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CLIMATE IMPACT OF TROPOSPHERIC OZONE CHANGES

Authors

F. Stordal, S. Bekki, D. Hauglustaine, M. Millan, R. Sausen, E. Schuepbach, D. Stevenson, R. van Dorland and A. Volz-Thomas

4.1 Background

4.1.1 Introduction

The present chapter focuses on the changes in tropospheric ozone and their impact on climate. Research on tropospheric ozone started about 150 years ago, very soon after its discovery. Having received wide interest as an indicator of clean air in cities during the second half of the 19th century, tropospheric ozone regained the attention of European scientists and environmental policy makers in the early 1970s as an air pollutant, because its mixing ratio was found to increase well above toxic levels during photochemical smog episodes, which had first been observed in southern California. More than a decade ago evidence was presented that the ozone background in the lower troposphere over Europe had increased by a factor of two or more since the beginning of the 20th century [Volz and Kley, 1988]. The trend seemed to become less pronounced in the nineties, although no clear picture emerged from the many data sets of the different ozone monitoring networks that had been established, especially in the upper troposphere.

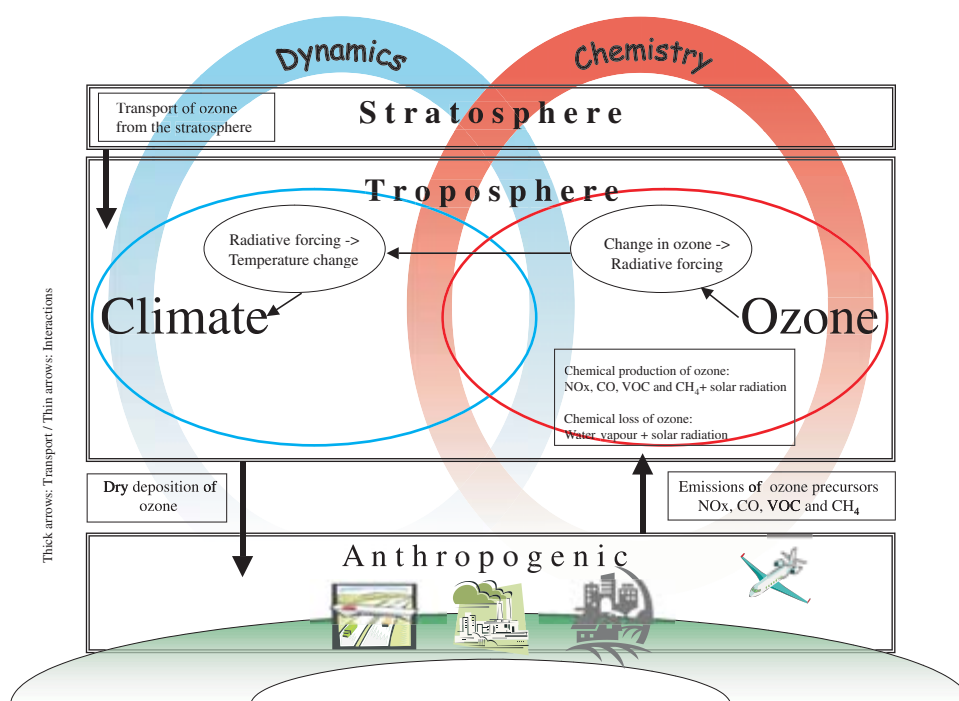


Figure 4.1 Processes governing the budget of tropospheric ozone and processes through which changes in tropospheric ozone impacts the climate

It is now well recognised that the large increase in the background tropospheric ozone concentrations during the previous century is mainly due to anthropogenic emissions of gases leading to ozone formation, the so-called ozone precursors (NO_x, CO, CH₄ and Volatile Organic Compounds

(VOCs), see Figure 4.1). Tropospheric ozone influences the radiative balance of our planet, both by absorbing solar radiation and, more importantly, by acting as a greenhouse gas (GHG) through absorption and re-emission of infrared radiation from the Earth's surface. As a result, the ozone increase in the troposphere has led to trapping of radiation in the troposphere, yielding a positive radiative forcing (see Chapter 1 for definition, and see Figure 4.1). Recent calculations show that the increase in tropospheric ozone represents the third largest positive radiative forcing since pre-industrial time [IPCC, 2001]. In this way the ozone increase has had a significant climate warming impact (see Figure 4.1). In addition tropospheric has an indirect climate impact as it decreases the concentration methane, through its chemical influence on OH.

Ozone has an atmospheric residence time in the order of weeks to a few months, depending on altitude, geographical location and time of the year. As a consequence, changes predominantly occur downwind of the industrial regions. The complexity of atmospheric chemistry, often critically depending on a variety of atmospheric conditions, leads to a large variability in space and time of the ozone levels.

