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Regional and Global Air Pollution from Aircraft

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REGIONAL AND GLOBAL AIR POLLUTION FROM AIRCRAFT

1 THE ATMOSPHERE

Aircraft exhaust can contribute to several air pollution problems, depending on the chemical and physical conditions in the part of the atmosphere where the emissions take place.

Based on temperature structure the atmosphere may be divided into several parts, as shown schematically in Figure 1. In the lowest region, the troposphere, the temperature falls with height. It extends through roughly the first 10-15 km, and is characterised by strong, vertical mixing caused by dissipation of heat from the surface. Currently commercial flights are mostly in the troposphere. A few flights take place in the lower part of the stratosphere, which is a 30-40 km thick layer above the troposphere. In the stratosphere, the absorption of ultraviolet light by ozone (O_3) causes warming and increasing temperature with height and suppresses vertical motions of air parcels.

Atmospheric motion is slowed down along the ground due to friction, and cooling or warming of the surface gives rise to a vertical flow of heat in or out of the overlying air. This drives the turbulence which carries heat as well as trace gases away from, or towards the underlying surface. The boundary layer has a variable depth ranging from a few tens of metres in stable conditions to about 2000 m in very unstable situations.

This mixing layer is a large reservoir for gases released from sources at the ground. Typically in middle latitudes the mixing layer can exist for several days (2-4 days) before it is broken down. Break-down may occur as a result of large scale weather phenomena, as e.g. when fronts are passing.

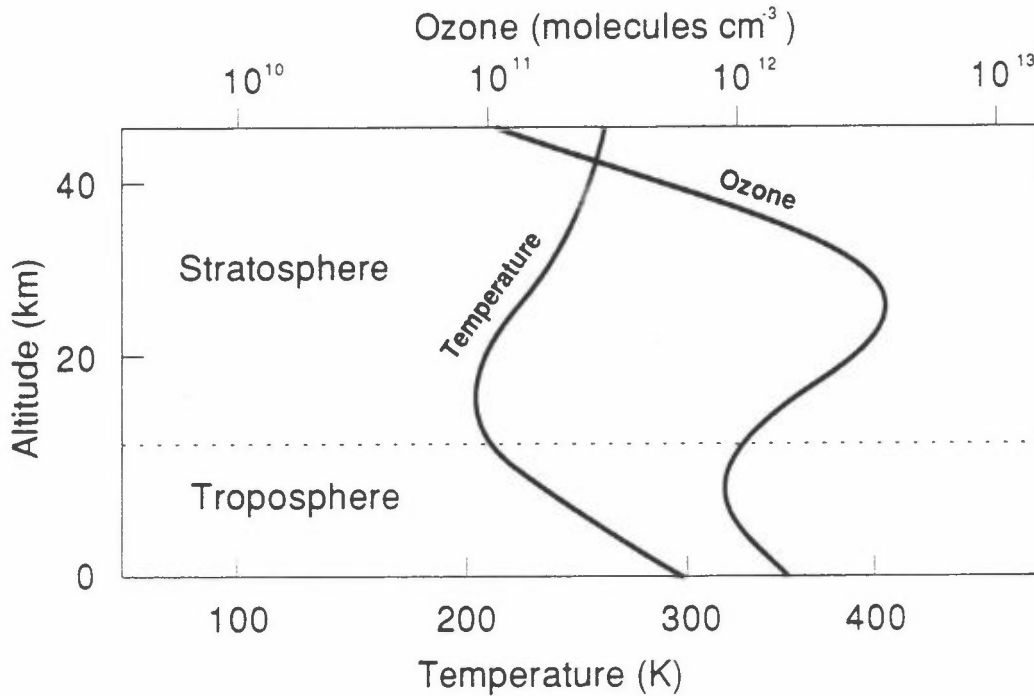


Figure 1: Temperature profile and distribution of ozone (molecules per cubic centimeter) in the troposphere and the stratosphere.

The depth of the boundary layer is largest over heated surfaces. During summer land surfaces are generally warmer than oceans, explaining the thicker boundary layer over continental Europe than over the oceans and the coastal regions (Figure 2). The situation is reversed during the winter when the oceans are warmer than most land surfaces (Figure 2).

Anthropogenic emissions may lead to pollution of the atmosphere on various horizontal scales. In this report only regional (defined here as scales above 100 km) and global problems are discussed.

Nitrogen is by far the most abundant chemical element in the atmosphere. Molecular nitrogen (N_2) constitutes slightly less than 80% of all the molecules in the atmosphere. It is stable under the temperatures and radiative conditions that prevail up to at least 100 km altitude. For this reason it does not take

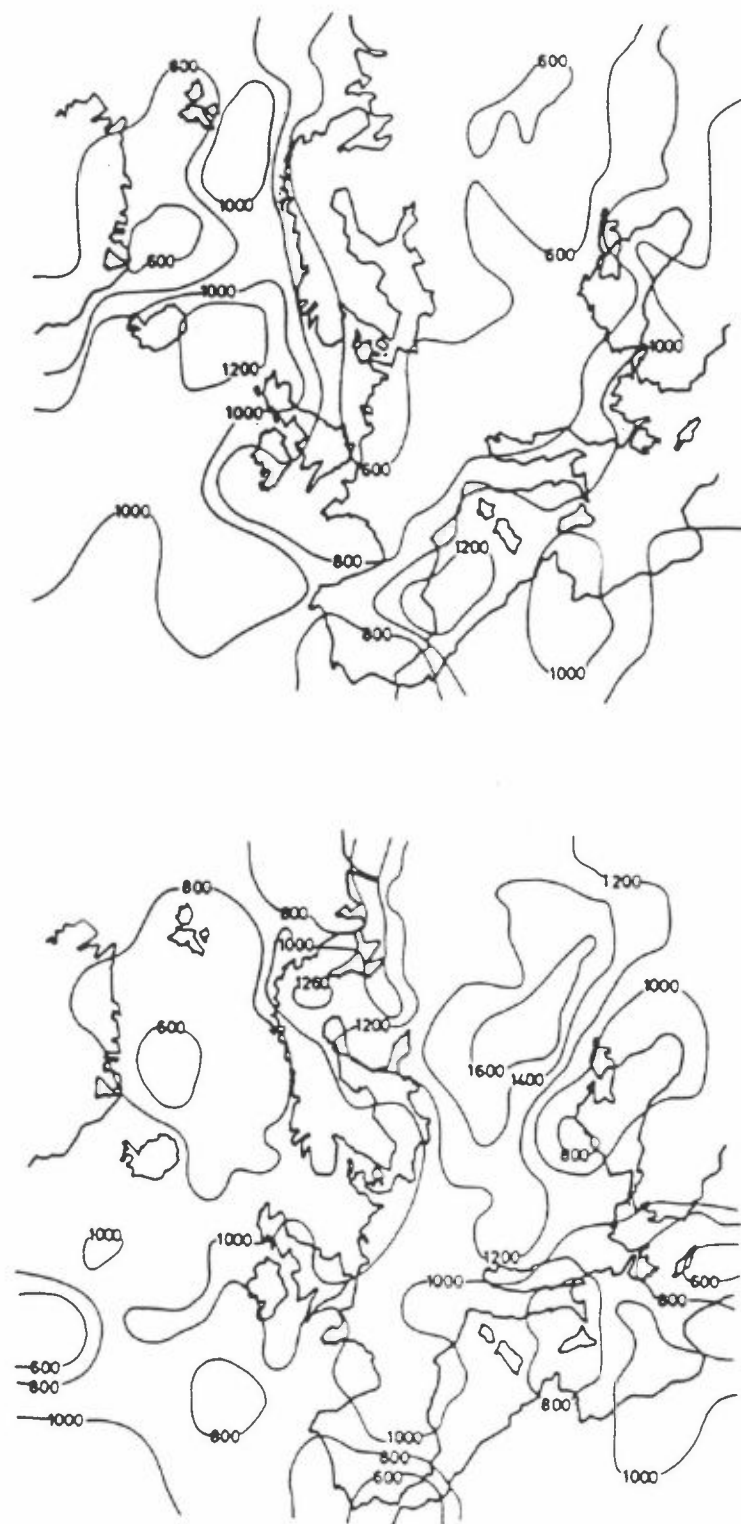


Figure 2: Average depth of the boundary layer at 12 GMT, February 1979 (upper) and June 1979 (lower), in meters, from Eliassen and Saltbones (1983), based on radiosonde data.

part in the lively photochemical activity involving e.g. the production and destruction of ozone, and the oxidation and decomposition of pollutants that are emitted into the atmosphere.

Several other nitrogen containing species, however, play important roles in the photochemistry of the atmosphere, including all the oxides of nitrogen (NO_x) and other oxygen-containing nitrogen compounds.

The second most abundant chemical element is oxygen. Molecular oxygen (O_2) constitutes about 21% of the atmospheric molecules. O_2 is also very stable in the troposphere and the stratosphere. A slow loss takes place in the stratosphere, where O_2 is dissociated by ultraviolet radiation, leading to the formation of a gaseous constituent which is of great importance; ozone. The concentration of ozone is very low compared to those of N_2 and O_2 . Most of the ozone is located in the stratosphere (Figure 1) in the so called ozone layer, containing about 90% of the atmospheric ozone. The ozone layer protects us from large doses of ultraviolet solar radiation, and it strongly influences the temperatures and the meteorology of the stratosphere.

