THREE-DIMENSIONAL MODELLING OF ATMOSPHERIC CHEMISTRY

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A three-dimensional chemistry/transport model of the troposphere has been developed and used to investigate the chemical composition of the global troposphere. This model, named IMAGES (Intermediate Model for the Annual and Global Evolution of Species) extends over both hemispheres and from the Earth surface to the lower stratosphere (50 mb). The horizontal resolution is 5 degrees in longitude as well as in latitude. The geographical distribution of the pollutants release at the Earth surface is determined from a detailed inventory of the different contributions to the trace gases emissions, including the vegetation, biomass burning, fossil fuel burning, the soils and the oceans. Surface deposition velocity maps for several trace gases were also constructed from ecosystem and climatic datasets.

This model includes about 120 chemical reactions and photodissociations describing the sources and sinks of about 50 chemical species. These belong to the oxygen, hydrogen, nitrogen, carbon and sulphur families. Surface deposition of chemical compounds as well as wet removal of water-soluble gases are taken into account. Washout is determined using the observed precipitation rate and the climatological distribution and frequency of cloud occurrence determined by the International Satellite Cloud Climatology Program (ISCCP). The photodissociation coefficients of 20 species are interpolated at each time step and each point of the grid from tables giving these coefficients for discrete values of the altitude, the solar zenith angle, the ozone column above the point and the albedo of the underlying surface. This table was calculated using a 1-d radiative transfer model based on the delta-Eddington method.

Long-lived species are transported by winds, diffusion and cloud (Cumulonimbus) convection. In this 'intermediate' model, the effect of short-term variability of the winds is simulated as an eddy diffusion process. The advective transport is performed using monthly mean winds taken from the climatological analysis of the European Centre for Medium-Range Weather Forecasts

(ECMWF). Other fields such as the atmospheric temperature, water mixing ratio and precipitations are also taken from climatological datasets. Vertical convection in cumulo-nimbus clouds is parameterized as a 1-d process, using the ISCCP climatology.

The timestep used in the model simulations is variable : the diurnal cycle is treated at the beginning of each month, using a short timestep ; a timestep of 1 day or 12 hours is used during the rest the simulation.

The purpose of the model is to calculate the three-dimensional distributions and the global budgets of the most important chemically active trace gases in the troposphere. IMAGES is an ideal tool to assess e.g. the impact of human activities on the chemical structure of the atmosphere and to understand the role played by chemically active trace gases in such concerning problems as the greenhouse effect and the photochemical smog. The model has been validated by an extensive comparison with available observations of CH_4, CO, NO_y, O_3 and other species. Although IMAGES cannot represent properly the real short-term variability of the atmospheric composition, the comparison shows its ability to reproduce the large-scale features and climatological budgets of the major tropospheric compounds. The atmosphere during the pre-industrial period has been simulated with the model and compared with the present-day atmospheric composition, showing the important impact of the industrial revolution on the global budgets of greenhouse gases such as methane and tropospheric ozone.

References

Müller J.-F. 1992. Geographical distribution and seasonal variation of surface emissions and deposition velocities of atmospheric trace gases. J. Geophys. Res. 97 : 3787-3804.