- *Tropospheric ozone has increased and is expected to continue to increase in certain regions due to anthropogenic emissions of ozone precursors.*
- *Tropospheric ozone absorbs terrestrial as well as solar radiation resulting in a positive radiative forcing when ozone concentrations increase.*
- *Due to its short lifetime changes in the concentrations and the subsequent radiative forcing and climate effects are all very difficult to quantify accurately.*

4.1.2 Budget of tropospheric ozone

Quantifying the impact of natural and anthropogenic ozone as a climate gas requires quantitative understanding of the processes that control its abundances (see Figure 4.1). This is more complex for tropospheric ozone than for long-lived greenhouse gases. The concentration of long-lived greenhouse gases in the troposphere can be determined from a limited number of measurement stations at the ground. This is not the case for ozone, which varies very significantly in space and time throughout the troposphere.

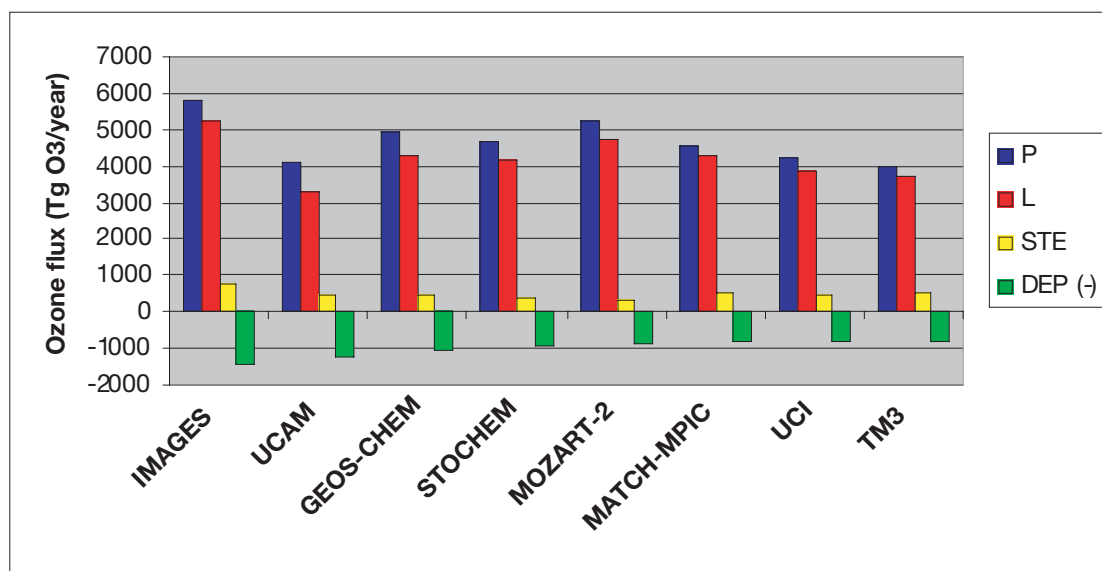


Figure 4.2 Tropospheric ozone budget based on 8 global chemistry transport models (CTM). The figure shows the chemical production (P) and loss (L), the Stratosphere-Troposphere Exchange (STE) and the surface deposition (DEP). See text for further discussion. The numbers represent global averages for 1990 conditions.

Ozone is formed *in situ* in the troposphere in photochemical reactions. It is also transported into the troposphere from the stratosphere, where the concentrations are much higher than in the troposphere. The chemical production in the troposphere is counteracted by other reactions that destroy ozone. Finally, ozone is also removed from the troposphere by deposition at the ground, most efficiently by vegetation. The total amount of ozone in the troposphere, which is about 370 Tg(O₃) [Park et al., 1999], is only slowly varying, and the amount for a given year (for example) is almost maintained as a near balance between the production and loss processes described above.

The influx of stratospheric ozone into the troposphere is very difficult to quantify. Based on observed correlations with other gases it has been estimated to be about 475 Tg(O₃)yr⁻¹ [Murphy and Fahey, 1994]. The uncertainty is large, and in current chemistry transport models (CTMs) the ozone influx from the stratosphere ranges from about 420 to 790 Tg(O₃)yr⁻¹ (from Figure 4.2). The ozone concentrations have decreased in the stratosphere over the last decades due to emissions of ozone depleting substances (see Chapter 2). Thus the influx has been reduced. In a recent study, which is based on a combination of model calculations and analysis of ozone observations, it has been estimated that the transport of ozone from the stratosphere to the troposphere has been reduced by as much as 30% from the early 1970s to the mid 1990s [Fusco and Logan, 2003].

