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Atmospheric effects of high-flying subsonic aircraft: a catalogue of perturbing influences

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KNMI

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ATMOSPHERIC EFFECTS OF HIGH-FLYING SUBSONIC AIRCRAFT; A CATALOGUE OF PERTURBING INFLUENCES

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PREFACE

Only recently, the scientific community has become to see air pollution by subsonic aircraft as potentially a serious threat to the environment. This may be illustrated by the fact that in the most recent bi-annual assessment of the ozone layer of the WMO (WMO 1991), the topic has for the first time been taken into account in a separate section.

In line with the attention paid by scientists to the topic, policymakers have also started to address the issue, both on a national and on an international level. In the Netherlands, pollution by aircraft is a government issue since the end of the eighties. In 1991 a covenant between several local, regional and national authorities was agreed on, in order to deal with possible environmental effects of an expansion of the Dutch main airport Schiphol, Amsterdam. In the covenant, a Dutch governmental memorandum was announced describing possible future government policies on air pollution related to air transport, based on a thorough research programme.
Apart from propositions for future government policies, the memorandum should provide policymakers with a comprehensive overview of all activities associated with air traffic influencing air quality. This overview will be based on several reports by specialists, each covering one aspect of air pollution by aircraft and published as part of the research programme.
The governmental memorandum is to be published by the end of 1994; the research programme, funded by the Dutch Ministries of Environment, Transport and Defense, should be executed in the period 1992 to 1994.

The underlying report is one of those specialists' reports. It focusses on the atmospheric effects of subsonic aircraft during the cruise operating mode. Thence, it will not deal with effects around airports, nor will it take into account effects of a possible supersonic fleet in the future.

1 INTRODUCTION

why this report?

Forecasts of global air transport demand predict an average annual growth rate between 5 and 7% until 2010. This implies that in 2005 the number of passenger-kilometres performed will at least be doubled compared to the situation in 1990. Since aircraft emit components which are both greenhouse gases and ozone precursors, this forecast of a substantial world-wide increase in air traffic aroused new interest, from both scientists and policymakers, in the atmospheric effects of aircraft exhaust. But although a great deal of work in both fields has been done on investigating -and how to deal with- the effects of air traffic at and around airports and of future supersonic flights in the stratosphere, little is known about the effects of subsonic aircraft during cruise.

This reports' objective is to contribute to fill this gap. By merging the results of many studies which directly or indirectly aimed at elucidating one aspect of the complex problem of how high-flying aircraft influence atmospheric processes, an attempt is made to give the readers some insight in, and sense of the seriousness of, the effects of high-flying aircraft.

what makes it worthwile to study high-flying aircraft independently?

For reasons of fuel economy[1] and flight time, aircraft during cruise[2] fly at or near tropopause level.

[1] When allowed to fly in and above the tropopause, the attained savings for 4 actual Lufthansa flights varied between 5.7-7.5% compared to the situation when tropopause contact is not permitted (Reichow 90);
The increase in fuel consumption per flight-kilometre when flying at sea-level is in the order of a factor two compared to the same flight at tropopause level as defined by the Standard Atmosphere (Peper 93).

[2] In this study, both "cruise (conditions)" and "high-flying" apply to aircraft flying at pressure levels corresponding to altitudes of between 8 and 15 km as defined by the Standard

From the viewpoint of climatic change, emissions of radiatively and chemically active trace gases at this level could be of particular interest because the Earth's radiative balance, which drives the climatic system, is most sensitive to changes in the concentration of radiatively active gases at tropopause level. From the viewpoint of ozone change, emissions of chemically active trace gases at tropopause level are of special interest also. Models have shown that the cross-over point between ozone destruction and ozone formation, both due to aircraft emissions of nitrogen oxides, is situated at this level. Emissions of these gases result in net formation of ozone below this level; they result in net destruction of ozone above this level.
Although many studies have yet been done on the effects of aircraft emissions, no study could be found aimed at elucidating the effects of aircraft during cruise in an integral approach, i.e., an approach in which both effects on climate and ozone are dealt with. This study has been initiated to meet this need.

how has this study been carried out?

The purpose of the underlying study is twofold. The first objective is to present an overview of the existing knowledge of possible atmospheric effects of emissions at cruise altitude. Three different approaches to the problem constitute the overview. They will be presented in separate sections:
1) In Chapter 2 will be indicated, by means of first-order calculations, whether the quantities of aircraft exhaust components emitted are such that they have the potential to perturb their respective background concentrations in the atmosphere significantly[3].

Atmosphere.

[3] In this study, when discussing calculated changes or perturbations in a certain variable with respect to an unperturbed situation, the term significant is used if a change or perturbation exceeds the, admittedly subjective, limit of 1% when steady-state is reached.

2) Possible consequences of emissions of nitrogen oxides for global ozone distributions will be obtained from published results of simulations with chemistry-transport models. They will be presented in Chapter 3.
3) Climatic consequences are obtained from published results of both detection studies and sensitivity studies with simple and more elaborate climate models. They are presented in Chapter 4.
Each approach constitutes a section largely independent of the other sections: the conclusion of each section can, thus, be considered as independent from the conclusions of the other sections. The only features the three different approaches have in common is that they all consider atmospheric effects of high-flying aircraft and that the situation in 1990 is considered as the base-case in each section.
The second objective is to present a general conclusion with respect to the seriousness of the possible atmospheric effects of high-flying air traffic. For this, the conclusions of the individual sections are merged and put into perspective in the general discussion and conclusion.

a reminder

This assessment study has for the largest part been based on studies by research institutes world-wide of which the results have been published in peer-reviewed journals. This implies that it stresses those topics of which information was readily and in large quantities available. This could have as result that problems which can be studied more easily get more weight -because more publications are available- than topics which are much harder to study. However, it cannot be said a priori that this approach gives a good account of the processes which in reality are most important. It is hoped that the reader keeps this in mind while going through the following.

2 ESTIMATES OF PERTURBATIONS IN BACKGROUND MIXING RATIOS DUE TO 1990 EMISSIONS OF HIGH-FLYING AIRCRAFT

2.1 ESTIMATES OF FUEL BURN DURING CRUISE

2.1.1 INTRODUCTION

Aircraft during cruise emit various materials into the atmosphere. Of these materials, engine exhaust components have the largest relative impact on atmospheric background concentrations, while contributions from leakage of fluids (hydraulic and sanitation), fuel dump, chipped paint, and erosion of other components are apparently trivial (Miake-Lye et al. 92).

In this section will be calculated how emissions of these exhaust components influence the concentrations of these components in the unperturbed atmosphere, i.e., their background concentrations. The (change in) atmospheric concentration is represented by a dimensionless number: the volume mixing ratio. The calculation will be done for different atmospheric layers or volumes of air. To what extent this ratio will be influenced is dependent on the magnitude of the emissions as well as on how long a component remains in the atmosphere. The latter is given by the residence time: the average time spent in a given part of the atmosphere by the molecules of a given gas. This is determined by both chemical and transport processes. In general, if a species does not react, i.e., it is chemically inert, transport processes will determine its residence time. On the other hand, if a chemical is reactive or can easily be dissociated by solar radiation, its residence time will be largely determined by its rate of reaction or the level of irradiance.

In reality, trace gas background mixing ratios as well as their residence times differ to a large extent. Moreover, for many trace gases both variables show strong gradients with latitude, longitude and altitude.

For a given emission, these differences in residence times and background mixing ratios result in significant differences in the perturbation of background mixing ratios, relatively as well as absolutely. In addition, for many dynamical and chemical processes there is a strong transition at the boundary between the troposphere and the stratosphere, the so-called tropopause. One of the effects of this change in characteristics which is important for this section, is that for some trace gases the gradient in background concentration in the vertical shows a sudden and marked change. Therefore, one should like to know which part of the cruise emissions is injected to the upper troposphere, and which part is emitted at tropopause level or directly above, i.e., in the lower stratosphere.

In this chapter, aircraft emissions in different parts of the atmosphere will be calculated for the situation in 1990. For this, the following values will be used as a starting point: in 1990 176 Tg of aviation fuel has been burnt world-wide. 20-25% of this amount has been burnt by military aircraft (Wuebbles et al. 93, Egli 90) which leaves 75-80% for all civil aviation including general aviation and airliners carrying freight and mail. For convenience, the distribution of flights by military aircraft as well as their emissions in a relative sense, both in space and time, will be considered as indistinguishable from the distribution of flights and emissions of civil aircraft.

2.1.2 GLOBAL FUEL BURN DURING CRUISE

In the following, it is assumed that 65-70% of the 1990 aviation fuel used has been burnt during cruise. From an emission inventory presented by NASA (Wuebbles et al. 93), accounting for 76% of 1990 fuel used, has been calculated that about two thirds of world-wide fuel use in 1990 occurred during cruise. From the emission inventories presented by McInnes and Walker (92) and Boeing (Baughcum et al. 93), accounting for 52% of 1989 and 1990 fuel used, respectively, has been calculated that nearly 70% of global aviation fuel use occurred during cruise.

2.1.3 FUEL BURN DURING CRUISE IN DUTCH AIRSPACE

As a special case, flights above the Dutch territory have been considered. From a study by Williams (90) can be calculated that about 45% of aviation fuel used in Dutch airspace in 1989 has been burnt during cruise. This is about 20% lower than the global average, which is no surprise considering the fact that relatively many landing and take-off cycles take place in the Netherlands due to the presence of Schiphol airport and that relatively many short-range flights occur which do not reach 8 km in altitude. About 40% of fuel used above the Netherlands has been burnt during cruise above FL 300 and highest Flight Level reached by overflying aircraft was FL 330. It is assumed that the situation in 1990 was the same.

2.2 ESTIMATES OF STRATOSPHERIC FUEL BURN

2.2.1 GLOBAL STRATOSPHERIC FUEL BURN

In the following, it is assumed that 30-50% of 1990 aviation fuel used has been burnt during cruise in the stratosphere. This estimate, which is 10-20% higher than figures found in other studies, is based on recent studies in which for the first time the percentage of fuel burnt in the stratosphere is calculated from real flight data with information about local tropopause heights.

Most results apply to the Northern Hemisphere only, but as nearly all long-range cruise occurs in that region -93% according to Ko et al. (92)- this will not introduce too large an uncertainty.

Ko et al. (92) used zonally averaged climatologies and estimated the fuel use in the stratosphere for the 1987 subsonic fleet to be 48% of total fuel use in the Northern Hemisphere, assuming an average flight altitude of 11k[4]. Due to the variability of the mean tropopause altitude with season, monthly percentages varied between 16% and 72% (see also Section 2.3).

Based on limited data about tropopause altitudes in January and July for the 1970 atmosphere, Schumann (92, 93) estimated that more than 50% of air traffic flying north of 50N[5] at 250 hPa, i.e., at Flight Level 340[6], occurs within the stratosphere. As flight levels for long-range flights are in general assigned in advance, captains of airplanes do not have the possibility to change their preference for a flight level during flight. Thus, results from statistical studies correlating flight levels and tropopause heights can be considered as reliable.

[4] k = kilometre in altitude as defined by the Standard Atmosphere (ICAO 64)

[5] N = degrees Northern latitude

[6] Flight Level can be defined as the altitude at which an aircraft is flying, in hundreds of feet, according to the cockpit reading of ambient atmospheric pressure, which is translated to flight altitude according to a standardized atmosphere: the Standard Atmosphere (ICAO 64).

As the estimates for total and stratospheric fuel burn, both during cruise, are made independently, this leaves a range for tropospheric fuel burn during cruise from 15-40% of all fuel used by aircraft world-wide.

2.2.2 STRATOSPHERIC FUEL BURN IN DUTCH AIRSPACE

1990 radiosonde data, for 12 GMT for De Bilt, which were taken as representative for the whole Dutch airspace, have been analysed. From these data has been calculated with data presented above that about 5 to 10% of high-flying air traffic in Dutch airspace occurs in the stratosphere (Figure 1.1). This is much lower than the global average.

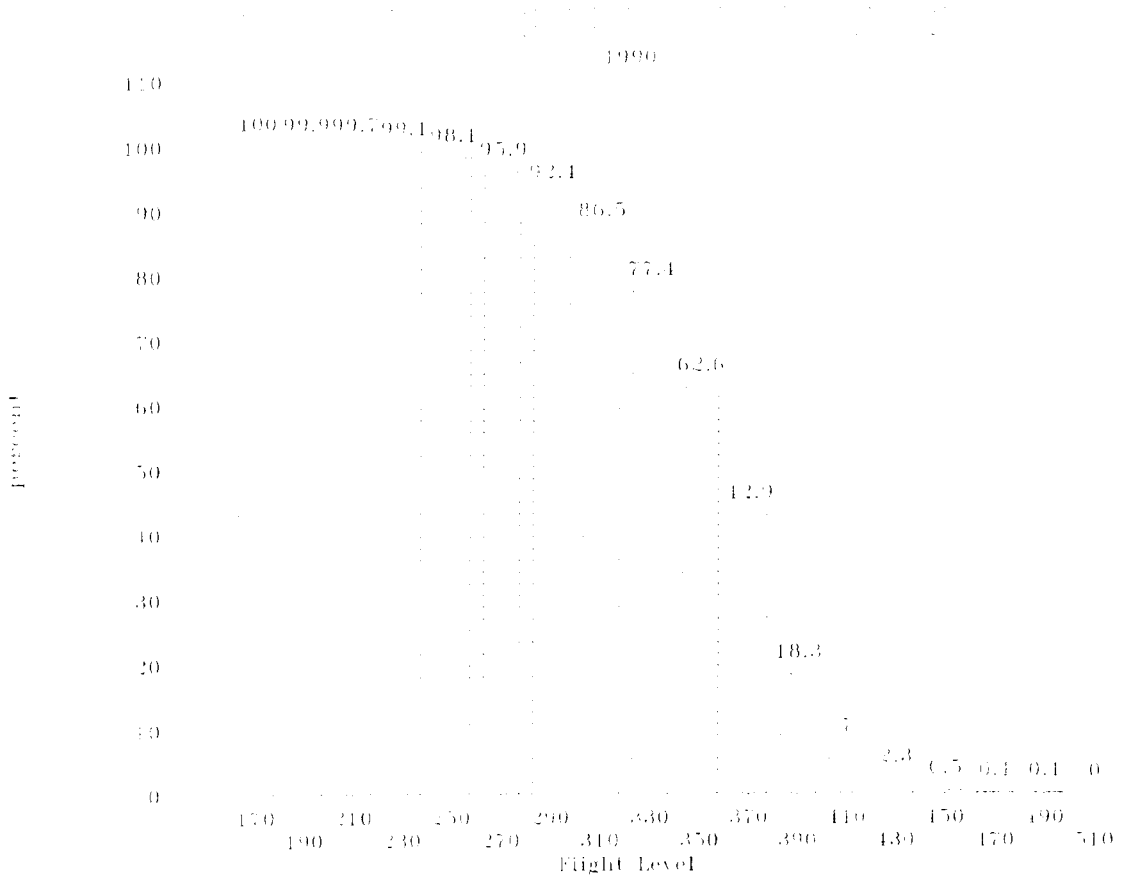

Figure 1.1 The 1990 fraction of planes in the troposphere if all planes would have flown at the same Flight Level (results presented by courtesy of Marc Allaart, KNMI)

2.3 FUEL BURN IN THE NORTH ATLANTIC FLIGHT CORRIDOR

2.3.1 INTRODUCTION

Increases in background mixing ratios in the North Atlantic Flight Corridor (NAFC) and in the exhaust plume of a jet aircraft flying in the NAFC have also been considered. The NAFC is the atmospheric region between western Europe and North America in which most aircraft connecting these two areas fly. The NAFC is bounded by a limited economical height band {Flight Levels 310 (9.5k) to 390 (11.9k) (ICAO 88)}, where aircraft fly most efficiently because of thermodynamic considerations (low ambient temperatures enhance the efficiency of combustion engines), low drag because of low ambient density and low turbulence due to the the prevailing meteorological situations. The NAFC is situated between about 45-60N. Its volume can approximately be represented by a box of 1000 km x 5000 km x 2k. With a mean air density of 0.35 kg/m^3, the NAFC accounts for 3.5 x 10^{18} g or about 0.07% of the mass of the Earth's atmosphere.

2.3.2 1990 FUEL BURN IN THE NAFC

Hoinka et al., cited in Schumann (93), analysed the distribution of all air traffic crossing the North Atlantic during the period from 1989 to 1991. They found that on average 512 aircraft per day (sum of both directions) passed that region. Fransen (93) calculated a fuel use in the NAFC of 8.5 Tg for 1992. This amount has been calculated with the help of fuel use figures given in Aircraft Operation Manuals for many types of aircraft, as provided by the aircraft industries. These should be considered as reliable. Information of flight movements in the NAFC (602 flights daily) was extracted from Oceanic traffic data recorded by Shanwick Oceanic Area Control Centre. Assuming the same aircraft mix for 1990 compared to 1992, but less flights per day -512 instead of 602- a fuel burn of 7.2 Tg has been calculated for the NAFC in 1990, or 4% of total 1990 aviation fuel use.

