NILU OR: 80/91

NILU OR : 80/91
REFERENCE : O-1565
DATE : DECEMBER 1991
ISBN : 82-425-0321-4

Environmental Effects of Emissions of Nitrogen Oxides

A General View and a Particular Emphasis on the Role of Aircraft

F. Stordal and U. Pedersen

CONTENTS

1 NITROGEN CHEMISTRY IN THE ATMOSPHERE AND IN PRECIPITATION

Nitrogen is by far the most abundant atom in the atmosphere. Molecular nitrogen (N_2) constitutes about 80% of all the molecules in the atmosphere. It is stable under the temperatures and radiative conditions that prevail in the atmosphere up to at least 100 km altitude. For this reason it does not take part in the lively photochemical activity in the atmosphere, 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. At extremely high temperatures, as in lightning strokes and in high-temperature combustion processes, such as in automobile and aircraft engines, molecular nitrogen is decomposed to form chemically active NO_x species.

1.1 ACID RAIN

Sulphates and nitrates are removed from the atmosphere by wet and dry processes. The removal is often referred to as acid rain because of the focus on the composition of rainwater. Since the removal takes place by both wet and dry deposition, the overall process is more properly termed acid deposition, a term that is used in the following.

The species that are removed from the atmosphere by acid deposition, are produced by chemical reactions of the gases sulphur dioxide (SO_2), nitric oxide (NO) and nitrogen dioxide (NO_2). These chemical reactions are driven by oxidizing agents also present in the atmosphere and derived from the gas ozone (O_3).

These oxidizing agents are :

Hydroxyl radical (OH), which plays a very important role in oxidation reactions of SO_2 and NO_2.

Hydroperoxyl radical (HO_2), which takes part in a cycle of reactions in polluted air which lead to the production of ozone and combined with other HO_2 radicals gives

Hydrogen peroxide (H_2O_2), which is an important oxidation agent for SO_2 in solution.

The oxidation of nitrogen oxides (NO_x) is much more complex than that for sulphur dioxide. The reaction in solution gives nitric acid as an end product, but since neither NO nor NO_2 are very soluble in water at the concentrations found in the atmosphere, this reaction is unimportant.

The gas phase reactions depend strongly on sunlight. In the presence of sunlight the reaction of NO with O_3 which gives NO_2, is balanced by the photolysis of NO_2 yielding NO and O_3. An equilibrium is thereby established (photostationary state). The NO_2 may react further with hydroxyl radicals (OH) to give gas phase nitric acid (HNO_3).

During daytime the reaction of NO_2 with O_3 is balanced by the photodecomposition of the product (NO_3) to give NO_2 again. At night the reaction of NO_2 with O_3 is not reversed and NO_2 reacts to give NO_3 which reacts further to give nitric acid.

The removal of nitrogen containing components from the atmosphere is by dry deposition of NO_x and HNO_3, or they can be incorporated into the precipitation by :

- nucleation scavenging of nitrate-containing aerosols in cloud formation

- absorption of gaseous nitric acid by cloud droplets
- below-cloud scavenging of nitrate-containing aerosols by rain.

The paths of NO_x leading to acid deposition are shown in Figure 1.

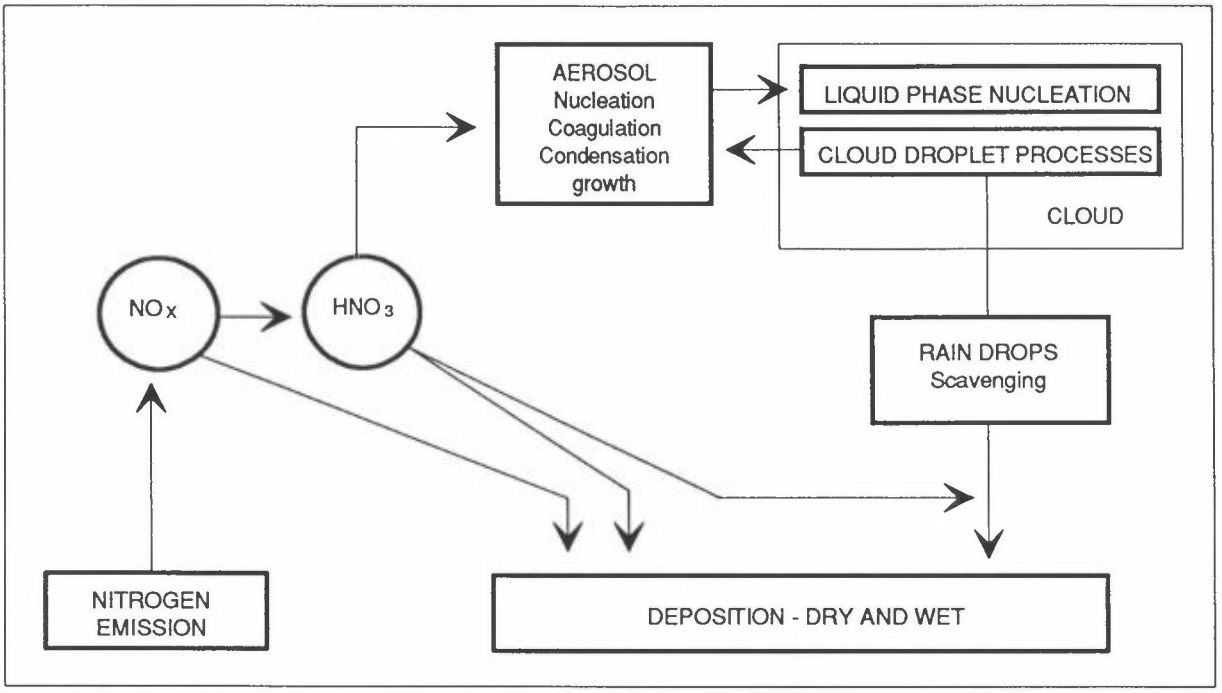


Figure 1: Atmospheric paths of NO_x leading to acid deposition

1.2 NITROGEN SATURATION IN SOILS

The input to the forest ecosystem may occur through

1) wet deposition of NO_3^-, NH_4^+, and other nitrogen species

2) dry deposition of nitrogen-containing aerosols or gaseous NH_3, NO_2, NO and HNO_3

3) N_2 fixation

Most important for the input of oxidized nitrogen is the wet deposition of NO_3^- and the dry deposition of aerosols and gases.