The remaining 10% reside in the troposphere, and is mostly formed from atomic oxygen released from photodissociation of nitrogen dioxide.

2 AIR POLLUTION PROBLEMS

Perhaps the first air pollution problems in man's history were what we today term "smog" and acid rain. The Spanish philosopher Moses Maimonides (1135-1204) described the air in the cities as "turbid, thick, misty and foggy", problems that he blamed on "all that pours forth from its inhabitants and their superfluities". After the industrial revolution these problems increased due to the widespread use of coal for heat and energy production.

In our own century, a number of severe pollution episodes in London have been described. These pollution episodes occurred during weather conditions leading to shallow boundary layer well separated from the free troposphere above, hindering the dispersion and dilution of the pollutants. The pollution was characterized by high levels of sulphur dioxide and other pollutants in smoke and fog, now often called smog ("London smog").

Deposition of acids, e.g. in acid rain, is an important part of the sulphur pollution. It is now realised that emissions of nitrogen compounds are adding to this problem, as the increased acidity is due to nitric as well as sulphuric acid. High sulphate and nitrate concentrations in rain and atmospheric aerosols are now frequently found far away from large sulphur emissions. The acid rain problem is therefore not only a local, but also a regional problem especially in Europe and North America.

A different kind of air pollution was first observed in Los Angeles in 1944. During the 1950's it was established that non-methane hydrocarbons (NMHC) and NO_x in the presence of sunlight would form ozone (O_3) and other pollutants, which caused irritations of eyes, reduced performance of athletes, and various other symptoms. Vegetation was also damaged. Because sunshine is a necessary driving force in the formation of this pollution, the phenomenon is usually called "photochemical air pollution" or "photochemical smog", although neither smoke or fog are key components.

In the 1970's it was recognized that the photochemical oxidant and acid rain problems were linked together through a series of chemical reaction cycles, involving such species as methane (CH_4), a number of other hydrocarbons, carbon monoxide (CO), NO_x , hydroxyl (OH) and hydroperoxy (HO_2) radicals, O_3 and sulphur compounds.

O_3 in concentrations only a little higher than the natural levels is known to affect human health, vegetation and certain

materials in a negative manner. It also plays a key role in the oxidation and removal of several gases from the troposphere. Furthermore, it plays a significant role as a greenhouse gas in the troposphere and lower stratosphere. Changes in the tropospheric ozone concentration level on a global scale can therefore affect the chemical and physical conditions of the atmosphere in several ways and potentially also the conditions for life.

In 1895, S. Arrhenius - the Swedish physical chemist - drew attention to the fact that certain gases, as carbon dioxide (CO_2), absorb infrared radiation emitted at the Earth's surface, hindering energy to escape to space and increasing the temperature of our planet by a mechanism similar to the one heating a greenhouse. The heating of the atmosphere by CO_2 was therefore called the greenhouse effect. And in 1938, the British chemist R. Callendar was already able to demonstrate that CO_2 emissions from the combustion of fossil fuels accumulated in the atmosphere. It is now known that the atmosphere contain several greenhouse gases. The most important ones are water vapour (H_2O), CO_2 , CH_4 , nitrous oxide (N_2O), chlorofluorocarbons (CFCs) and O_3 .

The greenhouse effect of the atmosphere is a natural phenomenon caused by the presence of water vapour and trace gases. It leads to a temperature difference of approx. 30°C between the radiation temperature of the Earth/atmosphere system (-19°C) and the near-surface temperature (approx. $+15^\circ\text{C}$) during the current interglacial period. The greenhouse effect is essential for human life on Earth, because without it, large parts of the Earth would be covered with ice. However, currently the concentration of greenhouse gases like CO_2 , CH_4 and N_2O are increasing on a global scale, increasing the greenhouse effect and therefore threatening to lead to global warming and climate change.

In May 1985, a surprising paper, published in Nature by three scientists from the British Antarctic Survey in Cambridge (Farman, Gardiner and Shankin), reported that the ozone amount measured in spring (September and October) at the Antarctic station of Halley Bay had exhibited a dramatic downward trend since the mid 1970's. The measurements indicated that the October mean vertically integrated ozone concentration was 40% lower in 1984 than 10 years earlier. This information produced a great deal of speculation about the causes of this rapid and perhaps unprecedented change in the atmosphere. It had been known for 10 years that CFC's had a potential to reduce the ozone layer, but the models simulating the effects of these chlorinated molecules were not predicting such dramatic changes in the ozone layer as a result of the releases of CFC's up to 1985. Moreover, at that time, no dramatic change in the ozone amount had been detected at mid-latitudes so that the CFC/O₃ theory was still disputed by several scientists, industrialists and policy makers.

However, the springtime ozone concentrations measured by satellite over Antarctica later confirmed the existence of an ozone hole over the South pole region and showed that this hole covered the entire Antarctic continent, i.e., an area as large as the United States.

Data gathered during several expeditions organized since 1986 gave evidence that the amount of chlorine (which efficiently destroys ozone) is 100 times higher in the Antarctic regions during winter and early spring than at mid- and low latitudes. These elevated concentrations of reactive chlorine result from complex processes affecting chlorine-containing molecules on the surface of ice particles in polar stratospheric clouds. Such clouds are observed in the lower stratosphere at high latitudes in cold airmasses. Since the presence of chlorine results from industrially produced CFCs, the appearance of the ozone hole is one of the most dramatic change in the atmosphere resulting from human activities.

Recent analyses of satellite data, balloon borne instruments, as well as ground based observations of O_3 by Dobson spectrometers, show that there has been a downward trend in O_3 over the last decade also in the Northern Hemisphere, at high as well as at mid-latitudes. Scientists suspect chlorine from CFCs to be responsible for the decline in the ozone layer also in the Northern Hemisphere.

3 EMISSIONS FROM AIRCRAFT

Aircraft exhaust contains a mixture of several pollution gases as well as particles. Emission of NO_x is a result mainly of decomposition of atmospheric N_2 by reaction with hot combustion gases in the engine. The emission rate depends on the fuel which is used and the efficiency of the burning in the aircraft engine, which both in turn depend on characteristics of the engine. These factors are not precisely known. Instead, empirical emission factors are available which are based on measurements on a few gas turbines under different operating conditions.

Table 1: Total NO_x emissions from aircraft in Norway, Denmark and Sweden. Numbers are given as tons of NO_2 per year, including international flights.

		HC tons	CO tons	NO_x tons	CO_2 tons
Norway	(1989)	700	3700	4400	1500 000
Denmark	(1985)	800	4100	3200	
Sweden	(1989)	3000	8000	7500	2000 000

Complete burning of hydrocarbon (HC) fuel in aircraft jet engines, like in automobile engines, would yield only H_2O and CO_2 as combustion products. However, in reality it is very difficult to obtain an ideal burning, and as a result aircraft exhaust also contains unburnt HCs, CO, sulphur dioxide (SO_2) and nitric oxide (NO). SO_2 is emitted when the fuel contains

sulphur. CO emissions take place when there is a deficit of oxygen (O_2) or low pressure in the burning chamber (idling). Unburnt HCs are emitted when there is a relatively low temperature in parts of the engine's burning chamber. Emission of NO_x occurs from burning of fuels with N containing compounds (fuel NO_x formation), but in addition in all aircraft engines due to decomposition of atmospheric O_2 to atomic oxygen at high temperatures. In subsequent reactions, starting with a reaction between N_2 and the oxygen atom, two NO_x molecules are formed.

Emissions of NO_x , CO and unburnt HCs cause ozone formation. With the current knowledge, which is incomplete, about the amounts of the various compounds in the aircraft exhaust, model experiments show that NO_x emissions are by far the most important for ozone generation (Johnson and Henshaw, 1991). As O_3 is an efficient greenhouse gas in the upper troposphere and the lower stratosphere, NO_x emissions also lead to an increased greenhouse effect, which could be as important as, or even more important than, the increase in the greenhouse effect due to the CO_2 emissions from aircraft (results of model calculations by Isaksen et al., 1990 and Johnson and Henshaw, 1991, combined with results from IPCC, 1990). It is therefore appropriate to focus particularly on the NO_x emissions in aircraft exhaust.

In the Nordic countries Denmark, Sweden and Norway aircraft emit a total of approximately 15 thousand tons per year (as NO_2 , equals 4.5 kton N/yr) of nitrogen oxides (Table 1, from Knudsen and Strømsøe, 1990). About 75% of the emissions are estimated to take place at cruise altitudes, i.e. in the free troposphere and in the lower stratosphere. In Norway 2% of the man made emissions originate from civil aircraft. On a global scale civil aircraft contribute with 600 kton N/yr, constituting about 1% of the total man-made NO_x -emissions of 40 000-60 000 kton N/yr. Natural sources of tropospheric NO_x are quite uncertain, and are believed to be 10 000-20 000 kton N/yr from soils, 2 000-8 000 kton N/yr from lightning - most of which is emitted in the free troposphere, and 1 000 kton

N/yr transported from the stratosphere. These figures for the global emissions are from the ongoing Stratospheric Ozone Assessment by UNEP/WMO (1991).

