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LONG-RANGE TRANSPORT AND CHEMICAL TRANSFORMATION OF POLLUTANTS IN THE SOUTHERN AFRICAN REGION

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ABSTRACT

The paper presents the first results of a long-range chemical transport model developed for Southern African conditions. The model utilizes a combined Eulerian-Lagrangian description of the transport and diffusion of pollutants, and incorporates a description of the chemical transformations, dry and wet deposition as well as an estimation of the pH value of precipitation. A unique feature of the model is the use of an atmospheric boundary layer dynamics model to account for the major role this layer plays in the turbulent diffusion process. The comparison of the model results with experimental data from the DEBITS (Deposition of Biogeochemically Important Trace Species) programme confirms its reliability and usefulness in the quantification of environmental problems typical for the region of Southern Africa. Result maps illustrating concentration and deposition fields allow for the development of regional environmental policies and corrective activities.

Key Words: long-range pollution model, atmospheric boundary layer, atmospheric chemistry, turbulent diffusion.

1. INTRODUCTION

Dispersion modelling of transport, diffusion and chemical transformation of pollutants and trace gases over the Southern African region which spans between 52^o South to 1^o North, 28^o West to 68^o East, presents a special challenge due to three major factors. The first factor is associated with the frequent occurrence of a stable anticyclonic environment. This environment inhibits the vertical exchange of air masses and stratifies the troposphere into persistent layers, in which residence times of pollutants are prolonged from several days to weeks over the region. The second factor stems from the different distribution of emission sources in Africa. Biogenic emissions from biomass burning, vegetation and soils are equal or substantially bigger than anthropogenic emissions over larger parts of the region. Thirdly, long-range transport is vital for the existence or destruction of many fragile ecosystems that receive nutrients or pollutants mainly from the atmosphere. In addition to these major factors, experimental studies on the tropical meteorological factors affecting the long-range transport and chemical transformation of pollutants are limited, and theoretical understanding of the atmospheric processes in the regions with negligible

Coriolis force, are still lacking. Special emphasis should be placed on the identification of key linkages between the physical, chemical and anthropogenic processes governing the functioning of the bio-geophysical and biogeochemical systems of Southern Africa that lead to significantly elevated ozone concentrations over considerable sections of the tropics.

The paper describes the development and application of an appropriate dispersion package for studying the peculiarities of the long-range transport, diffusion and chemical transformation of pollutants and trace gases in the Southern Africa region. Special attention is given to the transport of harmful substances from the highly industrialized regions to the predominantly rural areas of the region as well as wet and dry deposition over sensitive land and water ecosystems.

2. THE LAGRANGIAN-EULERIAN DIFFUSION (LED) MODEL

The Lagrangian-Eulerian Diffusion (LED) model (Djolov *et al.*, 1987) utilizes in a complimentary way the positive features of the Lagrangian and Eulerian description of hydrodynamic flows. It is well known that the essence of the Lagrangian method consists of studying the properties and variation of a fixed fluid volume during its motion. Using this idea in the model, any volume of polluted air is identified by the trajectory of its center of mass. The diffusion and transformation processes of pollutants are investigated on the basis of analytical solutions of the appropriate differential equations in Eulerian coordinates with origin at the center of mass of puffs. As part of the basic structural element of the model, the puff allows for approximation of any type of emission source by using proper puff volume and emission time intervals. Experimental and theoretical studies reveal the fundamental fact that the transport and diffusion of pollutants in the atmosphere can be studied by separating the horizontal and vertical processes (Syrakov e*t al.*, 1983). Indeed, the turbulence in vertical direction is small-scale and it would be appropriate to use the law of mass conservation (the diffusion equation) while in horizontal direction the turbulence is large-scale, the turbulent eddies are not limited by the earth surface and the diffusion process requires a statistical description. This idea is expressed by the critical key formula for the concentration C^K of pollutant K in a point (x, y, z) which reads:

$$C^K(x,y,z) = \sum_{i=1}^{M}\sum_{j=1}^{N_j} Q_{ij}^K(t_{ij}) q_h(x, x_{ij}^c, y, y_{ij}^c, t_{ij}) q_z(z, z_{ij}^c, t_{ij}) q_w(t_{ij}) \qquad (1)$$

where $Q_{ij}^K(0)$ is the quantity of the K^{th} pollutant in the j^{th} puff emitted by the i^{th} source in the moment $t_{ij} = 0$, M is the number of sources, N_j is the number of puffs, q_h and q_z are the horizontal and vertical distribution functions, q_w is the wash-out function and t_{ij} is the life time of the puff. The time variation of Q_{ij}^K is due to chemical transformations, dry and wet deposition processes. The analytical expressions for the functions q_z, q_z and q_w, and more details for the LED model are presented in Djolov *et al.* (1987). It should be pointed out that the vertical and horizontal diffusion functions are explicitly dependent on the atmospheric boundary

layer (ABL) turbulence through the eddy transfer coefficients in vertical and horizontal directions. Therefore, LED needs input from an appropriate simple enough and reliable boundary layer model which, allows for incorporation of new research developments in the ABL dynamics.