KNMI calculated daily tropopause levels at 10W-55N for 1992 from data provided by the European Centre for Medium-Range Weather Forecasting (ECMWF); from these data has been estimated by Fransen (93) that 46% of 1992 NAFC fuel use occurred in the stratosphere.
Hoinka et al. have also correlated their data set with tropopause altitudes as calculated from ECMWF data; they estimated that about 44% of 1990 NAFC fuel use has been burnt above the tropopause in the stratosphere. However, as mean tropopause altitudes have a seasonal dependence, stratospheric fuel burn exhibited large variations; in February 1990, for example, the stratospheric part reached 75%.
For flights between Frankfurt and New York JFK, Schumann (92) calculated an average of 30% of the total flight time above the tropopause, based on a limited number of cases.

2.3.3 FUEL BURN IN THE AIRCRAFT EXHAUST PLUME

For the exhaust plume, it is assumed that 10 g fuel per metre is burnt during cruise in the NAFC, a typical value for a Boeing 747 at cruise, and that its products of combustion are evenly distributed over a channel with an aereal cross-section of 1000 m^2 (Arnold et al. 92). This should correspond to the situation a few seconds after the plane has passed {10000 m^2 is assumed by Schumann and Reinhardt (91); 3600-5400 m^2 is calculated by Gerz cited in Schumann (93) for the situation reached within about 10 seconds, 21000-52000 m^2 for the situation reached within 3 minutes; 100 m^2 is assumed by Danilin et al. (92)}.

2.4 EMISSIONS BY HIGH-FLYING AIRCRAFT

2.4.1 INTRODUCTION

The major gaseous components of aircraft exhaust consist of products of combustion {carbon dioxide (CO_2), water (H_2O) and sulfur dioxide (SO_2)}, side products of combustion {principally nitric oxide (NO), but also nitrogen dioxide (NO_2)}, and products indicating combustion inefficiencies {carbon monoxide (CO), hydrocarbons (HCs) and soot[7] (C)}.

The emission of a specific trace gas X per unit fuel used is generally represented by an Emission Index (EI), defined by the International Civil Aviation Organization (ICAO) as the mass of X produced in g divided by the mass of fuel used in kg (ICAO 80). The Emission Index is, thus, a dimensionless number. By convention, it is derived from sea-level measurements for each flight stage, for instance take-off or approach, separately. In this context, a flight stage is defined by a certain associated thrust[8] level only, e.g., take-off indices are measured at 100% thrust setting, approach indices at 30% thrust setting.

With respect to CO_2, H_2O and SO_2, the combustion of kerosine can be considered to occur stoichiometrically, i.e., all substances involved in the chemical process react or form in the exact proportions which follow from theory. The EIs for CO_2, H_2O and SO_2 are thus dependent on the chemical composition[9] of the fuel only, and independent of engine thrust and ambient conditions.

[7] As the radius of soot particles, which consist of elemental carbon, varies between 0.01-0.1 μm (Pitchford et al. 91, Turco 92), their transport behaviour can be considered as gaseous.

[8] thrust; force directed forward in a jet-engine as a reaction to the ejection rearward of gases. By definition, 100% thrust is the maximum power, expressed in Newtons, an engine can deliver under ambient conditions. This has as consequence that, because of differences in ambient conditions like air density and temperature, 100% thrust high in the atmosphere means much less power than 100% thrust at ground level.

[9] Major changes in fuel composition become necessary only to increase its thermal stability, which becomes necessary for aircraft flying with speeds of around Mach 3 (Miake-Lye et al. 92).

Moreover, as the different types of aircraft considered in this study use fuels which can be considered to have the same chemical composition, there is no uncertainty assigned to these indices. The EIs for NO and NO_2, and for CO, HCs and C are dependent on thrust, ambient conditions as well as on type of propulsion (e.g., turbo-jets, turbo-props or piston-engined option) and engine. For this reason, they will each have a broad range.

2.4.2 CO_2, H_2O AND SO_2 EMISSIONS

The CO_2 EI can be calculated from the oxidizable carbon content of the fuel, which is about 86% by mass (Shell cited in Schumann 92). From this, an EI of 3.15×10^3 is calculated.
The same line of reasoning can be followed for water emissions. The fuels covered in this study have an EI of 1.24×10^3 for H_2O.
The SO_2 EI is also a function of the fuel composition only; all fuel sulfur is believed to be emitted as SO_2 (Miake-Lye et al. 92). Because the maximum allowable mass percentage of sulfur in fuel is 0.3%, i.e., 3 g/kg (ICAO 81), the maximum SO_2 EI is theoretically 6. With reference to studies on fuel sulfur content cited in Olivier (91) and Schumann (92, 93), and to one of the few in-situ measurements in the exhaust trail of a jet aircraft during cruise (Rosen and Greegor cited in Hofmann and Rosen 78), a SO_2 EI of 1.1 can be considered as a reliable best-estimate.

2.4.3 EMISSIONS OF NITROGEN OXIDES

2.4.3.1 Introductory remarks

EIs for NO_x, i.e., NO and NO_2, found in literature have an extensive range. At the same time, it is hard to find accountable estimates of residence times for NO emitted at cruise altitude. These matters introduce large uncertainties when calculating the perturbations in background concentrations.

Because of the importance of (perturbations in) background concentrations of nitrogen oxides for both ozone and climate (for this, see chapters 3 and 4), uncertainties associated with emissions of nitrogen oxides will be dealt with in more detail here.

- NO_x measurements have been reported as the total mass of NO_2, plus the mass equivalent of NO oxidized to NO_2. Thus, NO_x is reported as the NO_2 gram equivalent of the total measured species, independent of the oxidation state, even though it is, typically, mostly NO near the exit of gas turbine engines[10]. Unfortunately, this convention is not always stated explicitly, generating some confusion in interpreting these numbers[11].

- For more accurate emission indices of nitrogen oxides, needed for models used in climate research, EIs from measurements under sea-level conditions, following the recommendations by ICAO (81), are translated into flight-level figures by means of empirical relationships. According to Lister (93a), predictions on NO_x emissions at flight level versus combustor inlet temperature by nine different research groups, using two different methods with, altogether, nine different parameters, gave a spread which ranged from 25% to a factor of 3 for different types of calculations.

- In general, EIs used are calculated with the measurement technique recommended by ICAO (81), i.e., a probe sampling technique. Several scientists have compared this technique with corresponding in-situ spectroscopic measurements of NO. The spectroscopic values were higher than the corresponding probe values by factors of 1.3 to 6 (Hidalgo and Crutzen 77).

[10]The NO_2 fraction decreases from 18% at idle to less than 5% at more than 30% engine power setting, according to Spicer et al., cited in Schumann (92).
[11]In this study, NO, NO_2, and NO_x emissions will be reported as gram equivalents of NO_2.

Although it is acknowledged that the more recent measurements done by ICAO are much more accurate (Lister 93c), published results of recent comparisons with values from spectroscopic techniques could not be found.

- Residence times for NO_x directly below and above tropopause level are difficult to determine. Model calculations show that the residence time at sea level varies from less than one to more than ten days depending on the NO_x background mixing ratio and the NO_x to hydrocarbon ratio (Liu et al. 87, Lin et al. 88). At tropopause level, NO_x mixing ratios as well as temperatures are much lower. This probably results in significantly larger residence times when NO_x is emitted at these altitudes.
As only one reliable estimate for the residence time of NO_x at tropopause level was found, figures presented in tables 2.4 and 2.5 have to be considered as uncertain.

These four points should be kept in mind when considering NO_x EIs and the possible atmospheric effects of NO_x emissions as calculated with models.

2.4.3.2 Emission Indices of Nitrogen Oxides
i) For cruise, NO_x EIs between 6.0 and 26 were found. Egli (90) proposes 15, based on 1984 figures, while Lufthansa estimates for their 1988 fleet span the range 6.0 to 16.4 (Nüßer and Schmitt 90). Ko and Weisenstein (93) recommend 20.7 for subsonic cruise, to be used in all models contributing to the second programme of the National Aeronautics Space Administration (NASA) on the atmospheric effects of stratospheric aircraft.
Only few global aircraft NO_x-emission inventories have yet been published. From figures given by McInnes and Walker (92), an average cruise EI of 9.9 can be calculated. From figures given by Wuebbles et al. (93) and Baughcum et al. (93), EIs of 10.2 and 11.7 have been calculated, respectively.

Though both inventories are till today the most detailed with respect to the spatial distribution of the emissions, their actual EIs are calculated with the help of empirical formulas with as input ICAO figures, of which the accuracy is uncertain.
Only recently, the first in-situ measurements of NO and NO_2 abundances in a young exhaust-trail of a jet aircraft at cruise altitude have been published (Arnold et al. 92). From measurements in the exhaust trail of a DC-9 airliner flying at 9.5k, 1k below local tropopause altitude, a NO_2 EI of 26 was calculated. Compared to ICAO estimates, this is indeed a high figure.
Giving a large weight to the measurements of Arnold et al. compared to the values from the three emission inventories, 18 was taken for the NO_2 cruise EI, but with a large uncertainty range of ± 8.
ii) For flying at cruise above the troposphere, an EI of 15 ± 5 has been chosen for this study. At higher altitudes the ambient pressure is lower, while, following the standard atmosphere, the ambient temperature remains at a constant value. As NO_2 EIs are essentially square-root dependent on combustor inlet pressure and exponentially dependent on combustor inlet temperature, this leads to lower NO_2 EIs. Using this dependency, McInnes and Walker (92) calculated for four different aircraft types that flying at 11.3k instead of 10.1k results in a weighed-mean decrease of the NO_2 EI of a little more than 11%. Analysis of the emission inventory by Wuebbles et al. (93) leads to the same results.
iii) Values given for the average EI over all flight stages vary from 10 to 21.7. An EI of 10 can be calculated for the 1990 situation from information presented by Kavanaugh (88). Wuebbles et al. (93) and Baughcum et al. (93) calculate 10.9 and 12.6, respectively, for their 1990 emission inventories; McInnes and Walker (92) 11.6 for their 1989 inventory.

Other estimates are significantly higher. Egli (90) calculated an EI of 18 ± 3, based on 1984 figures. Swissair (Egli 94) reports for its 1989 fleet an EI of 21.7. The difference could be explained by older engines having lower EIs for NO_2 than modern ones caused by lower combustion temperatures. Taking into account that the Swissair 1989 fleet is more modern than the 1990 average and that an EI of 18 has been chosen for cruise, accounting for 65-70% of global aviation fuel used, an EI of 18 has been opted for. Again, a large uncertainty range of ± 8 is attributed to the estimate as to account for the emissions that took place during idle, taxi and approach (generally lower EIs for NO_x and lower fuel flows during longer periods), and take-off and climb (generally higher EIs for NO_x and higher fuel flows during shorter periods).

2.4.4 EMISSIONS OF CARBON MONOXIDE, HYDROCARBONS AND SOOT

CO and HC emissions from aircraft are quantitatively less important; both global emissions and emissions by fossil fuel combustion are orders of magnitude larger (Table 2.3). Calculated perturbations of their background mixing ratios, presented in tables 2.5 and 2.6, reflect this: these are not significant.
Calculated perturbations for soot are relatively large, while information about non-aircraft soot emissions could not be found.

8 g CO per kg fuel is emitted averaged over all flight stages; 7 g/kg is emitted during cruise. These values are primarily based on information by Wuebbles et al. (93), from which 8.4 for all stages and 7.4 for cruise conditions has been calculated. From Baughcum et al. (93), 6.3 and 3.4 are calculated, respectively. 0.7-2.5 has been calculated for the Lufthansa 1988 fleet at cruise (Nüßer and Schmitt 90) and 1.1 for subsonic cruise (Ko and Weisenstein 93). For the EI averaged over all flight stages, 14.6 has been calculated by Johnson and Henshaw (91) and 1.16-5.15 for the Swissair 1989 fleet with an average of 3.31 (Egli 94).

4 is calculated for the Lufthansa 1988 fleet (Reichow 90). EIs found for CO have a large spread. Thus, a large uncertainty margin is taken.

Little information exists on the EIs for hydrocarbons. The average EI for all HCs combined is estimated as 2.5 for all cases considered in this study. This estimate is, again, primarily based on information presented by Wuebbles et al. (93), from which EIs of 2.6 and 2.7 have been calculated, respectively. From Baughcum et al. (93), 0.8 and 1.7 are calculated, respectively. Swissair estimates are 0.19-2.55 over all fuel used, with an average of 1.05 (Egli 94). Johnson and Henshaw (91) use 2.6, Lufthansa 0.05-0.7 for cruise conditions (Nüßer and Schmitt 90). Four hydrocarbons seem to occur in relatively large amounts: ethene ($H_2C=CH_2$, about 15% of all hydrocarbons emitted), formaldehyde ($H_2C=O$, about 15%), methane (CH_4, about 10%) and the exotic species hexamethylcyclotrisiloxane and octamethylcyclotetrasiloxane {$(-Si(CH_3)_2O-)_{3,4}$, together about 10%} (Shareef et al. 88). Ethene is included in tables 2.4 and 2.5 to give an idea about its relative importance, should it be emitted at altitude in the same relative abundance. Recent measurements indicate that methane is emitted in smaller amounts than those present in ambient air (Lister 93b): in other words, methane present in ambient air is effectively burned.

Soot EI estimates are not abundant: they vary from 0.007-0.03 for cruise (Lufthansa cited in Nüßer and Schmitt 90), to 0.014 (Lufthansa cited in Reichow 90) and an assumed EI of 0.1 as average over all fuel used (Pueschel et al. 92). From in-situ measurements in the exhaust plume of a jet aircraft during cruise at 2.6k, Pitchford et al. (91) calculated an EI between 0.001-0.03. Giving a large weight to these measurements, soot EI is estimated as being 0.02 for all cases considered in this study.

2.5 CALCULATED PERTURBATIONS IN BACKGROUND MIXING RATIOS

With reference to the scope of this study and in accordance with both the Standard Atmosphere of the ICAO (64) and the tropopause level as defined by the World Meteorological Organisation[12], in the following the region between 8-10.7k (FL 260-350) is defined as the upper troposphere and the region between 10.7-15k (FL 350-490) is defined as the lower stratosphere including the tropopause. The upper boundary of 15k might seem a little high considering the real distribution with altitude of current civil aircraft. However, with respect to the Standard Atmosphere, it can be read as about 4k above tropopause level. From this point of view, 15k is a realistic flight level.

In this section, six tables are given presenting the amounts of exhaust components which in 1990 were injected in the global atmosphere and the portions of these emissions which were injected during cruise (i.e., between 8-15k), during cruise in the lower stratosphere and in the upper troposphere, and during cruise in the NAFC and in the exhaust plume while flying in the NAFC. Values given are calculated by multiplying the annual fuel use in the atmospheric volume of interest, e.g., the lower stratosphere, with the emission index for a specific exhaust component for the flight stage which applies to the volume of interest. The resulting figure can be considered as the emission rate, i.e., the emission in units of mass per unit of time, which in this case is the year 1990.

Tables 2.1 and 2.2 present the absolute 1990 emissions for several trace gases due to all air traffic movements and to aircraft during cruise.

[12] The tropopause: the first level above 500 mbar at which the lapse rate drops below 2 K/km and remains below that value for at least 2 km.

Table 2.1: Global 1990 emissions of all air traffic combined

TRACER	EMISSION INDEX AVERAGED OVER ALL FUEL USED	TOTAL 1990 AIRCRAFT EMSSIONS
CO_2	3.15×10^3	554 Tg
H_2O	1.24×10^3	218 Tg
SO_2	1.1	0.19 Tg
NO_2	18 ± 8	3.2 ± 1.4 Tg
CO	8 ± 5	1.4 ± 0.9 Tg
All HCs	2.5 ± 1.5	0.44 ± 0.26 Tg
$H_2C{=}CH_2$	0.4 ± 0.2	0.07 ± 0.04 Tg
C	0.02	0.0035 Tg

Total fuel used by aviation in 1990: 176 Tg.

a: Emissions in Teragram, i.e. 10^{12} gram.

Table 2.2: Global 1990 emissions by aircraft flying at pressure levels corresponding to altitudes of between 8 and 15 km[a] as defined by the Standard Atmosphere

TRACER	CRUISE EMISSION INDEX	AIRCRAFT 1990 CRUISE EMISSIONS IN Tg	SHARE OF TOTAL 1990 AIRCRAFT EMISSIONS
CO_2	3.15×10^3	374 ± 14	65-70%
H_2O	1.24×10^3	147 ± 6	65-70%
SO_2	1.1	0.13 ± 0.01	65-70%
NO_2	18 ± 8	2.0 ± 0.9 *b*	61-63%
CO	7 ± 5	0.85 ± 0.65	40-65%
HCs	2.5 ± 1.5	0.30 ± 0.19	65-70%
C	0.02	0.0024 ± 0.0001	65-70%

a: This altitude range corresponds to the Flight Level range 260-490.

b: Note that for this value different EIs for cruise in the upper troposphere and lower stratosphere have been taken, i.e., 18 and 15, respectively.