NO_x -emissions from aircraft constitute only a small fraction of the global man made emissions. Also, since most of the aircraft emissions take place above the cloud level, they do not play any significant role in acid deposition. On the other hand, since the emissions take place in the upper troposphere and the lower stratosphere, they can have an effect on O_3 and the greenhouse effect which is disproportionally larger than expected from the moderate fraction of the man-made emissions that they constitute. Such effects are described in the following sections.

4 ACID RAIN

Nitrogen is emitted from aircraft as NO_x , mainly NO , which is rapidly converted to nitrogen dioxide (NO_2) by O_3 :



Nitrogen dioxide is oxidized by the major atmospheric oxidizing agent, OH , to form gaseous nitrate (HNO_3):



HNO_3 is easily solved in rain water, where it is dissociated, forming positive H^+ ions, thereby increasing the acidity of the rain water:



The deposition of nitrogen to the ground in precipitation is called wet deposition. Nitrogen can also be deposited (dry deposition) to the ground directly from the gas phase species,

through absorption by vegetation, soil surfaces or water bodies. The paths of NO_x leading to acid deposition (as dry and wet deposition) are shown in Figure 3.

Sulphur emitted from aircraft as SO_2 , in principle can contribute to acid deposition, in processes similar to those described above for nitrogen compounds. However, the sulphur emissions from aircraft are small and do not contribute to any significant acid deposition.

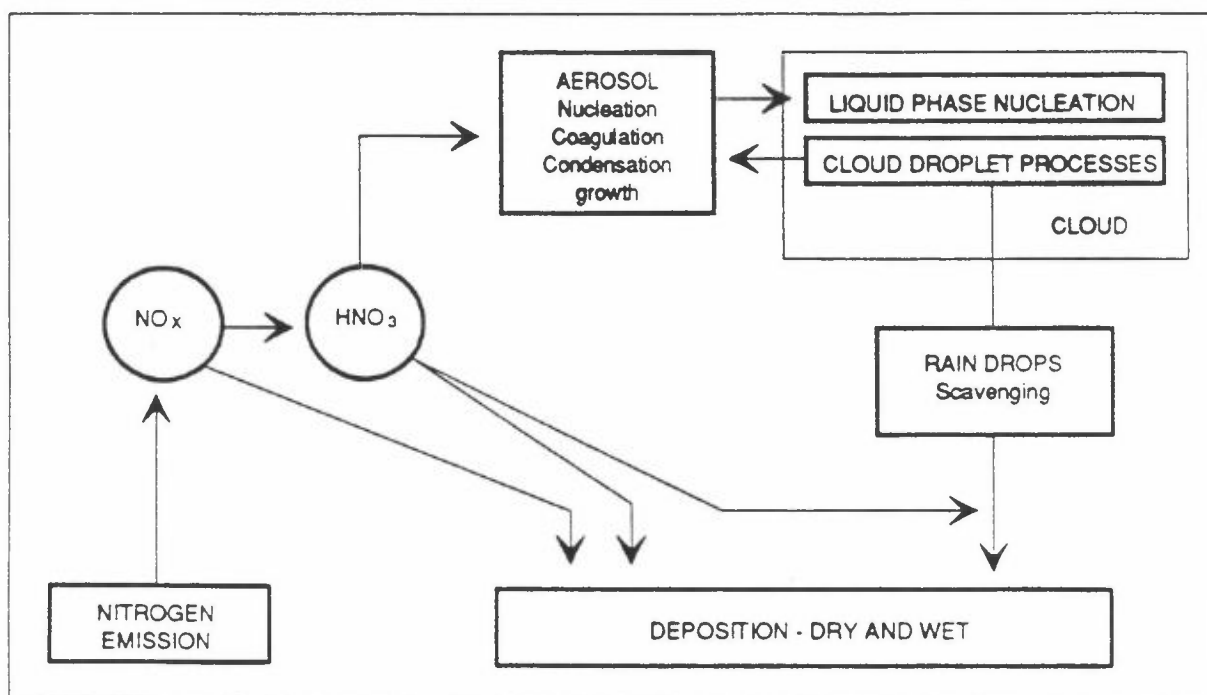


Figure 3: Atmospheric paths of NO_x leading to acid deposition.

Except in the tropics there are hardly any clouds in the upper troposphere. Nitrogen emitted at cruise levels in this altitude region does therefore not deposit directly to the ground. Still, after dispersion throughout the troposphere and also towards the ground, the nitrogen will be deposited. However, the deposition is distributed over large areas, and therefore does not contribute to a significant extent to the acid rain problem.

5 OZONE AND OXIDANT FORMATION IN THE TROPOSPHERE

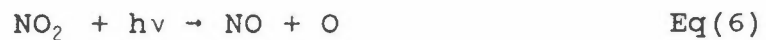
The chemical reactivity of the troposphere is driven by solar radiation. OH radicals, which play an important role in the oxidation of nitrogen (Eq 2) as well as several other compounds, are formed when O_3 is dissociated in the presence of H_2O . O_3 is dissociated by ultraviolet radiation (by a photon with energy $h\nu$, proportional to the frequency, ν , of the radiation)



forming an excited oxygen atom ($O(^1D)$) which reacts with H_2O to form OH:



O_3 is formed (section 4) in reactions following dissociation of NO_2



The oxygen atom, O, formed rapidly combines with O_2 to form O_3 :



The formation of ozone by this mechanism requires that NO is recycled to NO_2 so that the reactions (6-7) can proceed several times. The hydro peroxy radical (HO_2) and the methyl peroxy radical (CH_3O_2) (both formed e.g. from CH_4), can provide recycling of NO_2 in the reactions



A wide range of other peroxy radicals, denoted by the general term RO_2 , can also convert NO to NO_2 :



Such peroxy radicals are decomposition products of a variety of NMHC, emitted e.g. from aircraft. The reactions (6, 7 and 10) form a chain in which ozone is produced by the participation of NO_x and NMHC, which explains why O_3 is readily formed as a result of NO_x and NMHC emissions. The roles of NO_x and NMHC in the O_3 formation are different in the sense that NO_x is a catalyst (not consumed) while the NMHC are consumed. In the vicinity of aircraft, the emitted NO will initially deplete ozone, in reaction (1).

Tropospheric O_3 is central to the problem of the oxidizing capacity of the troposphere, and also the oxidation in the atmosphere as a whole, since photolysis of ozone is the primary source of the main oxidizing agent OH. Ozone is also in itself an efficient oxidizer of many chemical species. The oxidizing capacity determines the efficiency of the decomposition and removal of pollutants emitted into the atmosphere. NO_x , CO and CH_4 which are partly controlled by man made emissions, strongly influence the levels of O_3 and OH through a series of interconnected chemical reactions, some of which are explained above.

O_3 is produced in air containing NO_x in excess of ca 100 ppt through the reactions described above. In air with low concentrations of NO_x the situation is appreciably different in that reactions destroying ozone dominate over the production. A key loss mechanism for ozone is its reaction with HO_2 and OH:



which form a catalytic chain depleting O_3 without destroying OH and HO_2 .

As discussed above, man made emissions of NO_x over industrialized continents are the dominating source of NO_x . However, nitrogen oxides cannot be transported more than a few thousand

kilometers away from its sources, since their lifetime in the troposphere is only a few days. A large part of the troposphere is therefore presumably poor in NO_x , an assumption which is supported by measurements over the Pacific Ocean. An exception could be the upper troposphere, where lightning may constitute an important source of NO_x . Nevertheless, NO_x produced by aircraft engines in the upper troposphere can provide a significant source of NO_x , at least locally and regionally. As discussed below, significant ozone production is found to follow the aircraft emissions at these altitudes.

5.1 IMPACT OF NO_x IN THE UPPER TROPOSPHERE AND LOWER STRATOSPHERE

O_3 is formed more efficiently from NO_x in the upper troposphere than in the lower troposphere. Model calculations show that NO_x emitted from aircraft may form 20 times more O_3 than the same amount of NO_x at ground level (Isaksen et al., 1990; Johnson and Henshaw, 1991). Aircraft emissions may therefore have a potential to significantly influence the global oxidizing capacity in spite of its moderate contribution to the total manmade NO_x emissions.