Acidic deposition is attributed to anthropogenic emissions of SO_2 and NO_x which are converted by oxidation and hydrolysis to the acid anions SO_2^{--} and NO_3^- which are found in the precipitation.

The most important dry deposition compound appears to be nitric acid vapour (HNO_3). Deposition of the other gas phase nitrogen oxides (NO_2, NO) is known to be small compared to the deposition of HNO_3.

In the recent years the increased input of atmospheric nitrogen (wet and dry) has led to what is called the "nitrogen saturation hypothesis" (Nihlgård, 1985). The nitrogen saturation in a forest ecosystem has been defined as a situation where ecological resources other than nitrogen, have become limiting to forest productivity. In unpolluted areas forests get their main amounts of nitrogen from biological turnover of organic matter in the soil. Today forests in many regions get perhaps an equal supply of nitrogen from wet and dry deposition. This nitrogen deposition to acid forest soil may in the long run cause several negative effects both to the soil itself and the trees.

1.3 THE CONCEPT OF CRITICAL LOADS

The critical load concept was originally developed and used as a policy tool by the Canadian authorities in the early 1980's. This concept has been further developed and extended in workshops arranged within the work programme under the Convention on Long Range Transboundary Air Pollution.

The critical load for nitrogen can be defined in two ways. Both sulphur and nitrogen contribute to the total input of acidic compounds to an ecosystem, and the ratio of sulphur to nitrogen

can therefore vary without changing the critical load for acidic compounds.

The critical load for acid deposition to an ecosystem has been defined as: The highest deposition of acidifying compounds that will not cause chemical changes leading to longterm harmful effects on ecosystem structure and function.

Biological nitrogen transformation can generate and consume large quantities of acidity. A nitrogen cycle that is in balance will of course have no net production or consumption of acidity, but in a situation where nitrogen is no longer limiting the biomass growth, inorganic nitrogen may accumulate in the soil and increase the possibility for nitrate leaching.

We can therefore also define the critical load of nutrient nitrogen as: The maximum deposition of nitrogen compounds that will not cause eutrophication or induce any type of nutrient imbalance in any part of the ecosystem or recipient to the ecosystem.

The critical load for total acidity input indicates how much acidity that can be loaded onto the system. The critical load must however be defined in the terms of sulphur and nitrogen acidity, since emissions are in the form of sulphur and nitrogen compounds.

The eutrophication criterion that sets a maximum for nitrogen deposition is independent of the acidification impact, and it can be lower or higher than the maximum limit set for nitrogen deposition by the critical load for total acidity.

As we can see, the emissions of NO_x play an important role in the concept of critical loads since it contributes to the acidification as well as to the eutrophication of the ecosystem through acid deposition.

1.4 THE PRESENT SITUATION IN THE NORDIC COUNTRIES

Most of the NO_x deposited over the Nordic countries is of European origin. The emissions of gaseous nitrogen compounds in Europe are not distributed uniformly across the regions, and are advected out of the source areas by wind, undergoing chemical transformations to form secondary pollutants before they are depleted to the ground. Consequently, the deposition over the Nordic countries may vary a great deal from one region to another.

Variation in wet deposition between different areas is mainly a consequence of differences in precipitation amounts, but is also to some extent due to differences in the rain water composition. Dry deposition varies on an even finer scale, partly due to variations in air pollution load and partly depending on aerodynamic and absorbing characteristics of the receptor (forest, grassland, water etc.). For example a hillside facing the main wind direction or a forest edge, receives a higher deposition than a less exposed site, and a forest receives more deposition than an open field.

The deposition varies also in time, for example from year to year. In rural areas far from the emission source, this variation can be considerable. Such variations are mainly due to changes in large-scale weather patterns.

A map showing the deposition of oxidized nitrogen to the Nordic countries is shown in Figure 2. The deposition has been estimated using wet deposition monitoring results, throughfall data, air pollution concentrations as well as literature data.

The results show that the highest concentrations of oxidized nitrogen are found in Denmark and in the southern part of Sweden and Norway. The less exposed areas (< 0.2 g-N/m^2) are in the northern part of Finland, Sweden and Norway.