3. ATMOSPHERIC BOUNDARY LAYER MODEL (ABL)

A unique feature of LED is the use of an appropriate ABL model calculating its dynamics and turbulent characteristics. Usually, the long-range models are driven by the output of the best available meso-scale forecast models or in the diagnostic case by observed meteorological fields. In both cases however, the ABL dynamics are not properly represented due to the restrictions in the model vertical resolution or insufficient number and distribution of observations. This is a serious simplification since the changes in wind velocity and atmospheric stability occurring in the ABL influence dramatically the transport and diffusion processes. For example, the value of the vertical exchange coefficient changes by order of magnitudes depending on the stability conditions in the ABL. The magnitude and direction of the wind velocity vary considerably with height, and the angle between the geostrophic and surface wind can surpass 50-60 degrees. The wind variations in the ABL are even more complicated when the baroclinicity effects are present. Turbulent friction convergence creates vertical motions that in spite of their small value, lead to substantial displacement of the polluted air volumes because of their perseverance. Another important consideration is that most of the emissions of pollutants and trace gases are released from sources located near the earth's surface up to few hundred meters. The existence of frequent inversion layers at the top of ABL forces the diffusion and transport of pollutants to take place in the lower parts of the atmosphere for prolonged periods of time. These facts underline the importance of inclusion of an appropriate ABL model in any turbulent dispersion package aimed at modelling long-range diffusion and transport phenomena.

In the LED model the two-layer parametric ABL model proposed by Yordanov *et al.* (1983) is included. The ABL model is driven by the following meteorological variables: 1) the geostrophic wind vector $\vec{v}_g$; 2) the potential temperature ϑ_H at the top of the ABL; 3) surface temperature ϑ_s which can be calculated from the energy balance equation or taken from observation or numerical weather forecast model. Note that these are the boundary conditions for the turbulent ABL equations for momentum and heat exchange at arbitrary stratification.

From these external to ABL meteorological variables, and the local parameters: the Coriolis parameter - f, the roughness parameter z_0 and the buoyancy parameter $\beta = g / \overline{\vartheta}$, the following non-dimensional external parameters can be composed:

$$Ro = \frac{| v_g |}{f z_0} \tag{2}$$

the Rossby number, and

$$S = \frac{\beta(\vartheta_H - \vartheta_s)}{f \mid v_g \mid} \tag{3}$$

the external stratification parameter.

These parameters uniquely determine the turbulent regime in a horizontally homogeneous ABL. The details of the ABL model are presented in Yordanov *et al.* (1983).

4. CHEMICAL MODEL

The major anthropogenic atmospheric pollutants at present in Southern Africa are sulphur, nitric and hydrocarbon compounds. In this paper only the results for the sulphur and nitric oxide chemical transformations are presented due to their abundance, relatively long residence times and being the main precursors for the formation of acid deposition. The complicated and numerous chemical photochemical reactions of the hydrocarbons have a pronounced effect on the environment in the vicinity of the major sources.
The most important mechanism for the formation of acid deposition is the direct transformation of sulphur dioxide to sulphate:

$$SO_2 \xrightarrow{K_1} SO_4^{2-} \tag{4}$$

where $K_1(t,r)$ is the constant of transformation which is a function of the time and space. The K_1 value can be obtained assuming that it can be written as a sum of homogeneous and heterogeneous components. The rate of homogeneous transformation is taken from the Endlich *et al.* (1984) paper where it is calculated as a function of the solar insulation using data for a clear atmosphere. The heterogeneous component is chosen on the basis of the review presented by Möller (1980).

The chemical transformations that NO and NO_2 undergo are complicated. Smog chamber measurements of the typical reactions of NO compounds show that 90% of the reactions result in the formation of NO, NO_2, PAN (Peroxyacetyl nitrate) and NO_3. This allows the chemical transformations of these five components to be included in the model. The equations describing the rate of transformation of the nitrogen compounds due to chemical and physical processes are based on Brodzinsky *et al.* (1984). The rate equations used in the model are linear. The equilibrium of NO and NO_2 reflects the photolysis of NO_2 to NO and the photo and thermal pathways of NO to NO_2 conversion.

