6.29 MODELLING LONG-RANGE TRANSPORT AND CHEMICAL TRANSFORMATION OF POLLUTANTS IN THE SOUTHERN AFRICA REGION

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INTRODUCTION

The dispersion modelling transport, diffusion and chemical transformation of pollutants and trace gases over the region of southern Africa 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 that inhibits the vertical air masses exchange and stratifies the troposphere into persistent layers in which the residence times of pollutants are prolonged from several days to weeks over the region. The second factor is related to the totally different from the Northern Hemisphere distribution of emission sources. Natural emissions from fires, domestic fires, vegetation and soils are equal and bigger than fossil fuel emission over larger parts of the region. Thirdly, the long-range transport, diffusion and chemical transformation of pollutants and trace gases is vital for the existence of many fragile ecosystems which are receiving nutrients mainly from the atmosphere. Special attention deserves the identification of the key linkages amongst the physical, chemical and anthropogenic processes underpinning the functioning of the biogeophysical and biogeochemical systems of southern Africa which lead to the significantly elevated ozone concentrations over considerable sections of the tropics.

The aim of the paper is the development 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, wet and dry deposition over sensitive land and water ecosystems, the relative importance of natural and anthropogenic emissions in the atmospheric chemistry cycles, the mechanisms for the formation of elevated levels of ozone, the transport and deposition of contaminants over the adjacent Atlantic and Indian oceans.

THE LED MODEL

The Lagrangian-Eulerian Diffusion (LED) model 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 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 centre 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 the centre of mass. The basic structural element of the model is the 'puff' which permit proper time and space approximation of any type of emission source. Continuous surface and linear sources can be conveniently approximated by instantaneous sources, which periodically emit polluted air volumes shaped as rotational ellipsoids with a vertical axis and Gaussian concentration distribution (Syrakov at all (1983)). 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. 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 main formula for the concentration C^K of pollutant K in a point (x, y, z) which reads:

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

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_i 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. 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 all. (1987).

THE ATMOSPHERIC BOUNDARY LAYER MODEL (ABL)

The LED unique feature, amongst the long-range models known to the authors, is the resolving of the ABL dynamics. Usually, the long-range models are driven by the free atmosphere parameters (geostrophic wind velocity), which are available from the observations or appropriate meso-scale forecast model. The same approach is adopted as well in the meso-scale plume models. 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. 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. Thus, 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 because of the possible striking changes in the turbulent friction force due to the stratification conditions. The angle between the geostrophic and surface wind can surpass 50-60 degrees depending on the thermal and dynamic conditions. 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 surface up to few hundred meters. These facts underline the importance of inclusion of an appropriate ABL model in any 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_o, the buoyancy parameter $\beta = g/\overline{\vartheta}$, the following nondimensional external parameters can be composed:

$$Ro = \frac{|v_g|}{fz_o} \tag{2}$$

is the Rossby number and

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

is the external stratification parameter.

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

THE CHEMICAL MODEL

The chemistry model used for the transformation of the chemical species is based on a simplified version of the Master Chemical Mechanism (Jenkin *et. al.*, 2002). Details of the kinetic and mechanistic data used to construct the degradation Master Chemical Mechanism have been discussed in detail by Jenkin *et. al.*, 1997.

Emphasis is also given to inorganic reactions and specifically those leading to the formation of acid deposition as described by Pienaar and Helas (1997).

THE MODEL INFORMATION

Fig.1, Fig.2 and Fig.3 present the emission, surface roughness and typical wind field driving LED model during the SAFARI 2000 campaign. It can be noted that the roughness field are available for all seasons. The wind field presented in Figure 3 has persisted from 5th of August till 9th of August giving an idea of the closed anticyclonic circulation clearly visible in the upper part of the vector filed. This circulation pattern which normally embrace the whole region and are more intense and persistent up to two-thee weeks is a subject of special case study.