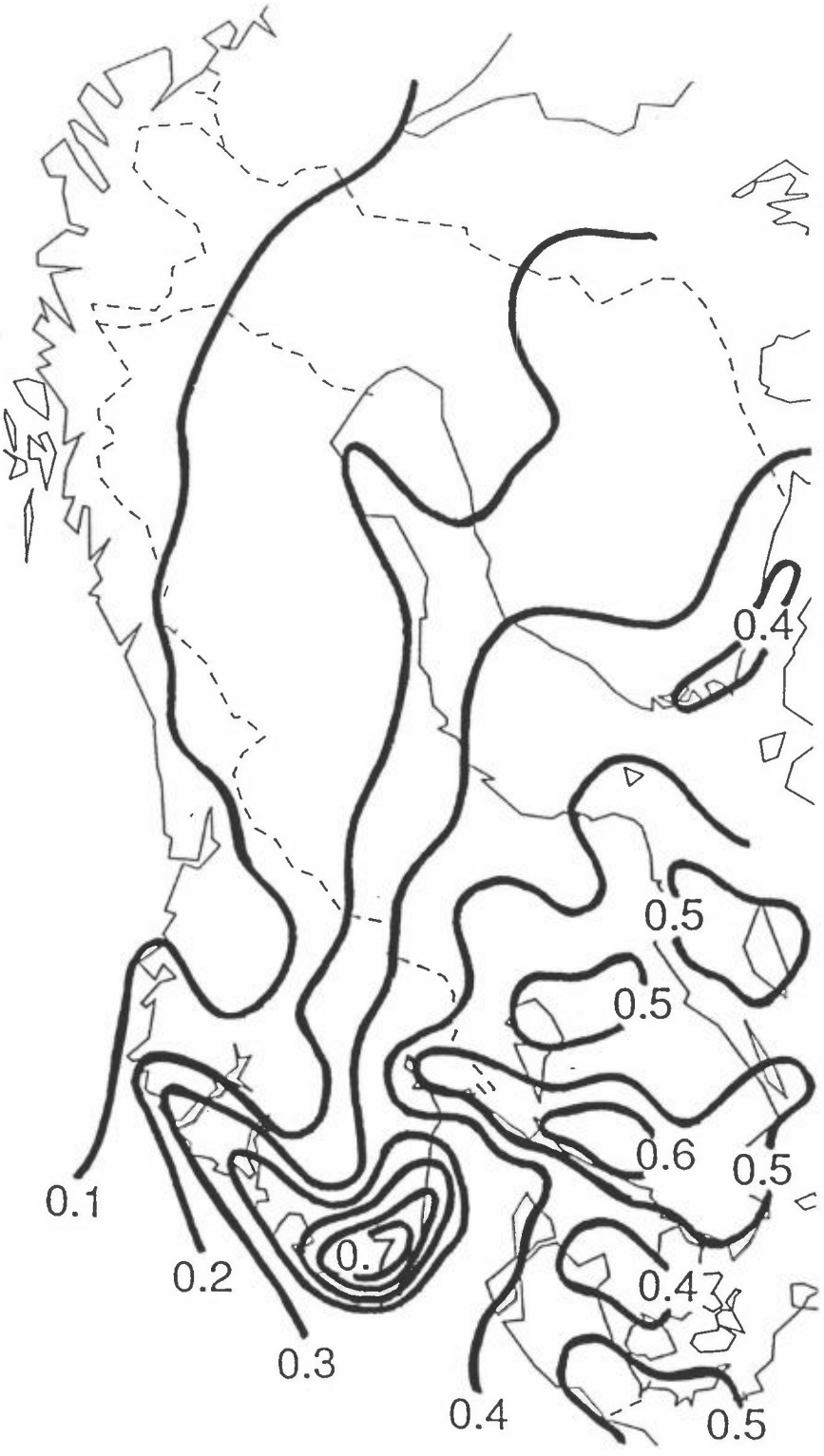


Figure 2: Deposition of oxidized nitrogen in the Nordic countries. From Lövblad et al. (1991).

2 SPECIFIC ENVIRONMENTAL CONCERNS RELATED TO EMISSIONS FROM AIRCRAFT

Aircraft exhaust contains NO_x resulting from decomposition of atmospheric N_2 as well as from combustion products from the fuel. 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. For civil aircraft these factors are not precisely known.

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 (1987) (11)	4100	5900	7600	
Sweden total (1989) (12)	3000	8000	7500	2000 000

Complete burning of hydrocarbon (HC) fuel in aircraft jet engines, like in automobile engines, would yield only water vapour (H_2O) and carbon dioxide (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, SO_2 and 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 ozone 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.

2.1 THE SIGNIFICANCE OF NO_x EMISSIONS FROM AIRCRAFT IN THE FREE TROPOSHPERE, COMPARED TO EMISSIONS FROM LAND BASED AND NATURAL SOURCES

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 orginate from civil aircraft. On a global scale civil aircraft contribute with 600 kton N/yr, constituting 1% or less 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. All the global numbers are taken from the ongoing Stratospheric Ozone Assessment by UNEP/WMO (1991).

NO_x-emissions from aircraft make only a small fraction of the global man made emissions. Furthermore, 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 ozone 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 following sections.

2.2 THE CHEMISTRY OF THE ATMOSPHERE IN AREAS WITH LOW BACKGROUND CONCENTRATIONS OF NO_x

The chemistry of the troposphere is driven by solar radiation. OH radicals, which play an important role in the oxidation of nitrogen and sulphur (section 1.1) as well as several other compounds, are formed when ozone is dissociated in the presence of water vapour. Ozone is formed (section 1.1) in reactions following dissociation of NO_2.

Tropospheric ozone 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 OH. Ozone is furthermore an efficient oxidizer itself. 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 ozone and OH through a series of connected chemical reactions.

Ozone is produced in air rich of NO_x through reactions described above. An efficient ozone generation requires that the NO molecule formed in the NO_2 photolysis is recycled to NO_2 so that the dissociation can take place several times. In NO_x rich areas the chemical conditions favour chemical reactions that provide such a recycling.

In areas with low NO_x the situation is quite different. Reactions destroying ozone dominate over the production. A key loss for ozone is then its reaction with HO_2. Under such conditions OH is depleted as well.