5. MODEL INFORMATION

A Cartesian grid with 60 x 50 grid cells, on a 50 km resolution was superimposed over the modelling domain of 10°E – 40°E to 10°S – 35°S. Model input parameters

(meteorological data, surface roughness lengths, emission database, etc.) used conform to the grid resolution.

The meteorological data (annual dataset for 2000) needed for input in the model was taken from the short-range forecasting Eta model, obtained from the South Africa Weather Service (SAWS), Pretoria, South Africa. The horizontal resolution of the model is 48 km with 38 Eta levels in the vertical dimension.

The wind field presented in Figure 1 has persisted for large parts of August giving an idea of the closed anticyclonic circulation clearly visible by the vector fields. This typical circulation pattern normally embraces the whole southern Africa region, and persists up to two to three weeks during the winter season.

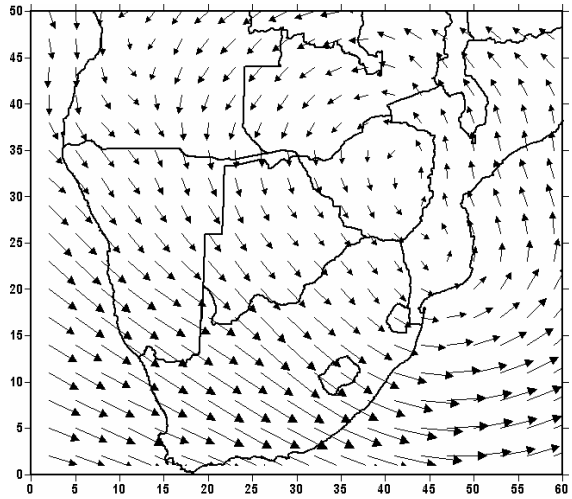

Figure 1.Typical anticyclonic circulation pattern experienced during winter seasons over the region.

The emission database utilized in the model was obtained from Fleming and van der Merwe (2000). The methodology followed and sectors included in the inventory were done according to the guidelines provided by the Intergovernmental Panel on Climate Change (IPCC) (1996). The database includes emissions from various sources on a 20 km grid resolution. The 20km resolution database was reworked to a 50km resolution database, and Figure 2 is a graphical presentation for total sulphur dioxide (SO_2) emissions emanating from the modelling domain.

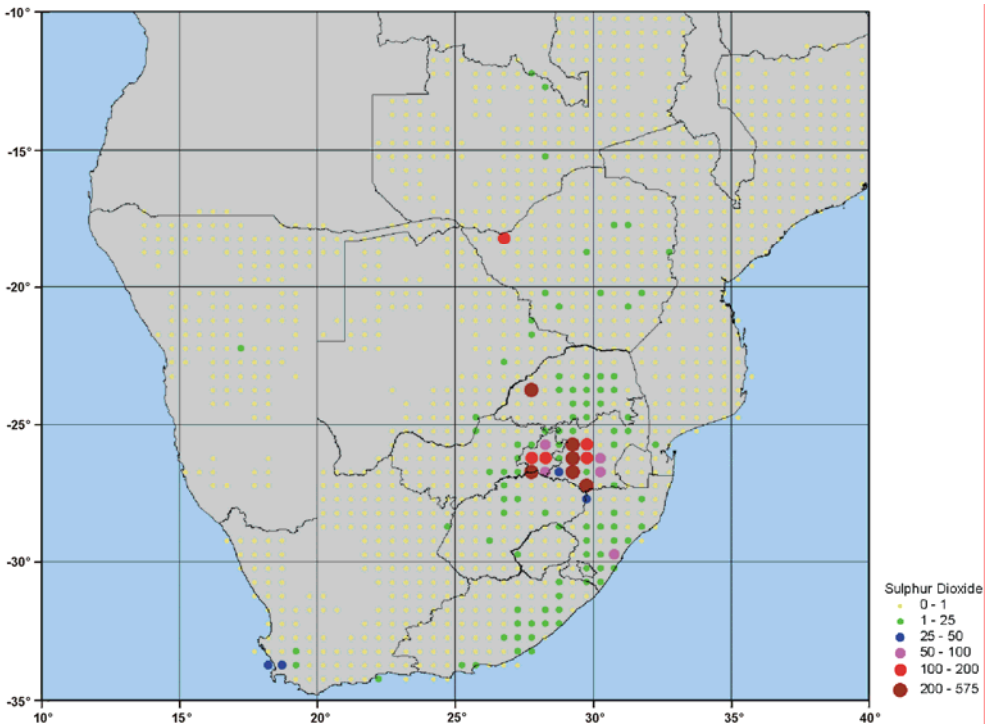

Figure 2.Total SO_2 emissions for the modelling domain (units in Gg per annum) (Fleming and van der Merwe, 2000).