Model studies of effects of aircraft emissions on tropospheric O_3 have been performed by about half a dozen groups. Recent results obtained with the global two-dimensional model of Isaksen and Hov (1987) shows that the present fleet of civil aircraft increase local concentrations of NO_x and O_3 by 65 and 7% respectively at the location of maximum influence, which is at the 10km level at 40°N (Figure 4, Fuglestvedt and Isaksen, 1991). Beck et al (1992) report increases of NO_x and O_3 to be 40% and 12% in a zonal two-dimensional (longitude-altitude) model, adopting somewhat different emission assumptions. The sensitivity of ozone to increases in NO_x at flight levels varies considerably between models, as demonstrated by a comparison (Figure 5) made by Johnson and Henshaw (1991). These differences are due to differences in the representation of the

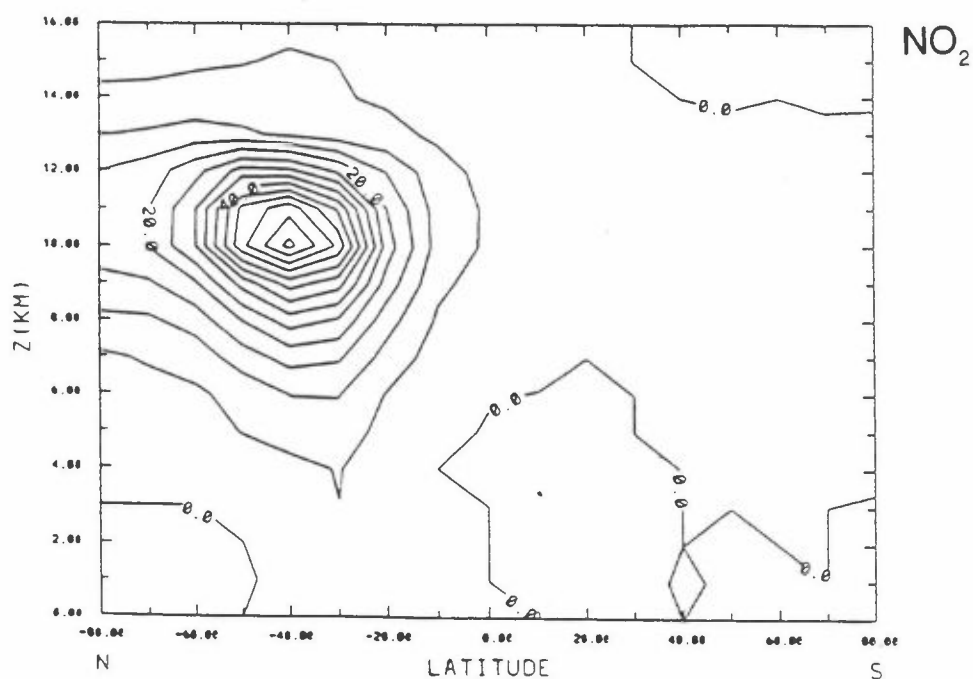
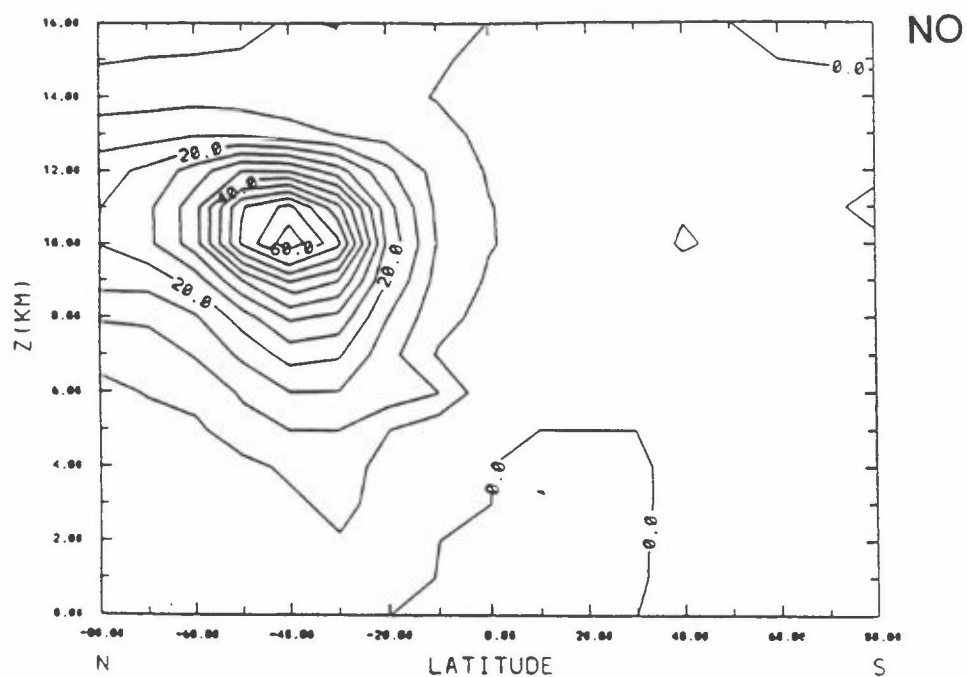


Figure 4: Changes in the concentrations (%) of NO_x, O₃ and OH at various latitudes and altitudes resulting from the current fleet of civil aircraft, as resulting from the model of Isaksen et al. (1990). The results are from Fuglestvedt and Isaksen (1991).

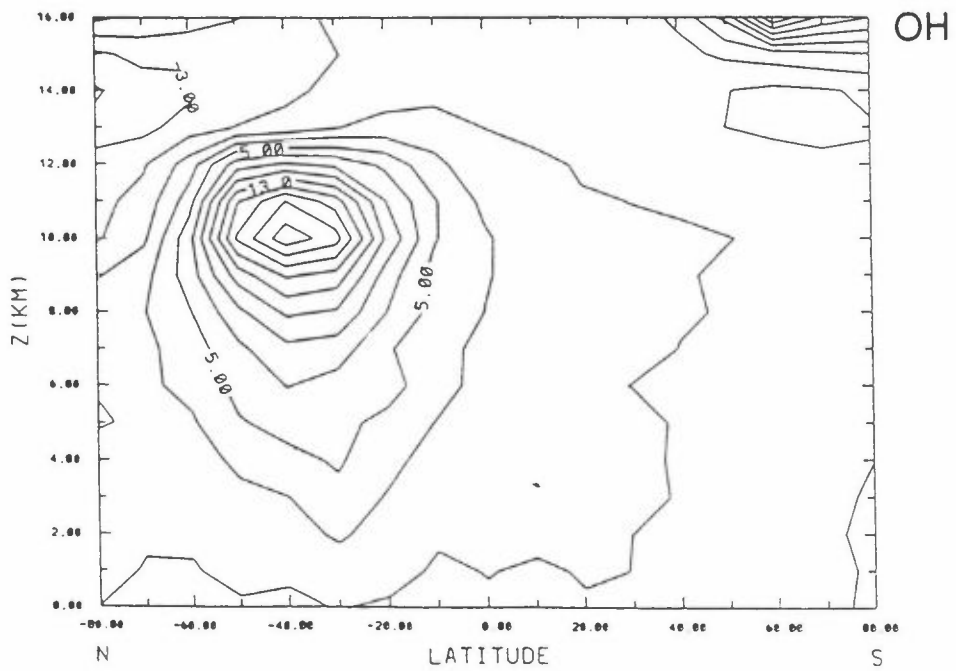
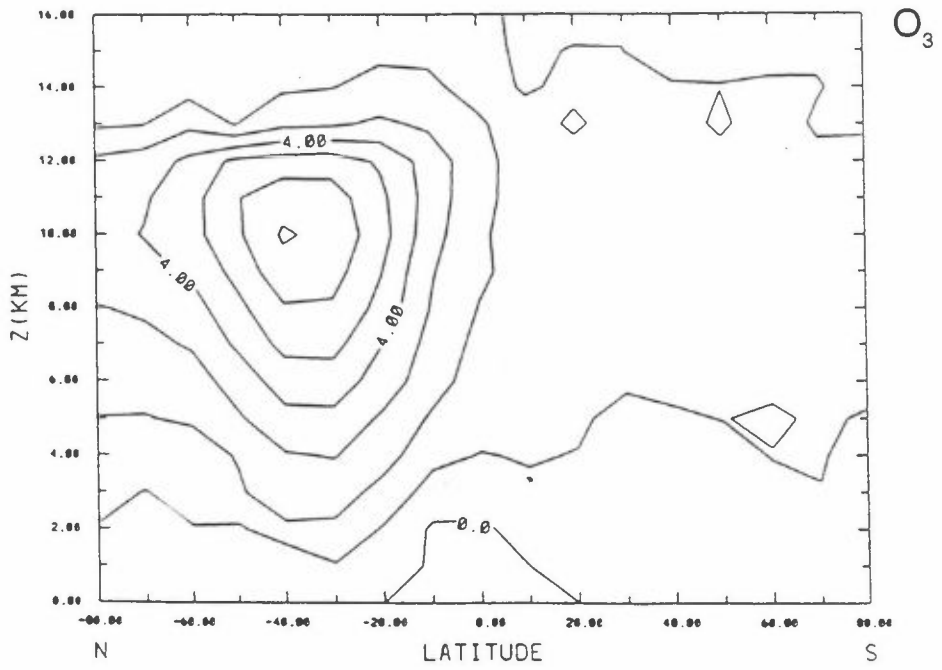


Figure 4 cont.