Table 2.3: Global aircraft emission data compared to *i*) global emission data of all biogenic and anthropogenic sources combined, and to *ii*) global emissions by fossil fuel combustion only

TRACER	TOTAL GLOBAL 1990 EMISSION *a*	AIR-CRAFT SHARE *b*	GLOBAL 1990 EMISSION BY FOSSIL FUEL COMBUSTION	AIR-CRAFT SHARE
CO_2	7×10^5 Tg	0.01%	*c* 21×10^3 Tg	3%
H_2O	*d* 5×10^7 Tg	$\approx$ 0	?	?
SO_2	16×10^1 Tg	0.1%	*e* 13×10^1 Tg	0.1%
NO_2 *f*	2×10^2 Tg	1%	82 Tg	4%
CO	3×10^3 Tg	0.01%	*g* 3×10^2 Tg	0.5%
HCs	$> 1 \times 10^3$ Tg	$\approx$ 0	*h* $< 12 \times 10^1$ Tg	> 0.4%
C	?	?	?	?

a: Biogenic and anthropogenic sources.

b: Percentages given only to indicate order of magnitude.

All figures taken from IPCC (90, 92), with the following changes:

c does not include emissions by cement production and gas flaring;

d is estimate of total water evaporation from the Earth's surface (Press and Siever 86);

e based on 1980 inventory by Spiro et al. (92);

f: figures given include revised estimate for aircraft emission, i.e., 3.2 Mt of NO_2 instead of 1.97 (this study);

g includes combustion by both stationary ovens, boilers and engines, and transportation (Warneck 88);

h is estimate of global man-made emission (Hough 91);

Table 2.3 gives an indication about the relative contribution of aircraft emissions compared to the estimated global emissions of all biogenic and anthropogenic sources combined, and compared to the global emissions from the combustion of fossil fuels.

In Tables 2.4 to 2.6, perturbations of background volume mixing ratios in the lower stratosphere and upper troposphere are presented for several exhaust components. They are calculated in a first-order approximation from Equation 1.1,

$$A = (B \times C \times D) / (E \times F) \qquad \text{(Eq. 1.1)}$$

in which

A = absolute change in volume mixing ratio of exhaust component Q in a volume within the Earth's atmosphere Y;

B = emission rate of Q, in units of mass per unit of time, in height interval Z in Y;

C = residence time[13] of Q in Z;

D = the molecular weight of air, i.e., 28.97;

E = mass of Y; and

F = the molecular weight of exhaust component Q for which the calculation is done.

The calculated absolute change in mixing ratio is a dimensionless number; in other words, it does not matter which units are used in the calculation, if only they are used consistently. The changes calculated, which are in this approach independent of current background mixing ratios, may occur if emissions continue in the same order of magnitude, for a period of time that is at least as long as the residence time. The residence time is defined here as the average time spent in a well defined part of the atmosphere by the molecules of a given gas after emission in this part.

[13]The average atmospheric lifetime or residence time of a species within a certain defined part of the atmosphere can be calculated in the simplest way as the concentration within that part divided by the rate of supply to that part. For residence times in the whole troposphere of shorter than about 10 years, significant latitudinal gradients can be expected. For extremely short residence times (0.001-0.1 years), gradients in the vertical plane may also be encountered.

Table 2.4: Steady-state perturbations in background volume mixing ratios in the upper troposphere due to high-flying aircraft

TRACER	RESIDENCE TIME *a*	UPPER TROPOSPHERIC 1990 AIRCRAFT EMISSIONS	LOCAL STEADY-STATE MIXING RATIO INCREASE *b*	BACKGROUND 1990 MIXING RATIO *a*
CO_2	see text	152 Tg	7 ppm	*c* 353 ppm
H_2O	0.07 yr	60 Tg	0.01 ppm	150 ppm
SO_2	*d* 0.03 yr	0.053 Tg	1 ppt	*e* 50 ppt
NO_2	*f* 0.03 yr	0.87 Tg	27 ppt	*g* 200 ppt
CO	*h* 0.25 yr	0.34 Tg	0.1 ppb	*i* 100 ppb
C_2H_4	*j* 0.1 yr	*i* 0.018 Tg	3 ppt	*j* a few ppt
C	*k* 0.03 yr	0.00097 Tg	0.05 ppt	*l* 7 ppt

a: A typical Northern-Hemispherical value for the upper troposphere, just under tropopause level is given;

b: Figures given are averaged over total mass of the upper troposphere. Here, the upper troposphere extends from 8-10.7k in accordance with the Standard Atmosphere. For this volume a mass of 0.6×10^{21} g is calculated, i.e., about 12% of total atmospheric mass; ppm = parts per million parts; ppb = parts per billion parts; ppt = parts per trillion parts;

c is taken from IPCC (92);

d is SO_2 residence time in the unperturbed marine atmosphere (Warneck 88);

e is taken from Warneck (88);

f is taken from Ehhalt et al. (92);

g is estimate for average value for 30-60N, from figures given by Ehhalt and Drummond (88);

h is taken from Lelieveld (94);

i is estimate for average value for 30-60N, calculated from figures given by Marenco et al. (89);

j values given for residence time, emissions and background mixing ratio (based on Warneck 88) are highly uncertain. All values, including this note, apply to formaldehyde also;

k is based on information given by Pitchford et al. (91);

l is taken from Pueschel et al. (92).

Table 2.5: Steady-state perturbations in background volume mixing ratios in the lower stratosphere due to high-flying aircraft

TRACER	RESIDENCE TIME *a*	LOWER STRATOSPHERIC 1990 AIRCRAFT EMISSIONS	LOCAL STEADY-STATE MIXING RATIO INCREASE *b*	BACKGROUND 1990 MIXING RATIO *a*
CO_2	see text	222 Tg	7 ppm	*c* 353 ppm
H_2O	1 yr	87 Tg	0.2 ppm	*d* 10 ppm
SO_2	*e* 1 yr	0.077 Tg	58 ppt	*f* 50 ppt
NO_2	0.1 yr	1.1 Tg	115 ppt	*g* 200 ppt
CO	*h* 0.6 yr	0.49 Tg	0.5 ppb	*i* 80 ppb
C_2H_4	*j* 1 yr	*j* 0.026 Tg	45 ppt	*j* a few ppt
C	1 yr	0.0014 Tg	6 ppt	*k* 2 ppt

a: A typical Northern-Hemispherical value of 1 year is given for the lower stratosphere, just above tropopause level, unless more specific information was available;

b: Figures given are averaged over total mass of the lower stratosphere. In this study, the lower stratosphere extends from 10.7 to 15 km in altitude with respect to the Standard Atmosphere. For this volume a mass of 0.6 x 10^{21} g is calculated, i.e., about 12% of total atmospheric mass;

c is taken from IPCC (92);

d is taken from Fabian (90);

e is estimate for average residence time of stratospheric aerosol, taken from Snetsinger et al. (87);

f is taken from Warneck (88);

g is estimate for average value for 30-60N, calculated from figures by Ehhalt and Drummond (88);

h is upper estimate taken from the range for tropospheric lifetime of ^{14}CO, calculated by Volz et al. (81);

i is estimate for average value for 30-60N, calculated from figures given by Marenco et al. (89);

j values given for residence time, emissions and background mixing ratio (based on Warneck 88) are highly uncertain. All values, including this note, apply to formaldehyde also;

k is taken from Pueschel et al. (92).

Table 2.6: Changes in background trace gas concentrations in the North Atlantic Flight Corridor due to high-flying aircraft: *i*) steady-state increases in background volume mixing ratios, and *ii*) initial volume mixing ratio increases in the aircraft wake

TRACER	AIRCRAFT 1990 NAFC EMISSIONS *a*	NAFC MIXING RATIO INCREASE		MIXING RATIO INCREASE IN EXHAUST		NAFC 1990 BACKGROUND MIXING RATIO		
CO_2	23	2	%	17	%	*b*	353	ppm
H_2O	8.9	0.2	%	1100	%	*c*	5	ppm
SO_2	0.0079	3	%	14000	%	*d*	100	ppt
NO_2 *e*	0.11	30	%	150000	%	*f*	180	ppt
CO	0.050	0.07	%	340	%	*g*	60	ppb
C	0.00014	9	%	46000	%	*h*	3	ppt

The residence time for all exhaust gases except CO_2 (see text) in the NAFC is one day: the characteristic wind speeds in the three dimensions at these altitudes are such that in one day the emissions are effectively transported out of the NAFC[14]. Because the NAFC is situated at the tropopause level, mixing ratio increases are compared with background mixing ratios in the lower stratosphere between 50-60N. Mixing ratio increases could here be given as percentages, which give a better subjective presentation of the perturbations, because background mixing ratios given can be considered as representative for the whole volume of interest.

a: in Tg, i.e., 10^{12} gram;

b is taken from IPCC (92);

c is based on information given by Graßl (90);

d is estimate based on figures given by Chatfield and Crutzen (84) and Warneck (88);

e: mixing ratio increases are calculated with the NO_2 EI for cruise under stratospheric conditions;

f is calculated from figures given by Ehhalt and Drummond (88);

g is taken from Marenco et al. (89);

h is based on measurements by Pueschel et al. (92).

[14]Note that in the long run stratospheric mixing ratio increases given in Table 2.5, which take chemical processes implicitly into account too, should be added for each value given.

No difference is made between removal by chemical or dynamical processes (whichever removes first). In addition, it is assumed that the emissions are distributed evenly over the volume of interest and that major transport and chemical processes remain the same on average in the course of time.
CO_2 is treated separately because it has an effective residence time of typically 100 years (IPCC 90). In this study, atmospheric perturbations are calculated with residence times of which the upper limit is based on the speed of vertical transport processes as determined by atmospheric dynamics. For CO_2, this approach eventually underestimates the perturbation by two orders of magnitude. So for CO_2, emissions of all aircraft have been distributed over the whole atmosphere. For a residence time of 100 years, this results in a perturbation of 7 ppm.

Before presenting conclusions, some remarks which put the values calculated into perspective should be made.
i) With current predictions on air traffic growth vis-à-vis current predictions on emission reduction measures, it is highly probable that yearly aircraft emissions continue to increase. If so, the calculated mixing-ratio increases will increase as well.
ii) Perturbations calculated give only a first indication about trace gases of which either the atmospheric abundance should be monitored or the atmospheric effects should be studied. If the projected perturbations in mixing ratio of two trace gases are of comparable magnitude, it does not imply that their atmospheric effects will be of the same strength. Moreover, even if they are, things may be quite different if both perturbations are doubled. This kind of behaviour is due to non-linearities present in chemical and physical processes. If, for example, the background mixing ratio of a certain trace gas is perturbed significantly, different chemical pathways may be chosen by this gas. This may lead to changes in residence times, making the initial predictions for this gas invalid.

To circumvent this, trace gases of which the background mixing ratios are calculated to perturb significantly by equation 1.1 should be studied more extensively with more comprehensive relationships, e.g., with the help of chemistry-transport models (Chapter 3). When the perturbations remain significant when calculated in a more accountable way, possible climatic consequences of these perturbations should be calculated. This could be done with the help of climate models (Chapter 4).

iii) Residence times used are often based on model simulations. However, not many models take full account of exchange processes between tropospheric and stratospheric air. In reality, this exchange is significant: by definition at tropopause level, in particular at mid-latitudes. Incorporation of these exchange processes in models may lead to shorter residence times. Smaller net differences between tropospheric and stratospheric perturbations than calculated in this study may result from this.

iv) Current background mixing ratios include perturbations due to anthropogenic emissions since pre-industrial times. If anthropogenic non-aircraft emissions remain much larger than aircraft emissions, as is currently the case, the relative aircraft contribution to the anthropogenically induced perturbations may remain small while in absolute terms it may have reached a high level. So, a method should be found to disregard the contribution of anthropogenic non-aircraft emissions when judging the perturbations due to air traffic.

A comparison of the calculated perturbations from aircraft with pre-industrial, and thus unperturbed, background mixing ratios may give a better indication of the aircraft effects. When judging possible perturbations by high-flying aircraft, this may lead to more accurate conclusions because perturbations by other human activities are not partly accounted for. As pre-industrial values have yet been established for some relevant gases only, calculated perturbations have in this study been judged with current background mixing ratios as reference.

2.6 DISCUSSION AND CONCLUSION

A first indication of the possible effects on background mixing ratios of aircraft emissions has been given in this chapter. From this, the following conclusions have been drawn. They should be considered as preliminary only because of the simplicity of the calculation method used for estimating possible perturbations. For the same reason, only qualitative information will be given in this section.

- Compared to all anthropogenic emissions from the combustion of fossil fuel in 1990, global air traffic contributes significantly to both CO_2 and NO_x emissions. It is not yet clear what the aircraft contribution to either emissions of water, specific hydrocarbons or soot is.
- From the calculations follows that, although the amount of fuel burnt in the upper troposphere is of the same order of magnitude as the amount of fuel burnt in the lower stratosphere, predicted perturbations in background mixing ratios are in an absolute sense for all exhaust trace gases an order of magnitude larger for the stratosphere than for the troposphere. The reason for this is the fact that residence times are generally higher in the stratosphere, a result of differences in chemical pathways chosen by species and other non-linearities.
- In the North Atlantic Flight Corridor, predicted changes in background mixing ratios are significantly different from when emissions are spread over a larger volume. This is mainly due to the fact that the residence time used for emissions in the NAFC is purely based on atmospheric dynamics. It is calculated as being one day, which is short albeit realistic. This approach results in significant, i.e., larger than 1%, increases in soot, sulfur dioxide and and nitrogen oxides on the short term. On the longer term stratospheric mixing ratios will add, resulting in significant increases for all exhaust components except CO.

- Subjectively, the atmospheric load may be considered higher in the NAFC than in the lower stratosphere as a whole because air traffic is by definition more dense in corridors. However, it follows from the calculation method used that the opposite is true. Moreover, the fact that interhemispherical exchange of air is a very slow process and that only very little air traffic takes place in the Southern Hemisphere has not even been taken into account in the calculations. This would enhance calculated lower-stratospheric perturbations for the Northern Hemisphere.
- Calculated relative perturbations in background mixing ratios show large differences for each exhaust component when all cases are considered. Large differences in calculated relative perturbations also exist between the different exhaust components. Relative perturbations are non-significant to (extremely) large for soot and H_2O, significant to (extremely) large for NO_2 and SO_2, (non-)significant to comparatively small for both CO and CO_2.
- More information is needed about emissions of exhaust constituents like ethene, formaldehyde and cyclosiloxanes and about their atmospheric abundance and residence times. Though emitted in relatively small quantities, they may be able to perturb their respective background mixing ratios to a high extent.

Assuming that there is a positive relation between the magnitude of a relative perturbation of background mixing ratios and the probability that this perturbation provokes environmental and climatic effects, calculations made in this section give a first indication of the exhaust components and processes that are important in this respect. Based on the information provided, interest should be primarily focussed on chemical and dynamical processes in the exhaust plume as well as on the atmospheric effects from emissions of nitric oxide, sulfur dioxide and soot in the lower stratosphere: for these cases perturbations calculated were the largest by a significant margin.

3 PERTURBATION EFFECTS: INFLUENCES ON THE OZONE DISTRIBUTION

3.1 INTRODUCTION

In Chapter 2 has been indicated how high-flying aircraft may influence the atmospheric abundance of several trace gases. The results are mainly of scientific interest, however, since society is primarily interested in the effects of these influences. These will be dealt with in this and the following chapter.
Although simple calculations can be used to indicate of which trace gases, emitted by aircraft, large perturbations may be expected in background mixing ratios, this route cannot be followed to elucidate the effects of these perturbations. On the contrary, because of the comprehensiveness of processes involved, the use of large physical and chemical models is mandatory when studying the effects of aircraft induced perturbations in background concentrations. These models are complex to such an extent that supercomputers are needed to run them.
For even a relatively small number of chemical constituents in a realistic, three-dimensional global atmosphere, the calculation of changes in ozone is usually extremely computation-intensive. Because of current limitations in computer resources, the chemistry implemented in these models is necessarily simplified; 3-D models to study ozone effects of short-lived tracers, like NO_X injected by aircraft, are still under development. 3-D transport-models, however, have become accurate in reproducing detected perturbations in background concentrations due to, e.g., emissions of inert tracers. These are now frequently used for calculating the distribution of aircraft NO_X emissions (e.g., Velders et al. 94, Van Velthoven et al. 94). Nonetheless, society is less interested in the actual perturbations, but rather in the effects of these perturbations for, ultimately, human health. Consequently, results of models which calculate ozone effects, important to life on Earth, provide more relevant information.

Two-dimensional models are often used for the stratosphere, where longitudinal mixing is rapid, so that the spatial variability is largely confined to the latitude and altitude coordinates. They provide useful information on the zonally averaged distribution, the seasonal evolutions and the budgets of species affecting either ozone or climate.

1-D models, in which latitude and longitude are fixed and only the altitude distribution is of interest, have been used for both stratospheric and tropospheric calculations, as they offer the advantage of rapid calculation even with fairly detailed chemistry. They can thus be used for climate-sensitivity studies over long simulation times. Dynamical exchanges are, however, poorly formulated in these models.

