

Global modelling of atmospheric trace gases: Application of a global three dimensional model

T.H.P.The, D.L.Veenstra, J.P.Beck

RIVM-LLO, P.O.Box 1, 3720 BA Bilthoven, The Netherlands

Abstract

The global MOGUNTIA model was applied in two studies: 'Aspects of atmospheric methane' and 'Effects of aircraft emissions'. Results of both studies with respect to the 'European Renaissance' and a 'Joint Implementation' scenarion will be presented.

Methane plays a key role in the atmospheric radiative balance and chemistry. Per mass unit it is 58 times more effective than CO₂. In a chemical sense is is the primary sink for OH and it therefore detemines the oxidising capacity of the atmosphere to a large extent. We studied model results from 3 methane scenarios representing a uniform regional growth, a joint implementation option, and the European Renaissance assumptions.

Previous studies on effects of aircraft emissions have indicated an increase in the upper tropospheric ozone concentration of 5 - 12% due to aircraft emissions. Since ozone at the upper troposphere level is an effective greenhouse gas, the atmospheric radiative balance may be (further) disturbed by this process. The additional amount of ozone at the cruising altitude is modelled to result in a radiative forcing of 0.03 - 0.07 W/m².

We assessed the importance of homogeneous chemical processes in aircraft plumes before large scale mixing of the emissions occurs. The exhaust-plume-model developed resulted in a parameterisation of the sub-grid plume effects which was finally implemented in the global MOGUNTIA model. The parameterisation is currently a conversion factor showing a 60% conversion of NO_x into NO_y when the plume reaches the grid dimensions of MOGUNTIA. Preliminary results with the the global model show that this conversion results in a lower ozone production (6%) than due to unconverted emission fields (8%).

1. Introduction

The broad objectives of this project are the production of the 'base-document CH₄' and a document on aircraft and air pollution, both also by Environment Ministry order. In order to make the necessary scenario runs and assessments we put into operation a global three-dimensional model (the MOGUNTIA model) at RIVM. With this MOGUNTIA model the relations between emissions and concentrations of trace gases relevant to atmospheric chemistry on a global scale are studied. In this extended abstract both the work on aircraft emissions and on methane will be described.

2. Aspects of atmospheric methane

Methane (CH_4) plays a key role in the atmospheric chemistry and radiative balance. The actual contribution to the *increase* of global warming or so called climate forcing cannot be disregarded. It has been estimated that the climate forcing during the past decade, caused by CH_4 together with CFCs and N_2O was nearly as large as that for CO_2 . Per mass unit, CH_4 is 58 times more effective as a greenhouse gas than CO_2 . This figure does not entirely express the contribution to the greenhouse effect due to increased emissions, because the lifetimes are not taken into account. Obviously, long-lived components contribute more than shorter-lived ones. Therefore, another parameter, known as the Global Warming Potential (GWP) was introduced. The GWP is defined as the time-integrated commitment to climate forcing of a kilogram of gas relative to a kilogram of CO_2 . The GWP values of CH_4 are 35 after a 20-years period, and 11 after a 100 years period.

Methane is the most abundant trace gas after CO_2 and opposed to the latter gas it is chemically reactive. As a result CH_4 together with CO is, even though it is not extremely reactive, the primary sink for hydroxyl (OH). These OH-radicals play a key role in atmospheric chemistry. Hydroxyl, O_3 and H_2O_2 are the most important oxidants in the atmosphere. Their total atmospheric burden is called the oxidising capacity of the atmosphere. They react with several other trace gases and radicals and control their concentrations. Since OH reacts in large quantities with CH_4 , a further major increase in atmospheric CH_4 could lead to a reduction of the atmospheric oxidising capacity. Furthermore, oxidation of CH_4 is an important source of formaldehyde (HCHO). About 810 Tg CO per year comes from the oxidation of CH_4 , compared to 1930 Tg/y from other sources Logan et al., 1981.

The effects of CH_4 in the atmosphere are not limited to the troposphere but also extend to the stratosphere. About 50 Tg CH_4 per year enters the stratosphere, mostly through the inter-tropical convergence zone (ITCZ). Oxidation of CH_4 in the stratosphere leads to the formation of HCl, acting as a reservoir of Cl, considered to be responsible for the destruction of stratospheric ozone. On its turn stratospheric O_3 determines the amount of ultraviolet radiation entering the troposphere. At higher altitudes, the greenhouse gas properties of methane are causing an additional cooling of the stratosphere, as methane acts as a radiator. As stratospheric CH_4 dissociates, water vapour is produced. Methane, thus becomes a major source of stratospheric water vapour, since there is little direct transport of water vapour from the troposphere into the stratosphere.

