ATMOSPHERIC TRANSPORT

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1. INTRODUCTION

Trace species with chemical lifetimes significantly longer than the local characteristic time for horizontal and vertical exchanges may be transported far away from their emission sources before they react with other chemical species. It is therefore of primary importance to understand the basic mechanisms governing the transport of chemicals and to formulate these processes as accurately as possible in numerical models.

The study of trace species dispersion in the atmosphere requires a detailed understanding of the basic transport processes on a wide range of spatial and temporal scales especially between the different atmospheric layers. Shown in Figure 1 is a schematic representation of various transport mechanisms below 30 km. Of particular importance are the exchanges between the atmospheric boundary layer (ABL) and the free troposphere, the ABL and the surface, including wet and dry deposition processes, and between the stratosphere and the troposphere. Several papers in this volume deal with some of these questions. The purpose of this report is to identify the important problems to be resolved in the next decade or so and to suggest research strategies in this context.

1.1. Spatial and Temporal Scales in Atmospheric Transport

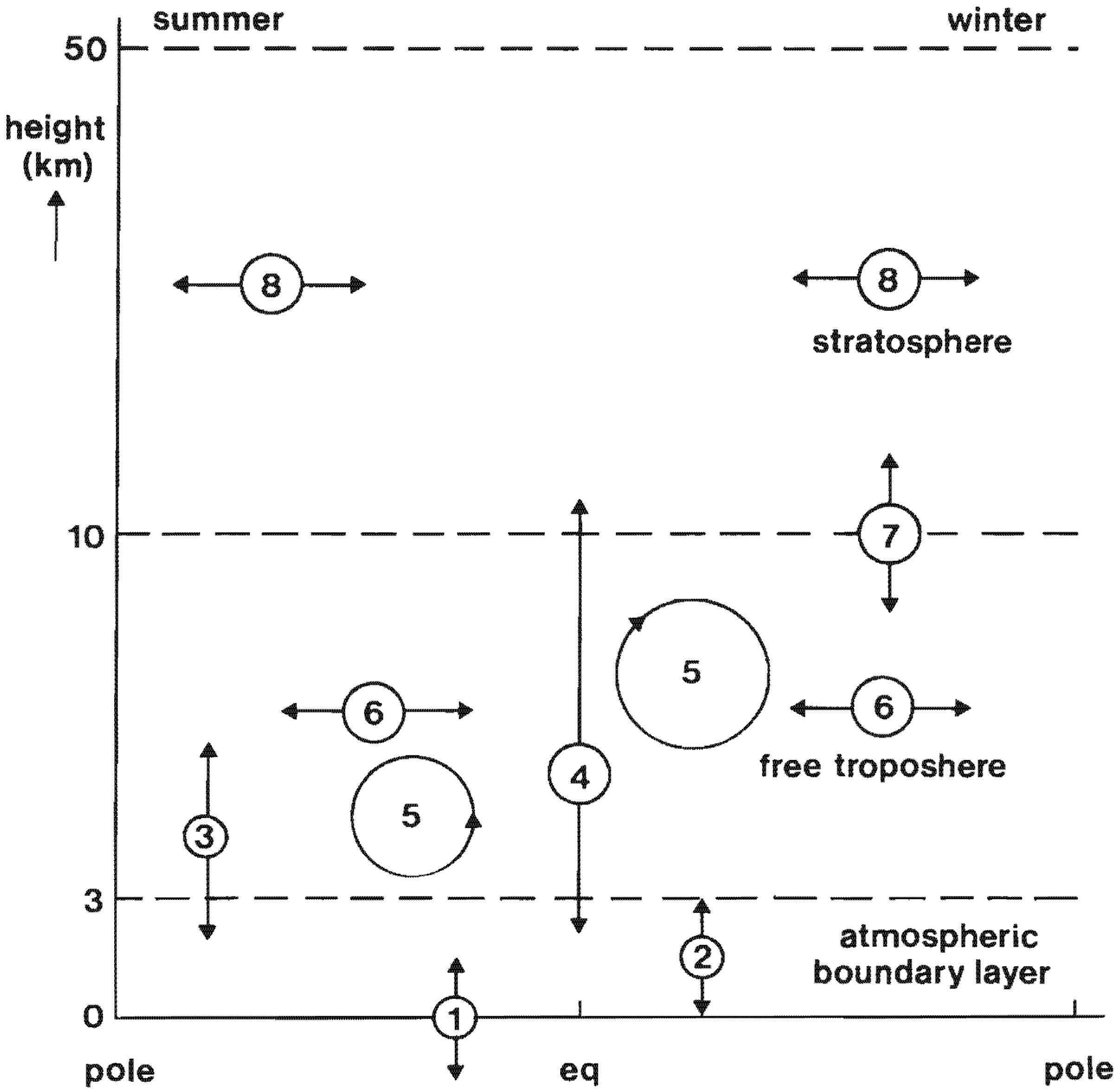
The type of transport affecting chemical species varies with the time scale involved. For some applications it may be desirable to know hourly values of concentrations; for other only monthly, seasonally or yearly averaged concentrations are required. Also the spatial resolution of observations vary widely. In the modelling of transport, therefore, it makes sense to distinguish these various scales.