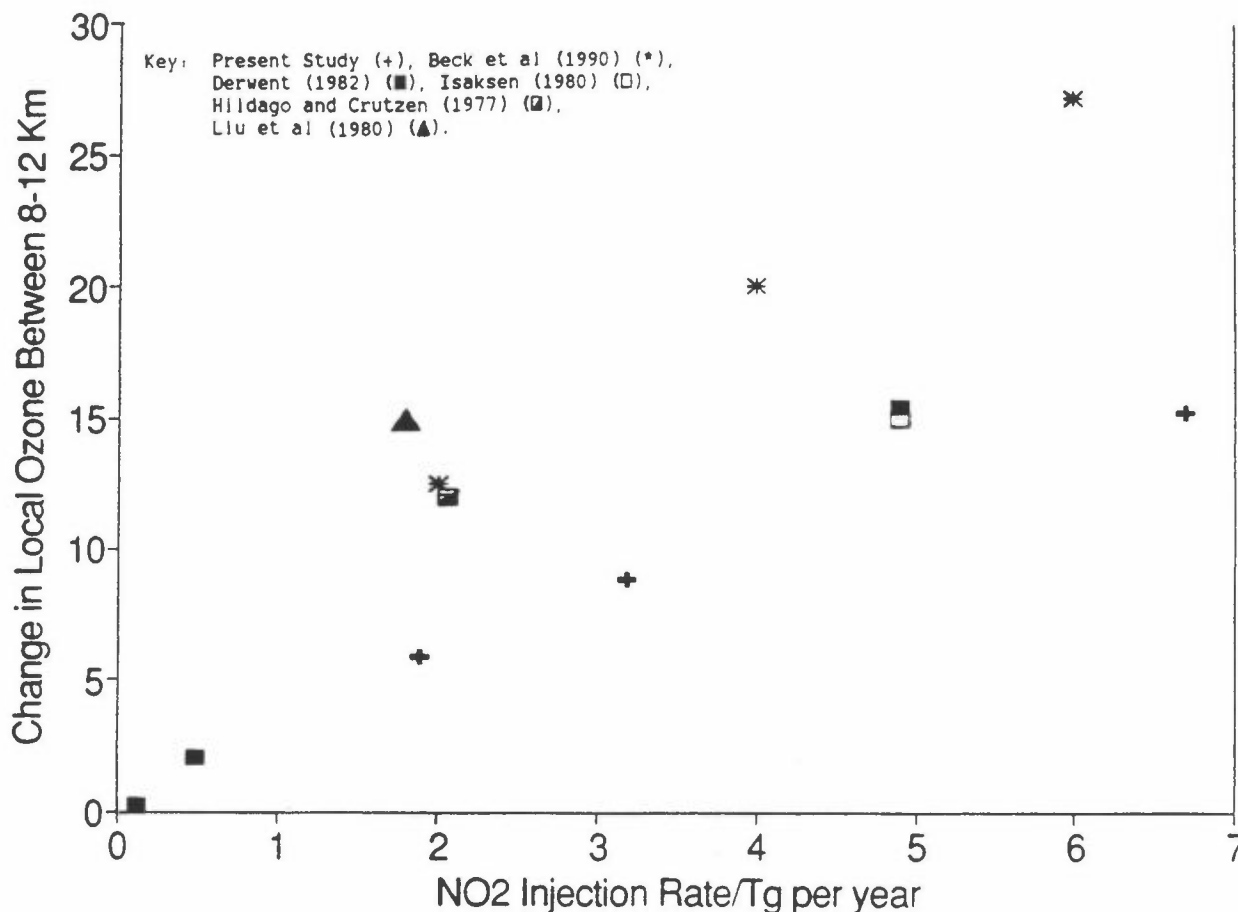


Figure 5: Percentage change in ozone concentrations in the 8-12 km region at 30°N versus NO_x aircraft emissions for various models. From Johnson and Henshaw (1991).

photochemical processes, in the transport of gases in the models as well as in the assumed altitudinal distribution of the aircraft emissions. E.g. in the model of Johnson and Henshaw (1991) vertical transport in the middle and upper troposphere is more efficient than in the Isaksen et al. (1990) model. The aircraft emissions are therefore more efficiently transported to higher altitude levels in the former model resulting in a stronger influence on O₃ above the flight altitude.

The effect of NO_x emitted by aircraft also depends on the assumed natural level of NO_x in the free troposphere. This level is not very well known, and depends e.g. on the strength of the lightning as a source of NO_x .

Model estimates of the impact of aircraft exhaust on the chemistry and physics of the atmosphere are uncertain due to incomplete knowledge about several topics. Firstly, estimates of NO_x emissions are uncertain, both total releases from aircraft and the spatial distribution of the releases. Secondly, the way several processes are treated in models is subject to improvement. There is limited knowledge about the chemical processes on a very small scale in gas and ice phases in condensation trails, and also how to incorporate such processes in large scale models. Heterogeneous chemical reactions in clouds are believed to play a significant role in the O_3 budget in the troposphere, but incorporation of such processes in models is presently at a preliminary stage. Quantification of parameters describing transport of atmospheric species need to be improved. In particular a better representation of the transport between the boundary layer and the free troposphere is needed. Improvements of models will help giving more realistic calculations of the impact of aircraft as well as changes in e.g. O_3 due to increased emissions of several precursor gases for which emissions are now known to be increasing.

Increased efforts in observation and monitoring of the chemical composition of the atmosphere are also needed in order to improve predictions of future changes in the chemical and physical properties of the atmosphere, including impacts of current and future aircraft operations. To understand the global O_3 budget, measurements of NO_x species are needed over remote areas, as over oceans, including vertical profiles throughout the troposphere. Similarly observations of O_3 at the surface as well as by vertical soundings is needed, to understand the processes governing tropospheric O_3 concentrations in general.

5.2 DIFFERENCES BETWEEN THE POLAR AND THE TROPICAL ATMOSPHERE

The troposphere is characterized by decreasing temperatures with increasing altitudes. Further upward from the top of the troposphere, the tropopause, the temperature gradient is

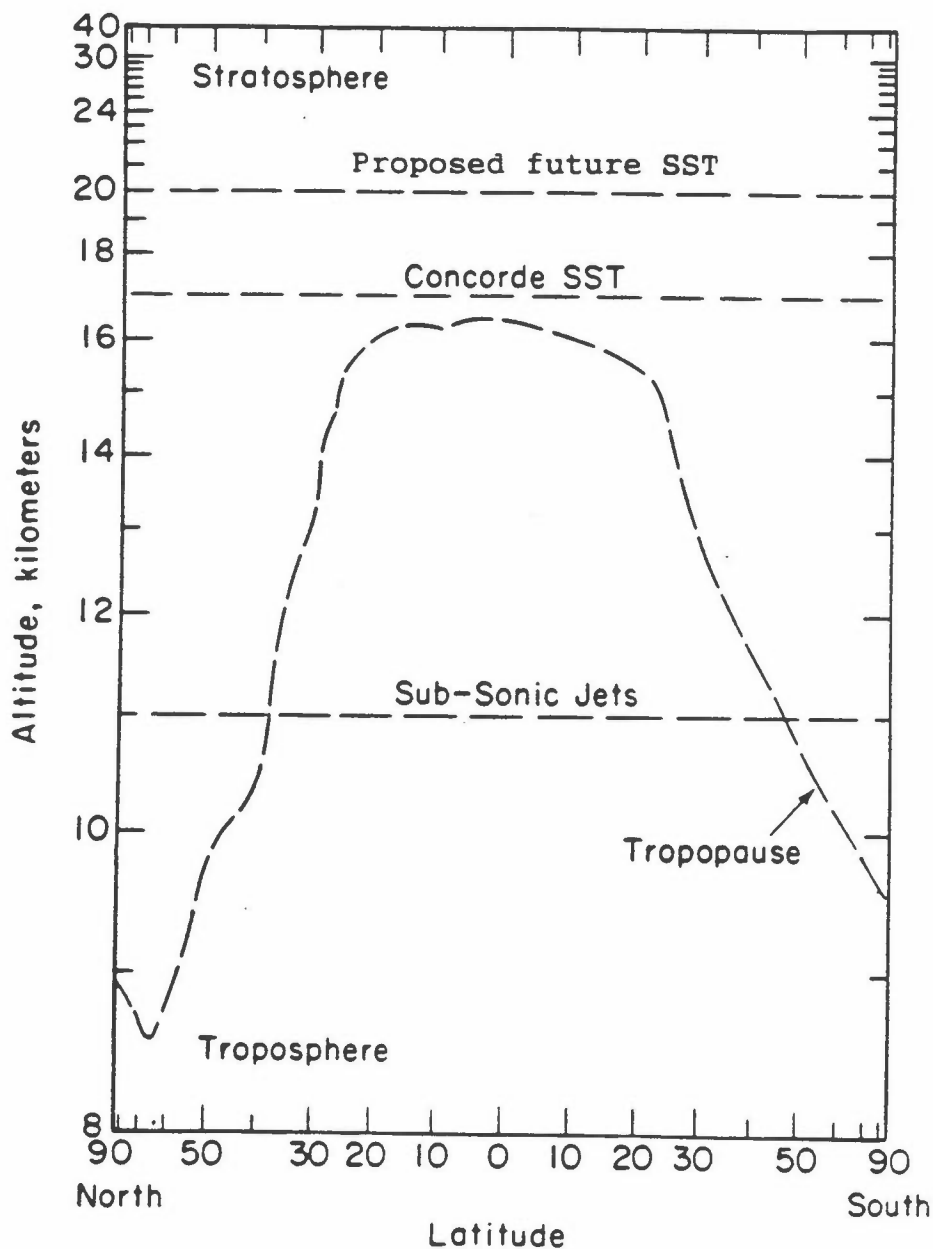


Figure 6: Tropopause altitudes at various latitudes, and typical cruise levels for presently flying subsonic jet and Concorde supersonic aircraft as well as future supersonic aircraft proposed by the Boeing and other companies. From Johnston (1990).

reversed, and the temperature increases with altitude. The tropopause is higher above the surface in the tropics than at high latitudes. As shown in Figure 6 typical cruise levels for commercial aircraft is in the troposphere at low and mid latitudes, while aircraft typically cruise in the lower stratosphere at high latitudes. Since the stratosphere is stable, strongly inhibiting vertical mixing, it takes much longer time to remove aircraft exhaust emitted in the stratosphere than exhaust emitted in the troposphere, where the vertical mixing is much stronger.