As discussed above (section 2.1) man made emissions of NO_x over industrialized continents constitute the dominating source of NO_x. Nitrogen oxides can not 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 become 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.

2.3 IMPACT OF NO_x IN THE UPPER TROPOSPHERE AND LOWER STRATOSPHERE

Except in the tropics there are no 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 does therefore not contribute to a significant extent to the acid rain problem.

Ozone 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 ozone than the same amount of nitrogen oxides at groundlevel (Isaksen et al., 1990; Johnson and Henshaw, 1991). Aircraft

emissions therefore has a potential to significantly influence the global oxidizing capacity despite its moderate contribution to the total man made NO_x emissions.

Model studies of effects of aircraft emissions on tropospheric ozone 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 3, 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 4) made by Johnson and Henshaw (1991). The differences are due to differences in the representation of the photochemical processes, the transport of gases in the models as well as 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 ozone above the flight altitude.

The effect of NO_x emitted by aircraft also depends on the 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.

Ozone is a greenhouse gas. Production of ozone from man made emissions of NO_x therefore increases the greenhouse effect through the ozone formation. This indirect greenhouse effect comes in addition to the direct greenhouse effect of the CO_2 which is produced in the combustion in aircraft engines. Ozone is most efficient as a greenhouse gas in the upper troposphere

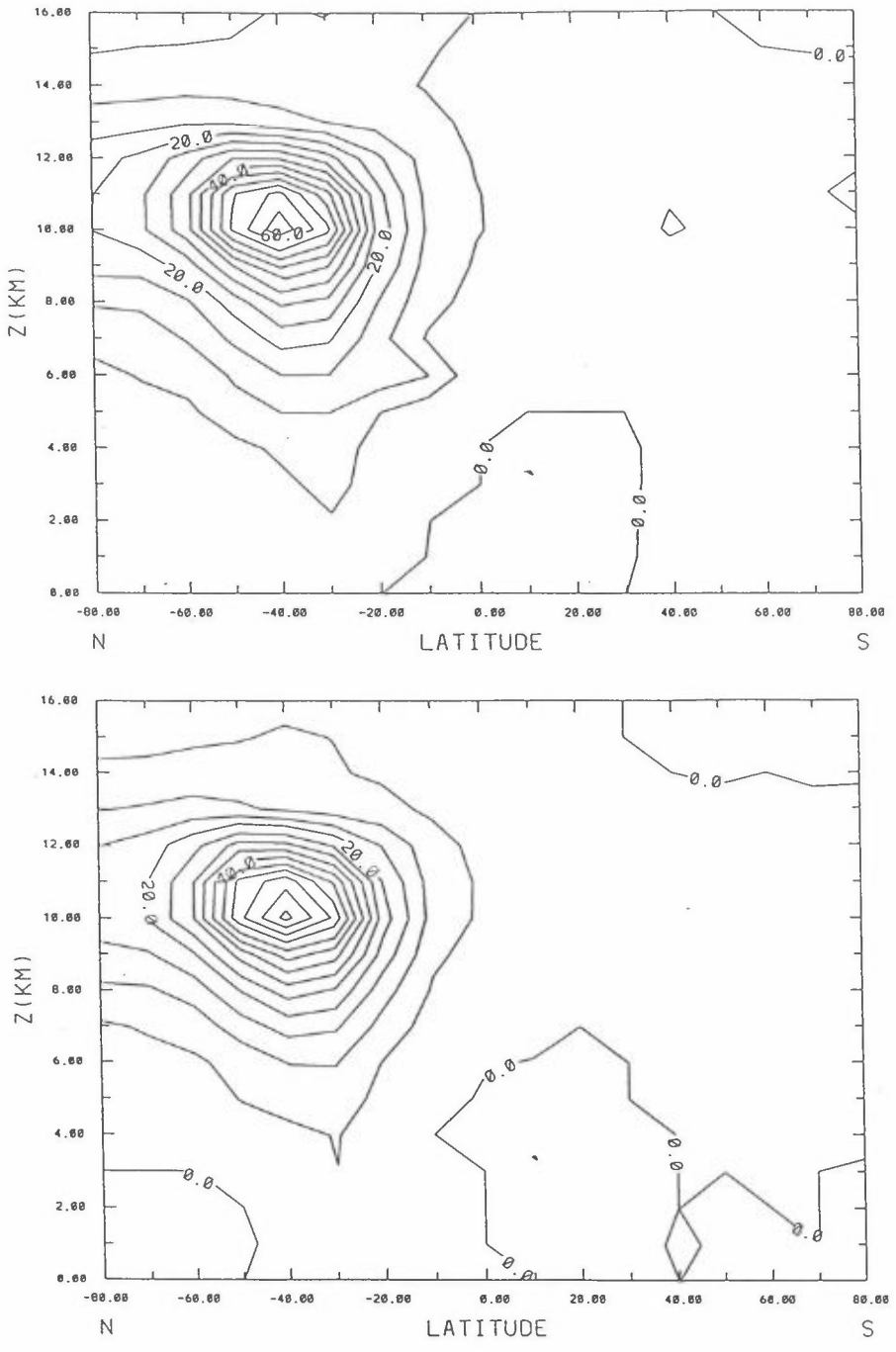
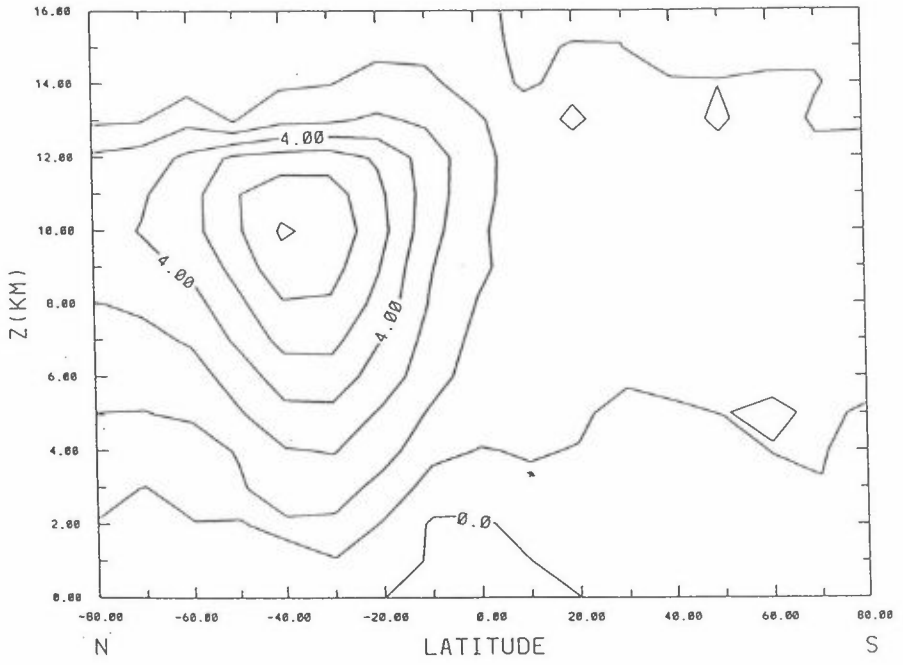