The magnitude of the surface deposition sink is difficult to establish from observations, and the range in the CTM calculations is again large, ranging from 670 to 1640 Tg(O₃)yr⁻¹. The difference between the flux from the stratosphere and the destruction at the surface is balanced by the net chemical production (NCP), which varies from 70 to 880 Tg(O₃)yr⁻¹ in current models. This shows that, from current CTM studies, it is not possible to accurately quantify the relative importance of stratospheric input, deposition at the surface and net chemical production in the global tropospheric ozone burden. Therefore, the evaluation of the mutual impact of tropospheric ozone and climate on each other remains a difficult task.

NCP of tropospheric ozone is the difference between chemical production (P) and chemical loss (L), which are typically one order of magnitude larger than the NCP (chemical production and loss: each ~4000-6000 Tg(O₃)yr⁻¹). Chemical production of ozone occurs (see Chapter 1) when NO reacts with a peroxy radical (HO₂, CH₃O₂, or RO₂ - a peroxy radical derived from a VOC other than methane). For this reason, hydrocarbons and nitrogen oxides are called ozone precursors. Ozone is chemically destroyed in the troposphere in a catalytic cycle involving OH and HO₂, and in a pathway involving the reaction of O(¹D), a product of ozone photodissociation, with H₂O. There are other minor loss processes; reaction of O₃ with hydrocarbons, and NO_y (the sum of NO and its atmospheric oxidation products) losses, which represent an indirect sink of O₃. A chemical loss of ozone in the Arctic winter/spring has been identified, involving halogen radicals and aerosols [e.g. Platt and Moortgat, 1999]. The contribution to the global loss is probably small. NCP also has a distinct distribution: it peaks in the polluted boundary layer, but is negative over much of the oceans and in the lower-mid free troposphere, before becoming positive again in the upper troposphere. This distribution mainly reflects the concentration fields of NO_x (typically a C-shaped profile, with maxima in the boundary layer and in the upper troposphere) and H₂O.

The large spatial variability in ozone abundances in the troposphere reflects the wide range of scales in the distributions of ozone precursors. The representation of all the relevant scales in global models is a major challenge. For example, urban, biomass burning and aircraft plumes (and convection) are small scale phenomena which cannot be fully resolved in global models. Since the ozone chemistry is strongly non-linear, the crude parameterisation of such

processes in global CTMs makes it difficult to establish reliable regional and global budgets for tropospheric ozone.

- *The content of ozone in the troposphere is maintained as a balance between transport from the stratosphere, deposition at the ground and in situ chemical production and loss. Figures representing these processes can be established by use of chemistry-transport models. The crude parameterisation of certain processes (in particular small scale processes) in global CTMs makes it difficult to establish an accurate global budget for tropospheric ozone.*
- *Increased chemical production due to emissions of ozone precursors is not the only process that has changed the content of ozone in the troposphere. Combination of model calculations and analysis of ozone observations has revealed that the transport of ozone from the stratosphere to the troposphere has been reduced by as much as 30% from the early 1970s to the mid 1990s.*

4.2 Ozone precursors: anthropogenic emissions and concentrations

Most of the ozone in the troposphere is formed as a result of emissions of precursor gases, both natural and anthropogenic emissions. Since anthropogenic emissions are acknowledged as being the main driver behind the increase in tropospheric ozone since pre-industrial time, they are our main concern here. The magnitude, variability and trends of anthropogenic emissions of ozone precursors are described in this section. Natural emissions of ozone precursors are described in Chapter 5, as changes in natural emissions of ozone precursors can occur as a result of climate changes.

- *Anthropogenic emissions are acknowledged as being the main driver behind the increase in tropospheric ozone since pre-industrial time.*

4.2.1 Methane (CH₄)

Methane is one of the major greenhouse gases. In addition, like other hydrocarbons, its oxidation in the presence of significant amount of NO_x and sunlight leads to photochemical production of ozone. The total emission of methane has been estimated to be about 600 Tg of CH₄ per year. About 60% of it is of anthropogenic origin with the most important sources being related to fossil fuel use, ruminants and agriculture. Its global burden is of the order of 5000 Tg of CH₄ and its lifetime is about 9 years [IPCC, 2001].

The tropospheric distribution of methane is relatively well characterised, especially through surface measurements in global monitoring networks (vertical gradients are very small in the troposphere). Ice core measurements indicate that pre-industrial levels of methane were about 40% of the present-day levels. This appears to match well the estimated fractions of anthropogenic (60%) and natural (40%) emissions. CH₄ levels have been increasing since pre-industrial time. However, the increase seems to have been slowing down in the last two decades (see Figure 4.3) [Dlugokencky et al., 1998]. The exact causes for this apparent long-term decline in the CH₄ growth rate have not yet been clearly identified. There are also some large year-to-year fluctuations of the growth rate superimposed on the declining trend. The explanations put forward for these fluctuations invoke inter-annual variations in biomass burning, gas, coal, or wetlands emissions, or in the OH sink (see Chapter 5). Note that even the largest of these inter-annual fluctuations corresponds to an annual change in the atmospheric CH₄ burden of less than 1% of the total burden.