We studied three different CH_4 emission scenarios:

- 1- a uniform regional emission growth, in all source categories and regions minus 0.5%/year;
- 2- Joint Implementation assumptions: A focus on Non_OECD countries, where options for emission reduction are cheapest; all sources and regions differentiated;
- 3- the European Renaissance scenario: reference case; all sources and regions different.

Furthermore, we compared our results to the TM2 model (Hein, 1994). The results will be discussed extensively in our final NRP report. Briefly, a comparison of the results of scenarios 1 and 2 shows that CH_4 concentrations are, after 10 years simulation, more than 7% lower in the northern hemisphere in the Joint Implementation case. Ozone is about two percent lower in the global atmosphere and the OH concentrations are about three percent higher.

3. Aircraft emissions

3.1 Considerations with respect to aircraft emissions

Aircraft emissions affect the chemical composition of the atmosphere in several different ways, depending on flying altitude and location. The conventional subsonic fleet grew extensively in the 1970s and 1980s and is expected to grow by a further 100% during the next two decades. This has led to several model studies focusing on effects of aircraft emissions on ozone in the upper troposphere (Hidalgo and Crutzen, 1977; Isaksen, 1980; Derwent, 1982; Ehhalt *et al.*, 1992; Beck *et al.*, 1992). These studies have predicted an increase in upper tropospheric ozone levels due to emissions of NO_x varying between 5 and 12%. The aircraft-induced changing ozone profile could have major implications for the atmospheric radiative balance, since ozone is an effective greenhouse gas at the tropopause level. Model calculations show that a radiative forcing of 0.04 - 0.07 W/m^2 may result from the predicted additional amount of ozone at the cruising altitude (Mohnen *et al.*, 1993; Fortuin *et al.*, 1994).

The previous modelled estimates of the ozone increases probably suffer from considerable errors due to uncertainties in the magnitude and distribution of emissions. Furthermore, a number of simplifications were made in the representation of aircraft emissions in atmospheric chemistry models.

Firstly, in these studies it was assumed that the present subsonic fleet emits all exhaust gases below the tropopause. However, new estimates predict that a considerable part (25 to 50%) of the emissions from the subsonic fleet are deposited directly into the lower stratosphere. Obviously, the effects of emissions in the lower stratosphere will be different from those in the upper troposphere.

Secondly, in the previous work it was assumed that immediate large-scale mixing of all emissions occurs. For this reason, the impact of flight corridor effects with the occurring specific chemistry has not been assessed. Again, in the previous work, the chemical composition of the source gases during large-scale mixing is considered identical to that of the emission at the tail pipe. Model studies of the first 4 seconds of the lifetime of a plume show that not more than 1% of the emitted NO_x underwent conversion to HNO_3 . However, this does not exclude that larger conversions from NO_x to NO_y will be found if a longer period of the lifetime of a plume is modelled.

Thirdly, another process omitted is the possible involvement of non-linear reactions involving NO_y species and heterogeneous chemistry occurring on ice particles in the contrail. The response to emitted NO_x is expected to change if heterogeneous chemistry occurs. The NO_x - O_3 chemistry is very non-linear and results in a greater ozone production per unit of NO_x for lower NO_x concentrations (Liu *et al.*, 1987). A part of the emitted NO_x had probably already been converted to NO_y before the plume reached grid dimensions. Therefore the ozone production caused by the NO_x emissions may be overestimated in the older work.

Focusing on the considerations mentioned above, in our work we assess the importance of the chemical processes in an aircraft plume in the upper troposphere before large-scale mixing occurs. Therefore an exhaust plume model is developed and the plume model study is expected to result in a parameterisation of the sub-grid, in particular $\text{NO}_x \sim \text{NO}_y$, chemistry of the plume. Using this parameterisation the global NO_x emission fields of aircraft can be translated into new processed fields of effective emissions, in which the sub-grid effects have been taken into account. These new emission fields were used as input for a MOGUNTIA study on the effects of aircraft emissions at cruising altitudes on the global atmosphere.

3.2 The aircraft exhaust plume model

The model consists of two sets of differential equations representing the mass balance equations in and outside the exhaust plume. The FACSIMILE package (Curtis and Sweetenham, 1985), which applies a version of Gears' method, has been used to calculate the arithmetic solutions of the chemical equations.