The area of concern in this study ranges from the atmospheric boundary layer through the tropopause, i.e. the vertical scale ranges from 1 to 20 km. Numerical models which are able to resolve the structure of transport over the vertical range should have a grid which is at least an order of magnitude smaller. In a consistent three-dimensional model, horizontal scales should be compatible with vertical scales and can be inferred from the vertical scale by equating their corresponding time scales. Vertical transport velocities vary from $\sim$ 0.01 m/s

I. S. A. Isaksen (ed.), Tropospheric Ozone, 383–392.

Fig. 1 A schematic view of transport processes in troposphere and stratosp

1. Earth/ocean-atmosphere exchange processes
2. dry-convective ABL-mixing (Ekman-layer pumping)
3. Mid-latitude vertical exchange processes associated with frontal systems and moist convection
4. Deep convection in tropical storms
5. Mean tropospheric circulation
6. Horizontal exchange by breaking of Rossby waves
7. Troposphere-stratosphere exchange (tropopause folding)
8. Horizontal eddy diffusion in the stratosphere



(subsidence) to $\sim$1 m/s (convection), which leads to time scales of 1000 - 100.000 s. Realizing that horizontal transport velocities are of the order of 1-100 m/s, the horizontal mesh size should be somewhere between 0.1 and 1000 km. (It should be noted here that chemical processes have their own timescales which may set different requirements. This problem, however, is not addressed here).

A typical model with a grid size of 100 km, a vertical resolution ranging between 100 and 1000 m and a timestep of three hours should resolve the (horizontal) Rossby planetary waves, most circulations associated with cut-off lows, orographically induced vertical motions by the large mountain ridges (Alps, Andes, Himalya, Rocky mountains), large scale subsidence, cyclones and anticyclones.

They however do not resolve small scale turbulent disturbances (boundary layer and clear air turbulence),cumulus cloud convection and associated precipitation, local circulations such as nocturnal jets, valley circulation, land-sea breeze and drainage flow, incidental cumulonimbus entrainment of stratospheric air in the tropics and fronts.

Features that are partly resolved include tropopause folds, mesoscale flow over mountains and mesoscale organization of convection in the tropics.

1.2. Potential Applications of Transport Modelling

The modelling of transport in the troposphere may contribute to our understanding of the distribution and trends of atmospheric constituents. In particular, it may contribute to the following fields:

- regional modelling of ozone

Causal relationships may be found between various sources of primary pollutants and the occurrence of high ozone concentrations.This can be done by episodic transport models, i.e. models which describe transport (and chemistry) over a period of a few days only.

Models which have proved their ability to predict concentration fields can be used either to supply or to interpolate data in regions where measurements are sparse.

Simpler and faster means to identify sources are trajectory analyses. Such analyses (and forecasts) may be helpful in guiding experiments and examining chemical transformations during field experiments.

- global modelling of ozone

For the study of the global distribution and trends of ozone and other relevant trace constituents, measurements will provide only a very limited and incomplete picture. They are, especially at elevated levels, difficult to carry out and relatively expensive. Models provide global fields and may realistically represent the spatial and temporal variability of trace gases.