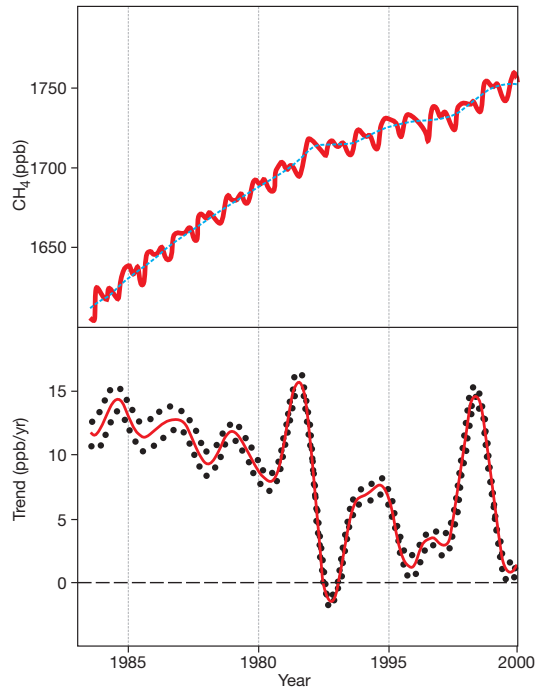


Figure 4.3 CH_4 mixing ratio (upper panel) and growth rate (lower panel).

- Methane is a greenhouse gas yielding a radiative forcing. In addition emissions of methane leads to an indirect radiative forcing through formation of ozone which takes place in the presence of NO_x .
- An observed upward trend and inter-annual variations in the growth rate are influenced by the history of emissions and removal of methane by OH. Anthropogenic emissions account for 60% of the total emissions.

4.2.2 Carbon monoxide (CO)

Presently most of the direct emissions of CO are man-made (almost evenly distributed between emissions from biomass burning and fossil fuel). The other important anthropogenic source of CO is the oxidation of hydrocarbons, in particular methane, emitted during industrial activities. As a result the northern hemisphere contains about twice as much CO as the southern hemisphere. Even within the northern hemisphere there are large gradients, due to the relatively short lifetime (1-2 months) and heterogeneous emission patterns. Its global burden is estimated to be about 360 Tg [IPCC, 2001].

As pointed out for ozone in the previous section, there are uncertainties in establishing the distribution and the budget of CO because of the spatial and temporal variations (see Figure 4.4) and the lack of observations to resolve the variability. It is worth noticing that satellite retrieval of the CO column have been achieved with Measurement of Air Pollution from Satellite (MAPS) and Interferometric Monitor for Greenhouse gases/Advanced earth Observing System (IMG/ADEOS) data [Clerbaux et al., 1998; Connors et al., 1999]. These data, along with the satellite observations with Measurements Of Pollution In The Troposphere (MOPITT), should help to reduce some of the uncertainties. Surface observations have shown that CO increased slowly on a global scale until the late 1980s. Then, there has been a decrease of about 2% per year for the period 1991-1997 [Novelli et al., 1998], possibly due to decrease in motor vehicle emissions as a result of catalytic converters. A trend in OH would also impact the trend in CO.

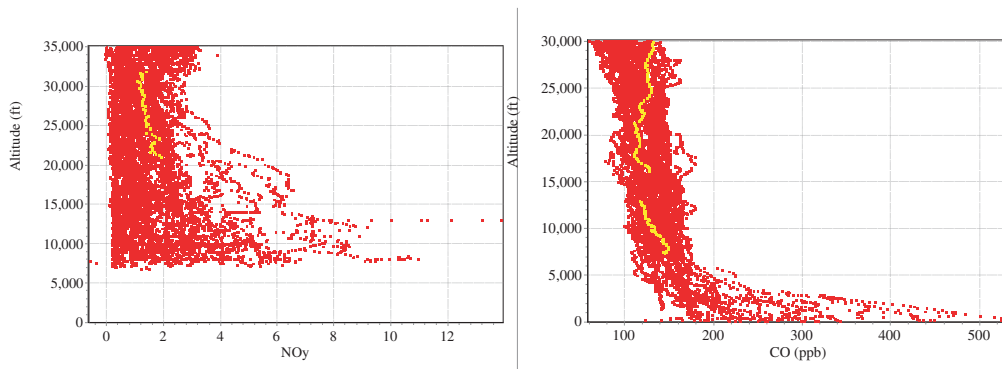


Figure 4.4 Vertical profiles of NO_y (left) and CO (right) over Frankfurt in spring 2002, collected by the EC funded MOZAIC project [Source: P. Nedelec and A. Volz-Thomas, MOZAIC data base].