Even though aircraft cruise in the stratosphere at high latitudes, they presently do not fly high enough to cause depletion of the ozone layer. Model results show that the present fleet of aircraft causes ozone generation at all levels at all latitudes (Figure 4, Fuglestvedt and Isaksen, 1991). In the lower troposphere an increase in the OH concentration accompanies the O₃ increase, as OH is formed from ozone. However, in the upper troposphere and lower stratosphere OH is depleted by the NO_x injected in the atmosphere by aircraft (Figure 4).

5.3 HISTORIC TRENDS AND PROJECTIONS CONCERNING BACKGROUND OZONE CONCENTRATIONS IN THE TROPOSPHERE

Since the levels of ozone precursors - NO_x, CO, CH₄ and other hydrocarbons - have been observed to increase over the last decades, an upward trend in ozone could be expected. Observations of surface ozone seem to support an expected upward trend in regions of the Northern Hemisphere. An increase of about 1-3%/yr has been detected in several European and Japanese stations in modern data since the 1960's and 1970's. The situation is different in Canada and in the Southern Hemisphere, where even a very weak negative trend have been found in some locations.

In addition to the recent surface ozone measurements, there are few historic records of ozone. One is a series of data collected near Paris in the period 1876-1905. These data have been reanalyzed and compared with modern measurement methods, and indicate values of ozone corresponding to about half of the present-day values (Figure 7).

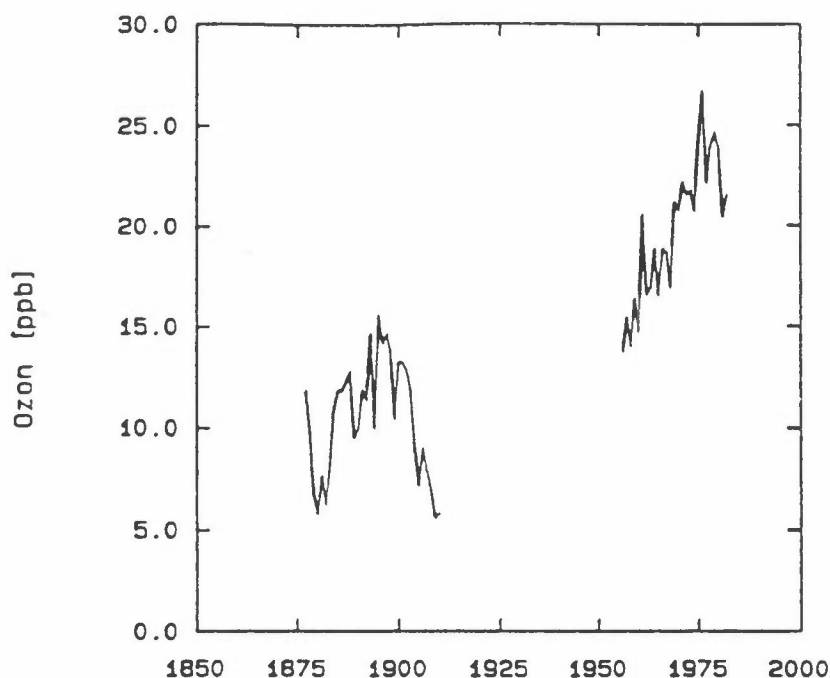


Figure 7: Annual averages of ozone mixing ratios at Montsouris near Paris around the last turn of century (Volz and Kley, 1987), compared to modern data measured at the clean air station Arkona in northern Germany (Feister and Warmbt, 1987).

Nine stations have since the 1960's and 1970's provided measurements of ozone throughout the troposphere, by balloon borne instruments. Again European (Figure 8) and Japanese measurements exhibit positive trends, and the Canadian exhibit negative trends. This discrepancy may, or may not, be due to a change of the instruments used in the Canadian measurements.

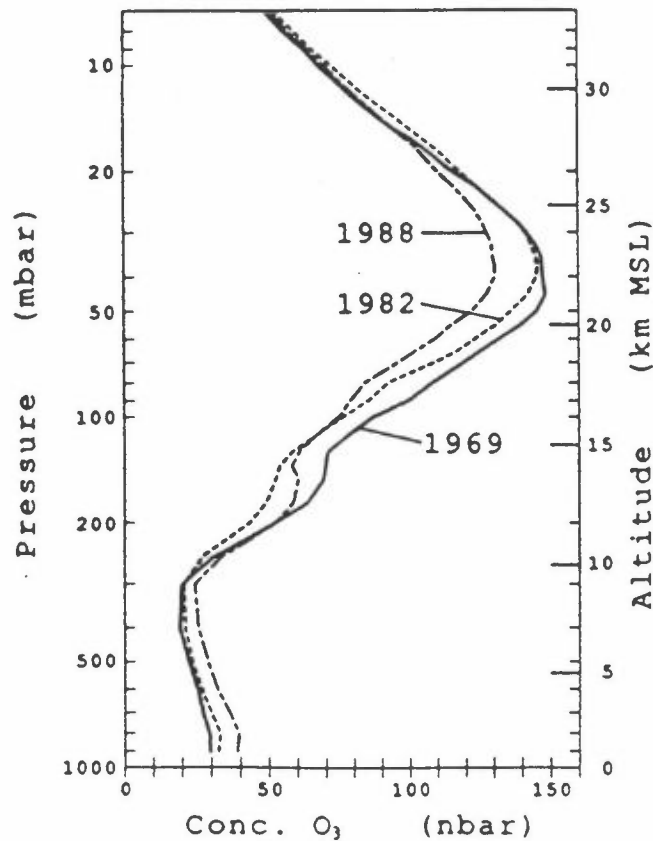


Figure 8: Selected annual means of the ozone balloon soundings from Payerne in Germany (from Staehelin and Schmid, 1991).

6 THE STRATOSPHERIC OZONE LAYER

The stratosphere is vulnerable to human impact because the air is stable against vertical mixing. Pollutants introduced in the stratosphere will therefore remain there for at least several months, possibly a few years. During this time interval the horizontal mixing is considerable, and pollutants will spread on a nearly global scale. Furthermore, the air in the stratosphere is thin, again making it vulnerable to pollution.

In the beginning of the 1970's supersonic stratospheric transport (SST) aircraft were planned. Some scientists were then worried that emissions of NO_x from such aircraft would damage the ozone layer. As the scientific understanding of the processes governing the O_3 concentrations improved, and as the

size of the fleet was dramatically reduced compared to the first plans, it is now believed that the currently flying fleet of SST do not influence the ozone layer to any significant degree.

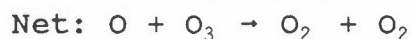
In the mid 1970's scientists voiced concern that CFC's were capable of reducing the ozone layer, a theory that has now been proven to be true in the Antarctic ozone layer. Recently, it has been suggested (WMO/UNEP, 1992) that ozone reductions observed in the atmosphere above the Northern Hemisphere are also, at least partly, due to the accumulation of chlorine from CFC's and bromine from halon gases in the atmosphere.

Ozone formation in the stratosphere is initiated by the photo-dissociation O_2 . The mechanism is simple: solar ultraviolet radiation is sufficiently energetic to split the O_2 molecule into two oxygen atoms:

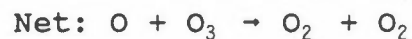


When colliding with other O_2 molecules, these oxygen atoms combine to produce O_3 . (Eq 7).

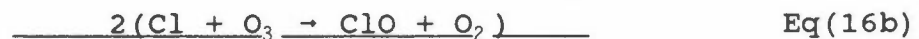
O_3 in the stratosphere is destroyed by several chemical reactions. Species in the nitrogen, hydrogen, chlorine and bromine families are involved in many chemical cycles destroying O_3 . It is characteristic for these cycles that they are catalytic, i.e. ozone is destroyed without destruction of the other reactants in the reactions. This explains that relatively small amounts of e.g. nitrogen oxides and chlorine can have such a pronounced effect on O_3 . The major catalytic cycle involving nitrogen oxides is



This chemical cycle is responsible for about half of the natural O_3 loss in the stratosphere. A similar catalytical chain involving chlorine is



A cycle initiated by the self reaction of ClO is known to be a major cause of the Antarctic ozone hole:



The participation of the ClO self reaction (in reality a sequence of reactions yielding two Cl atoms as end products, Eq 16a) explains the rapid development of the Antarctic ozone hole since the rate of the reaction is proportional to the ClO concentration squared.