2. FUNDAMENTAL PROCESSES IN THE TROPOSPHERE

2.1. Vertical Stratification

Away from the boundary layer, the potential temperature of the atmosphere increases with height. Its vertical gradient determines the stability against convective mixing and therefore the potential for laminar, stratified flow. In the stratosphere, where the potential temperature increases rapidly with height, laminar flow along isentropic surfaces dominates, and any trace gas released as a parcel into the lower stratosphere spreads rapidly while remaining on the surface. The stratification is not as pronounced in the troposphere, but is nevertheless present, particularly in high pressure systems where subsidence inhibits convection.

Laminar flow of this kind has several implications for atmospheric chemistry which are not well understood. Most obviously, if reactive tracers lie in adjacent thin laminae a few hundred meters thick, their chemistry will be most active at the interface - which can be very sharp. Thus, in stable flows the inhomogeneous mixing of tracers of different origin suppresses chemistry and permits long-range transport of reactive trace constituents. Modelling the transport of reactive species becomes very hazardous under these conditions - the familiar expression of the law of mass action,

$$d(A)/dt = -k\ (A)\ (B)$$

with (A) and (B) averaged over some model box, will clearly be a misrepresentation. Fundamental research into the extent and the properties of laminar flow, as well as its impact on chemistry, is required.

2.2. Wind Shear and Parcel Integrity

Related to this problem is the effect of wind shear on tracer distributions. A parcel of air, initially of a regular shape, perhaps 100 km square, will be extruded by wind shear over a period of a few days to a long thin, coiled parcel. In doing so, parcel integrity is eventually lost, since the surface area increases enormously and diffusion permits exchange of tracers with adjacent air parcels. Again, Eulerian type models are sensitive to this process of differential advection since it can act to increase gradients by bringing together air parcels of very different origins. Furthermore, when the extruded air parcel becomes thinner than the grid of the model, its identity within the model is lost and its contents immediately mixed into its surroundings. Thus Eulerian models may be expected to underestimate the long-range transport of trace constituents.

Isentropic trajectory calculations based on numerical forecast models are a powerful tool for calculating air transport in the free atmosphere. But these too are sensitive to wind shear - both from the standpoint of parcel integrity, and because of the sensitivity of the calculations to initial conditions. Wind shear is very large near jet streams, and trajectory calculations for longer than two or three days in the free troposphere should be undertaken only for specific meteorological situations.

2.3. Stratosphere-Troposphere Exchange

The problems of describing exchange of air between the troposphere and the stratosphere are magnified in situations with tropopause foldings, where stratospheric air flows in a lamina beneath a northwesterly jet stream as a response to frontogenesis. The fraction of the ozone flowing into the fold which remains in the troposphere (rather than returning isentropically to the stratosphere) is not known. It is important to know this fraction, since any air returning to the lower stratosphere is likely to contain a tropospheric component introduced by the vigorous mixing at the base of the fold. This would then re-enter the troposphere in the next fold downstream, so that all the air entering the troposphere in a folding event would not be of stratospheric origin.

Calculations of the global flux of ozone from stratosphere to troposphere, based on experimental data, have so far consisted of extrapolation from a number of case studies, mostly in the U.S. For the Southern Hemisphere, in particular, the flux is not known, even in the mean, let alone in its variability. Global estimates of stratosphere-troposphere exchange have been calculated with GCMs, and agree quite well with those based on case studies, but the results need to be confirmed by global studies using real data before confidence can be expressed in flux estimates. An uncertainty of a factor of two, particularly in the Southern Hemisphere, is likely.

2.4. Fronts

The mesoscale dynamics of frontal systems are not well understood by meteorologists. Some structure is well-known - e.g. the conveyor belt mechanism ahead of a cold front and the extensive slow ascent of air above a warm front. Embedded in these are mesoscale rainbands where enhanced precipitation and vertical transport is found, often in a highly organised structure. Fronts also behave erratically over land, especially in mountainous regions where they occasionally disappear, perhaps reforming on the other side of the mountains.

