and less by variations in concentrations of acidic species in the rain. The wet deposition flux is determined by calculating the measured concentration of the species of interest in the

TABLE 1: Calculation of average seasonal inferred dry deposition velocities (cm/s) of SO_2 and NO_2 for this study based on previously determined velocities (1996 – 1998).

Compound	Location	Autumn		Winter		Spring		Summer	
		Day	Night	Day	Night	Day	Night	Day	Night
SO_2	Palmer	0.260	0.120	0.140	0.100	0.240	0.120	0.370	0.150
	Elandsfontein	0.260	0.120	0.150	0.100	0.220	0.110	0.350	0.150
	Mean	**0.260**	**0.120**	**0.150**	**0.100**	**0.230**	**0.120**	**0.360**	**0.150**
NO_2	Palmer	0.130	0.040	0.050	0.040	0.110	0.040	0.240	0.080
	Elandsfontein	0.140	0.100	0.050	0.040	0.120	0.050	0.260	0.080
	Mean	**0.135**	**0.070**	**0.050**	**0.040**	**0.115**	**0.045**	**0.250**	**0.080**

Source: Mphepya[33]

rain and the precipitation rate.[33] For this study, measured precipitations were available from the meteorological station network of the South African Weather Service.[48]

To compile the study-specific estimate of the cumulative acidic deposition, wet deposition was estimated by using a 10-year volume weighted mean precipitation chemistry record.[34] Ion concentrations for the compounds of interest were extracted (Table 2). This included ammonium, which on deposition enters into reactions involving plants, thus contributing to net acidification.[49]

The wet deposition calculation followed an inferential process where the measured compound of interest (acidic ion concentration) was multiplied by the measured (per site) precipitation for mean annual volume:

$$F = C \times P \qquad \text{[Eqn 2]}$$

In this equation F is wet flux (mol/m^2 per year), C is measured concentration in precipitation (mol/m^3) and P is precipitation rate (mm/year). For this study, sulphate, nitrate and ammonium were included to obtain the overall inorganic – sulphur and nitrogen – deposition loads. Dry and wet deposition results were combined in order to derive a study-specific *total acidic deposition* estimate from sulphur and nitrogen (excluding organic acids) species. Although no aerosols were calculated separately they were indirectly calculated and included from the modelled soil dust deposition and the measured rainfall chemistry composition data.

Critical loads exceedance

This section is based on the methodology published by Kuylenstierna et al.[20] However, several modifications were made for this study, which tested the possibilities based on other reports.

Base cation deposition estimation

Dry base cation deposition: Kuylenstierna et al.[20,21] set the carbonate content in wind-blown dust at 10%.[50] Elsewhere, percentages have been cited within the range 3% – 20% .[50,51,52] Because of the relatively wide range and high uncertainty of the soil deposition data, it was decided to test an upper and a lower limit, within which the acidic loads could be evaluated against the soil or ecosystems sensitivity. These range limits were the lowest and highest percentages plausible. So the base cation (BC) dry deposition rates were estimated on the one hand to be 20% of the wind-blown and deposited soil dust, and 3% on the other hand. Two concentration ranges of soil BC deposition, 10 meq/m^2 – 25 meq/m^2 per year and 25 meq/m^2 – 50 meq/m^2 per year (Kuylenstierna et al.[20]), were based on the model by Tegen and Fung[53] for the study region (Figure 1). The midpoint of each range was used as a mean estimate of BC deposition from soil dust, representing an estimate of 10% calcium carbonate content. This mean value was then used to calculate two extreme values, 3% and 20%, of soil dust BC estimates.