- CO contributes to generation of tropospheric ozone. The majority of direct emissions are anthropogenic. An increasing trend during much of the previous century was followed by a slight decline over the final decade, possibly due to decrease in motor vehicle emissions as a result of catalytic converters.

4.2.3 Volatile Organic Components (VOC)

As for CH₄, VOCs (including non-methane hydrocarbons), produce ozone in the presence of NO_x and sunlight (Figure 4.1). In general the natural emissions are larger than the anthropogenic. However, this is only true for global budgets and for the VOC group as a whole. The fraction of anthropogenic emissions varies considerably between individual species and also geographically. The largest anthropogenic sources are from motor vehicles due to evaporation and incomplete fuel combustion, as well as biomass burning. VOCs include a large number of species, most with a very short lifetime. Their global distributions and budgets are only poorly known from a sparse network of observations. Anthropogenic emissions occur in heavily populated and industrialised regions where natural emissions are relatively low. Consequently they have a major impact on ozone formation on local and regional scales.

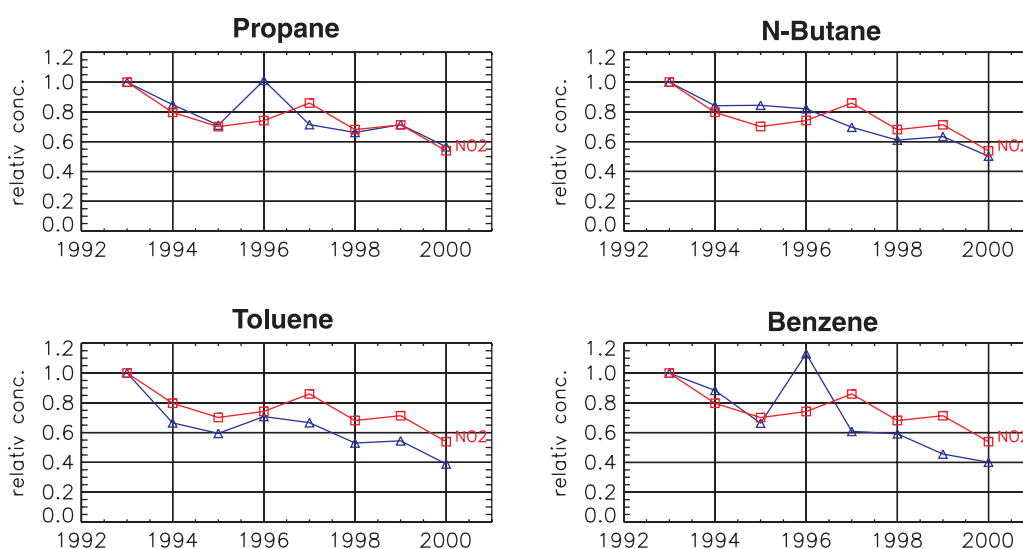


Figure 4.5 Winter medians (Jan-March) of selected hydrocarbons (blue) and NO₂ (red) at Waldhof from 1993-2000. The NO₂ data are 24-hour samples and only from those days when hydrocarbon samples were collected. All data are relative to the medians in 1993 [Solberg et al., 2002].

A few VOCs, such as ethane and acetone, have lifetimes long enough to impact ozone formation on a hemispheric scale. Data for ethane are most comprehensive. Ethane levels increased in Europe in the 1950s and through most of the 1980s [Ehhalt et al., 1991]. Since then they have been decreasing (-2.7% per year in the column at Jungfraujoch [Mahieu et al., 1997] somewhat weaker trends in Arizona/US and New Zealand). Figure 4.5 shows that the decrease in ethane is accompanied by similar negative trends in other VOC components (and NO₂) in the 1990s at a rural German station (20-50% decline in the period 1993-2000). This is indicative of covariance in the VOC and NO_x sources in this region.

- A wide range of VOCs contribute to ozone formation in the troposphere. On a global scale the natural emissions dominate, but on local and regional scales anthropogenic emissions produce significant amounts of ozone. The largest anthropogenic sources are from motor vehicles and biomass burning.