Aircraft flying at higher speeds also fly at higher altitudes, and have specialized fuel requirements. Supersonic aircraft typically need to fly in the lower stratosphere. For comparison, the scheduled airtime between Los Angeles and Sydney (7 500 miles) is 14.5 hours for current aircraft flying at Mach 0.85 at 11 km, using Jet-A fuel (Mach 1 equals the speed of the sound, about 1 200 km per hour). For a proposed aircraft, the airtime is 5.1 hours at Mach 2.4, at 20 km, also using Jet-A fuel. Aircraft at higher Mach numbers (4, 6, 12) would fly at even higher altitudes (25, 30, 35 km) with shorter airtimes (3.7, 3.0, 1.7 hours) but with specialized fuel and engine requirements.

The nitrogen oxides (NO and NO_2) are involved in photochemical production of O_3 , (Equations 6-7) but also in catalytic destruction of O_3 (Equations 14a-b) (Figure 9). The former process dominates in the troposphere and the lower stratosphere, and

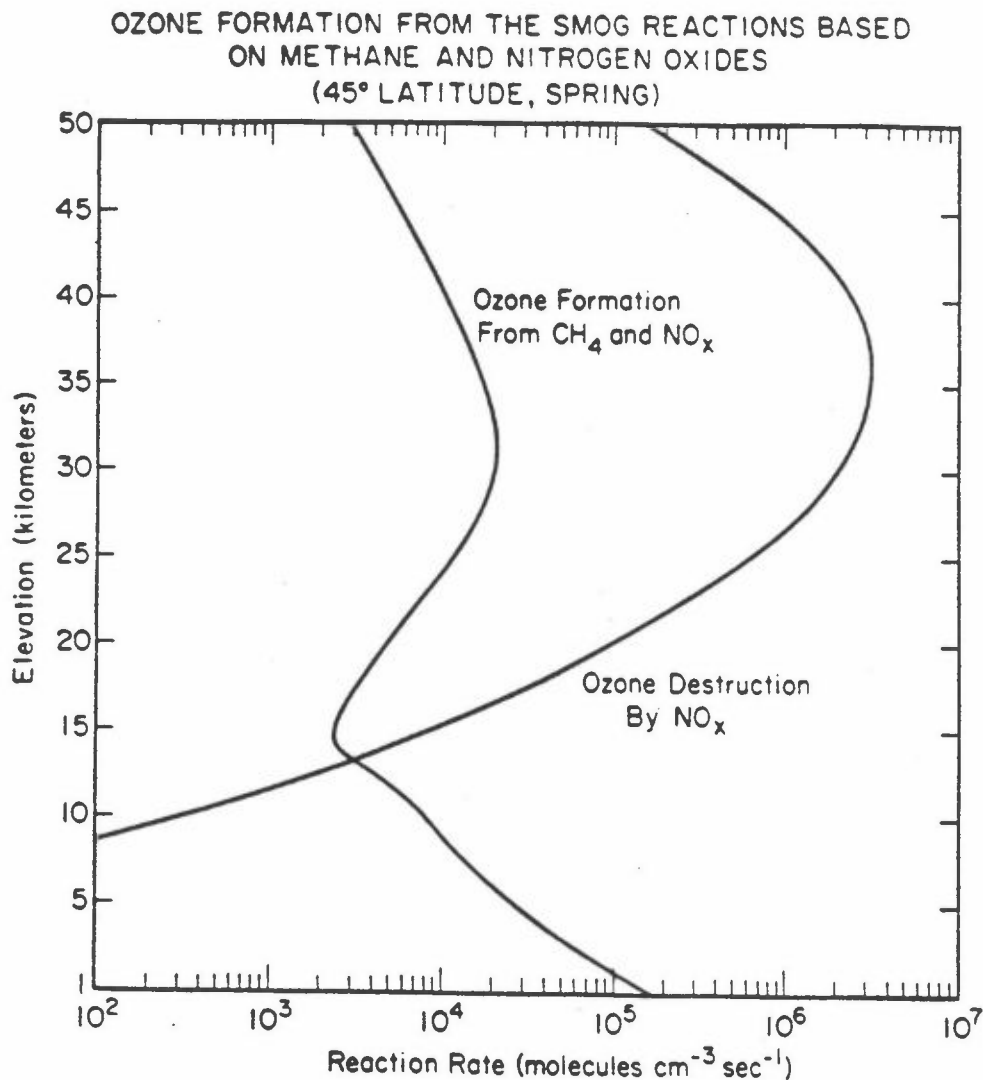


Figure 9: Rates of ozone formation and ozone destruction due to photochemical reactions involving NO_x species in the troposphere and the stratosphere. From Douglas et al., 1991).

the latter in the middle and upper stratosphere. The crossover point, the altitude where the two effects cancel, is at approximately 15 km (depending on latitude and time of the year). Only very few aircraft presently fly above this level. Only the Concorde SST's fly at levels where the NO_x emitted in the exhaust deplete ozone (Figure 6). The presently flying Concor-

des to a very little degree deplete the stratospheric ozone layer, since the number of aircraft is very low, and since they fly very close to the crossover point. A substantial fleet of supersonic aircraft flying even faster and at higher levels than the Concorde are now being proposed (Figure 6). Ongoing research shows that if such aircraft are flying high enough they can significantly deplete the ozone layer (WMO/UNEP, 1992), especially at middle and high latitudes.

A recent independent assessment of the impact of a projected fleet of supersonic aircraft on stratospheric O₃ (Douglas et al., 1991) has concluded that the ozone loss increases with the amount of nitrogen oxides emitted. These models used gas-phase chemistry and assessed ozone loss for the case of 500 aircraft flying at Mach 2.4 between 17 and 20 km with an annual fuel use of 7×10^{10} kg/yr. The annual-average loss of column ozone at middle latitudes in the northern hemisphere is predicted to be 2-6%. For a comparable fleet operated at Mach 3.2 between 21 and 24 km, the comparable column ozone losses are 7-12%. However, recent experiments have shown that reactions on sulfate aerosols can change the partitioning of nitrogen oxides. Two model studies incorporating this heterogeneous chemistry have recently reexamined the Mach 2.4 case and found substantially less ozone change (-0.5% to +0.5%). These implications are being examined as part of a separate assessment (WMO/UNEP, 1992).

7 THE GREENHOUSE EFFECT

Greenhouse gases are defined as atmospheric gases whose main absorption bands are in the wave-length range of heat radiation, i.e. in the infrared range; as their concentrations increase, they enhance the greenhouse effect of the atmosphere and may thus trigger a rise in surface temperatures.

Table 2 lists the current and pre-industrial concentrations of some greenhouse gases, as well as their atmospheric lifetime, the current annual growth rates of their concentrations, and the specific greenhouse potential of one molecule (impact of one additional molecule released into the atmosphere at present concentrations relative to carbon dioxide).

Table 2: Summary of key greenhouse gases influenced by human activities.

Parameter	CO ₂	CH ₄	CFC-11	CFC-12	N ₂ O
Pre-industrial atmospheric concentration (1750-1800)	280 ppmv	0.8 ppmv	0	0	280 ppbv
Current atmospheric concentration	353 ppmv	1.72 ppmv	280 pptv	484 pptv	310 ppbv
Current rate of annual atmospheric accumulation	1.8 ppmv (0.5%)	0.015 ppmv (0.9%)	9.5 pptv (4%)	17 pptv (4%)	0.8 ppbv (0.25%)
Atmospheric lifetime (years)	(50-200)	10.5	55	116	132
Greenhouse potential of one molecule, relative to CO ₂	1	21	12400	15800	206
GWP, 100 year time horizon (direct effects only)	1	11	3400	7100	270

H₂O and O₃ hold a special position among the greenhouse gases, because they have relatively short lifetimes, and hence, their concentrations vary considerably in space and time.

The greenhouse effect is caused by the fact that, due to IR absorption by trace gases, the effective radiant layer of the atmosphere (i.e. the layer in which the IR emission by atmospheric gases and clouds into space is at a maximum) is cooler than the Earth's surface. Currently, H₂O, CO₂ and high clouds account for 90 percent of the natural greenhouse effect. The remaining 10 percent is contributed by other trace gases. These are above all O₃, N₂O and CH₄ as well as in recent years increasingly CFC's. These greenhouse gases very effectively warm

the Earth's surface and the lower atmosphere, since they are almost transparent to solar radiation, while they absorb a rather large proportion of the IR radiation from the Earth's surface.

Because of the greenhouse effect, a change in the atmospheric abundance of trace gases which absorb infrared radiation will have a direct impact on surface temperatures. Currently, the greenhouse effect is increasing, because of increasing atmospheric concentrations of greenhouse gases due to human activities. As a result, an increasing proportion of the radiation from the Earth's surface is absorbed by atmospheric greenhouse gases and radiated back to Earth. Consequently, the radiation balance at the top of the atmosphere becomes positive, the Earth loses less energy and its surface warms up. This rise in temperature is also controlled by numerous feedback mechanisms in the climate system and can only be estimated, therefore, by means of extensive and complicated model calculations.