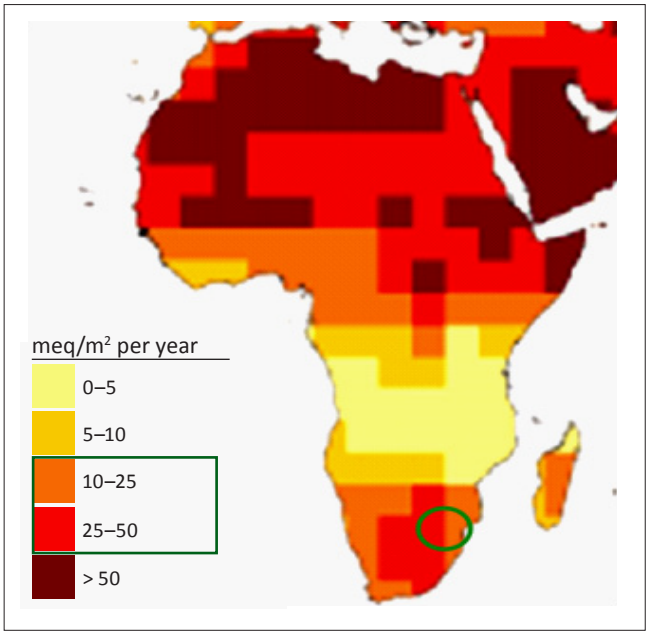
Wet base cation deposition: An estimate for wet deposition of BC, using long-term measurements of rain chemistry base cations in the study region,[34] was included to infer a total BC deposition and to improve the net acidic load estimate (Table 3).

Having four long-term precipitation chemistry sets for four different geographic regions provided the rain chemistry data needed to estimate rain chemistry concentration for the sites in this network. Matching of long-term rain chemistry sets depended on geographical proximity to the rain chemistry measurement site and proximity to the pollution source region or relative remoteness in the background. The positions of the long-term rain chemistry sites in relation to

TABLE 2: Ten-year volume weighted mean precipitation ion concentrations (µeq/l).

Ions	Long-term precipitation chemistry measurement sites			
	Louis Trichardt	Amersfoort	Warden	Vryheid
Cation				
NH_4^+	11.3	25.0	23.1	21.1
Anions				
SO_4^{2-}	15.3	57.2	46.8	40.5
NO_3^-	7.9	24.6	23.1	18.4

Source: Turner and de Beer[34]



Source: Kuylenstierna et al.[20]

FIGURE 1: Base cation deposition (meq/m^2 per year) extracted from modelled dust deposition and based on depositions of soil dust alone, with an assumed 10% calcium content.

TABLE 3: Ten-year volume weighted mean base cation concentrations (µeq/l).

Cations of importance	Long-term precipitation chemistry measurement sites			
	Louis Trichardt	Amersfoort	Warden	Vryheid
Ca^{2+}	13.0	22.8	20.5	19.9
Na^+	12.1	9.7	11.5	16.6
K^+	3.9	4.2	8.3	14.0
Mg^{2+}	4.6	5.5	6.8	6.0
Total	**33.6**	**42.2**	**47.1**	**56.5**

Source: Turner and de Beer[34]

the passive sampling network of this study are indicated in Figure 2.

The BC ion concentrations (excluding NH_4^+) were summed and then multiplied by the total annual recorded precipitation. The annual results were then averaged and the means used as the wet BC component. This BC component was not apportioned for the industrial and sea-source BC. On this basis, two cumulative BC estimates were taken into account when the net acidic deposition was calculated. The lower cumulative BC deposition (3% soil dust BC content with wet BC), as the worst-case scenario, was plotted and interpolated into isopleths for a spatial distribution overview (Figure 3).

Soil data

An international registry of soil information is updated and maintained by the International Soil Reference and Information Centre (ISRIC), Food and Agriculture Organization (FAO) and United Nations Environment Programme (UNEP) under the International Union of Soil Sciences (IUSS) in a compilation named *Soil and Terrain Digital Database* (SOTER). Each SOTER database is comprised of two main elements: a geographical component and an attribute component. For this study, the regional SOTER base for southern Africa was used, SOTERSAF (SOTER for Southern Africa, release 1.0, FAO and ISRIC 2003).[35] Both geographical and attribute data were then handled by a geographic information system (GIS) programme to create polygons of soil types over the study area. The geographical data were related to an attribute database containing cation exchange capacity and base saturation. These values were then combined to calculate the sensitivity classes using the sensitivity allocation as per the Kuylenstierna et al.[20] method.