These fronts are not regions of uniform precipitation and ascent, and must be treated especially carefully in chemical models. By their nature, they are important for exchanges from the boundary layer to the free troposphere in the extratropics - so that boundary layer trajectory calculations, for instance, should terminate when a front is encountered. Trajectory calculations through frontal regions are also difficult to perform in the free troposphere. Experiments with releases of non-reactive chemical tracers in the vicinity of fronts would greatly aid chemists and meteorologists in studies of the transport in these regions.

2.5. Cloud Sheets

Extensive stratocumulus sheets are a feature of winter anticyclones in the high midlatitudes. These layer clouds often interact very little with the rest of the atmosphere during daytime, so that trace constituents trapped in them can be transported up to 1000 km. Wet chemistry occurs in these clouds, but there is seldom any precipitation-the clouds usually evaporate.

2.6. Cumulus Cloud Transport

One area of fundamental uncertainty concerns the transport properties of convective clouds. There are strong indications that the inflow, outflow and trajectories of molecules depend on the depth and breadth of the cloud, the environmental wind shear, the rainfall process, etc... Observations and modelling studies are now only beginning to define quantitatively the answers to these fundamental questions. Carbon monoxide and hydrocarbons provide tracers of air motions that are free of the measurement and interpretative difficulties associated with conventional meteorological analyses. Cloud water and cloud air may take different trajectories in cumulonimbi. Soluble species with low vapor pressures are expected to follow the water droplets, but many important species that are soluble have a high vapor pressure and may move from drop to drop.Soluble conserved tracers may illuminate these processes.

Nitric oxide may have important sources within thunderstorms. The impact on NO distribution in the global troposphere depends on the transport through and out of the clouds as well as complex heterogeneous chemistry. Currently, all of these processes are poorly understood.

Cloud droplets are very efficient for scavenging particles and soluble gases. Complicated cloud models including interactions between dynamical, chemical and microphysical processes in the gaseous, liquid, ice and particle phase are difficult to incorporate into even the highest resolution mesoscale models. Further complications arise from the complexity of radiative transfer and heterogeneous chemistry in this environment. Nucleation is the main in-cloud scavenging process whereas impaction dominates below cloud. The stochastic nature of precipitation fields makes it difficult to compare model results with rain chemistry observations because individual rain gauge data are not representative enough. In fact large discrepancies are often observed.

2.7. Transport in the Atmospheric Boundary Layer

Processes within the lowest part of the ABL, say 5-50 m (the surface layer), are well represented by th Monin-Obukhov similarity theory which provides a framework for parameterizing the aerodynamic resistance to deposition of particles and weakly reacting gases. Closer to the surface ($\sim$1 mm), molecular and laminar resistance is generally parameterized on the basis of models tested by laboratory measurements. How well these laboratory conditions represent the real world remains to be demonstrated.

The case of particle deposition over water is more difficult due to the growth of particle size as they approach the water surface. Effects of bubbles, spray and foam should also be clarified since these mechanisms may apply to a large fraction of the earth's surface.

More data (surface type, moisture content, chemical/biological state) are required in order to assess the surface resistance to transfer for trace gases over a wide variety of surface conditions. It should be mentioned that some species have their main resistance to transfer in the water, so that mixing and molecular diffusion in the upper layer of the ocean should be coupled to the meteorological model.

The horizontal transport of species is performed by the advecting wind (longitudinal diffusion being at most 10 % except in stagnant wind conditions). Under unstable conditions, the upward mixing of ground-emitted species together with the relatively large gridsize of models make the assumption of a vertically homogeneous wind field reasonable in the ABL. However, the 925-mb surface should be more representative of ABL-wind than the often used 850-mb surface generally located above the ABL top. The interfacial layer at the top of the ABL is often characterized by a strong jump in wind speed and direction.

In the case of the stable ABL, there is a large velocity and directional shear throughout the ABL so that the horizontal transport of species should be height dependent close to the source as they rise slowly (due to weak turbulence) from the surface.

The collapse of the continental daytime well-mixed ABL at sunset leaves boundary layer air in a residual layer aloft, subsequently transported elsewhere. This type of exchange has not been properly addressed in many existing transport models.