The current situation is that only few model runs to study the effects on ozone and climate due to emissions from high-flying subsonic aircraft have yet been made. This picture changes if supersonic aircraft is also taken into account. Because of the upcoming interest in commercial supersonic air traffic in the beginning of the seventies and a renewal of this interest at the beginning of nineties, their effects have been, and are being, documented extensively. However, as this report focusses on emissions of trace gases at and near tropopause level, these studies will not be considered as supersonic aircraft generally inject species at substantially higher altitudes.

In all studies with chemistry-transport models considered the focus has been on the effects of emissions of NO by aircraft on the distribution of ozone. The argument for this is as follows:

i) background NO_x mixing ratios are will be disturbed relatively strongly (e.g., Tables 2.4 to 2.6 and Section 2.6);

ii) NO_x is part of the ozone generation-destruction cycle (Warneck 88); and

iii) the distribution and total amount of ozone is important to life on Earth (UNEP 91).

3.2 RESULTS FROM CHEMISTRY-TRANSPORT MODELS

Results of the studies considered can be classified into changes in total ozone[15], and into changes in tropospheric and stratospheric content. The last separation can be explained by the fact that model studies indicate that NO_x results in net ozone formation when emitted in the troposphere and in net ozone destruction when emitted in the stratosphere. This is nicely illustrated in figure 3.1: according to the 1-D model used, the cross-over point is situated between about 12k and 14k. More recent studies with 1-D models give essentially the same result. Both decreases in stratospheric ozone and increases in tropospheric ozone may eventually create a situation that is potentially damaging for ecosystems in general (UNEP 91). For reasons of convenience, in this study this separation between stratospheric and tropospheric effects is kept.
Four tables of the results of studies done to date on the effects of high-flying subsonic aircraft will be presented in this chapter. In all studies considered, NO_x injection rates of the same order of magnitude as those calculated in this study for 1990 are used, i.e., an injection of 1.1 Tg in the lower stratosphere and 0.9 Tg in the upper troposphere (tables 2.4 and 2.5). All results apply to local situations; under the header "Remarks" information about the situation to which the calculated change refers can be found. Values given refer most of the time to an ozone perturbation for a specific altitude during a specific season at a specific latitudinal band in the Northern Hemisphere. The percentage change in ozone calculated, refers to the state when a new equilibrium is reached (steady-state conditions).

[15] The column ozone is the total amount of ozone in a column of air above a monitoring station extending from the Earth's surface to the Sun. The abundance of ozone in this column is expressed in Dobson units. 100 Dobson units corresponds to an ozone layer thickness of 1 mm if all ozone were at 1 atmosphere pressure and 0°C. The annual mean value above The Netherlands for the period 1971-92 is about 330 DU.

Results from studies by Brühl and Crutzen (88), Beck et al. (90. 92), and Johnson and Henshaw (91) and Johnson et al. (92) are presented in the tables as reference points: they are known among a relatively large public. They have not been taken into account in the conclusion, however, since they put a large fraction of the emissions in the layer between 0-8k. The last point also counts for the study of Wauben et al. (94), of which the results have also been presented in this chapter. Due to the presence of vertical transport processes in the models, it is impossible to judge from the information given to what extent perturbations at a certain altitude can be associated with cruise emissions, the focus of this study, or with emissions during, for instance, idle and taxiing at airports.

The studies by Luther et al. (79) and Derwent (82) are borderline cases. They have been taken into account because in both studies the largest part of the emissions, both over 93%, have been injected into the layer of the atmosphere this report focusses on, i.e., the layer between 8-15k as defined by the Standard atmosphere.

Table 3.1 presents the calculated stratospheric perturbations from high-flying aircraft. Only low net relative ozone destruction values are found. It should be noted, that most of the ozone is found at the calculated altitudes of maximum destruction. Tropospheric perturbations can be found in Table 3.2. Perturbations calculated are generally an order of magnitude higher than for the stratospheric case; one should know, however, that ambient mixing ratios are an order of magnitude smaller in the upper troposphere compared to stratospheric values. The study by Danilin et al. (92) has a somewhat different approach compared to the other studies represented in Table 3.2. It studied the short-term response (from seconds to hours) of the atmospheric trace gas composition to perturbations in the aircraft plume region.

They calculated a steady-state increase in ozone of about 8% in the exhaust plume, independent of the assumption that all NO_x injected consists of NO only or of NO_2 only. What could be of interest for future modelling studies is that Danilin et al. think that it is perhaps possible to study the global effect of aircraft emissions by extrapolating perturbations calculated for the plume-chemistry level to the level of grid-cells. According to this approach, it is possible to assess with a 2- or 3-D model the impact of aircraft on the global atmosphere by taking the plume effect of a single aircraft and combining it with an inventory of aircraft emissions, which for every grid cell of the model accounts for the geographical and diurnal distribution of air traffic, flight altitudes, et cetera.

Tables 3.3 and 3.4 present perturbations in total ozone. 1-D models calculate global average perturbations (Table 3.3), 2-D models are able to calculate perturbations in total ozone for different latitudinal bands (Table 3.4). The results of Kinnison et al. (89) and Johnston et al. (89) have been presented primarily in Table 3.4 to make clear once again what the effects are of a change in model parameters. Both models are identical, only the residence time of NO_x has been changed. This results in a change of the sign of the change in total ozone. As the emissions are injected at altitudes which are beyond the scope of this study, i.e., from 15k upward, they have not been taken into account in the conclusion.

Table 3.1: Local steady-state perturbations in ozone content in the stratosphere due to high-flying aircraft

Study	Type of Model	Injection Rate (Tg NO_2/yr)	Stratospheric O_3 Change	Remarks @
Hidalgo and Crutzen (77)	2-D	2.06 at 10.8k (1990) 0.46 at 14.5k	- 0.2% - 0.4%	45N, S, 20-40k
Ko et al. (91)	2-D	3.05+ (2015)	- 1-2%	30-60k
Brühl and Crutzen (88)	1-D	# (1985) # (2050)	negligible - 4%	NH, 20k

@: N = degrees northern latitude; S = summer; k = kilometre in altitude; NH = Northern hemisphere;

+: Between 8k and 14k.

#: In the study by Brühl and Crutzen, projections by Wuebbles et al. (84) were used. No information about injection rates or absolute emissions were provided, however, in the study by Brühl and Crutzen.

Table 3.2: Local steady-state perturbations in ozone content in the troposphere due to high-flying aircraft

Study	Type of Model	Injection Rate (Tg NO_2/yr)	Tropospheric O_3 Change	Remarks @
Hidalgo and Crutzen (77)	2-D	2.06 at 10.8k (1990)	+ 12%	10.7k, 45N, S,
		0.46 at 14.5k	+ 3%	13.6k
Liu et al. (80)	1-D	1.84 above 9k		NH
		hi trop. NO_x	+ 40%	
		lo trop. NO_x	+ 80%	
Derwent (82)	2-D	0.13$^+$ (1960)	+ 0.25%	30N,
		0.49$^+$ (1975)	+ 2.0%	S,
		4.86$^+$ (1990)	+ 15.4%	8-10k
Ko et al. (91)	2-D	3.05$^\#$ (2015)	+ 4-8%	NH, <15k
Danilin et al. (92)	1-D	0.2 g NO_x/m *	+ 8%	50N, 10.7k
Brühl and Crutzen (88)	1-D	? (1985)	+ 5%	NH,
		? (2050)	+ 60%	10k
Beck et al. (90, 92)	2-D	2.06	+ 12%	30-60N,
		4.12	+ 19%	8-12k
		6.18	+ 25%	
Johnson and Henshaw (91), Johnson et al. (92)	2-D	1.94 (1990)	+ 5.9%	30N,
		3.18 (2000)	+ 8.8%	S,
		6.69 (2015)	+ 15.2%	8-12k
Wauben et al. (94)	3-D	1.94 (1990)	+ 3%	globally
			+ 8%	in NAFC
			+ 9%	locally

@: k = kilometre in altitude; N = degrees northern latitude; NH = Northern hemisphere, S = summer;

$^+$: Between 6k and 14k. Derwent includes the emission inventory of Bauer (79) which has a maximum between 10-11k, between 30-40N;

$^\#$: Between 8-14k;

*: Study of Danilin et al. concerns aircraft wake only.

Table 3.3: Global steady-state perturbations in total ozone due to high-flying aircraft, calculated with 1-D models

Study	Injection Rate (Tg NO_2/year)	Total O_3 Change (%)	Remarks @
Luther et al. (79)	1.2 at 9k	+ 0.40	79a chem
	1.2 at 11k	+ 0.64	79a chem
	1.2 at 13k	+ 0.84	79a chem
	1.1 (79)	+ 0.5	
	3.08* (90 lo)	+ 1.29	79a chem
	4.86* (90 hi)	- 0.31	75 chem
	4.86* "	+ 1.43	77 chem
	4.86* "	+ 1.86	79a chem
	4.86* "	+ 2.01	79b chem
Liu et al. (80)	1.84 above 9k		NH
	hi trop. NO_x	+ 4	
	lo trop. NO_x	+ 8	
WMO (81)	2.5 at 9k	+ 0.5	Chang
	2.5 at 11k	+ 0.8	"
	2.5 at 13k	+ 0.9	"
	2.5 at 13k	- 0.7	Brasseur
Kinnison et al. (89), Johnston et al. (89)	0.6 at 12k	+ 0.1 to + 0.5	#
	1.2 at 12k	+ 0.2 to + 0.9	#
	1.9 at 12k	+ 0.2 to + 0.4	#
	2.5 at 12k	+ 0.2 to + 1.8	#
	4.9 at 12k	+ 0.1 to + 3.2	#

@: NH = Northern hemisphere;

#: Percent change in ozone column strongly dependent on assumed Cl_x mixing ratio at 50k; values varied between 1.1 (weakest net ozone depletion due to Cl_x, resulting in weakest ozone formation due to NO_x) and 21.6 ppbv (strongest net ozone depletion due to Cl_x, resulting in strongest ozone formation due to NO_x), except for the 1.9 Tg case where Cl_x values varied between 3.1 and 7.9 ppbv;

*: Between 5.5 and 14.5k, with a maximum at about 10.5k.

Table 3.4: Global steady-state perturbations in total ozone due to high-flying aircraft, calculated with 2-D models.

Study	Injection Rate (Tg NO_2/year)	Total O_3 Change (%)	Remarks @
Hidalgo and Crutzen (77)	2.06 at 10.8k 0.46 at 12.7k 0.46 at 14.5k	+ 0.19 + 0.03 - 0.05	
Derwent (82)	0.13^{+} (1960) 0.49^{+} (1975) 4.86^{+} (1990)	+ 0.06 + 0.28 + 1.81	30N
Ko et al. (91)	3.05$^{\#}$ (2015)	+/- 0.25	NH only
Kinnison et al. (89)	1.8 at 16.5k* $t(NO_x)$ = 0.3 yr	+ 0.01 + 0.04 - 0.02	 SH NH
Johnston et al. (89)	$t(NO_x)$ = 0.5 yr	- 0.7 - 0.4 - 0.9	 SH NH

@: NH = Northern hemisphere; SH = Southern hemisphere;

*: Between 15k and 18k and 37-49N;

$^{+}$: Between 6 and 14k. Derwent includes the emission inventory of Bauer (79) which has a maximum between 10-11k and 30-40N;

$^{\#}$: Between 8 and 14k.

Concerning the perturbations in ozone by aircraft NO_x emissions, it should be recognized that models with which these perturbations were calculated have developed rapidly the past two decades. However, due to lack of measurements of the atmosphere's composition which have sufficient detail in both time and space, it is still unknown how accurate these models reproduce reality.

Figure 3.2 is presented to give an idea about changes in model results for a given NO_x perturbation by supersonic aircraft due to changes in model chemistry or kinetic parameters. This is done for illustration purposes only, as the present study does not take supersonic aircraft into account.

A recent development is the inclusion of heterogeneous chemical processes, for instance on the surface of contrail ice particles or aerosols which form from soot and sulfur dioxide emitted by aircraft, in chemistry-transport models. The inclusion of this kind of processes may alter model results significantly, particularly those due to stratospheric emissions by subsonic aircraft. It is still a matter of debate, however, if perturbations will increase or decrease by the inclusion of heterogeneous chemistry. Recent results from models which include heterogeneous reactions on aerosol surfaces show that calculated stratospheric ozone losses may increase due to heterogeneous conversion of inactive forms of chlorine in the stratosphere into active forms (e.g., Brasseur and Granier 92), and they may decrease due to heterogeneous conversion of NO_x to HNO_3, reducing the efficiency of the NO_x catalytic cycle (e.g., Bekki et al. 91). This last process may result in higher abundances of active chlorine (less NO_x is available to form reservoir species with ClO_x), with the afore mentioned result. As it is not yet clear whether heterogeneous processes increase or decrease calculated stratospheric ozone losses due to high-flying aircraft, we will not take these into account in our conclusion in a quantitative sense.

If it turns out that heterogeneous chemical reactions mitigate the direct chemical influence of nitrogen oxides emissions on ozone significantly can it be foreseen that the discussion about ozone effects due to aircraft emissions of water vapour in the lower stratosphere will be reopened. This discussion grew silent after it had become clear that NO_x emissions were expected to have a much larger effect (Johnston 92).

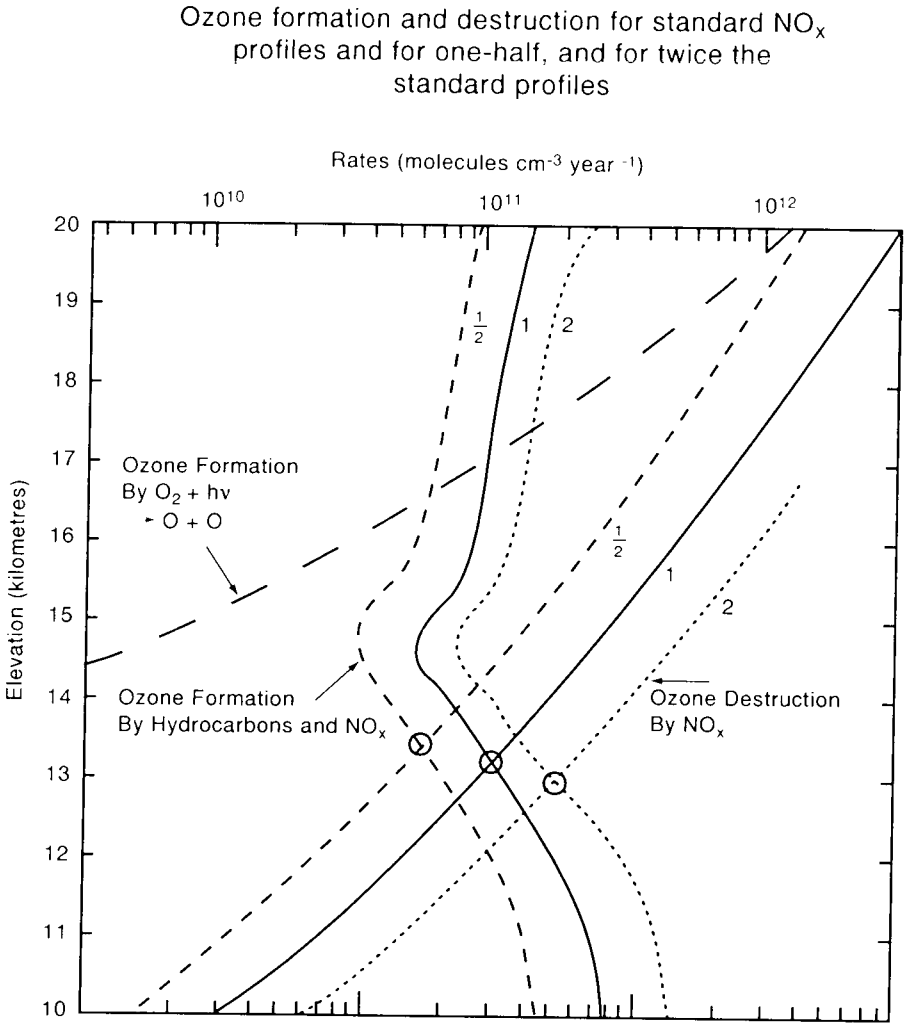

Figure 3.1.

The rate of ozone formation from the so-called smog reactions equals the rate of destruction from the NO_x catalytic cycle at about 13 km. For half and for double the assumed natural background of NO_x, the cross-over point is essentially unchanged. The altitude dependence of the rate for the reaction by which the ozone layer is primarily formed is also given (adapted from Johnston and Quitevis, 1975).