Determining critical loads from soil sensitivity classes

The 2-year mean sulphur and nitrogen total deposition rates (total; dry and wet) were converted from deposition rate (kg/ha per year) into units suitable for critical loads assessment, i.e. meq/m^2 per year.[54] Critical loads are defined in relation to the buffering rate of soils according to the Kuylenstierna method.[20] In terms of this definition, critical loads are therefore highly correlated to soil buffering rates, which are in turn related to weathering rates. A categorical classification of soil sensitivity has been adapted from Kuylenstierna et al.[20] as an intermediate step in combining acidic deposition rates with soil vulnerability. The ranges of acidic deposition at which each of these five soil classes are susceptible to critical load exceedance are indicated in Table 4.

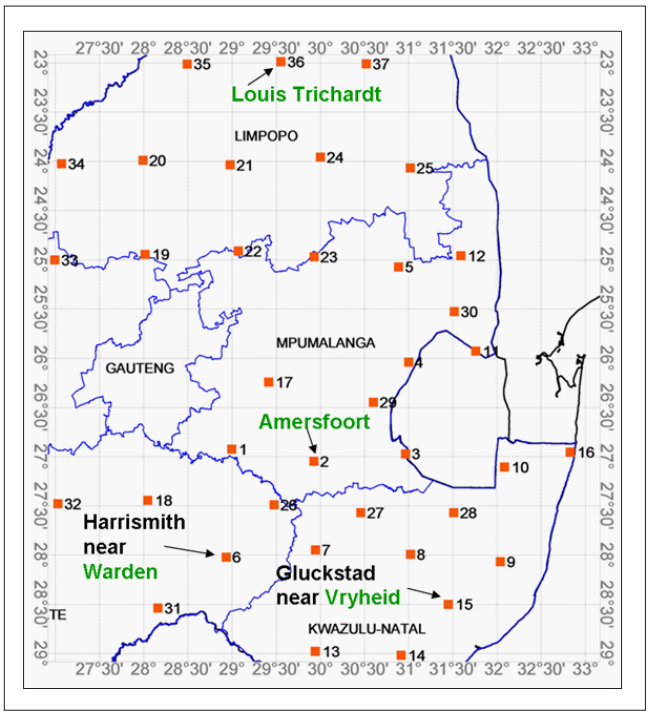


FIGURE 2: Extent of the study area within South Africa with several sites of measurement indicated.

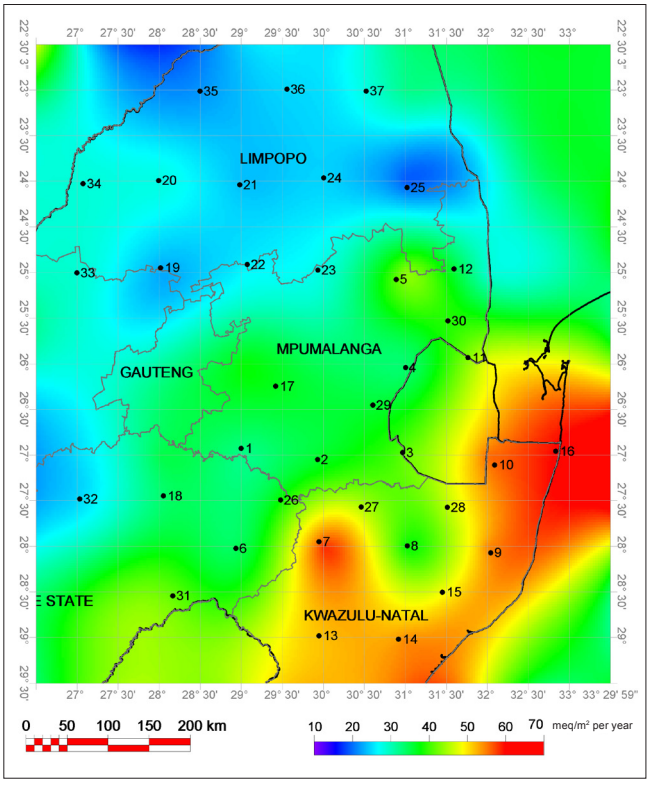


FIGURE 3: Total (dry plus wet) lower base cation estimated deposition rates (meq/m^2 per year).