The chemistry of the plume is calculated using a set of reactions given by Beck *et al.* (1992). This set consists of 42 species, 73 gas-phase reactions and 16 photolytic reactions. Background concentrations are based on the measurements of the STRAT0Z III campaign (Drummond *et al.*, 1988). Currently, the set of reactions does not contain any heterogeneous reactions, but it will be extended with heterogeneous chemistry in a short while. Also, for extension to the lower stratosphere the tropospheric chemistry will be replaced with a stratospheric reaction set.

The diffusion part of the model was based on the theory of Gelinas and Walton (1974), which states that the kinetics and chemistry of an exhaust plume can be described separately. In this approach, the plume is described by a box of variable volume, which always encompasses the exhaust species. This 'gaussian' approach seems justified since detailed model exercises have shown that interaction of airplane wing vortices with the exhaust trail is of secondary importance in the dispersion of the plume (Louisnard *et al.*, 1994). The modelled dimensions of an exhaust plume are about 3 km high and about 100 km wide after 24 hours expansion.

The O₃ concentration in the plume rapidly decreases after the injection of emissions, in particular caused by the reaction of emitted NO with O₃. The O₃ concentration is re-established to a level similar to the background concentration in about an hour due to mixing with background air. Subsequently, some production of ozone occurs and the resulting concentration in the plume is somewhat higher than the background level for a few hours.

Only minor chemical changes of emissions during the first 4 seconds of the lifetime of a plume - 'the vortex regime' - were found by Louisnard *et al.* (1994). However, our work predicts that if the plume is followed for several hours the high NO_x concentration occurring immediately after injection decreases rapidly due to mixing with background air. A part of the NO_x is also converted to NO_y components. At first, HONO is formed; this then undergoes rapid photolytical decomposition. Subsequently, HNO₃ is formed and after some hours also HO₂NO₂, from which formation was initially inhibited through a lack of HO₂ (caused by the reaction of NO with HO₂).

The calculated NO and NO₂ concentrations in the plume are quite similar to measured amounts in a young plume (Arnold *et al.*, 1992). But the measured perturbations of HONO and HNO₃ are 10 to 20 times greater than calculated in the exhaust plume model. A reason for this discrepancy may be our limited set of homogeneous reactions.

The exhaust plume model was shown to be a good instrument to study plume chemistry. Generally speaking, maximum perturbations (caused by the emission of an aircraft) of the plume concentrations exist only in the first few hours after injection and disappear after about a day. We found that these perturbations strongly depend on the amount of emission, hour of the day and the season when injection takes place, as well as the background concentrations.

3.3 Translation of aircraft emission fields

The parameterisation of sub-grid chemistry was initially in the form of a simple conversion factor: an emission of x molecules of NO_x is converted after expansion to grid-dimension into y molecules of NO_x and z molecules of NO_y (HNO₃, HONO, HO₂NO₂, N₂O₅,

PAN), with $x = y + z$. The transformation of NO_x into NO_y components depends strongly on the altitude of emission and background concentration (e.g. NO_y , OH and HO_2) components, and less on the time of injection (time of day, season) and amount of emission. The higher the altitude of aircraft emissions the slower the conversion of NO_x into NO_y . Here the lower temperatures are the primary reason, followed by the lower OH and HO_2 concentrations.

As a first parameterisation, it is assumed that after one day the exhaust plume has reached grid dimensions and the plume model predicts that in the new emission fields the NO_x emissions should be differentiated into NO_x (40%) and NO_y (60%) components, the latter group divided in specific components contributing to NO_y .

3.4 MOGUNTIA-model runs; effects of aircraft

The processed emission fields are used as input for the MOGUNTIA model. MOGUNTIA is a global tropospheric 3D-model incorporating transport and chemistry and it was originally developed at the Max Planck Institute in Mainz (Zimmermann, 1988). It has grid dimensions of $10^0 \times 10^0 \times 100$ hPa, starting at the surface and extending to the 100 hPa level. The dynamics of the model are calculated with ECMWF fields, assuming turbulent mixing. The reaction set used for the chemistry is from Dentener (1993).

The calculations are performed with aircraft emission fields and corresponding anthropogenic NO_x emission fields based upon data of McInnes and Walker (1992) and Müller (1992), processed by Olivier (1994). Preliminary results of unprocessed 1990 emission fields show an increase in the ozone concentration at the location of the North-Atlantic flight corridor at the cruising altitude of 8%. The processed emission fields show an increase of about 6% at the similar location.

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