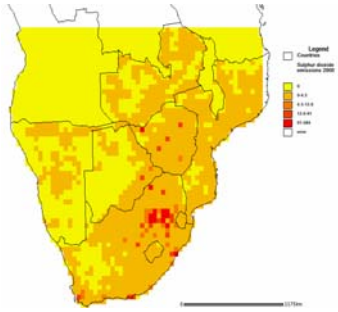

Figure 2. Emission distribution

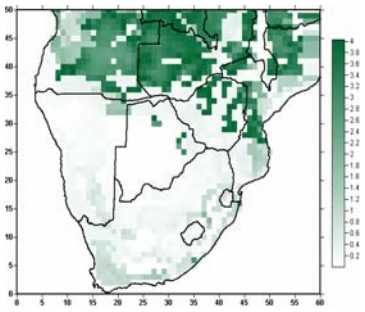

Figure 2. Roughness field

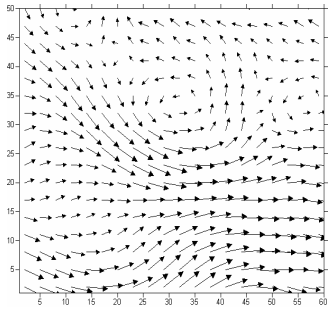

Figure.3 Typical wind field

RESULTS

Fig.4, Fig.5 and Fig.6 show SO_2 concentration, SO_2 deposition and NO_3^- deposition fields for August 2000. The NO_3^- deposition field illustrates the transport of potentially useful substances for the functioning of the tropical ecosystems. The first evaluation of the model performance is done using data from the DEBITS (Deposition of Biogeochemically Important

Trace Species) programme which was initiated in 1990 under the IGAC (International Global Atmospheric Chemistry) framework. Surface sulfur dioxide (SO_2), nitrogen dioxide (NO_2), ammonia (NH_3) and ozone (O_3) concentrations are monitored at remote sites in South Africa. The locations of the monitoring sites are shown on the base map in Fig.4-6 and the values of the monthly concentrations in Table 1.

Table 1. SO2 concentrations in $\mu g / m^3$ *measured* during *August 2000 at DEBITS sites in southern Africa*

ETOSHA (Namibia)	**1.05**
LOUIS TRICHARDT	1.58
AMERSFOORT	6.84
CAPE POINT	1.05
ELANDSFONTEIN	27.89
PALMER	10.00
CEDARA	2.63
SKUKUZA	2.10

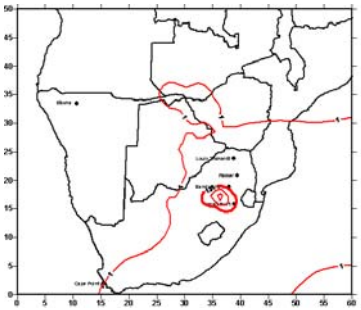

Figure 4. SO_2 *concentration*

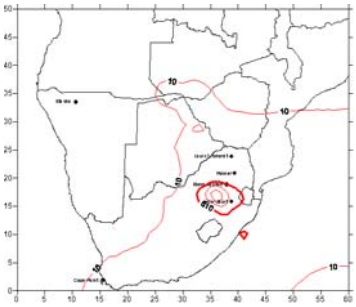

Figure 5. SO_2 *deposition*

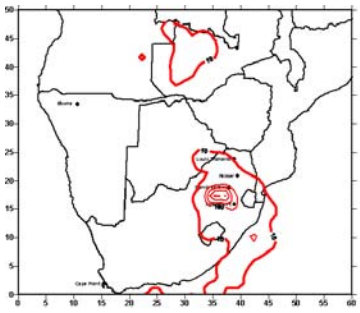

Figure 6. NO_2 *deposition.*

DISCUSSION

The model results obtained with the Southern Hemisphere version of the LED model, which incorporates ABL model and new comprehensive chemical model, 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 for SO_2 exhibited in Table.1, which fall under the concentration field are of the same order of magnitude, the model giving higher concentration near by the sources. There are several projects undertaken to be studied with the model, namely, runs with the natural emissions in order to partition the contribution of anthropogenic and natural sources; study the formation of ozone over the tropics; quantify the pollution episodes during a persistent gyre (anticyclonic) circulation and others.

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