Surface emission can be treated in the same way as deposition. However, all models suffer from the absence of temporal distribution in the emission data (both anthropogenic and biogenic). Incorporation of time-dependent emission input would significantly improve transport models capability.

3. USING METEOROLOGICAL/DYNAMICAL MODELS TO SIMULATE TROPOSPHERIC TRANSPORT

High resolution global general circulation (climate) and weather prediction models (100 km grids and 20-40 levels in the vertical) can simulate realistic synoptic meteorology down to and including such important mechanisms for free tropospheric transport as extra-tropical cyclones and tropical waves. Their precipitation patterns are accurate on a regional scale, but not quantitatively correct on the mesoscale. Medium resolution meteorological models ($\sim$300 km grids and 10-30 vertical levels) are also able to simulate such important features but show only qualitative agreement with observation. Meteorological fields from global models can simulate the global climatology of distribution and deposition of chemical trace species and provide 3-5 day forecasts for particular experiments. Unfortunately, at their highest resolution, these models tax the limits of current computers. We can reasonably expect that the next generation of computers (approx.1990) will support high resolution global transport/chemistry models which transport 1-5 chemically reactive species or groups. We should be planning to develop the medium resolution transport/chemistry models (1-5 transported species) which are consistent with current computer resources and, at the same time, begin to plan for the future.

Current regional and meso-scale (50-100 km sq) meteorological models have only been used to study specific event on a 1-2 day time scale though their use has been proposed for climatological studies. Such models produce more realistic simulations of synoptic scale phenomena as they evolve over 1-2 days. Beyond that time, the results rapidly deteriorate. The precipitation fields are more realistic than the highest resolution global models, but they do not yet capture the observed local variability.

Models which transport 10-20 reactive species during a particular meteorological event are currently under development.While these models simulate the gross distribution and deposition features for reactive trace chemicals, there are serious deficiencies in the detailed structure. Beyond 2 days the current model meteorology rapidly degrades. The continued improvement of such meteorological models and their extension in space and time is clearly needed.

An alternative approach is the nesting of a mesoscale model in a coarse global model. This is just in the development stage and the key scientific question is whether the result will be a regional meteorology that has been seriously degraded by the deficiencies of the coarse global model or a coarse global model that has a meteorology in a region that is of meso-scale quality. If it works, this may provide a framework for generating a realistic meso-scale chemical climatology, not just a 2 day forecast.

All these models exhibit two major common difficulties: the inability to resolve the important boundary layer turbulent transport processes and convective vertical transport in the free troposphere. While realistic meteorological models of these processes do exist, it is not possible, as discussed in 2., to include them explicitly in the transport/chemistry models. Therefore it is necessary to parameterize their effects. Development of realistic parameterizations for both boundary layer turbulence and cloud transport and precipitation is a critical need for both meteorology and atmospheric chemistry.

4. MODELLING SUB-GRID TRANSPORT PROCESSES

4.1 Parameterization of Subgrid-Scale Convective Transports

The underlying transport functions describing the transport by clouds of material from each level of the troposphere to every other level are not clearly defined by theory or experiment. Consequently, we cannot be very confident of our parameterizations. Traditional parameterizations for convection have errors in their conceptual underpinnings. New parameterizations for weather-forecast and climate models will probably await more fundamental understandings of cloud-scale and mesoscale meteorology. Air chemical applications cannot wait.

There are good reasons why air chemistry makes different demands on transport models. The parameterization of convection in meteorological models addresses the problems of latent heat release and momentum generation, but does not pay particular attention to material transport. This problem will remain a very difficult one that requires the cooperation of atmospheric chemists and dynamical meteorologists.

4.2. Parameterization of Subgrid-Scale ABL Transport

Exchange between the ABL and the free atmosphere occurs by entrainment in the case of a well-mixed ABL (warm continental daytime conditions or cold air outbreaks over warmer seas). Since this is a small-scale process, it has to be parameterized by using resolved or calculated quantities. In the case of a stable ABL (nocturnal continental case or spring/summer case over mid- and high latitude seas), the weaker entrainment, resulting from internal wave breaking or downward bursts of turbulence driven by strong velocity shear, has not been properly parameterized. Synoptic scale downward motion (subsidence) modulates the growth of the ABL, both in the stable and unstable cases.