4.2.4 Nitrogen oxides (NO_x)

Nitrogen oxides catalyse ozone formation in the troposphere. The lifetime ranges from about one to up to ten days, and there is an extremely large spatial and temporal variability in the concentrations of NO_x (see for example profiles of NO_y, a group of species strongly correlated to NO_x, from MOZAIC in Figure 4.4), ranging from a few pptv in remote oceanic regions to a few hundred ppbv in polluted urban regions. The global distribution and budget of NO_x is thus very difficult to establish, although column abundances of NO₂ are now measured from satellite platforms. The Global Ozone Monitoring Experiment (GOME) satellite data show maxima of NO₂ over industrialised regions in North America, Europe and Asia (Figure 4.6). It is important to note that in polluted situations surface layers contribute most to the total columns, and in this altitude region derivation of satellite data is often difficult due to e.g. clouds and aerosols. Potential future retrievals of NO_x profiles from satellite observations are of extreme importance for the understanding of ozone budget since NO_x is a major driver of ozone chemical production in the troposphere.

The major anthropogenic source of NO_x is fossil fuel combustion (approximately 33 Tg(N) per year), with 40% from the transportation sector. The emissions and concentrations (see Figure 4.5) have been slowly decreasing in Europe since the early 1990s. However, emissions were increasing in south-east Asia in the 1980s, and are possibly still increasing, although there are indications that the emissions of some greenhouse gases are now reduced in this region. Another anthropogenic source, which is more uncertain, is biomass burning which can be seen in the satellite data over Africa in Figure 4.6. However, parts of the emissions from biomass burning and soils are of anthropogenic origin (see also Chapter 5).

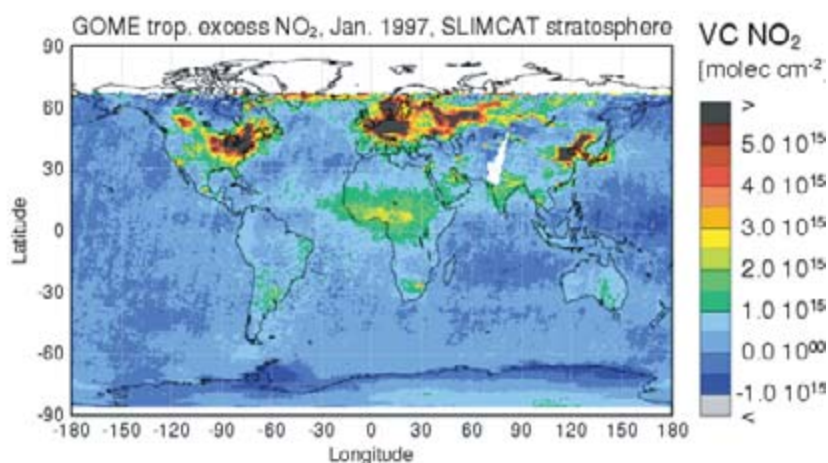


Figure 4.6 Global distribution of the tropospheric NO₂ column derived from the GOME instrument (TROPOSAT Annual Report 2001). The stratospheric SLIMCAT model has been used to remove the stratospheric part of the column.

- NO_x play a special role in formation of tropospheric ozone since they catalyse ozone production. The major anthropogenic source is fossil fuel combustion with approximately 40% from the transport sector. NO_x is short-lived and there is a large spatial variability in the concentration and the ozone formation.

4.2.5 Aircraft emissions

Aircraft are the only anthropogenic source that emits outside the planetary boundary layer. This increases the effect of species with a short atmospheric lifetime like NO_x . The primary emissions of aircraft are CO_2 and H_2O as direct product of the combustion of kerosine. Due to its rather long lifetime (many decades) the location or altitude of the CO_2 emission is of negligible importance for the associated climate impact. In the case of water vapour emissions location matters. In particular, water vapour emissions from a large fleet of supersonic aircraft would have a noticeable climate impact [IPCC, 1999].

Beside the primary emissions, aircraft also emit various other species like NO_x , VOCs, sulphur components or soot (at rates at least two orders of magnitude smaller than the primary emissions). With respect to an impact on ozone the aviation NO_x are of particular interest. At cruise altitude the life time of NO_x is at least an order of magnitude larger than close to the surface. Therefore, significantly more ozone can be produced per NO_x molecule released by aircraft than by NO_x from surface emissions. In addition the lifetime of ozone produced in the upper troposphere is significantly longer than at lower altitudes, making the potential ozone change larger.

Several 3-D inventories of these emissions exist for the period around 1992 [Baughcum et al., 1996; Schmitt and Brunner, 1997; Gardner, 1998]. All have similar total amounts and geographical and vertical distributions. Figure 4.7 shows the zonal mean aircraft NO_x emissions according to the DLR-2 inventory [Schmitt and Brunner, 1997]. Obviously, a large fraction of the emissions occurs at cruise altitude (10-12 km ~260-190 hPa) in the northern extra-tropics. The global mean emissions are 0.56 Tg N per year.