Figure 3: Changes in the concentrations (%) of NO, NO_2, O_3 and OH at various latitudes and altitudes resulting from the current fleet of civil aircraft, as resulting from the model of Isaksen and Hov (1987). The results are from Fuglestvedt and Isaksen (1991).



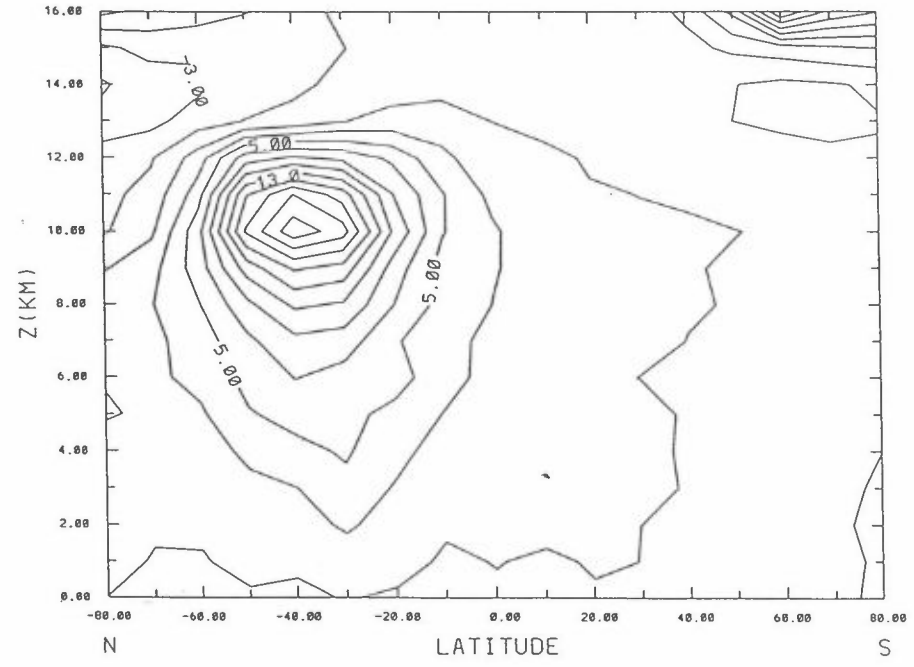


Figure 3 cont.

and lower stratosphere where, as we have already pointed out, NO_x very efficiently produces ozone. 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.

Johnson et al. (1992) have estimated the amplification to be about a factor 30. It must be noticed that such a number should be used with caution, not as much because the effect of aircraft emissions are uncertain, but particularly as the effect of surface NO_x emissions is difficult to assess. The change in ozone resulting from surface emission of NO_x depends on the photochemistry in the region of the emissions, and varies considerably between various locations.

The nitrogen oxides (NO and NO_2) are involved in photochemical production of ozone, as explained above, but also in catalytic destruction of ozone (Figure 5). The former process dominates in the troposphere and the lower stratosphere, and the latter in the middle and upper stratosphere. The crossover point, the altitude where the two effects cancel, is at approximately 15 km. Only very few aircraft presently fly above this level. Only the Concorde SSTs fly at levels where the NO_x emitted in the exhaust deplete ozone (Figure 6). The presently flying Concorde aircraft deplete the stratospheric ozone layer to a very small degree, 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 Concordes are being proposed from several aircraft manufacturers (Figure 6). Ongoing research shows that if such aircraft are flying high enough they can significantly deplete the ozone layer (Stordal et al., 1991), especially at middle and high latitudes.