Convective and mechanical production of turbulence have been mainly observed over the sea together with the occurrence of an extended capping layer of stratocumulus clouds. Radiative destabilisation at the top of the cloud layer, enhancing mixing inside the cloud, should be included.

Most parameters required to describe surface and ABL- top processes in transport models (surface heat flux, friction velocity, convective velocity and ABL- height) are provided by modern meteorological models (e.g. European Centre for Medium Range Weather Forecasts).

5. MODEL STRATEGY

Numerical models are important tools for studying the behaviour of trace-species in the atmosphere and for assisting in ozone control strategy formulation. Despite much progress made in recent years, a number of difficulties in model formulation remain to be solved.

The type of model to be used for regional or global studies should be governed by the particular problem to be addressed. For example, the chemical behaviour of fast reacting species for a given solar illumination can be derived from a simple zero-dimensional model if the concentration of the long-lived species, especially the source gases is specified either from field measurements at the given location or from a comprehensive transport model. Trajectory models (Lagrangian models) with a coupled chemical code may consider the changes of chemical composition within an air parcel which encounters different solar and meteorological conditions. These models are useful for the interpretation of trace species observations at selected measurements sites or for the assessment of the distribution of pollutants emitted at a fixed point. They are only reliable if the flow is not disturbed by wind shear, small-scale turbulence, strong convection or fronts. Trajectory calculations have shown that the origin of air parcels above a give location can vary strongly with altitude. Such simulations can only be performed over a limited period of time (a few days).

Regional or global distributions of trace species calculated over long integration times are usually provided by Eulerian models. The simplest approach involves one-dimensional models in which the vertical exchanges are parameterized by eddy diffusion. This method oversimplifies the representation of the transport processes which are advective rather than diffusive in nature and therefore other empirical parameterizations have been used. One-dimensional models can still be useful to give order of magnitude estimates of global budgets and to study the local details of chemical processes. Because of large horizontal inhomogeneities in both dynamical and chemical processes in the troposphere, the interpretation of the results given by such models is severely limited and the calculated vertical distributions cannot be regarded as global average conditions.

Two-dimensional models are in principle capable of accounting for latitudinal and seasonal dependence of solar illumination, and boundary conditions and allow a rudimentary representation of meridional advection. The limitation of such models arises from the difficulty of representing zonal averages of mass, momentum and energy fluxes, which are the result of complex dynamical processes involving large zonal assymmetries. The net fluxes resulting from these processes are usually specified through empirical parameters. However by using transport coefficients based on observed meteorological variability and dispersion of tracers, one may simulate the meridional distribution of chemical tracers.

A detailed representation of the transport of trace species on any scale (e.g. ABL turbulence, convective cloud, front, cyclone or interhemispheric) should be based on a full three-dimensional representation. The success of such models depends on the spatial and temporal resolution adopted in the dynamical formulation as well as on the completeness of the chemical scheme included. Despite the fact that the simulation of the real atmosphere is significantly improved by such models, some parameterization is still required to account for sub-grid transport and eventually to ensure numerical stability. With the rapid development of more powerful computers, the resolution of three-dimensional models can be expected to improve in the next five to ten years. Interaction with observations is important in model development, both for validating model predictions and for designing complex field experiments. Chemical tracer measurements have an important part to play in this interaction.

The most comprehensive chemical transport models of the troposphere should be linked to general circulation models, have the highest possible resolution, extend to the stratosphere and be capable of eventually studying the climatic impact of natural and anthropogenic perturbations.

A practical approach to the three-dimensional transport/chemistry problem is to use the assimilation fields from numerical forecast models to represent atmospheric dynamics. Chemical calculations may then be conducted in an "off-line" mode, but care should be taken to extract the full range of information required from the forecast model, not just temperature and wind fields.

Although less complex models remain quite useful for addressing a wide range of specific questions and for trying out new ideas, a major effort should be undertaken to develop high resolution three-dimensional chemical transport models. Because of the magnitude of this task, such projects require international cooperations and a long-term committment.