A simple quasi 2-D model is used to study the zonal distribution of NO\textsubscript{x}. The model includes vertical transport in form of eddy diffusion and deep convection, zonal transport by a vertically uniform wind, and a simplified chemistry of NO, NO\textsubscript{2} and HNO\textsubscript{3}. The NO\textsubscript{x} sources considered are surface emissions (mostly from the combustion of fossil fuel), lightning, aircraft emissions, and downward transport from the stratosphere. The model is applied to the latitude band of 40°N to 50°N during the month of June; the contributions to the zonal NO\textsubscript{x} distribution from the individual sources and transport processes are investigated. The model predicted NO\textsubscript{x} concentration in the upper troposphere is dominated by air lofted from the polluted planetary boundary layer over the large industrial areas of Eastern North America and Europe. Aircraft emissions are also important and contribute on average 30\%. Stratospheric input is minor about 10\%, less even than that by lightning. The model provides a clear indication of intercontinental transport of NO\textsubscript{x} and HNO\textsubscript{3} in the upper troposphere. Comparison of the modelled NO profiles over the Western Atlantic with those measured during STRATOZ III in 1984 shows good agreement at all altitudes.

1. INTRODUCTION

A major fraction of the available measurements of NO in the upper troposphere were made during the Stratospheric Ozone aircraft campaign STRATOZ III in June 1984 (Drummond et al., 1988). The flight track of that campaign was placed mainly along the coastlines of North America, South America, western North Africa and Europe between 70°N and 60°S latitude and from 0 to 12 km altitude. The most conspicuous feature in the individual vertical profiles (see for example figure 1) and in the two dimensional representation of all measurements (figure 2) is the large hump of high NO-Concentrations in the upper troposphere at 10-11 km altitude extending all over the northern hemisphere.

To explain this result, all possible NO-sources must be considered, namely stratospheric input, emissions by high flying aircraft, lightning and fast vertical transport from the planetary boundary layer. Unfortunately, the observed patterns in the NO-distribution were by themselves not sufficient to provide a quantitative estimate of the contribution of the various sources. Also the hope of using correlations with the concentrations of other trace gases measured during STRATOZ III, like O\textsubscript{3} and CO, proved vain so far. So we attempted to use a simple quasi-2-D model to quantify the impact of the sources mentioned. The present paper summarizes the salient findings of our earlier work (Ehhalt et al., 1992).
2. MODEL DESCRIPTION

The model includes three types of transport: vertical transport by deep convection and eddy diffusion, and horizontal transport by vertically uniform wind. The fast vertical transport induced by deep convection is treated like a random process and superimposed onto a continuous transport by eddy diffusion. Selected by a random number, a fixed fraction of air at a specific altitude level is replaced by air from the boundary layer followed by a downward shift from each of the lower levels to the next one to maintain mass balance. This random process is adjusted such that the product of transport frequency and exchanged air fraction matches the exchange rate profiles (fig. 3) taken from the convection statistics of the GCM developed at the Goddard Institute of Space Studies (M. Prather, private communication 1990).

Horizontal transport is introduced by moving the whole air column (0-14 km altitude) with a horizontal displacement given by a 8 m s⁻¹ westerly wind (Houghton, 1985). The tropopause is located at 12 km altitude. Temperature and density profiles are those of the U.S. Standard Atmosphere (1976). All parameters apply to the summer month at 40°-50°N latitude.

The model allows the nitrogen compounds NOₓ=NO+NO₂ and HNO₃ to be transported. Those species are interlinked by reactions (1) to (8).

\[
\begin{align*}
\text{NO} + \text{O}_3 & \rightarrow \text{NO}_2 \\
\text{NO}_2 + \text{hv} & \rightarrow \text{NO} \\
\text{NO}_2 + \text{OH} & \rightarrow \text{HNO}_3 \\
\text{HNO}_3 + \text{OH} & \rightarrow \text{NO}_2 \\
\text{HNO}_3 + \text{hv} & \rightarrow \text{NO}_2 \\
\text{NO}_2 + \text{O}_3 & \rightarrow \text{NO}_3 \\
\text{NO}_3 + \text{H}_2\text{O} & \rightarrow \text{HNO}_3 \\
\text{NO}_3 + \text{hv} & \rightarrow \text{NO} \\
\end{align*}
\]

The vertical concentration profiles of OH (Volz et al., 1981) and O₆ (Marenco and Said, 1989) and of the photolysis rates (Roeth, 1986) are held fixed at their diurnally averaged values. The rate constants used are those in Jet Propulsion Laboratory (JPL 1990).

The nitrogen compounds are removed from the atmosphere by dry and wet deposition. HNO₃ is deposited on all surfaces with a deposition velocity of 2 cm s⁻¹ (Huebert and Robert, 1985), NO₂ only on land surfaces with 0.5 cm s⁻¹ (Boettger et al., 1978). Wet deposition of HNO₃ occurs in two modes. One is associated with the fast vertical transport events which are thought to proceed through convective clouds removing half of the HNO₃ lofted by the event. Additionally, HNO₃ is removed with a time constant of 10 days at altitudes of 1-12 km and with a time constant of 2 days below 1 km.