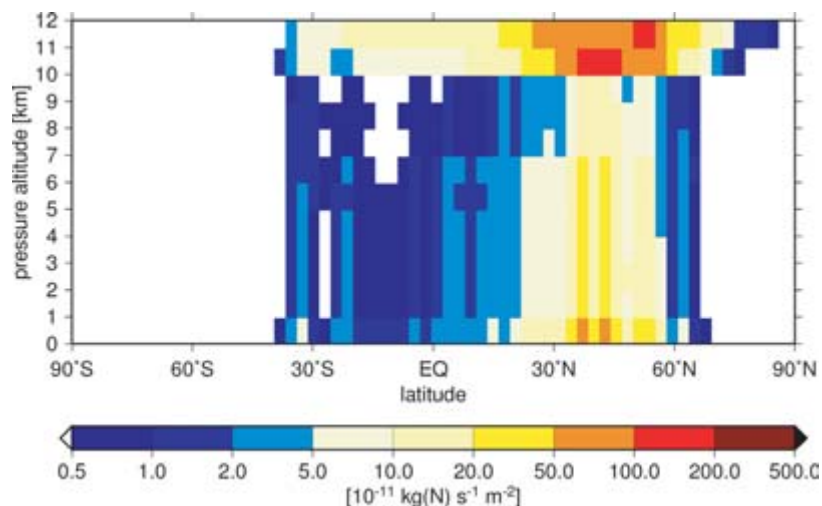


Figure 4.7 Zonal mean aircraft NO_x emissions according to the DLR-2 inventory for the year 1992. Note that the contour scaling is approximately logarithmic.

- Climate impacts of aircraft through formation of ozone from NO_x emissions are important as the emissions take place in a region with large climate impact of ozone changes and because NO_x has a long lifetime in this region.

4.2.6 Future emissions

The future distribution of ozone, and thus its impact on climate, will be controlled by the evolution of emission strengths of ozone precursors, which is very uncertain. In the few next years, and

perhaps decades, it is likely that certain trends in emission patterns of ozone precursors will persist. For example, it is likely that the shift in growth of emissions of NO_x , CO and VOC from north America and Europe to southern and eastern Asia will continue. This will in turn significantly affect the spatial distribution of ozone. This is in contrast with long-lived GHGs, as shifting the location of their emissions has little impact on their distribution.

IPCC have developed several scenarios for GHG emissions, including ozone precursors, until 2100 (IPCC SRES – Special Report on Emission Scenarios). One of the scenarios has been used by the global tropospheric ozone modelling community to estimate the ozone distribution, and the resulting radiative forcing, in the year 2100 [the OxComp study, Prather and Ehhalt, 2001]. The scenario chosen, A2p [see IPCC/SRES; and IPCC, 2001, Annex II] has the highest emissions of ozone precursors throughout most of the 21st century. Therefore, the results of the OxComp calculations, which are used to discuss the future impact of tropospheric ozone on climate in this chapter (see Sections 4.3.2 and 4.4.1), should be regarded as upper estimates of this impact.

In the scenario adopted in OxComp [Prather et al., 2001] anthropogenic NO_x and CO emissions increase by a factor 3.4 and 2.4, respectively, from 2000 to 2100. Relative increases in north America (1.8 for NO_x and 1.6 for CO) and Europe (2.0 and 2.0) are lower than the global average, whereas the increases in India (5.9 and 4.4), south-east Asia (5.7 and 4.6), Africa (7.7 and 6.0) and South America (8.7 and 6.5) are substantially higher.

Aircraft emissions are assumed to grow particularly rapidly during the next decades. IPCC [1999] provided several scenarios which correspond to the IS92 IPCC scenarios for greenhouse gas emissions. Until 2050 air traffic increases by factors of 3.6 to 10.1 relative to 1990 if only the FSEG scenarios are considered. The EDF scenarios assume a more rapid growth. (For definition of the scenarios, see [IPCC, 1999]). The corresponding NO_x emissions grow less rapid due to improved aircraft efficiency and cleaner engines. In the case of the FSEG scenarios the factors range from 2.0 to 5.7. Note that these growth factors only cover a 60 year period, i.e. the aircraft emissions increase faster than the total anthropogenic emissions.