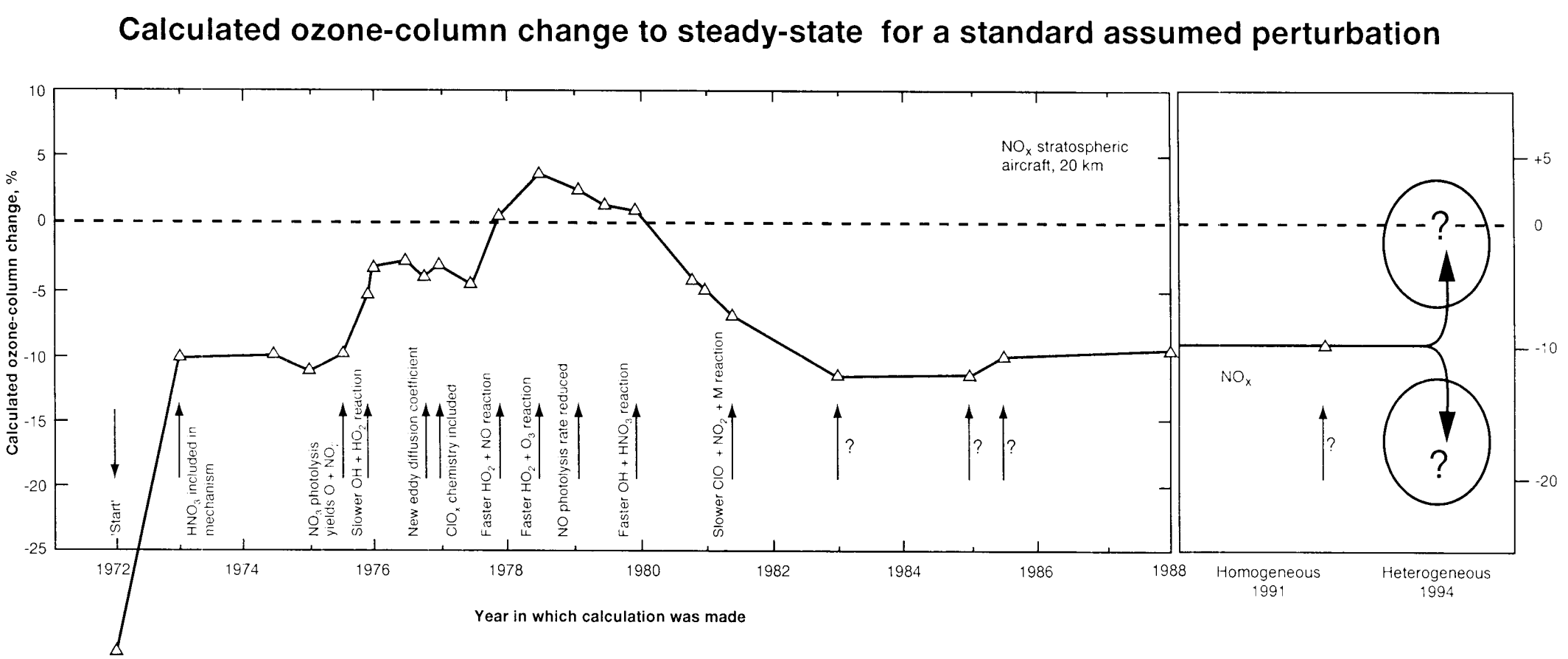

Figure 3.2.

The historical evolution of the calculated change in total ozone due to a NO_x injection of 2000 molecules $cm^{-3}s^{-1}$ over a 1-km thick layer centered at 20 km in altitude resulting in an emission of 2.5 Tg NO_2 yr^{-1}. All values are from the Lawrence Livermore National Laboratory 1-D model. Major changes in model parameters are indicated.
Figure adapted from Johnston (1993) with information from Luther et al. (1979) and WMO (1981).

3.3 DISCUSSION AND CONCLUSION

The outcomes of model studies on the effects of emissions of nitrogen oxides on the distribution of ozone vary to a large extent. Because of differences in how chemical and transport processes at tropopause height are parameterised or described in the models used, it is difficult to intercompare these outcomes. This situation is worsened by the fact that each research group uses different emission figures as input: NO_2 injections vary in magnitude, in altitude and in the way they are distributed over the grid, i.e., the simulated world. This makes intercomparison of the different model results, qualitative as well as quantitative, difficult at present. If this spread in input data remains, it will in the future, when atmospheric measurements are at hand, also be difficult to estimate which model uses the most realistic chemistry-transport scheme or, in other words, to give value judgments. This can be considered as a strong plea for an intercomparison of models by means of using the same input (e.g., climatology, aircraft emissions, or other anthropogenic perturbations). Comparison of model results is also problematic because the areas of interest differ. Studies concern global or local changes, in the Northern or Southern Hemisphere, in winter or in summer, et cetera.

In spite of the above-mentioned criticisms, a general picture can be extracted from the model results:

- Not one model run predicts that no change will occur in the global distribution of ozone by the injection of NO_2.
- High-flying subsonic aircraft will deplete stratospheric ozone. If the mean flight altitude increases, this effect will become stronger. Based on estimates for 1990 emissions by aircraft in the lower stratosphere (Table 2.5) and a limited number of model outcomes (Table 3.1), the depletion of stratospheric ozone in the Northern Hemisphere will not exceed 1% if aircraft emissions remain on the 1990 level.

- (Upper) tropospheric ozone is predicted to increase significantly due to aircraft emissions. Based on model studies (Table 3.2) and considering the 1990 emissions by high-flying subsonic aircraft in the upper troposphere (Table 2.4), an increase in current ozone concentrations in the Northern Hemispheric upper troposphere due to air traffic in the order of magnitude of 10% should locally already be observable. That is to say, if nitric oxide emission rates by aircraft are still at the 1990 level or above.
- It is not yet clear whether total ozone either decreases or increases on a global scale. Although by now enough information is available about the distribution of air traffic movements to perform this kind of calculations, they have not yet been done.

From present emissions of nitrogen oxides by high-flying aircraft, the emission inventories by McInnes and Walker (92) and NASA (Wuebbles et al. 93), and model results (Tables 3.3 and 3.4), is inferred that there is likely to be a small positive, but non-significant contribution to the globally averaged ozone column. As the stratosphere accounts for 90% of total ozone and the troposphere for 10%, this is in line with the conclusions drawn above concerning changes in stratospheric and tropospheric content.

Total ozone above the surface area situated under the North Atlantic Flight Corridor is bound to decrease a little because of higher mean flight-altitudes compared to the global mean situation. Above Europe and North America a significant increase in total ozone may be expected mainly because of generally lower mean flight altitudes resulting in strong increases in upper tropospheric ozone.

At this point, it should be stressed that even if models calculate zero change in total ozone, it does not mean that there could not be a change in the distribution of ozone and for that reason in the Earth's radiation budget.

4 PERTURBATION EFFECTS: INFLUENCES ON CLIMATE

4.1 INTRODUCTION

In science, assessing the result of a process of change in nature usually means performing an identical experiment. But what if the issues are complex or if the scale of the experiment would have to be unmanageably large? This is the problem atmospheric scientists are confronted with. How can we anticipate what is going to happen in the atmosphere, if no meaningful experiment can be performed? For the moment the answer is the use of a computer. The atmospheric system can be represented by a mathematical model. This model can be introduced into the computer. This being done, scientists can perform their experiments.
Mathematical models translate conceptual ideas into quantitative statements. There is a range of models, from those that simply treat one or two processes in detail to large-scale, multi-process simulation models. Such models are not faithful simulators of the full complexity of reality, but they are expected to tell us to a certain extent what are the logical consequences of explicit sets of plausible assumptions. Here, the assumptions consist of high-flying aircraft related increases in:
i) atmospheric concentrations of greenhouse gases;
ii) high-cloud coverage due to the occurrence of contrails; and
iii) increases in aerosols.
The consequences society is interested in concern climatic change.
Unfortunately, only few studies on the climatic effects of high-flying aircraft have yet been done. Those that have been done deal with temperature effects only, while society should be more interested in climatic variables which are more important to ecosystems, like, for instance, changes in precipitation or in prevailing wind patterns.

In addition, in all cases considered for this study but one, these effects are calculated for emissions of only one of the constituents of aircraft exhaust. For instance, the temperature effect of ozone perturbations due to NO_x emissions is studied, or the temperature effect of increases in high-cloud cover due to H_2O emissions. This effectively inhibits an integral approach when discussing effects on climate and environment of aircraft emissions. For this reason, results concerning influences on climate and environment will for each major exhaust component be presented separately.
Another route to estimate the climatic effects of aircraft emissions is via the detection of changes that already have occurred, if any. Some detection studies have yet been done and their results will here be presented. These concern changes in high-cloud coverage due to the occurrence of contrails.

Contrary to the foregoing two chapters, the separation between emissions in the stratosphere and in the troposphere, when discussing the effects, is not kept in this chapter. For most climatic effects, the underlying radiative phenomena which result from atmospheric perturbations are independent from whether these perturbations occur in the lower stratosphere or the upper troposphere.
The results of the different studies will be put in a broader context in the discussion and conclusion (Section 4.7) in order to come to an integral assessment of the climatic effects of high-flying aircraft.

4.2 CARBON DIOXYDE EMISSIONS

CO_2 is chemically inert: it has a long residence time and is well-mixed throughout the atmosphere. Its radiative effects can therefore be considered independent from where and when it is emitted. So, in a first approach, the aircraft contribution to the climatic effect from CO_2 emissions from fossil fuel combustion will be similar in magnitude as the relative contribution of aircraft CO_2 emissions to all fossil fuel CO_2 emissions. Hence, the contribution of high-flying aircraft CO_2 emissions to climatic change can be assessed easily.

According to aircraft manufacturers and aviation organisations, world-wide air transport demand in passenger-km (Pkm) performed will double between 1990 and 2002; there is no indication of saturation in demand after the year 2000[16] (Nüßer and Schmitt 90). This forecast implies a growth rate of about 6% annually. Between 1973-89 the growth rate in Pkms for the scheduled services of airlines of ICAO contracting states was about 7% annually, while the world demand for aviation fuels increased by 2.4% annually in the same period (OECD 91). This improvement in fuel efficiency was largely driven by rising fuel costs during the seventies, resulting in larger planes and higher load factors, together with the continuing advance of aircraft technology.

For reasons of enhanced combustion efficiency, higher load factors and larger planes, the amount of aviation fuel used will increase at about half the rate of increases in Pkms performed, i.e., 3%. This rate of efficiency improvement is less than which has occurred during the seventies and eighties because of the high level of efficiency already obtained.

[16] At this point we disregard the fact that the comparable increases in the number of flights are generally smaller (factor 0.8-0.9) {e.g. Report of ICAO meeting NAT TFG/26 (ICAO 92)} and the forecast that the air transport performance in freight traffic will increase stronger than passenger traffic (Nüßer and Schmitt 90).

Improvements will inherently cost disproportionately more money. In 1990 military aircraft accounted for about 25% of the total aviation fuel use (Wuebbles et al. 93, Egli 90). It is assumed here that in absolute terms this fuel use will stay at the same level till 2025. A similar forecasts, albeit with a shorter range, is made by ICAO (92): military traffic is projected to be at the 1989 level from 1992 to 1997. It follows that the annual growth rate in aviation fuel use for the period till the arbitrary year 2025 will be 2.5%. This implies a specific growth factor of 2.4 in the year 2025 relative to 1990, in line with Kavanaugh (88). Furthermore, total anthropogenic CO_2 emissions by fossil fuel combustion, including aircraft emissions, will increase annually by about 1.7% until 2025 according to the IPCC IS92a scenario (IPCC 92), i.e., a growth factor of 1.8. Then, the contribution of air traffic to global fossil fuel CO_2 emissions will increase from 2.6% in 1990 to 3.5% in 2025. The military share of air traffic CO_2 emissions will then be about 11%.

Around 2025 CO_2 will be responsible for 63% of the total radiative forcing[17] from pre-industrial values according to the IPCC 90 Business-as-Usual scenario. This figure is cumulative and includes land-use changes as well. In addition, aircraft have not been contributing significantly before, say, the sixties. Meanwhile, CO_2 emissions are increasing exponentially. From these figures[18] can be concluded that in 2025 the contribution of aircraft CO_2 emissions alone to the committed climatic change, i.e., the climatic change that would occur instantaneously if the time lag of the climatic system would be zero, will be significant, i.e., in the order of magnitude of 1%.

[17] There are several natural factors which can change the balance between the energy absorbed by the Earth and that emitted by her in the form of longwave infrared radiation; these factors cause the radiative forcing on climate.

[18] Recent results suggest that changes in tropospheric ozone may well have a far larger influence on the enhancement of the Greenhouse Effect than thought previously (Wang et al. 93, Van Dorland and Fortuin 94). If so, the relative contribution of CO_2 should be lowered accordingly.

4.3 WATER EMISSIONS

4.3.1 INTRODUCTION

Water vapour is by far the most important greenhouse gas of the atmosphere: it is responsible for more than two thirds of the natural Greenhouse Effect. When climatic change is concerned, the enhancement of this effect should be taken into consideration. Water emissions from aircraft may also contribute to this enhancement and in this respect two processes of which the importance is generally acknowledged, including their possible climatic impact, will be discussed:

i) the atmospheric input of water in the form of water vapour will be discussed; and

ii) the atmospheric input of additional water in the form of ice crystals by the formation of condensation trails, in the following called contrails, will be dealt with.

Other processes influenced by aircraft water emissions, but this time specifically by aircraft flying in the stratosphere, are the formation of polar stratospheric clouds, PSCs (Peter et al. 91, Arnold et al. 92), and of clouds consisting of sulfuric acid (Arnold et al. cited in Schumann 93). The saturation temperature for the formation of PSCs strongly depends on the local partial pressure of nitric acid and water vapour. Though most stratospheric aircraft do not generally fly in those parts of the atmosphere where PSCs normally occur, i.e., the polar stratosphere, large-scale transport processes will transport part of the aircraft emissions to those parts. Thus, PSC formation temperatures are also sensitive to NO_x and H_2O emissions from stratospheric aircraft. Consequently, aircraft flying in the stratosphere may increase the PSC-formation probability, and may enhance the potential ozone destruction by chlorine radicals accordingly. Whether this process is already occurring is still a matter of debate.

For instance, one may argue that both maximum cruise altitudes of present subsonic aircraft and the absolute stratospheric NO_x and H_2O emissions are too low for this effect to be of any importance on large scales. With respect to smaller-scale processes, however, another conclusion could be drawn. The emitted water vapour causes supersaturation in the wake, which, together with the rapid cooling of the exhaust air and the high HNO_3 content, initiates the formation of HNO_3-ice particles. Hence, PSC formation could be more likely in the plume of the aircraft than in the undisturbed environment.

Supersaturation is also used as an explanation for the recent finding that contrails may be formed composed of sulphuric acid aerosols. Measurements in the wake of an aircraft show that about 20% of the SO_2 emitted is converted into H_2SO_4. Sulphuric acid aerosols formed do not evaporate after plume dissipation because of supersaturation of the background atmosphere with respect to liquid H_2SO_4-H_2O.

As only preliminary results of the last two cloud forming processes presented have yet been published, possible influences on climate of these processes will not be discussed, nor will they be taken into account in the conclusions.

4.3.2 WATER VAPOUR EMISSIONS AND CLIMATE CHANGE

In general, aircraft water emissions are not significant compared to water emissions from other sources. However, the absolute amount that is possibly emitted in the stratosphere by aircraft, i.e., 87 Tg per year (Table 2.5 on page 27), is of the same magnitude as the water vapour emissions by stratospheric oxidation of methane, i.e., about 95 Tg/yr (Warneck 88).

For the stratospheric perturbation of water vapour background mixing ratio due 1990 emissions from high-flying aircraft as presented in Table 2.5, a steady-state warming at the Earth's surface in the order of 0.01°C is calculated from the mathematical expression given by Luther et al. (79).

In flight corridors water vapour mixing ratios will increase even stronger (Table 2.6 on page 28) than in the surrounding air masses. This may cause larger local perturbations of the radiation balance resulting in larger (local) changes in surface temperatures.
The above calculation is just a simple exercise based on old model results (see also Section 4.5). Moreover, local temperatures are only to a small extent determined by incoming radiation. Advection of air from other places by the prevailing circulation has a far larger influence on mean temperatures. In addition, if it turns out that a large fraction of the water emitted turns into ice (clouds) the above considerations will have to be mitigated. Whether this will amplify or mitigate the net radiative effect of water emissions cannot be said in advance.

4.3.3 CONTRAIL FORMATION AND ITS CLIMATIC EFFECTS

4.3.3.1 Introduction

An increase in contrails, caused directly by increases in air traffic and indirectly by an enhancement of contrail formation[19], may influence the climatic system via two different physical mechanisms, which are believed to have an opposite effect:

i) Added cirrus clouds, as which contrails can be considered, upset the radiation balance depending on the reflectivity, emissivity and temperature of the clouds.
ii) Added cirrus clouds dry the upper troposphere and lower stratosphere by the combined effect of two processes:

[19] This enhancement is due to higher cruising levels and the cooling of the lower stratosphere, in turn due to enhanced radiative cooling to space caused by increases in greenhouse gases.

- The sublimation of ambient water vapour into ice on the surface of ice crystals. The ice crystals, formed spontaneously in the exhaust wake, initially grow fast by this process and finally subside to lower altitudes.
- An enhancement of the hydrological cycle by precipitation increases in the areas underlying flight corridors. A quicker initiation of the precipitation process is expected than would otherwise occur, because falling ice crystals can nucleate supercooled water clouds into which they fall (SMIC 71).