The emission rates of NOₓ listed in Table 1 were calculated from Erhard and Drummond (1988) and Erhard and Drummond (1982). The emissions by lightning were coupled to the fast vertical transport by convective clouds allowing NOₓ-injection for transport events which exceeded 8 km over the continents and 4 km over the oceans. The total emission rate by lightning was adjusted to match the value given in table 1.

<table>
<thead>
<tr>
<th>Source</th>
<th>Emission Rate (10⁶ t N yr⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surface source</td>
<td>total: 8.63</td>
</tr>
<tr>
<td></td>
<td>fossil fuel burning: 7.45</td>
</tr>
<tr>
<td></td>
<td>Lightning: 0.29</td>
</tr>
<tr>
<td></td>
<td>Aircraft, civil: 0.081</td>
</tr>
<tr>
<td></td>
<td>Stratosphere: 0.037</td>
</tr>
</tbody>
</table>

The aircraft emissions were derived from an average emission index of 10 g NO₂/kg fuel and a fuel consumption in 1984 of 113*10⁶ t fuel/yr (Nuesser and Schmitt, 1990). The vertical and longitudinal distribution was adopted from the Climatic Impact Assessment Program (CIAP, 1975).

3. RESULTS

The model was allowed to cycle three times around the globe to assume steady state. The zonal two dimensional distribution of NO and two sequences of vertical profiles of NO and NOₓ are shown in figures 4 and 5. The contributions of the various sources in figure 5 were derived by model runs in which the individual source was switched off. Both figures demonstrate the development of a maximum in NO concentration at 10 km altitude as the air column passes over strong continental source areas. This maximum is a result of high surface concentrations carried upward by the fast vertical transport process. When the air column moves from a high source area out over the ocean (figure 5b), NO and NOₓ are very quickly removed from the lower troposphere, but remain in the upper troposphere due to their long lifetime of about 10 days at those altitudes.

These results were tested against experimental results from Drummond et al. (1988). Figure 6 shows the comparison of the model predictions for the vertical NO-profiles at 50°W, 40°-50°N and an average of the NO measurements between 50°N and 40°N about the south bound leg of STRATOZ III. The two profiles agree surprisingly well indicating that all four sources are needed to produce the high NO-concentrations observed in the upper troposphere.
Fig 5: Calculated vertical mixing ratio profiles of NO and NO$_y$.
 a: over continental western Europe
 b: over the western North Atlantic
4. CONCLUSIONS

Despite its simplicity the model used is capable of obtaining results similar to the salient features of the observed vertical NO-profiles at 40°-50°N in June 1984. In particular, it reproduces the high values and relative maxima in the vertical NO-profiles in the upper troposphere over coastal areas. The reason for this may be that for these latitudes and this time of year processes are indeed simple. The model facilitates a first examination of the contributions from various sources to the NO$_x$-concentration in the upper troposphere. Aircraft emissions which contribute on average about 30% and NO$_x$ lofted from the planetary boundary layer are the major sources of NO$_x$ in the upper troposphere at northern mid-latitudes during summer. The quantitative conclusions are of course uncertain. They depend on the assumed source strength which have errors of the order of a factor of 2. Work which has been done by using an improved 2-D model showed that the qualitative and quantitative conclusions given here did not depend on the assumption of vertically uniform wind speed or on the lack of horizontal eddy diffusion.

REFERENCES

Boettger, A., D.H. Ehhelt and G. Gravenhorst
KFA Juel Bericht 1558, p 1-96, Forschungszentrum Jülich, 1978

Climatic Impact Assessment Program (CIAP) 1975,
Monograph 2, Rep. DOT-78T-75-52,

Drummond, J.W., D.H. Ehhelt and A. Volz
J.Geophys.Res. 93, 15631-15645, 1988

Ehhelt, D.H. and J.W. Drummond
Proceedings of NATO Advanced Study Institute, Corfu, Greece,

Ehhelt, D.H. and J.W. Drummond

Ehhelt, D.H., F. Rohrer and A. Wahner
J.Geophys.Res. 97, 3725-3738, 1992

Houghton, D.D.
Handbook of Applied Meteorology, John Wiley, New York, 1985

Huebert, B.J., and C.H. Robert
J.Geophys.Res. 90, 2085-2090, 1985

Jet Propulsion Laboratory (JPL), Evaluation 9, JPL 90-1, 1990

Mareno, A., and F. Said
Atmos.Environ. 23, 201-214, 1989

Neeb, H.-G., and A. Schmilt

Roeth, E.P.
KFA Juel Bericht 2098, p 1-171, Forschungszentrum Jülich, 1986

J.Geophys.Res. 66, 5163-5171, 1961