Results

Total (dry and wet) deposition loads

As expected, total deposition estimates were highest for the site with high dry deposition and relatively high wet deposition. Elandsfontein (Site 17), at the centre of the industrial Highveld, had the highest deposition with an overall total acidic deposition of 23.2 kg/ha per year for the first year and 15.8 kg/ha per year for the second year. Sites downwind of the industrial Highveld (Site 1, Standerton and Site 2, Amersfoort) had the second highest cumulative deposition results for both annual cycles. The lowest total acidic depositions were for Site 25 near Phalaborwa (3.1 kg/ha per year in the first year and 2.9 kg/ha per year

TABLE 4: Allocated soil sensitivity classes against critical loads of acidity.

Sensitivity class	Critical load range (meq/m² per year)	Colour code
1[a]	0 – 25	Red
2	25 – 50	Yellow
3	50 – 75	Green
4	75 – 100	Blue
5[b]	>100[c]	Grey

[a]Most sensitive.
[b]Insensitive.
[c]No critical load.

in the second year), which had low dry and wet deposition, specifically as a result of low precipitation affecting the wet deposition contribution. When the results for all the sites were compared year on year, substantial differences were noted; for certain sites the differences varied by up to 8.6 kg/ha per year (Table 5). This variation can be directly attributed to the differences in precipitation from one year to the next, which directly affects the wet deposition estimate and thus the cumulative (dry and wet) deposition for certain sites.

Deposition of acidic compounds over the study region

The 2-year mean results for all sites were interpolated into isopleths through a GIS programme for a spatial distribution view of the total (dry and wet) acidic deposition loads (Figure 4).

Both the upper and lower BC deposition results were subtracted from the mean total (dry and wet) acidic deposition values to obtain the net lower and higher acidic deposition values, respectively. The lower net acidic deposition had no anion deposition greater than the cation deposition, except in the industrial Highveld, and represents the best-case scenario. It was not used for further assessment. The higher net acidic deposition result represents the worst-case scenario and was used for further assessment (Table 6).

The fourth column in Table 6 (higher net acidic deposition) was plotted and interpolated to create a spatial layer (Figure 5). The levels of acid deposition occur over the central industrialised Highveld, close to the major sources. There seem to be two regions of deposition: the first is an intense ovoid-shaped area over the source regions, pointed towards the south-east as expected, covering a 2° by 2° area (~50 000 km²). This region can be considered the most vulnerable to adverse effects from acid deposition. A

TABLE 5: Total (dry plus wet) acidic deposition rates (kg/ha per year).