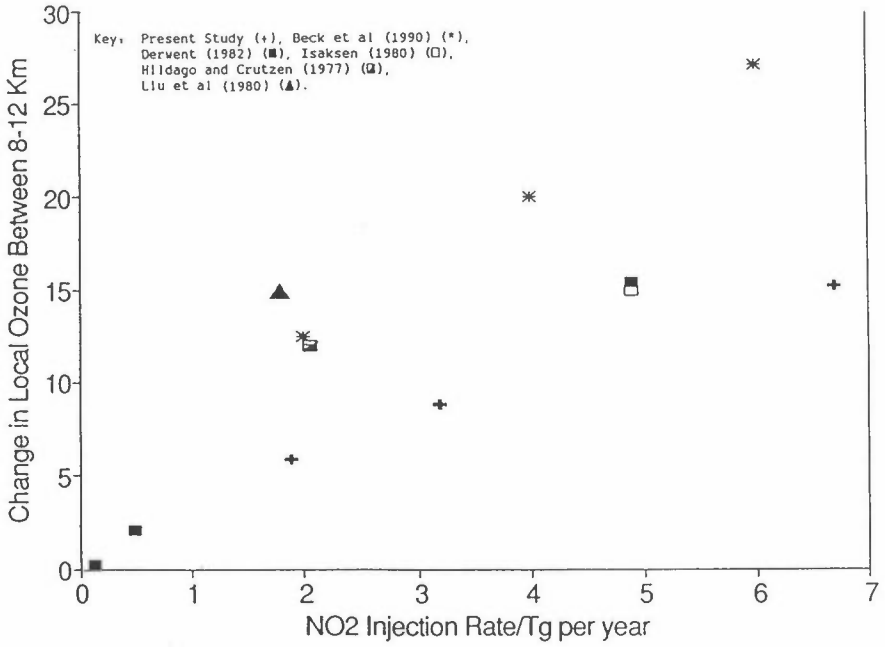


Figure 4: Percentage change in ozone concentrations in the 8-12 km region at 30° N versus NO_x aircraft emissions for various models. From Johnson and Henshaw (1991)

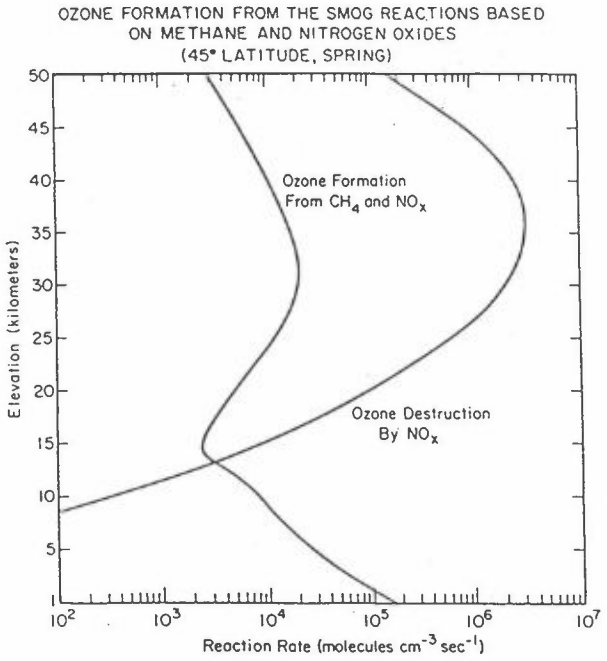


Figure 5: Rates of ozone formation and ozone destruction due to photochemical reactions involving NO_x species in the troposphere and the stratosphere. From Douglas et al. (1991).

2.4 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 reversed, and the temperature decreases with altitude. The tropopause has a larger vertical extent 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 3, Fuglestvedt and Isaksen, 1991). In the lower troposphere an increase in the OH concentration accompanies the ozone increase, as OH is formed from ozone. However, in the upper troposphere and lower stratosphere OH is removed by the NO_x injected in the atmosphere by aircraft (Figure 3).

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

Since the levels of ozone precursors - NO_x, CO, CH_4 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 trends have been found in some locations.

In addition to the recent surface ozone measurements, there are a 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 show values of ozone that were only about half of the present day values (Figure 7).