Aircraft contribute to an increased greenhouse effect in two ways, by emissions of the greenhouse gas CO_2 and by emissions of NO_x which can produce the greenhouse gas O_3 . In aircraft engines burning hydrocarbons, CO_2 is a combustion product. As CO_2 is a greenhouse gas itself, this gives rise to a direct greenhouse effect. CO_2 is very stable in the atmosphere, and is well mixed. CO_2 -emissions from aircraft therefore contribute equally to the greenhouse effect regardless of where the emissions take place. All phases of a flight contribute to the increased greenhouse effect. What matters is the amount of fuel that is burnt. However, since aircraft is only consuming 2-3% of current carbonaceous fossil fuel on the global scale, the impact on CO_2 -driven climate change is correspondingly small. Implications with respect to changed O_3 is potentially more important, since O_3 is also a greenhouse gas. Production of O_3 from man made emissions of NO_x therefore increases the greenhouse effect through the O_3 formation. Since NO_x itself does

not contribute to the greenhouse effect directly, and its effect on the greenhouse effect is through the formation of ozone, the effect is denoted as indirect.

In the IPCC (1990, 1992) reports the effect of the various greenhouse gases were compared using the concept of Global Warming Potential (GWP). GWP has been defined as the increased greenhouse effect resulting from the release of 1 kg of a greenhouse gas relative to the increased greenhouse effect caused by emission of 1 kg of CO_2 . Numbers for direct (see Table 2) as well as indirect greenhouse effect of greenhouse gases were given in IPCC (1990). Further work has revealed that a GWP may prove inapplicable for the shortlived gases, which are not well mixed in the atmosphere. In particular, GWP numbers for NO_x are not recommended by IPCC (1992). Near the ground it is believed that NO_x emissions can produce O_3 , but also produce OH and hence result in decreased concentration of CH_4 and some HCFC's. NO_x may therefore contribute both to an increase and to a decrease in the greenhouse effect. In the middle and upper troposphere, at cruise level for most current aircraft, the O_3 production is believed to dominate, contributing to an increased greenhouse effect.

O_3 is most efficient as a greenhouse gas in the upper troposphere and lower stratosphere where, as we have already pointed out, NO_x from aircraft very efficiently produces O_3 . There is therefore a twofold amplification of the indirect greenhouse effect caused by NO_x emissions from aircraft sources as compared to the emissions at the ground.

According to Johnson and Henshaw (1991) the present fleet of civil aircraft contribute with an increase in the greenhouse effect which is almost 20% of the increase in the greenhouse effect due to increased CO_2 in the decade 1970-1980. However, these numbers are uncertain, and more research is needed to quantify the greenhouse effect from aircraft emissions of NO_x .

Along with industrial activity, biomass burning and volcanic eruptions, aircraft emissions contribute to the formation of aerosols (airborne particles), which can absorb as well as reflect radiation. It is believed that the aerosols as a net effect cools the Earth's surface. It is not known to which extent aircraft emissions in this way lead to a cooling, which can partly counteract the increased greenhouse effect explained above.

8 REGIONAL AND GLOBAL AIR POLLUTION BY NORWEGIAN AIRCRAFT

In this report we have described air pollution problems related to the operation of aircraft, focusing on the effects on regional and global scales. An estimate of environmental effects only on regional and global scales due to the Norwegian fleet of aircraft is given in the following.

The Norwegian aircraft are presently not flying at altitudes in the stratosphere high enough to destroy ozone, this problem can therefore be excluded. The two major environmental issues in which the Norwegian fleet of aircraft can play a role is the increasing greenhouse effect and the changing of the oxidizing capacity of the atmosphere.

When numbers are given, one will find that the Norwegian contributions are small. Nevertheless, they may be significant when e.g. the size of the Norwegian population is taken into account.

8.1 CHANGING THE OXIDATION CAPACITY

In the case of changing the oxidation capacity of the atmosphere, the emissions of NO_x is of greatest importance (Johnston and Henshaw, 1990). The aircraft emissions of CO ,

methane and other hydrocarbons (which play a significant role in the oxidant formation in many situation at the surface) are not large enough to play a significant role.

As shown in Section 5., changes in NO_x , O_3 and OH as a result of aircraft NO_x emissions maximize around the cruising level (about 10 km) at our latitudes. The maximum increases of NO_x and O_3 at 60 deg N are about 30 % and 5 % respectively. These changes are due to the global fleet of aircraft, with local emissions having the largest impact and remote emissions an impact which decreases with increasing distance. Attributing changes in NO_x , and especially O_3 , to the Norwegian aircraft specifically, is not trivial. This is due both to the fact that the ozone formation is not directly proportional to the NO_x emissions, and the fact that the excess NO_x at one particular location (e.g. latitude) does stem from aircraft emissions at other locations as well.

One way to crudely estimate the effect of Norwegian aircraft only is firstly to assume that NO_x emissions from areas of different extent will influence the NO_x levels at our latitudes, as shown in Table 3. On a global scale only 0.2 % of the total aircraft NO_x emissions are due to Norwegian aircraft. Out of the total emissions in the region 60 - 80 deg N (approximately the Norwegian region) the Norwegian emissions make a fraction of 6-9 %. The lower of these numbers assumes Norwegian NO_x emissions given by Knudsen and Strømsøe (1990), while the the higher number assumes emissions given in SFT- 1990. The latitudinal distribution of aircraft NO_x emissions are taken from Isaksen et al. (1990). However, aircraft NO_x emissions from other latitudes also influence NO_x levels in the Norwegian region. One may therefore also want to estimate the fraction of the Norwegian emissions relative to emissions from somewhat larger regions. The Norwegian fraction is approximately 1.2%, 0.5% and 0.3% when emissions from the regions 50-90 deg N, 40-90 deg N and 30-90 deg N are considered, respectively (see

Table 3). On the other hand, the Norwegian emissions will be transported to other latitudes and contribute to the increase in the NO_x concentrations there.

Secondly, to estimate the effect on ozone formation, one could assume that there is a linear relationship between NO_x emissions and ozone formation, i.e. emissions of a given amount of NO_x (e.g. the Norwegian aircraft contribution) will have the same effect on ozone regardless of the size of any other given source (e.g. aircraft emissions from non-Norwegian aircraft). According to Figure 5, this is not a perfect, but quite reasonable assumption. The fractions of the Norwegian emissions relative to emissions in various regions (from Norwegian sector up to global scale) will then transform directly into the contribution of the Norwegian aircraft emissions to ozone formation in our areas.

Table 3: Relative contribution of Norwegian aircraft to the total aircraft emissions (emissions are taken from Knudsen and Strømsøe, 1990 and SFT, 1990 denoted as NILU-1990 and SFT-1990, respectively).

Emission area considered	Norwegian contribution (%)	
	NILU-1990	SFT-1990
60-80 deg N	6.4	8.6
50-90 deg N	1.1	1.4
40-90 deg N	0.5	0.6
Global	0.2	0.3

8.2 INCREASED GREENHOUSE EFFECT

As discussed above, the aircraft emissions contribute to an increased greenhouse effect both due to emission of CO_2 (direct climate effect) and NO_x , through formation of ozone (indirect effect). Estimating the relative CO_2 -effect from Norwegian aircraft is straightforward, since CO_2 is well mixed in the atmosphere. Emissions of CO_2 therefore contribute equally to the

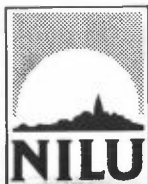
bulid up of CO₂ regardless of where they occur. The Norwegian aircraft contribute with 4.3% (5.5%) of the total Norwegian CO₂ emissions from burning of fossil fuel, and with 0.03% (0.04%) of the total global emissions (taken from IPCC, 1990) of CO₂ from fossil fuel combustion (the lower numbers assume Norwegian emissions from Knudsen and Strømsøe, 1990, the higher numbers - in parentheses - from SFT, 1990). Perhaps more important is the increased greenhouse effect of ozone produced from NO_x emissions. As discussed in Section 7., NO_x emitted at the aircraft cruise level is more efficient in producing ozone than surface emissions. Furthermore, ozone at this altitude is more efficient as a greenhouse effect than at the surface (Johnson et al., 1992). At present reliable numbers quantifying the climate effect of aircraft emissions of NO_x cannot be given. As the effect is currently believed to be potentially important, modelling projects, e.g. a European project involving science groups from several European countries, aimed at quantifying this effect are now under their way.

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TITLE Regional and Global Air Pollution from Aircraft
ABSTRACT The regional and global air pollution problems discussed in this report are ozone formation in the troposphere, acid deposition, depletion of the stratospheric ozone layer and increasing greenhouse effect. Currently, the two most important air pollution problems related to aircraft emissions are oxidant formation and increasing greenhouse effect, in both cases mainly due to emissions of NO _x . The acid deposition from aircraft emissions is not believed to be significant on global or regional scales. Depletion of the stratospheric ozone layer is neither a problem with the current fleet of aircraft. A proposed fleet of high flying aircraft in the future may however threaten the ozone layer.

* Kategorier: Åpen - kan bestilles fra NILU A
 Må bestilles gjennom oppdragsgiver B
 Kan ikke utleveres C