Site number	Sampling site	Sept 2005 – Aug 2006	Sept 2006 – Aug 2007	Mean rate	Change in rate
1	Standerton	15.20	11.40	13.30	3.80
2	Amersfoort	19.00	10.40	14.70	8.60
3	Piet Retief	10.30	6.70	8.50	3.60
4	Badplaas	6.00	5.80	5.90	0.10
5	Sabie	8.30	5.50	6.90	2.80
6	Harrismith	10.40	8.10	9.25	2.30
7	Newcastle	14.30	10.30	12.30	4.00
8	Gluckstad	7.10	8.50	7.80	-1.40
9	Hluhluwe	10.40	11.40	10.90	-1.10
10	Ingwavuma	10.70	11.60	11.15	-0.80
11	Masibikela	8.80	7.10	7.95	1.70
12	Skukuza	5.10	3.60	4.35	1.50
13	Escourt	11.60	9.20	10.40	2.50
14	Kranskop	11.90	10.10	11.00	1.80
15	Melmoth	9.40	9.00	9.20	0.30
16	Kosi Bay	10.90	13.70	12.30	-2.70
17	Kriel	23.20	15.80	19.50	7.40
18	Heilbron	13.00	7.10	10.05	5.90
19	Warmbaths	4.20	3.30	3.75	1.00
20	Vaalwater	5.10	3.80	4.45	1.40
21	Potgietersrus	5.00	4.70	4.85	0.30
22	Marble Hall	4.80	4.30	4.55	0.60
23	Steelpoort	5.10	4.90	5.00	0.20
24	Haenertsburg	4.00	3.60	3.80	0.50
25	Phalaborwa	3.10	2.90	3.00	0.30
26	Memel	14.50	7.90	11.20	6.60
27	Utrecht	10.40	9.60	10.00	0.70
28	Louwsburg	4.90	6.40	5.65	-1.50
29	Amsterdam	6.00	6.80	6.40	-0.70
30	Malelane	9.40	4.90	7.15	4.50
31	Fouriesburg	14.10	9.40	11.75	4.70
32	Kroonstad	9.30	3.50	6.40	5.80
33	Pilansberg	5.30	5.30	5.30	0.00
34	Thabazimbi	6.90	3.70	5.30	3.20
35	Tolwe	3.50	3.30	3.40	0.10
36	Louis Trichardt	4.00	3.70	3.85	0.40
37	Thohoyandou	5.80	4.50	5.15	1.30

Total deposition rate includes: SO_4^{2-} and SO_2 as S and NO_3^-, NO_2 and NH_4^+ as N.

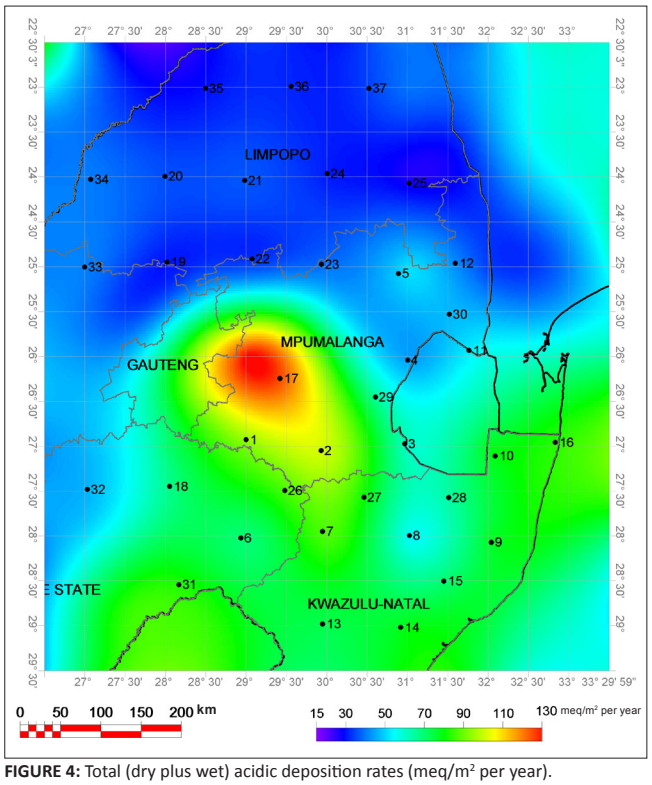


FIGURE 4: Total (dry plus wet) acidic deposition rates (meq/m² per year).

secondary zone of deposition of lower intensity extended to the south and south-south-west, over the eastern Free State and Lesotho (Figure 5).

Soil sensitivity

The next stage of the analysis was to compare the net acidic deposition with calculated critical loads for the soils in the study area. The soil sensitivity polygons are presented in Figure 6. These were then assigned critical loads as described above. Overall, most soils fell into the least sensitive class. However, certain regions are characterised by sensitive soils (Classes 1 and 2, respectively).