  Both processes are basically triggered by ice crystal growth in contrails.

Added cirrus clouds may have a warming effect at the Earth's surface (Arking 91), while drying of the upper troposphere and lower stratosphere may have a cooling effect at the Earth's surface (Lindzen 90).

In the following, interest is firstly directed towards the relative occurrence of contrails[20] and in the change in this occurrence that might be expected if air traffic increases. This should be known first before something can be said about the climatic implications of both effects, which is the ultimate goal.

An inadvertent enhancement of contrail formation will make it more difficult to attribute changes in contrail occurrence unequivocally to changes in air traffic.

Finally, the possibility of precipitation increases due to contrails will be discussed shortly.

[20] If aircraft fly into already existent clouds, holes may form (Zwart 69, 93). The occurrence of these holes, so-called distrails, can be explained by adiabatic cooling of the aircraft exhaust due to which the water present in the exhaust finally freezes. As a result, cloud water will be transported from the aqueous fase (cloud water droplets) to the ice fase. Thus, the ice crystals formed initially in the aircraft wake will grow, locally removing water from the cloud. Finally, they fall out of the cloud and a distrail forms. As there is no indication that this phenomenon occurs either frequently or on large spatial scales, distrails are not taken into account in the following.

4.3.3.2 When will contrails form and how long will they remain?
Appleman (53) studied the conditions under which contrails will form. If this information is combined with the fact that most high-flying aircraft fly at Flight Levels 290-390 can it be stated that contrails form if aircraft fly in air with temperatures of below -40°C. These temperatures prevail at and near tropopause level. For three areas near water in Western Europe, a high probability has been calculated for contrails to form in altitudes between 9.5-12k when radiosoundings for a day with contrail occurrence were inserted in Appleman's diagram for the forecasting of contrails (Betancor Gothe and Graßl 93).
Miake-Lye et al. (91), have applied the analysis of Appleman to the standard atmosphere (ICAO 64) as a function of altitude and latitude. From their results it can be estimated that aircraft flying at flight levels FL290 and lower, and at flight levels FL750 and higher, in general will not induce the formation of contrails. Between FL290-750, there are regions in which contrails will always occur and regions in which contrails are likely to occur. Note, however, that this is not a statical situation: for instance, a reduction of lower stratospheric temperature, as is predicted to occur due to the enhancement of the Greenhouse Effect (IPCC 90, 92), will strongly enlarge the region in which contrails may form.
Important with respect to the effects of contrails is how long they contrails persist. The residence time of contrails depends strongly on the relative humidity of the ambient air -the higher the humidity, the longer-lived the contrail- and, inversely, on the turbulence of the ambient air which causes mixing.
The relative humidity is higher in the troposphere, but the mixing will also be more rapid in the troposphere. Stressing the importance of the relative humidity, with present knowledge it is thought that the upper troposphere is more sensitive to contrail formation than the lower stratosphere (Schumann 93).

From the above studies, it can be concluded that a large part of high-flying aircraft fly at flight levels suitable for the formation of contrails and that contrails formed will probably remain longer in the upper troposphere than in the lower stratosphere. In the following paragraph study results will be given that may underpin this conclusion.

4.3.3.3 Detection studies of contrails

Angell (90) studied the cloud cover and sunshine duration over the whole USA: the cloud cover in the period 1970-88 compared to 1950-68 increased by 3.5%, but the sunshine duration decreased by only 1.2% in the same period. From this, Angell concludes that the observed additional cloud cover may largely consist of thin cirrus clouds which are too thin to turn off the sunshine recorder, but thick enough to be counted as cloud cover. More evidence, direct and indirect, for an increase in cirrus cloudiness has been reported by references cited in the same article.

Liou et al. (90) detected a significant increase of the mean annual high-cloud cover over Salt Lake City, Utah: 19.6% in 1965-82 compared to 11.8% in 1948-64. According to Liou et al., this increase appears to coincide with the jet aircraft traffic increase during that period. Analysis of the annual surface temperature for Salt Lake City shows a noticeable increase on the mean annual basis for the period 1965-82 of about 0.06°C/yr. The correlation between the two trends appears to suggest that the temperature increase could be caused by the increase in high-cloud amount associated with jet aircraft traffic during that period.

Changnon (81) finds a sharp anomalous increase in cloudiness and a decrease in sunshine duration in the central sections of the Midwest of the USA from about 1960 to the end of the study period, i.e., 1977, and he correlates this with the increasing fuel consumption by air traffic. Moreover, at weather stations in the east to west directed area of the central Midwest, moderated temperature occurrences (i.e., less extreme minimum and maximum monthly averages) have shown a sudden upswing since 1960, largely in fall and summer. This relates well with the decreased sunshine and increased cloudiness noted in this area and period, the last effect having occurred largely in fall and summer also. Changnon concludes his findings by saying that although the anomalous change in cloudiness is restricted to that area of the Midwest where jet traffic is the largest (more than 345 flights daily of high-level jet aircraft in 1979 over an area of about 400 km x 950 km), it may still represent only a natural fluctuation of sky conditions over the region.

Reinking (68) has measured a sunshine duration reduction of 20% on the first of two consecutive days. The measurements were made at the same time and in nearly identical weather situations with as major difference that on the first day thin cirrus was cast over the region. Contrary to the findings of Angell (90), in this case the thin cirrus must have been thick enough to turn off the sunshine recorder. More results of studies about changes in sunshine duration and the persistency of contrails are presented in Schumann and Wendling (90).

Processed satellite data on contrail coverage were presented in two studies. For the area between Frankfurt and Genua, the area fraction of contrails on average over 142 days from 12 months in the period 1989-1991 amounted to 0.4% according to Schumann and Reinhardt (91).

Bakan et al. (93) assessed longterm changes of contrail cloud cover over Europe and the eastern part of the Atlantic Ocean, i.e., the region 30W-30O and 35-75N. The analysis covered two periods of a few years duration, i.e., the periods 1979-1981 and 1989-1992, separated by one decade. The average value for the whole scene for the entire analysis period is 0.5%, with a maximum contrail cloudiness of almost 2% over western Europe and the Eastern North Atlantic along the NAFC. No significant long term trend could be detected for the area as a whole: average cloudiness decreased over central Europe and increased over the North Atlantic Flight Corridor.

Information about contrail life-time was also provided by Bakan et al. Only 2% of the contrail areas (regions with a larger group of contrails and a typical diameter of 1000 km) could be followed for less than about 6 hours, 62% of the contrail areas could be followed for more than one day, and 24% for more than two days.

If the annual mean unperturbed coverage of cirrus above the area of interest is considered as being 10 to 20% {e.g., Woodbury and McCormick (86), Koenig et al. (87), and London et al. (91)}, the increase in cirrus coverage which can be inferred from both studies is 2 to 20%. Such a change can result in a significant regional enhancement of radiative forcing towards the Earth's surface (see next paragraph).

Table 4.1.: Detected climatic changes, possibly due to high-flying aircraft

Study:	Bakan et al. (93)
Area of interest:	30W-30O; 35-75N
Results:	*i*) Significant averaged contrail-cloudiness decrease in central Europe; *ii*) Significant averaged contrail-cloudiness increase over the NAFC; Both changes occurred in early summer only and apply to 1989-92 compared to 1979-81
Associated with:	Either a systematically changed flight pattern or secular changes in the atmospheric circulation pattern (Europe case); increased flight activities (NAFC case)
Study:	Angell (90)
Area of interest:	Whole USA
Results:	*i*) Cloud cover 1970-88 compared to 1950- 68: + 3.5%; *ii*) Sunshine duration 1970-88 compared to 1950-1968: - 1.2%
Associated with:	Increases in thin cirrus clouds
Study:	Liou et al. (90)
Area of interest:	Salt Lake City, Utah, USA
Results:	*i*) Mean annual high-cloud cover 1965-82 compared to 1948-64: + 66%; *ii*) ΔT 1965-82: 0.06°C/yr
Associated with:	Jet aircraft traffic increase
Study:	Changnon (81)
Area of interest:	Midwestern states of the USA
Results:	*i*) 10-yr clear day values 1968-77 compared to 1901-10: - 26%; *ii*) 10-yr cloudy day freq's 1968-77 compared to 1901-10: + 41%; *iii*) 10-yr sunshine values 1968-77 compared to 1921-30: - 3.5%; *iv*) sharp increases in months with moderated temperatures during 1961-70
Associated with:	Increased air traffic fuel consumption

Only studies based on relatively long records of climatic variables have been presented in this table. Only then can the statement "the climate has changed" put forward with some confidence.

Kuhn (70) made in situ radiometric observations, by measuring IR and solar fluxes directly above and under contrail sheets, to analyse the effects on the environment of any alterations in the radiation budget in regions of heavy jet traffic. It was found that a 500 m thick contrail sheet between 10.7k and 12.2k increases the IR emission below the sheet and decreases the solar power below the sheet. The IR increase cannot make up for the decrease in solar radiation, resulting in a net decrease in radiative forcing of 12% at the contrail base. This results, in turn, in a 0.15°C decrease in the surface temperature beneath contrails with 5% contrail persistence, i.e., contrails are present for 5% of the time, and with 33% cloud cover on annual basis. This result is the opposite of the results presented above. A possible explanation for this is that the sheet was optically thicker than average. In that case a contrail produces a cooling effect, rather than a heating effect.
From analysis of observations of a specific contrail, a cooling effect was also found by Schumann (92). In this case, however, the cooling effect was not directly detected but calculated with a simple radiative transfer model. These contradicting results illustrate the complexity of estimating the climatic effects of (increases in) clouds. Another way of estimating the climatic effects of contrails is performing calculations with the help of more elaborate radiative-transfer models. In the following paragraph, results of this kind of studies will be given.

4.3.3.4 Modelling studies on contrails

Cirrus clouds, which contain a significant amount of large, non-spherical ice crystals in low concentrations, are normally optically thin and non-black. Because of their high location in the atmosphere and low reflection of the incoming solar fluxes, it has been physically recognized that the presence of cirrus clouds will normally produce a net increase in radiative forcing compared to a cloudless sky.

This results from the fact that the Greenhouse Effect of cirrus clouds, i.e., their netto downward IR forcing, is larger in magnitude and opposite in sign compared to their mitigating influence on solar radiation, i.e., their albedo effect (Arking 91). In the following, it is assumed that contrails exert the same kind of behaviour on the radiative balance as cirrus clouds, though recent results show that optical properties of contrails may in fact be very different from cirrus (see next paragraph).
What is of interest for this study, is the change in the radiative effect of the steady-state cirrus coverage due to (increases in) the occurrence of contrails as was described in last paragraph. To date, all pertubation experiments involving an increase in high-cloud cover indicate increases in atmospheric and surface temperatures, caused by a positive greenhouse feedback from the initial increase in high-cloud cover and a further amplification in high-cloud cover increase by increasing specific humidity (Liou 92). Three important model studies will be presented to provide some quantitative information on this topic.
Liou et al. (90) performed some calculations with a 2-D cloud climate model which included both long-wave and short-wave cirrus cloud effects to investigate the effect of an increase in contrail cover on cloud formation and surface temperature. Three experiments were carried out for the region between 20-70N, roughly corresponding to the region where high-flying air traffic is concentrated: a 5% increase of high clouds with all cloud covers and liquid water contents (LWCs) fixed in the model (case 1) and a 5% (case 2) and a 10% (case 3) increase of high clouds with interactive cloud covers and LWCs generated from the model. In all three cases, surface temperatures increase across the latitudes where cloud cover perturbations take place and, thus, specific humidity changes, i.e., a positive greenhouse feedback.

For the case in which the clouds are fixed in the model (case 1), surface temperatures increased by 2-6°C with the largest increase occurring at 60-70N. The surface albedo feedback included in the model used turned out to be the primary reason for this increase. When the cloud formation is interactive in the model the surface temperature is about 1°C (2.5°C) in the case of 5% (10%) high cloud cover increase but varies with latitude. The decrease in the sensitivity of the temperature between case 1 and case 2 is due to the reduction of surface albedo feedback.

Arking (91) studied the radiative effect of changes in cloud parameters with the radiative transfer model and cloud parameterization of Peng et al. (82), and a cloud classification and parameterization based on annual zonal mean cloud cover data. Arking calculated a globally averaged annual mean change in the net radiative flux, i.e., both short-wave and long-wave effects taken into account, into the Earth-atmosphere system from a 10% increase in cirrus cloud amount of 1.7 Wm^{-2}. This is of comparable magnitude to the effect of a CO_2 doubling on the net flux into the Earth-atmosphere system when equilibrium is reached, i.e., an increase of 4 Wm^{-2}. To put the effects of an increase in cirrus coverage in perspective: a 33 hPa reduction in altitude (for fixed temperature profile) produces the same change but with opposite sign. Reducing the emissivity, i.e., the ratio of the radiation emitted by a surface to that emitted by a black body at the same temperature, from 0.8 to 0.65 causes a reduction in radiative forcing of 3.7 Wm^{-2}.

Betancor Gothe and Graßl (93) calculated infra-red fluxes and heating rates for an atmosphere with a 500 m thick contrail. At a given ice content of 0.004 g/m^3, a far stronger atmospheric warming is found for a contrail with relatively small ice crystals than for a cirrus uncinus with large crystals: more than an order of magnitude stronger in the lower cloud half (10 K/day versus 0.8 K/day).

With increasing ice content both effects get stronger in an absolute sense, while the differences become smaller in a relative sense (88 K/day versus 10 K/day for an ice content of 0.05 g/m^3). However, ice clouds reflect, for a given ice content, stronger in the solar spectral range if they contain smaller crystals. Thus, contrails will reflect more sunlight (albedo effect) than regular cirrus. Due to the absence of specific model calculations in this spectral range, Betancor Gothe and Graßl could not determine to what extent the albedo effect is enhanced by the contrails found. This effect will compensate for the enhancement of the Greenhouse Effect by contrails to a yet unknown extent.

4.3.3.5 Precipitation changes due to contrail occurrences

Contrails start as water clouds and are rapidly transformed into ice clouds. The size of the contrail ice crystals varies depending on the water vapour saturation pressure over ice and on the evolution of the contrail. To study the possibility of seeding by falling contrail crystals, information about the exact size of the ice crystals occurring in contrails.

The only in-situ measurements of contrail crystal sizes have been done by Knollenberg (72) and he observed average sizes of nearly 500 μm. There are, however, strong reasons to expect significantly smaller crystals.

First of all, shortlived water cloud droplet size spectra peak in the 2-10 μm range (Grassl 90). Secondly, there are more cloud condensation nuclei in the exhaust plume than in the ambient air, in which natural cirrus clouds form, leading to smaller typical water droplet sizes and thus smaller ice crystals. Thirdly, from a comparison of simulated data and real data, Betancor Gothe and Graßl (93) derived that optical properties of contrails can only be simulated with crystal size distributions not typical for natural cirrus clouds; for calculations of contrail heating rates they used distributions with effective radii of 4 and 6 μm.

From this information, it is concluded here that it is unlikely that precipitation increases under areas with persistent contrail occurence will be measured or occur on significant scales. The argument for this is as follows:

- owing to the small crystal sizes the falling speeds of the ice particles are low,
- the distance that has to be covered until a particle reaches a supercooled low cloud is generally relatively large, and
- the chance that a supercooled low cloud is present underneath a contrail at the right time is probably not so large.

Furthermore, from the observation that contrail areas in general remain more than one day (Bakan et al. 93), it could be deduced that relatively few ice crystals grow to the critical mass which is needed in order to fall down. In addition, even if they do, the possibility remains that the ice crystals during their trip downward simply sublime or melt and subsequently evaporate.

Taken altogether it does not seem realistic to expect significant increases in precipitation in areas under contrails. Next to difficulties of (micro-)physical origin, this may explain why modelling of precipitation changes due to increases in flight movements, now and in the future, has not been done.

4.4 AEROSOL EFFECTS

4.4.1 INTRODUCTION

SO_2 and soot emitted in the ambient air form aerosols, which can be defined as dispersed systems containig solid or liquid particles suspended in a gas. SO_2 form aerosols by oxidation to sulfates. Soot particles form aerosols mainly by condensation and coagulation processes. The impact of aerosol particles on the radiation budget is manifold, either directly through scattering and absorption in the solar and thermal infrared spectral ranges or indirectly by modification of the micro-physical properties of clouds which affects their radiative properties. Thus, (increases in) aerosols from aircraft could have climatic implications.
Both aerosols could also influence the radiative balance via heterogeneous chemical processes influencing the distribution of radiatively active trace gases like, for instance, ozone. But as the processes likely involved have not yet been assessed thoroughly, this feature of aerosols will not be dealt with in this study.

4.4.2 AIRCRAFT RELATED TRENDS IN THE ABUNDANCE OF ATMOSPHERIC AEROSOL CONTENT

The total mass of non-volcanic stratospheric sulfate aerosol may be increasing at a rate of about 5% per year (Hofmann 90). Measurements indicate that during the last decade the aerosol optical depth over the Northern and Southern Hemisphere increased by about 40-50% (WMO 91).
The increase in non-volcanic stratospheric sulfate aerosol is believed to result from an increase in sulphuric acid in the stratosphere, possibly derived from sulphurous gases emitted naturally and anthropogenically at the surface. The most likely candidate, carbonyl sulphide, is not increasing sufficiently to explain the apparent increase in the background stratospheric sulphate {Rasmussen cited in Hofmann (91)}.