6. RESULTS & DISCUSSIONS

Figures 3 to 5 illustrates modelling results of ambient SO_2 concentration, total dry deposition of SO_X *as S* and total dry deposition of NO_X *as N* experienced during the winter season of 2000. The total *N* deposition field illustrates pollution caused by the emissions emanating from industry, transport and domestic biofuel use.

The first evaluation of the model performance is done using data from the DEBITS (Deposition of Biogeochemically Important Trace Species) programme. Surface sulfur dioxide (SO_2), nitrogen dioxide (NO_2), ammonia (NH_3) and ozone (O_3) concentrations are monitored at several sites in South Africa in the DEBITS (Deposition of Biogeochemically Important Trace Species) programme. Five stations were used for the validation of the model in this study. Cape Point was selected for its proximity to marine ecosystem, Louis Trichardt for its dry savanna ecosystem, and Amersfoort, Palmer and Elandsfontein as industrial highveld sites. The location of the monitoring sites is shown on the base map in Figures 3 to 5.

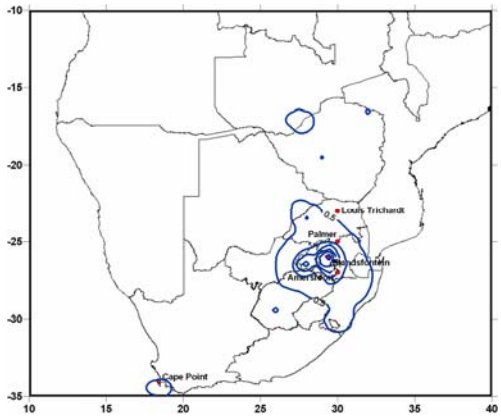

Figure 3.Ambient SO_2 concentrations predicted for the winter season during 2000 (Isopleths units in $\mu g.m^{-3}$).

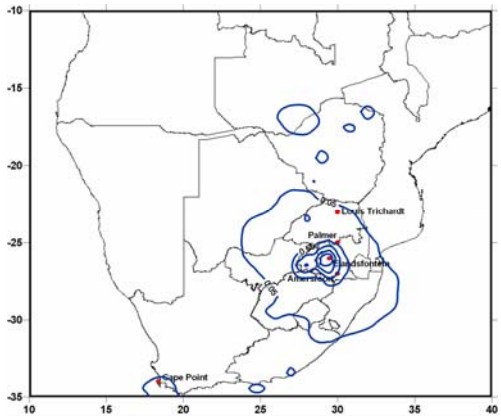

Figure 4.Total dry deposition SO_X *as S* accumulated during the 2000 winter season (Isopleths in $kg.ha^{-1}.3\ months^{-1}$).

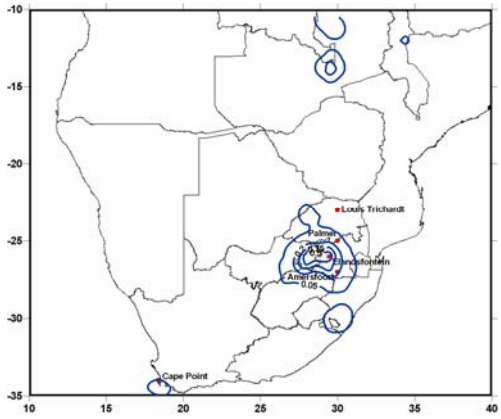

Figure 5.Total dry deposition NO_X *as N* accumulated during the 2000 winter season (Isopleths in $kg.ha^{-1}.3\ months^{-1}$).

Table 1 is a summary of the mean ambient SO_2 concentrations at the DEBITS sites in Southern Africa, compared to simulated results by the LED model, while Table 2 specifies the comparison of dry deposition of SO_X *as S* measured at the DEBITS stations with simulated results by the LED model respectively.