Acidification risk assessment in the study region

The higher net level acid deposition layer was laid over (subtracted from) the soil sensitivity values map corresponding to the higher (25-50-75-100) soil sensitivity estimate as per Table 4. The output is shown as the exceedance of critical loads of acidity (meq/m² per year) for the soils (Figure 7).

Exceedance of buffering capacity: exceedance of critical loads

The exceedance map shown in Figure 7 indicates the impacted areas and the degree to which net acidic deposition exceeds critical loads. These areas indicate ecosystems at risk, where acidification and consequent impacts to terrestrial and aquatic ecosystems have occurred or may occur. In contrast, the grey areas indicate areas not at risk from critical load exceedance.

The exceedance map shows that only the central industrial Highveld source area has the potential for high levels of exceedance. Two smaller areas have estimated exceedances of the soil buffering capacity in the intermediate range (51 meq/m² per year – 75 meq/m² per year) – one just east of Amersfoort and the other north of Standerton (Figure 7). A relatively large zone of potential exceedance in the lowest range (1 meq/m² per year – 25 meq/m² per year) occurred in the KwaZulu-Natal Midlands (in a triangle, south-east from Newcastle, north-east from Escourt and north-west from Kranskop). Several scattered small areas of sensitive soils are identified as intermediate and low levels of potential exceedance.

Discussion and conclusions

This study investigated the risk of acidification from both sulphur and nitrogen non-organic acidifying species, anthropogenic and natural, through the method of critical loads mapping. It follows axiomatically that changes in the ecosystem composition and function could occur as deposition continues. The results presented indicate that the upper limit estimate of acidic deposition may exceed

TABLE 6: The higher net acidic (total cumulative dry and wet sulphur and nitrogen minus total dry and wet estimated base cation) deposition rate for the study region based on the lower base cation estimate.

Site number	Total dry and wet deposition (meq/m² per year)		
	Acidic anions	Base cations	Net acidic deposition
1	88.5	32.3	56.2
2	97.1	36.6	60.5
3	56.1	42.0	14.1
4	39.4	33.6	5.8
5	46.5	43.2	3.3
6	65.9	35.0	30.9
7	83.2	58.0	25.2
8	51.5	38.3	13.2
9	72.9	54.1	18.8
10	74.2	55.5	18.7
11	53.3	48.1	5.2
12	29.0	28.9	0.1
13	69.3	51.9	17.4
14	75.8	54.3	21.5
15	61.2	44.8	16.4
16	82.1	61.3	20.8
17	130.8	37.6	93.2
18	66.3	34.9	31.4
19	25.2	22.8	2.4
20	30.4	27.8	2.6
21	31.1	23.7	7.4
22	30.2	26.5	3.7
23	34.8	28.6	6.2
24	25.5	24.7	0.8
25	21.1	19.3	1.8
26	77.6	39.4	38.2
27	66.8	46.2	20.6
28	37.5	29.9	7.6
29	43.0	34.1	8.9
30	50.2	42.3	7.9
31	83.2	44.0	39.2
32	42.4	25.3	17.1
33	35.6	28.7	6.9
34	35.3	28.5	6.8
35	23.2	21.2	2.0
36	25.8	25.4	0.4
37	34.5	32.2	2.3

the critical loads capacity of the local soils in several areas. The level of exceedance is comparable to the level of acidic deposition exceedance in other regions of the world with high emission and deposition rates.[49,55,56]