Additional sources must therefore be responsible for the increase. One of these could be sulphur emitted in the exhaust of jet aircrafts flying in the 8-15k region. Performing the following simple calculation {method taken from Hofmann (91)}, adapted for the situation in 1990} gives an indication of the probability that the stratospheric sulphate increase is due to high-flying air traffic. The current mass concentration of the background stratospheric aerosol is about 0.025 $\mu g/m^3$ of sulphur for the approximately 0.75 H_2SO_4/0.25 H_2O composition (Hofmann 90). For a global layer 5k thick and a lifetime of 1 year, which is typical for volcanic stratospheric aerosol, a yearly sulphur source of 0.06 Tg is required to sustain the current background aerosol concentration[21]. World-wide aviation fuel consumption was 176 Tg in 1990 which, for a sulphur concentration of 0.055%, amounts to a sulphur source of 0.097 Tg. In this study, it is assumed that about 30-50% of the air traffic takes place in the stratosphere, giving a direct sulphur source of about 0.029-0.048 Tg/yr. Of the remaining 0.07-0.05 Tg, about 30% is emitted above 8k. A part of this will enter the stratosphere through dynamical processes. This amount, which is latitude and model dependent, may be as large as 25%. This would result in a total stratospheric sulphur source of about 0.034-0.052 Tg/yr. As these values are 57-87% of the required source strength, high-flying aircraft appears to represent a stratospheric sulphate source to contend with.

This result is partly confirmed by model simulations by Bekki and Pyle (92). Their figures for 1990 aviation fuel use, the sulphur EI and the percentage of fuel burnt in the stratosphere resulted in a global 1990 stratospheric aircraft SO_2 emission comparable to this study's estimate, i.e., 0.049 Tg.

[21] Natural phenomena, e.g., volcanic eruptions, have as result that the background aerosol concentration shows large variations. Estimates of the background aerosol concentration are, thus, to a large extent uncertain.

They concluded that aircraft sulfur emissions may have represented a significant source of sulphate of 10-30% in 1990 in the stratosphere below 20k. The major discrepancy between their model results and measurements concerned, according to Bekki and Pyle, changes in aerosol size distribution.
For a global sulfur emission from stratospheric aircraft in 2025, which is based on the aviation fuel use scenario presented in Section 4.2 (fuel sulfur content remains the same as in 1990 and percentage of stratospheric fuel use is assumed to be 50% in 2025) and calculated with the Hofmann method, which results in 0.13 Tg S, Bekki and Pyle calculated perturbations in the background aerosol mass of up to 200% locally in the Northern Hemisphere compared to the 1979 atmosphere.

4.4.3 DIRECT AEROSOL EFFECTS

Radiative forcing calculations and simulations with climate models indicate that an increase in stratospheric aerosol content will exert a significant negative but due to the relatively short life-times of aerosols transient, i.e., short-lived, radiative forcing on the Earth's surface (IPCC 92). This should result in a small and temporary cooling effect, influencing the surface temperature record until a few years after the increase in aerosol content has occurred. Consequently, as long as the increase in stratospheric aerosol content continues the cooling effect will persist.
Black carbon aerosol (soot) has, in contrast to other aerosols, a uniquely high absorption cross-section. It dominates the absorption of light in most environments, and can play a role in radiative transfer and in aerosol effects on climate. In particular, it can offset a cooling attributed to anthropogenic sulfates and accelerate atmospheric warming by its greenhouse behaviour.
Quantitative results on these effects in relation to air traffic movements could not be found.

4.4.4 INDIRECT AEROSOL EFFECTS

In addition to the direct radiative effect of aerosols which form from SO_2 and soot emitted by aircraft flying in the stratosphere, it is also possible that aerosols might indirectly affect the climate by acting as nuclei for ice crystals in upper tropospheric cirrus clouds. Aircraft aerosols enter the upper troposphere directly from injections of SO_2 and soot by aircraft flying in the upper troposphere or they may enter the upper troposphere from the lower stratosphere by sedimentation and tropopause folding.

Jensen and Toon (92) showed that the presence of sulfuric acid aerosols may increase the concentration of ice crystals nucleating by as much as a factor of 5. The concomitant change in radiative properties may result in a significant increase in net radiative forcing (up to 8 W/m^2, resulting in surface warming) of cirrus near the tropopause. This indirect effect can be much stronger than the direct effect on a local scale. However, because contrails form in general over relatively small areas, on a local scale this indirect effect will be much weaker.

The same type of behaviour may be expected from soot particles. It is, however, difficult to determine whether soot aerosols may quantitatively have the same effect. Soot is emitted in quantities which are about two orders of magnitude smaller by mass. On the other hand, soot particles will probably be smaller, so hat they may be emitted in similar quantities by number. Incorporated in cirrus clouds and contrails, these soot nuclei like sulfur nuclei may alter the cloud optical properties, and thus their climatic impact.

Studies on these effects in relation with aircraft emissions have not yet been done.

4.5 EMISSIONS OF NITROGEN OXIDES

Concerning aircraft emissions during cruise, most interest has been addressed towards the inventory and effects of NO_x emissions. The reason for this is twofold.

i) Background NO_x mixing ratios are low at cruise altitude; it can be estimated by simple calculation that air traffic enhances NO_x abundances at these altitudes by orders of magnitude if no mixing would occur and if NO and NO_2 were inert gases with lifetimes of, say, 50 years (e.g., Tables 2.5 and 2.6 of this study, and Fabian 90).

ii) The Earth's radiative balance, which determines global mean surface temperature, is most sensitive to changes in tropospheric ozone at altitudes of between 8-12k (Figure 4.1), where aircraft emissions of nitric oxide are at a maximum and where model sensitivities of ozone to emissions of nitrogen oxides are enhanced.

Having made clear the importance of studies towards the effect of aircraft NO_x emissions on the ozone distribution and the subsequent translation of this effect into a radiative effect, one may observe at the same time that not much research has yet been carried out towards these indirect effects of NO_x emissions, let alone the climatic effect of NO_x emissions.

Johnson et al. (92) have calculated the decadal change of the surface temperature due to tropospheric ozone changes induced by aircraft NO_x emissions. Their figure of 0.01°C is of comparable order of magnitude as the change in surface temperature over 1970-80 due to observed CO_2 increases, i.e., 0.067°C, calculated with the same model. However, the global warming potential (GWP) of NO_x may be significantly reduced because of associated increases of tropospheric OH concentrations which may reduce mixing ratios of important greenhouse gases like HCFCs and methane.

Moreover, aircraft emissions between 0-8k also have been taken into account in the study of Johnson et al. For this reason, it is not taken into account in the conclusion of this section.

The photoactivity of NO_2 is of major importance to atmospheric chemistry in addition to that of ozone. NO_2 absorbs solar radiation throughout the entire visible and near ultraviolet spectral region. Thus, increases in NO_2 background mixing ratio may also have a direct radiative effect and, consequently, a climatic effect.
In Luther et al. (79), two mathematical expressions can be found which relate changes in stratospheric ozone and in stratospheric NO_2 to changes in surface temperature. These expressions can be used to estimate an approximate value for ΔT. For the stratospheric change in NO_2 due to high-flying air traffic (Table 2.5 on page 27), ΔT is in the order of -0.01°C. For the concomitant stratospheric ozone change as calculated by Hidalgo and Crutzen (77) (Table 3.1 on page 38), ΔT is in the order of -0.01°C. It should be noted at this point that the model used is old. Recent models will probably give different results. They could not be found, however. Furthermore, changes in NO_2 and O_3 are chemically strongly coupled to each other and to changes in H_2O by the ozone generation-destruction cycle. In other words, changes in atmospheric concentrations of water vapour, ozone and nitrogen oxides should not be calculated independently and without chemical models.

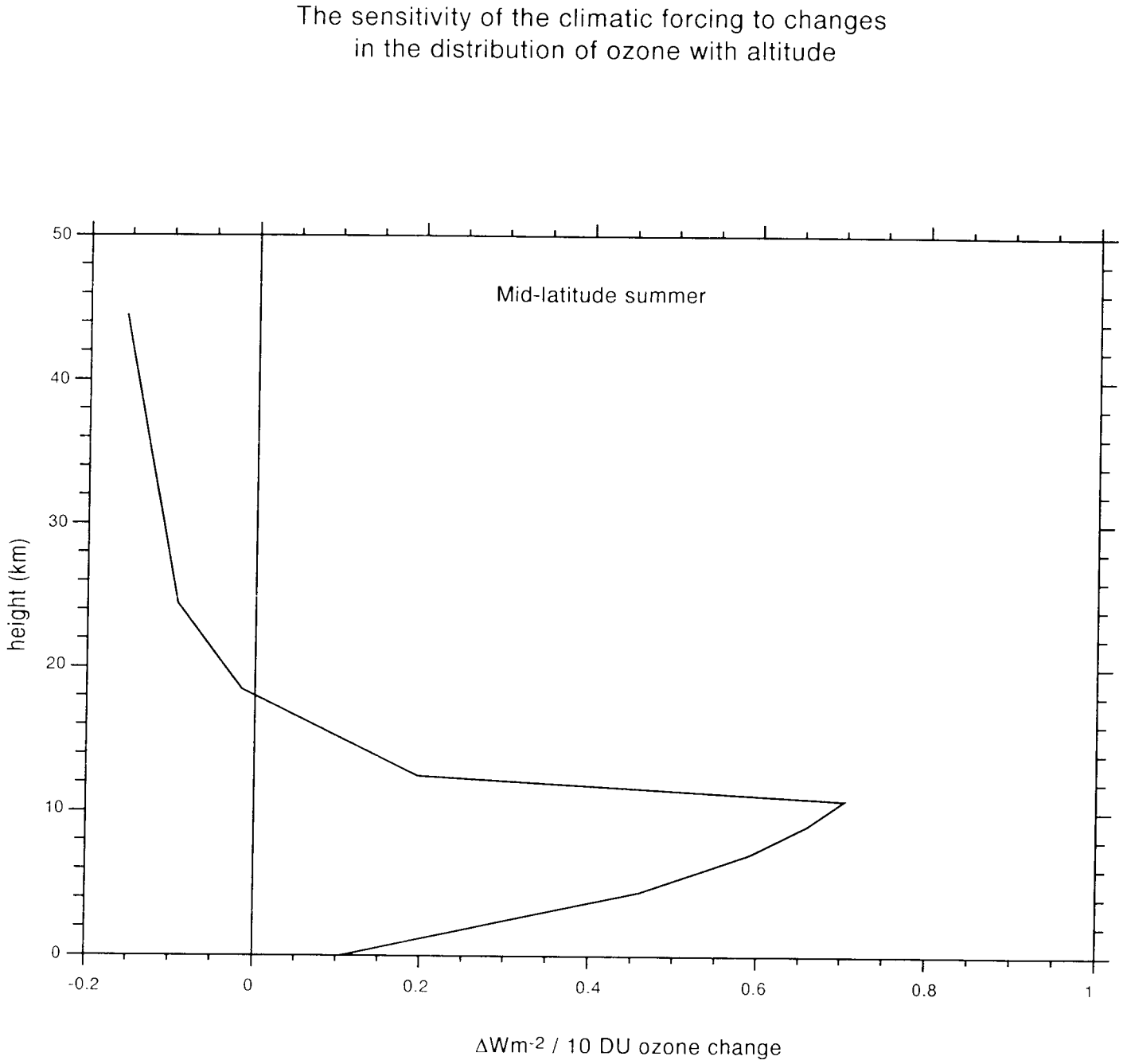

Figure 4.1.

The change in net radiative forcing at the tropopause due to changes in the vertical distribution of ozone as calculated with a 1-D radiative-convective model for mid-latitude summer under the assumption of fixed dynamical heating. The forcing due to a 10 Dobson-unit increment in one model layer is calculated. This is done for all model layers successively. No feedback effects are included in the radiative forcing.

According to the model used and for mid-latitude summer, ozone increases below about 18 kilometres in altitude warm the Earth; they cool the Earth if they occur above 18 kilometres. From the model calculations follows that the Earth's radiative balance is most sensitive to ozone changes around the tropopause (which is situated at about 12 kilometres during summer at mid-latitudes) where most modern jet aircraft fly during long-range cruise. This is nicely illustrated in the figure (results presented by courtesy of Paul Fortuin, KNMI).

4.6 EMISSIONS OF CARBON MONOXIDE AND HYDROCARBONS

No results have yet been published of studies on the climatic effect of emissions of either CO or HCs by aircraft. Nonetheless, atmospheric effects of these gases are well documented and some characteristics of those gases will be given here.

CO is not radiatively active in the infra-red; it will, thus, not have a direct effect on climate. Changes in CO concentrations, however, may in the presence of NO influence the tropospheric O_3 distribution. In addition, it may affect tropospheric hydroxyl concentrations. CO could thus indirectly affect concentrations of methane and some of the halogenated hydrocarbons. By acting in this way may CO exert an indirect radiative effect.

Hydrocarbons in the stratosphere react with hydroxyl as their primary sink. Some of the hydrocarbons have strong longwave absorption bands, but current atmospheric concentrations are too small, i.e., 1 ppb or less, to be important. Substantial increases in global emissions of hydrocarbons could lead to changes in the tropospheric ozone distribution, as well as affecting tropospheric and lower stratospheric HO and CO concentrations. Three HCs are presented in Table 1.1: ethene has not yet been detected in the stratosphere; formaldehyde and methane play a role in ozone chemistry.

4.7 DISCUSSION AND CONCLUSION

High-flying air traffic has the potential, by its scale, its predicted growth, and the physical and chemical characteristics of those areas in the atmosphere where kerosine is burnt in substantial amounts, to induce considerable perturbations in background concentrations of many trace gases (see chapters 2 and 3). These perturbations can influence surface temperatures via a change in the Earth's radiative balance. Based on studies published on this matter and on calculations with respect to CO_2 and SO_2 emissions presented in this chapter, the following is concluded:

- carbon dioxyde emissions

CO_2 emissions by air traffic are significant compared to total CO_2 emissions from all fossil fuel combustion processes. This situation will remain in the foreseeable future. From this information, values given in Section 4.2 and information provided by IPCC 92 has been calculated that for IPCCs' most widely used scenario, i.e., the IPCC-IS92a scenario, CO_2 emitted by aircraft alone may be responsible for a significant fraction of the committed climatic change[22] in 2025. This change for the arbitrary chosen year 2025 is a result of the accumulation of anthropogenic emissions of all greenhouse gases combined.

- water emissions

H_2O emissions by aircraft may have important climatic implications as far as increases in high-cloudiness are concerned. Compared to CO_2 effects, the implications of high-cloud cover increases for the radiative balance will be more severe. On the other hand, they will be of a more local character, because increases in high-cloud coverage will be restricted to those areas were high-flying air traffic is dense.

[22]The committed climatic change is the climatic change that would occur instantaneously if the time lag of the climatic system would be zero.

It is currently believed that a persistent increase in high-cloud coverage due to contrails leads to a net warming of the Earth's surface. According to both detection studies and model calculations presented in paragraphs 4.3.3.3 and 4.3.3.4, a substantial increase in net radiative forcing from contrail coverage due to high-flying aircraft may already have occurred in central Europe and the eastern part of the North Atlantic Flight Corridor. This radiative effect will be of the same order of magnitude, though confined to a smaller area, as the one induced by a CO_2 doubling with respect to pre-industrial concentrations. To put things in a timely perspective: though a radiative effect of this magnitude may already have occurred locally due to contrail coverage, the CO_2 doubling should not occur until the first decades of the next century according to the IPCC IS92a scenario. A radiative effect of this magnitude will lead to a climatic effect.

With today's emissions of water in the stratosphere, high-flying aircraft may contribute significantly to a warming of the Earth's surface as added water influences the radiative balance by its greenhouse behaviour, which is strongest where ambient temperatures are lowest, i.e., at tropopause level. If stratospheric water emissions by aircraft remain at the 1990 level, their long-term global warming effect may be several percents of the warming induced by a CO_2 doubling, given several assumptions (Section 4.3.2) and assuming the IPCC best-estimate climate sensitivity.

In addition, it could be stated that climate has already changed since the occurrence of high-flying air traffic in areas where this traffic is dense. Mean cloud coverage can be considered as a climatic variable and both the studies of Schumann and Reinhardt (91) and Bakan et al. (93) indicate that averaged cloud coverage due to contrails is 0.4 to 2% over relatively large areas and for longer periods. This statement will be of academic interest only. Society is primarily interested in how this change will influence ecosystems or human life. This information is not accounted for by the statement above.

- emissions of sulfur dioxide and soot

Simple calculations and model results indicate that SO_2 emissions by high-flying aircraft are contributing substantially to the stratospheric aerosol layer (Paragraph 4.4.2); the same can probably be said about soot emissions. No information about whether or not upper tropospheric aerosol is perturbed significantly by high-flying aircraft could be found.