An evaluation of the data in Table 1 indicate that the LED model under predict ambient air concentrations for the majority of DEBITS sites in 2000. The highest under prediction occurs at the Louis Trichardt site. A possible reason for this might be the complicated topography around these stations which is not properly accounted for in the ETA meso-scale forecast model. Moreover, data in Table 2 indicates an over prediction of dry deposition at the different DEBITS stations respectively. Once more only Louis Trichardt demonstrated an under prediction of the total dry SO_X *as S* deposition simulated against measured vales at the site.

Table 1. Mean ambient SO_2 concentrations in $\mu g.m^{-3}$ measured during 1996 to 1998 at DEBITS sites in southern Africa compared to simulated ambient concentrations for 2000.

Site	Value	Autumn	Winter	Spring	Summer
Cape Point	**Measured**[1]	1.2	1.3	1.9	1.2
	Modelled[2]	1.7	1.2	1.3	1.6
Amersfoort	**Measured**[1]	5.9	9.6	7.7	10.5
	Modelled[2]	4.4	5.0	7.3	3.7
Louis	**Measured**[1]	2.9	4.1	3.0	2.1
Trichardt	**Modelled**[2]	0.5	0.5	0.8	0.5
Elandsfontein	**Measured**[1]	20.0	33.5	20.0	19.5
	Modelled[2]	16.6	15.3	19.5	14.7
Palmer	**Measured**[1]	10.3	14.9	5.4	3.2
	Modelled[2]	7.8	6.1	7.8	7.2

[1]Mean values measured from 1996 to 1998.
[2]Modelled LED values for 2000.

Although the results presented are preliminary, a conclusion can be made that the deposition velocities for SO_2 utilized in the model should be reduced, based on the discussion above. Tables 1 and 2 confirm that LED turbulent diffusion package is capable of producing reliable results.

Table 2. A comparison of total deposition of SO_X *as S* / kg.ha^{-1}.3 months^{-1} measured during 1996 to 1998 at DEBITS sites in southern Africa with simulated total S deposition values for 2000.

Site	Value	Autumn	Winter	Spring	Summer	Annual Total[3]
Cape Point	**Measured[1]**	n/a	n/a	n/a	n/a	n/a
	Modelled[2]	0.27	0.18	0.20	0.24	0.89
Amersfoort	**Measured[1]**	0.43	0.47	0.55	1.08	2.52
	Modelled[2]	0.79	0.72	1.06	0.74	3.31
Louis	**Measured[1]**	0.22	0.19	0.21	0.21	0.84
Trichardt	**Modelled[2]**	0.15	0.11	0.16	0.13	0.56
Elandsfontein	**Measured[1]**	2.12	2.08	2.09	3.06	9.36
	Modelled[2]	2.77	2.02	2.43	2.52	9.74
Palmer	**Measured[1]**	0.83	0.72	0.50	0.43	2.47
	Modelled[2]	1.44	0.89	1.18	1.27	4.79

[1]Mean values measured from 1996 to 1998.
[2]Modelled LED values for 2000.
[3]Total deposition as kg.ha^{-1}.yr^{-1}.

7. CONCLUSION

The model results obtained with the Southern African version of the LED model, which incorporates ABL model and new comprehensive chemical scheme, give assurance that it can be used as a diagnostic and prognostic tool for air pollution studies at different time and space scales. The comparison with the available experimental data demonstrates that the results are reliable. For example, the values of the monthly average concentrations field of SO_2 in Figure 4 and deposition field of S in Figure 5 are of reasonable order of magnitude and compare favorably with the measured values at the three DEBITS stations. The results by LED are expected to improve by upgrading the ABL model on the basis of the contemporary understanding and parameterization of the convective process in the tropics that is dominated by non-local turbulent transport. This also means taking into account the baroclinicty effects on the ABL dynamics. A refinement of deposition velocities utilized for the different species in the model is also needed.

Figures 4 to 6 indicates that industrial pollution emanating from South Africa contributes significantly to its own and neighbouring countries pollution problem. Reliable information aimed at regional environmental policy and planning, as well as adequate and effective remedial actions for the individual countries and the region as whole, can be obtained by using sufficiently monthly, seasonal and annual meteorological records. Special attention also deserves case studies of the typical for the region prolonged anticyclonic circulation.

There are several applications with the model, namely, simulations with the natural emissions (vegetation, forest, soils) in order to partition the contribution of anthropogenic and natural sources to the pollution in the region; study the formation of ozone over the tropics; quantify the pollution episodes during a persistent gyre (anticyclonic) circulation and the fluxes of pollutants to the adjacent oceans and continents.

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