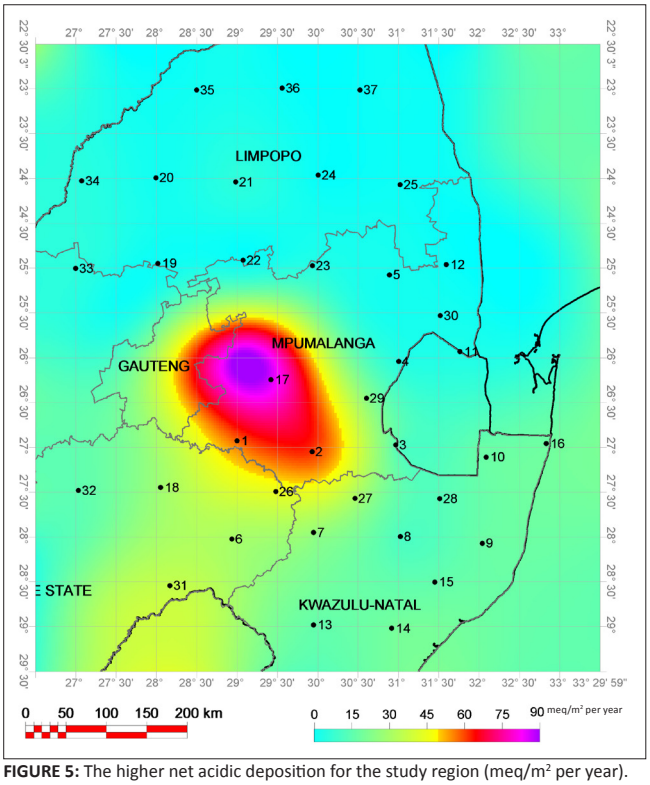
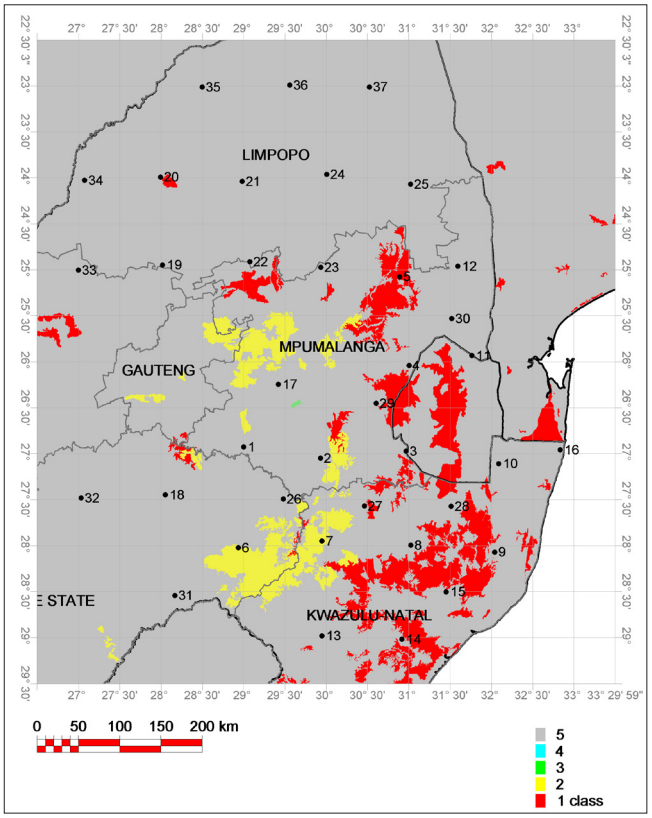


FIGURE 5: The higher net acidic deposition for the study region (meq/m² per year).

A high critical loads (for surface deposition) exceedance was determined in the western and central Highveld industrial region and adjacent area to the north and not, as anticipated, downwind towards the escarpment and the major forestry areas. Several smaller areas located downwind from major sources showed exceedance, although the levels of exceedance were low. Unsurprisingly, these areas of exceedance are on the Highveld plateau. Fieldwork is suggested to test soil acidity and check the effects of acidification in the ecosystems of the areas that showed exceedance.

Critical loads are based on a steady-state concept. Many ecosystems are not in equilibrium with present or projected depositions, because there are processes which delay the reaching of steady state for years or decades or longer. Soil acidification is also a natural process and it is difficult to quantify the extent to which this process is being accelerated by atmospheric acidic deposition. It is also extremely difficult to demonstrate in the field a clear connection between critical load exceedance, declining base cation to aluminium ratios in the soil solution and biological damage resulting from such changes. Therefore dynamic modelling is suggested for estimation of durations involved in attaining a certain chemical state in response to deposition scenarios.[57]

Of course, the method applied in this study is not the only method available. The suitability of other methods, particularly direct methods such as the determination of an index of acid neutralising capacity,[58] in local circumstances should be explored.