Currently, it is assumed that the net direct radiative effect of an increase in background aerosol, due to high-flying aircraft or else, is negative. This negative effect is, in the case of aircraft aerosol emissions, probably composed of a smaller positive effect due to soot increases and a larger negative effect due to increases in sulfate aerosol. This implies a net cooling effect at the Earth's surface. However, as aerosols are relatively short-living, they do not accumulate in the atmosphere to the same extent as, for instance, CO_2 or CH_4. This results in a shorter duration of the radiative influence exerted on the climatic system by aerosols compared to the effect from longer living trace gases.

Aersols may exert an indirect net positive radiative effect by changing the optical properties of high clouds. Recent results indicate that this effect may be larger in magnitude than the direct net negative effect. However, this positive indirect effect, which will result in a warming of the Earth's surface, will exist only where, and as long as high-clouds occur. Consequently, it is more local and more short-lived than the direct effect.

Both the direct cooling effect and indirect warming effect associated with emissions of soot and sulfur dioxide by aircraft may be significant when globally averaged. However, both effects have not yet been quantified. Thus, it cannot be said at present what the net radiative effect will be and whether the Earth will warm or cool due to high-flying aircraft emissions of soot or sulfur dioxide.

- emissions of nitric oxide and nitrogen dioxide

The net direct radiative effect of NO_x increases related to enhanced absorbtion of solar radiation is probably minor.

It is yet unclear what the net indirect radiative effect of emissions of nitrogen oxides by high-flying aircraft is. These will perturb ozone background mixing ratios at tropopause level. Because the radiative balance is sensitive to changes in the abundance of ozone at tropopause level, this perturbation is expected to have a significant radiative effect. However, calculations with the help of radiative models to estimate this effect have not yet been done specifically for aircraft during cruise. Indeed, aircraft during cruise fly at altitudes where the cross-over between ozone destruction and formation, both due to NO_x emissions, is situated. So, to calculate the change in ozone due to aircraft during cruise, detailed emission inventories of high-flying air traffic, with a good spatial and timely evolution, should be used in chemistry-transport models. This has not yet been done. Because of this, even the sign of the change in ozone is unclear (Chapter 3). Consequently, no qualitative information, for the global situation as well as for the situation in a specific area, of the climatic effect due to NO_x emissions from aircraft during cruise can yet be given.

- emissions of hydrocarbons and carbon monoxide

Climatic effects due to emissions of either hydrocarbons or CO by high-flying aircraft have not yet been studied. These emissions are quantitatively of small proportions, but as they play a role in ozone chemistry it cannot yet be ruled out that their indirect climatic effects may be significant at cruise altitudes.

From the above follows that aircraft during cruise has the potential to induce significant radiative effects. These effects, however, differ in sign as well as in magnitude (compare for instance the direct and indirect radiative effects of aerosol increase). The lifetimes of the various perturbations show large differences. Consequently, perturbations have a longer lasting and, thus, more global (longer lifetimes, e.g., CO_2) or shorter lasting and, thus, more local (shorter lifetimes, e.g., contrails) impact. Not enough information is yet available, quantitatively as well as qualitatively, to give an integral overview on the climatic effect of emissions from high-flying aircraft. This counts for any preferred time or local scale.
The conclusion above is strenghtened by a recent study of Fortuin et al. (1994). This study is the first study in which the direct and indirect radiative effects of perturbations in background concentrations of several trace gases due to aircraft emissions have been calculated integrally. Emissions of carbon dioxide, water (both clouds and vapour), sulfur dioxide and nitrogen oxides (influencing ozone) have all been taken into account. From the calculations follows that aircraft emissions may have perturbed the Earth's radiative balance significantly since pre-industrial times, be it that the effect calculated for the situation in 1990 is opposite in sign during winter compared to the effect during summer. Fortuin et al. calculated an absolute radiative effect due to aircraft emissions of up to 20% of the enhancement of the natural Greenhouse Effect since pre-industrial times, which is about 2,5 W/m^2 according to IPCC (90). This may have occurred by either enhancing (up to 0.4 W/m^2 during winter) or mitigating (up to -0,5 W/m^2 during summer) the natural Greenhouse Effect.

5 GENERAL DISCUSSION AND CONCLUSION

Results on the atmospheric effects of high-flying air traffic from three different approaches have been presented more or less independently in the foregoing three chapters. The study has been started with possible perturbations of background mixing ratios due to aircraft emissions from simple calculation. Then, possible influences from aircraft emissions on the distribution of ozone as calculated with chemistry-transport models have been elucidated. The last chapter comprised possible influences on climate. Each chapter was completed with a conclusion. At this point the results and conclusions of the three chapters will be merged in order to come to a general conclusion with respect to the atmospheric effects of high-flying air traffic.

Perturbation calculations make clear that emissions of nitric oxide in particular, but also the emissions of soot and sulfur dioxide, have the potential to increase the respective background volume mixing ratios significantly. Especially in the lower stratosphere, where residence times are larger in general and where the ambient air is thin, the calculated increases are considerable from a chemical point of view. Large increases in background mixing ratios may change the patterns of prevailing chemical reactions, leading to yet unknown effects. For example, according to the calculation method used in Chapter 2 perturbations in background mixing ratios due to lower stratospheric emissions by high-flying aircraft at 1990 levels will be the very large for soot and sulfur dioxide (background mixing ratios of both components will be more than doubled), and for nitrogen dioxide (increase by a factor of a half). Effects of perturbations in background mixing ratios for these three species will be dealt with first, before aircraft emissions of other trace gases will be considered.

It is yet unclear what the magnitude of the net radiative effect of an increased burden of the aerosol precursors soot and sulfur dioxide at tropopause level will be. Theoretical estimates of the direct radiative effects of aerosols are strongly dependent on the assumptions made about the optical properties of the aerosols, but at present it is generally accepted that increased burden of lower stratospheric and upper tropospheric aerosol will have a net negative radiative effect leading to a net cooling of the Earth's surface. This cooling is probably composed of a smaller warming effect due to soot increases and a larger cooling effect due to sulfur dioxide emissions. Note that the radiative effect of aerosol increases will in general be of shorter duration than radiative effects due to increases in other trace gases, because the aerosols are relatively short-lived.

With regard to the indirect radiative effect of aerosols it can be stated that many (micro-)physical and chemical effects have not yet been fully elucidated. For instance, there are indications that aerosols may significantly change the optical properties of high clouds by acting as cloud condensation nuclei. This may lead to a strong positive radiative effect leading to a surface warming. Increased aerosol burden may also enhance high-cloud formation; current knowledge indicates that this will also result in a net positive radiative effect.

Aerosols may also influence chemical processes in which reactions in, and at the surface of aerosols play a role, i.e., heterogeneous processes. These processes have not yet been taken into account in studies with chemistry-transport models which consider atmospheric effects of aircraft emissions. Preliminary results indicate that the incorporation of heterogeneous chemistry in chemistry-transport models focussing on the stratosphere results in a lowering of ozone destruction by nitrogen oxides and in an enhancement of ozone destruction by chlorine oxides present in the atmosphere due to emissions of CFCs.

Thus, the net effect of the inclusion of heterogeneous processes on the magnitude of ozone depletion as calculated with homogeneous chemistry only is yet unclear. There are no indications, however, that the incorporation of these processes will alter the sign of the change in stratospheric ozone. This sign is negative, leading to depletion of stratospheric ozone. Whether and, if so, how the inclusion of heterogeneous processes may influence the formation of ozone in the upper troposphere due to aircraft emissions is yet unravelled. This leads to the conclusion that the indirect radiative effect of emissions of soot and sulfur dioxide via a change in ozone background concentrations by influencing atmospheric chemistry is unclear.

Mixing ratios of nitrogen oxides are also expected to be perturbed significantly by aircraft during cruise. However, the net long-term effect of NO_x increases by high-flying aircraft on global total ozone is yet unclear. This inconclusive result can for the largest part be attributed to the fact that aircraft during cruise fly in general at altitudes where the cross-over point between net ozone formation and net ozone destruction, both due to emissions of nitrogen oxides, is situated. To determine the netto effect on ozone both a detailed flight inventory with a good spatial and timely evolution, and sophisticated chemistry-transport models, in which tropospheric and stratospheric chemical and transport processes are well described, should be combined. This has not yet been done.
Another source of uncertainty with respect to emissions of nitrogen oxides can be attributed to lack of fundamental knowledge about variability in tropopause heights and about troposphere-stratosphere exchange processes. Both associated processes influence the distribution between the troposphere and stratosphere of aircraft emissions, and thus the fraction of NO_x emissions which will lead to either ozone destruction or formation.

Taking into account the above mentioned uncertainties and the large spread in model assumptions and results, which makes interpretation of the data difficult, the following conclusion concerning the effect of high-flying aircraft NO_x emissions on ozone can be drawn. A long-term change in the distribution of ozone due to aircraft during cruise is likely to occur, with a preference for a small, but just significant increase in Northern Hemispheric total ozone -in the order of 1%- if emissions remain on the 1990 level. This change is most likely constituted of an increase in upper tropospheric ozone (up to 10% with 1990 aircraft emissions) and a decrease in lower stratospheric ozone (up to 1% with 1990 aircraft emissions). Perturbations may be much larger locally and on the short-term, depending on the density of air traffic and the prevailing meteorological situation.
Although the net impact on total ozone will probably be rather small, the effect on the radiative balance may well be very strong. First of all, there is less uncertainty about the fact that due to aircraft NO_x emissions upper tropospheric ozone will increase, probably strongly, and lower stratospheric ozone will decrease. It is also known from theory that the Earth's radiative balance is very sensitive to changes in the ozone distribution at tropopause level. Radiative-convective models have calculated that the above kind of perturbations may result in a relatively strongly enhanced radiative forcing towards the Earth's surface. This will on average result in a warmer climate.

Comparing the calculated perturbations in background mixing ratios for all exhaust components may lead to the conclusion that CO_2 and H_2O emissions are of less importance. However, we have to keep in mind that both species are strong greenhouse gases and, thus, that changes in the background mixing ratio of both gases influence the Earth's radiative balance.

If CO_2 emissions by aircraft were considered only, it would have been concluded that the contribution of high-flying aircraft to the total committed climatic change due to the combined emissions of all greenhouse gases from all anthropogenic influences combined will be significant, i.e., in the order of one percent, in the near future.

Water emitted in regions of the atmosphere where temperatures are low, for instance at altitudes between 8 and 15 km, and where the air is subsaturated with respect to ice, can exist in all possible phases. It can remain in the gas phase as (water) vapour, in the liquid phase as (liquid) water and in the solid phase as ice. This adds to the complexity of the evaluation of the amounts emitted by high-flying aircraft. Put simply: if no contrails are visible, the water emitted is in the gas phase as vapour, while if contrails are visible the water emitted is, at least partly, in the solid phase as ice. During the evolution of water vapour into ice crystal it will also pass time in the liquid phase.
Contrails are high-clouds and even minute changes in high-cloud cover or occurrence, emissivity or height, may result in significant influences on the Earth's radiative budget and, thus, on climate. In other words, when considering water emissions, calculating the radiative effect of changes in water background mixing ratios only is not conclusive if one wishes to predict the atmospheric effect of these emissions. This is stressed by the fact that studies done towards climatic effects of high-flying aircraft have relatively often considered the influence of contrails on the radiation budget.

Modelling studies indicate that enhanced high-cloud coverage leads to a net enhanced radiative forcing. The radiative effect and the temperature effect calculated from a 10% increase in cirrus coverage may locally be of the same order of magnitude as the effect which can be expected globally from a doubling of the CO_2 mixing ratio from the pre-industrial value of 280 ppm to 560 ppm, which will occur halfway next century under the IS92a scenario (IPCC 92)[23].

Recent detection studies indicate that above central Europe and the eastern part of the North Atlantic (where air traffic is relatively dense) at least 0.4% of the sky is covered with contrails on a yearly average basis. Over a smaller area this value may increase to 2%; over large areas pronounced maxima during spring and summer of 2% have been detected. Based on this information, and if high-cloud coverage is accepted as a climatic variable, it can be stated that it has been proven that high-flying aircraft have changed climate. This statement is of pure academic interest, however, as society is primarily interested in the impact of this change on ecosystems and human life.

If we combine the information about high-cloud coverage due to contrail formation with the fact that average zonal cloud coverage by cirrus is 10 to 20% over the area of interest, can it be stated that the increase of cirrus cloud coverage by contrails may range from about 2% to about 20%. According to the above mentioned result from a radiative transfer model, the enhancement of radiative forcing since the beginning of high-flying air traffic by the occurrence of contrails might locally already be significant.

[23] The climatic forcing associated with a doubling of carbon dioxide, i.e., 4 watt per square metre, will, with respect to the pre-industrial situation, manifest itself during the second decade of the twentyfirst century. This is because greenhouse gases other than carbon dioxyde are normally also accounted for when discussing a CO_2 doubling.

At present, there is no reason to believe that hydrocarbon and CO emissions by high-flying aircraft have a strong impact on atmospheric processes and, thus, on climate. But as these species are precursors for peroxy radicals, which play an important role in ozone chemistry, they may yet not be overlooked.

Summarizing, it can be said that high-flying air traffic is likely to have altered the chemical composition of the atmosphere at cruise altitude. The precise climatic implications of this perturbation are yet to be established.
At the same time can be said that there is circumstantial evidence that emissions of trace gases by high-flying aircraft may already have induced significant climatic effects by influencing, directly and indirectly, the Earth's radiative balance. Not much is yet known about these climatic effects, both qualitatively and quantitatively, but from preliminary assessments of the possible order of magnitude of the perturbation of the radiative balance follows that they may locally be of the same order of magnitude as those induced globally by all recent increases in the concentrations of radiatively active trace gases due to anthropogenic emissions at the Earth's surface taken together.
If the climatic effects due to high-flying aircraft will be clarified in an unequivocal way in the future it will probably be impossible to detect these which are of importance to ecosystems and human life. By the nature of the climatic system, relevant climatic effects like changes in precipitation, sunshine duration and temperature due to aircraft will prove to be inseparable from both natural fluctuations in these climatic variables and the changes in these variables due to anthropogenic activities other than air traffic.

6 NARROWING THE UNCERTAINTIES

In the foregoing, many uncertainties concerning the atmospheric effects of air traffic have been identified. What should be done to reduce those uncertainties in the most effective way? With respect to present subsonic air traffic, some processes can be identified which need to be resolved with priority:

- The evolution of contrails and their effect on chemical and radiative processes in the atmosphere.

As contrails may have a large, though local, influence on the Earth's radiative budget, (micro-)physical studies into the evolution of contrails and the optical properties of contrail ice particles, in addition to observational studies of contrail occurrences and properties will be rewarding. Moreover, contrail formation may significantly influence ozone formation in the upper troposphere (Lelieveld and Crutzen 90, 91) and in the lower stratosphere if contrails occur as Polar Stratospheric Clouds. Thus, a comprehensive quantitative description of ozone chemistry in clouds is needed.

- The assessment and modelling of stratosphere-troposphere exchange processes.

Extratropical cyclones, tropopause folds, gravity waves and deep convection all are responsible for the mixing of tropospheric and stratospheric air. These processes have not yet been elucidated comprehensively. As the uncertainty in the atmospheric effects of emissions at tropopause level can be attributed to a yet unknown extent to a lack of knowledge about how this emission will be distributed between the troposphere and stratosphere, exchange processes should be studied with priority.

- The development of comprehensive 3-D chemistry-transport models.

3-D models in which the dynamical and chemical processes in both stratosphere and troposphere are represented comprehensively, in which both spheres are fully coupled, and which have a high spatial resolution in the vertical at typical tropopause altitudes, i.e., between 8 and 16 kilometres should be developed. Such models are needed to calculate the overall effect of aircraft emissions on, for instance, ozone, and to study how emissions in flight corridors are distributed over the globe by large scale transport processes.

- The development of a model to study chemical conversions in the exhaust of aircraft in the plume region.

Chemical and diffusion processes in the plume, with timescales of up to a few hours, determine how the chemical composition of the ambient air is perturbed initially. This perturbation should be introduced into the larger chemistry-transport models, rather than the direct emissions by aircraft engines, determined by the respective emission indices, as they may cause large initial errors because of the large uncertainties attached. However, global chemistry-transport models cannot (yet) resolve these processes; it can thus be considered as a sub-grid-scale phenomenon which has to be parameterized. This paramerization should be based on a sound description of what is happening in reality. For this, a model of plume effects is needed.

Two further points of interest should also be mentioned. Firstly, the assessment of contrail climatology and the subsequent incorporation of this climatology in current GCMs in a perturbation run in order to quantify the climatic effects of contrails. Secondly, higher vertical resolutions at typical tropopause altitudes in current climate models should be aimed at.

A general point is that long-term scenarios on developments of air-traffic demand are needed from aircraft industries or from elsewhere. They are needed for the simulation of future climatic change due to aircraft emissions. Not many are available and one may ask whether they are reliable considering recent economical developments, affecting both airlines and the aircraft industries, and predictions on economic developments in the near future. For the same reason is more information needed about the likelihood of a future supersonic fleet and new propulsion technologies.

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