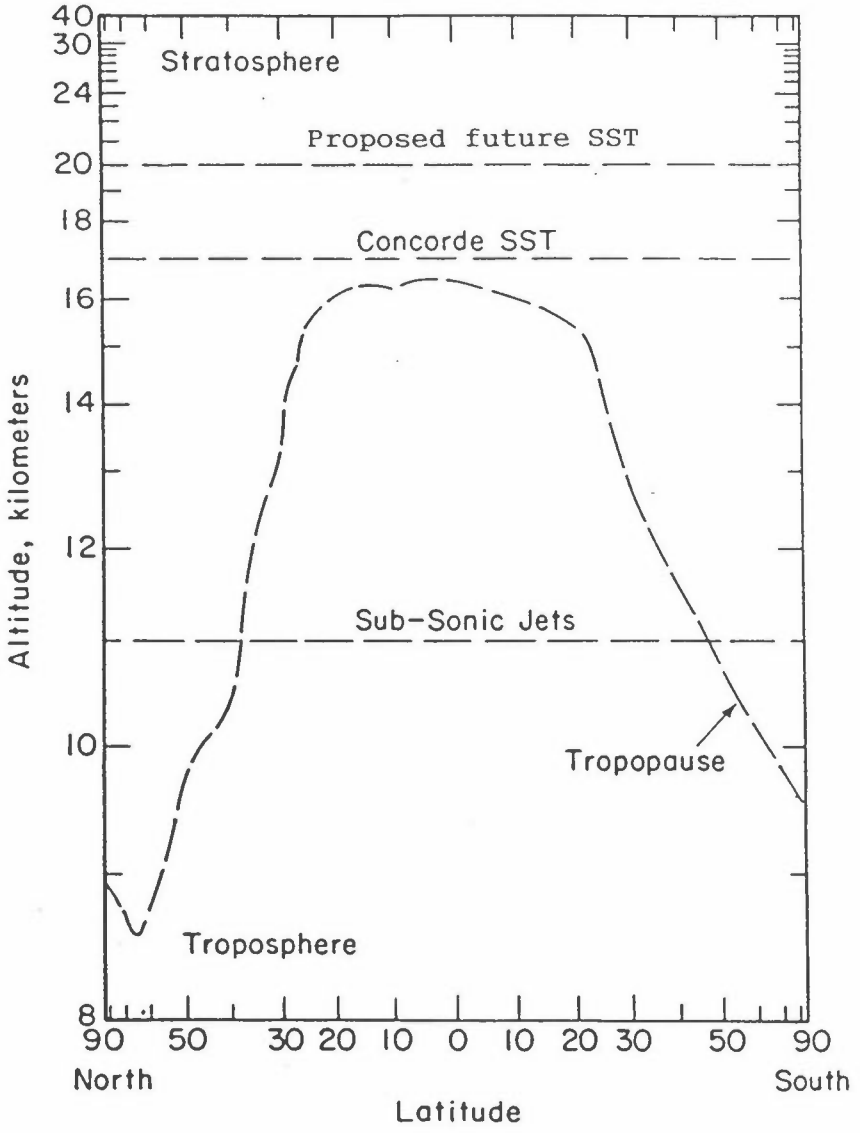


Figure 6: Tropopause altitudes at various latitudes, and typical cruise levels for presently flying subsonic jet and Concorde supersonic aircraft as well as future supersonic aircraft proposed by the aircraft industry. From Johnston (1990).

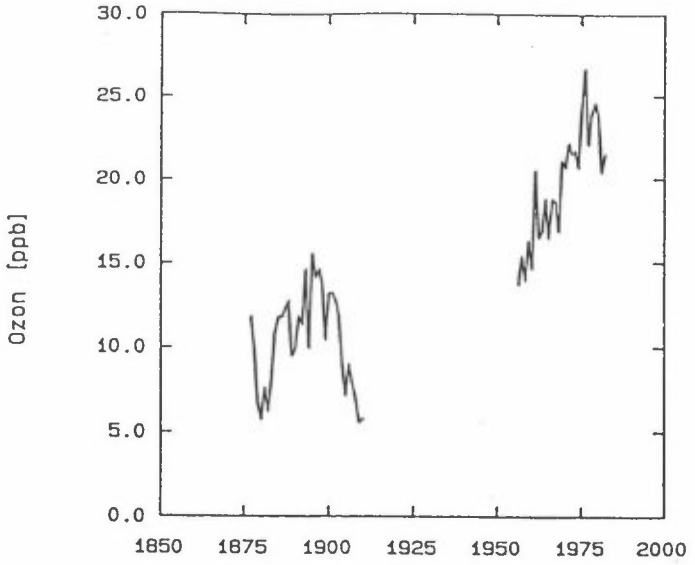


Figure 7: Annual averages of ozone mixing ratios at Montsouris near Paris around turn of the last century (Volz and Kley, 1987), compared to recent data measured at the clean air station Arkona in northern Germany (Feister and Warmbt, 1987).

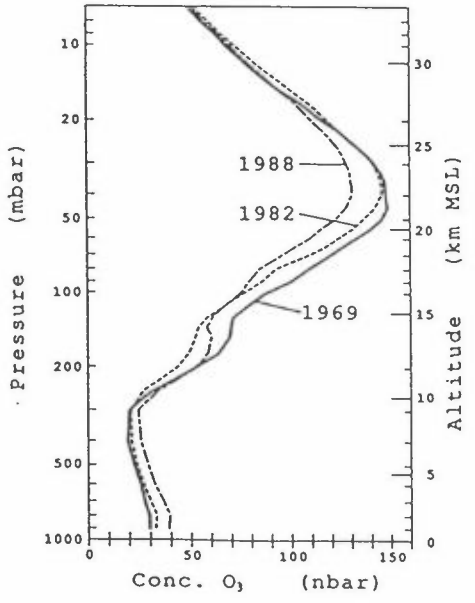


Figure 8: Selected annual means of the ozone balloon soundings from Payerne in Switzerland.

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. The Canadian data might be affected by an instrument change that occurred in the early 1980's.

3 CONCLUSIONS

Nitrogen species is known to contribute significantly to the acid deposition. As the sulphur emissions in some regions are currently being reduced, as in the US and Europe, the contribution from nitrogen species is becoming increasingly important. Deposition of anthropogenically released nitrogen to acid forest soil may in the long run cause several negative effects to trees as well as to the soil.

Nitrogen is particularly important in the assessment of critical loads, as it regards critical loads with respect to acidification as well as eutrophication of the ecosystem. In the Nordic countries there has been an increase in the deposition of oxidized nitrogen during the last decade. The deposition is largest in the southern parts of the region, i.e. Denmark and southern parts of Norway and Sweden.

Several gases are emitted in aircraft exhaust. Of particular interest are NO_x compounds which produce ozone, which controls the oxidizing capacity of the atmosphere, and which is a significant greenhouse gas. The comparatively small emissions of carbon monoxide and unburnt hydrocarbons have probably little effect on ozone.

Aircraft emissions of NO_x constitute only a small fraction of the total man made emissions. In Norway aircraft contribute with about 2%, while the number is 1% or less on a global scale. Despite the moderate contribution to the total anthropogenic emissions, NO_x emitted by aircraft is estimated to signi-

ficantly influence the atmosphere, since NO_x released in the middle troposphere is more efficient in producing ozone than surface emissions, and since ozone in the middle and upper troposphere is more efficient as a greenhouse gas than ozone further below.