Sources: Kuylenstierna et al.[20] and ISRIC[35]

FIGURE 6: Sensitivity classes of the soils based on soil attributes, with Class 1 being the most sensitive.

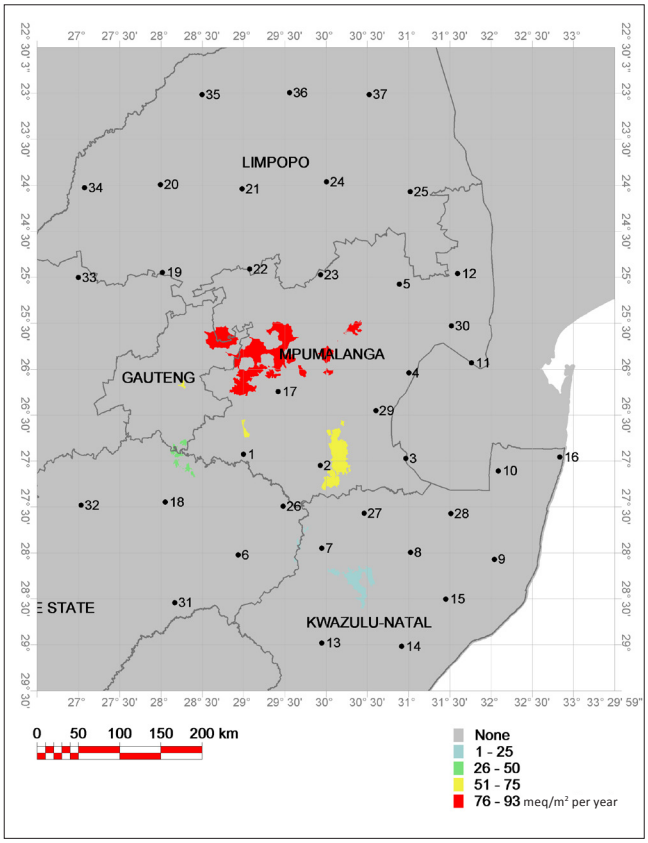


FIGURE 7: Exceedance of acidity critical loads, using the higher net acidic deposition rate and the more critical, higher level of soil sensitivity.

Our conclusion challenges the current understanding of the status of acid deposition from Highveld industrial emissions. However, uncertainties in the BC deposition data, wet acidic deposition estimates and soil database attributes remain. In this respect, measurements of soil dust deposition, with specific attention to BC content, is needed. There is a further need for direct wet deposition measurements on an extensive spatial and temporal scale. Such additional measurements would provide a greater level of confidence and would be needed to validate and implement costly control and mitigation policies on trace gas emissions, or to substantiate a decision that such mitigation is not justified on ecological grounds. In either case, the location of sensitive soils with respect to the deposition pattern is a step in the right direction towards identifying and monitoring ecosystems at risk.

Acknowledgements

The study was based on a joint project of the Department of Geography, Environmental Management and Energy Studies, University of Johannesburg; the Climatology Research Group, University of the Witwatersrand, Johannesburg; and the Atmospheric Chemistry Research Group, North West University, Potchefstroom. Thanks are extended to all those involved. The study was funded initially by Anglo American Corporation, and then jointly by Eskom and Sasol, with a matching contribution from the Department of Trade and Industry THRIP programme. M. Josipovic acknowledges the support of a National Research Foundation (NRF) Grantholder's Bursary, an Oppenheimer Memorial Trust Fellowship (16068) and a Sasol Fund supplementary bursary through the Faculty of Science, University of Johannesburg. The work was supported in part by an NRF Focus Area Project grant to HJA - Sustainability Studies using GIS & Remote Sensing FA2005040600018.

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