Ozone is formed in areas rich of NO_x, but is chemically destroyed in NO_x poor environments. Regions with enhanced NO_x due to man made emissions is probably confined to the continents and aircraft routes in the vicinity (within a few thousand kilometers) of emissions.

Ozone concentrations are presently increasing in the troposphere over certain regions in the Northern Hemisphere, both at the surface and throughout most of the troposphere. A positive secular trend has so far been documented over Europe and Japan.

4 PROPOSED FUTURE WORK

Although the main patterns of acid deposition is known, more observations are needed to get a sufficient understanding of the deposition of nitrogen species. In particular better estimates of dry deposition of nitrogen components is needed.

More knowledge of the nitrogen cycle in soils is needed. The complicated interplay between several processes should be studied and integrated in numerical models.

The concept of critical loads has only recently been introduced. Further work with determination of critical loads is needed if the concept shall be useful as a policy tool.

A better understanding of the dry deposition processes would give a better estimation of the total deposistion of nitrogen.

Model estimates of the impact of aircraft exhaust on the chemistry and physics of the atmosphere is uncertain due to

incomplete knowledge about several topics. Firstly, estimates of NO_x emissions are uncertain, both total releases from aircrafts 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 ozone 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 boudary 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. ozone due to incresed emissions of several source gases known to take place at present. Further model studies of infrared radiative (greenhouse) effects of increased ozone due to aircraft emissions is also needed.

Increased efforts in observations and monitoring of the chemical composition of the atmosphere is 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. To understand the global ozone budget, measurements of NO_x species are needed over remote areas, as over oceans, including vertical profiles throughout the troposphere. Similarly observations of ozone at the surface as well as by vertical soundings is needed, both to understand the processes governing tropospheric ozone and to establish secular trends.

5 REFERENCES

Beck, J.P., Reeves, C.E., De Leeuw, F.A.A. and Penkett, S.A. (1992) The effect of aircraft emissions on tropospheric ozone in the Northern Hemisphere. Atmos. Environ., 26A, 17-29.

Derwent, R.G. (1982) Two-dimensional model studies of the impact of aircraft exhaust emissions on tropospheric ozone. Atmos. Environ., 16, 1997-2007.

Douglas, A.R., Carroll, M.A., DeMore, W.B., Holton, J.R., Isaksen, I.S.A., Johnston, H.S. and Ko, M.K.W. (1991) The atmospheric effects of stratospheric aircraft: A current consensus. Washington D.C., National Aeronautics and Space Administration (NASA Ref.Publ.1251).

Fuglestvedt, J. and Isaksen, I.S.A. (1991) Calculations performed in connection with the ongoing Assessment of Stratospheric Ozone, by United Nations Environment Programme (UNEP) and World Meteorological Organization (WMO) (private communication).

Hidago, H. and Crutzen, P.J. (1977). The tropospheric and stratospheric composition pertubed by NO_x emissions of high-altitude aircraft. J. Geophys. Res., 82, 5833-5866.

IPCC (1990) Intergovernmental Panel on Climate Change, Scientific Assessment by Working Group 1. Ed. by Houghton, J.T., Jenkins, G.J. and Ephraums, J.J. United Nations Environment Programme (UNEP) and World Meteorological Organization (WMO).

Isaksen, I.S.A. (1980) The tropospheric ozone budget and possible man-made effects. Proceedings of Quadrennial Ozone Symposium, WHO, Boulder 4-9 August.

Isaksen, I.S.A. and Hov, O. (1987) Calculation of trends in the tropospheric concentration of O_3, OH, CO, CH_4 and NO_x. Tellus, 39B, 271-285.

Isaksen, I.S.A., Stordal, F. and Berntsen, T. (1990) Model studies of highflying supersonic commercial transport on stratospheric and tropospheric ozone. University of Oslo, Institute of geophysics (Institute Report no.76).

Johnson, E.E., Henshaw, J. and McInnes (1992) Impact of aircraft and surface emissions of nitrogen oxides on tropospheric ozone and global warming, Nature, 355, 69-71.

Johnson, C.E. and Henshaw, J. (1990) The impact of NO_x emissions from tropospheric aircraft. AEA Environment and Energy, Harwell Laboratory, Oxfordshire (Report AEA-EE-0127).

Johnston, H.S. (1990) Global ozone balance and currently proposed supersonic aircraft. Paper presented to American Chemical Society, August 1990.

Knudsen, S. and Strømsøe, S. (1990) Kartlegging av utslipp til luft fra norsk sivil luftfart. Lillestrøm, Norwegian Institute for Air Research (NILU OR 88/90) (in Norwegian).

Liu, S.C., Kley, D., McFarland, M., Mahlman, J.D. and Ley. H. (1980) On the origin of tropospheric ozone. J. Geophys. Res., 85, 7546-7551.

Nihlgård, B. (1985) The Ammonium Hypothesis - An Additional Explanation to the Forest Dieback in Europe. Ambio, 14, 1.

Lövblad, G., Andersen, B., Joffre, S., Pedersen, U., Hovmand, M., and Reissell, A. (1992) Mapping Deposition of Sulphur, Nitrogen and Base Cations in the Nordic Countries. Nordic Councils of Ministers (in press).

Stordal, F., Rognerud, B. and Isaksen, I.S.A. (1991) Impacts on the stratospheric ozone layer from highflying aircraft. Invited abstract: EGS XVI General Assembly, European Geophysical Society, Wiesbaden, April 1991.