- *Future emissions of ozone precursors are of course uncertain. In the next few decades it is likely that the current shift in surface emissions of NO_x , CO and VOC from Europe and north America to southern and eastern Asia will continue. Aircraft emissions are assumed to grow particularly rapid during the next decades.*
- *IPCC has developed several scenarios for the current century. One of these, A2p, which has the highest emissions of ozone precursors throughout most of the century, has been chosen for studies of future ozone and its climate implications discussed in this chapter.*

4.3 Distribution and trends of tropospheric ozone

Surface measurements made in the tropics in the late 19th century suggest fairly constant mixing ratios of only 5-10 ppbv throughout the year [Pavelin et al., 1999]. There are valid questions about the quantitative reliability of early surface ozone measurements [e.g. Lefohn et al., 2001]. However, they represent the only observational constraints on pre-industrial ozone levels. The lack of global information on pre-industrial tropospheric O_3 distributions is, therefore, the major uncertainty in the evaluation of the climate impact of changes in this key gas [Mickley et al., 2001; Hauglustaine and Brasseur, 2001; Shindell and Faluvegi, 2002].

To understand and quantify the impact of changes in tropospheric ozone on climate, it is therefore necessary to use models that describe complete global distributions, both for the pre-industrial and present atmosphere. The modelling intercomparison exercise conducted by Kanakidou et al. [1999] describes the state-of-the-art in global model resolutions, chemical scheme and per-

formances. However, joint scientific efforts are necessary in the future to combine observations and model results to establish and assess trends in ozone leading up to the current distribution.

- *Due to the large spatial variations in tropospheric ozone a global distribution of its concentration is difficult to establish. Thus one has to rely also on models to establish the global tropospheric ozone distribution and its trend. A joint observational and modelling effort is still needed to quantify changes in tropospheric ozone and the subsequent climate impact.*

4.3.1 Observed changes in tropospheric ozone

Due to the relatively short residence time of ozone in the troposphere the concentration of ozone is varying strongly within the troposphere on a global scale. As opposed to e.g. other well mixed greenhouse gases, as CO₂, observations are therefore needed at many locations and at many altitudes.

Surface ozone

Even within Europe the surface ozone concentrations are spatially varying to a large extent. A north-west to south-east gradient in current surface ozone concentrations can be observed in Europe [Scheel et al., 1997]. Similarly, a gradient towards higher concentrations from west to east in the United States [Logan et al., 1999] has been found. Current background ozone concentrations over Europe are between 19-27 ppbv on the Continent and 32 ppbv on the western European border [Scheel et al., 1997]. Levels of 35 ppbv are reported for unpolluted air masses at ground-level Mace Head (north-western Ireland) [Simmonds et al., 1997] and for the European Alps (Arosa, 1840 m asl) [Pochanart et al., 2001]. In the lower free troposphere over continental Europe (Jungfraujoch, 3,580 m asl), current background ozone levels are at 40 ppbv [Schuepbach et al., 2001]. In the Mediterranean regions background values are up to 60 ppbv, but in polluted areas ozone concentrations are often much higher. Not only the ozone concentrations, but also their seasonal cycle varies [Schuepbach et al., 2003].

Many factors control the variations in surface ozone, including emissions of ozone precursors, solar radiation to drive the photochemistry, moisture and transport processes. There is some observational evidence (see Figure 4.8) that ozone is formed as a result of ozone precursors during summer (the high NO_x percentiles in Figure 4.8), whereas ozone is reduced during winter, notably by emissions of NO_x and subsequent titration of ozone [Solberg, 2001]. This is most clearly demonstrated at locations at the outskirts of the major emission regions, experiencing the contrast between relatively clean and polluted air masses.

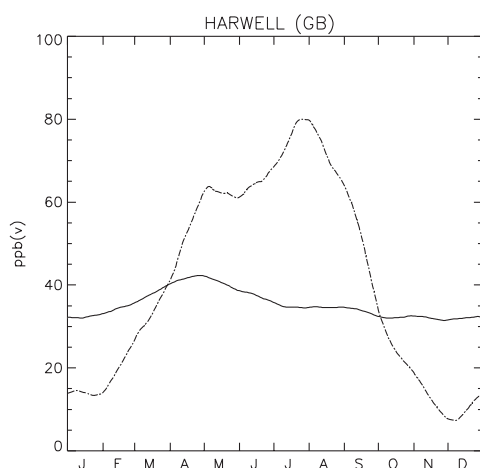


Figure 4.8 Seasonal cycles of ozone at Harwell, UK, as a function of the NO_x emissions integrated along back trajectories from the time of measurement. The solid curve represents back-

ground conditions and the dash-dotted curve polluted air masses (the lowest 25% and the highest 10% of the integrated NO_x emissions, respectively) [from Solberg et al., 2002].

Observed surface ozone concentrations in Europe show an increase by more than a factor of two over the 20th century [Staehelin et al., 1994; see also WMO, 1999; IPCC; 2001]. Ozone records dating back to the late 19th century are few, and the evidence for a positive trend in lower tropospheric ozone is strongest in Europe due to the first historical, quantitative ozone record at Montsouris (Paris). Volz and Kley [1988] established 11 ppbv as the observed background level at this site; the value is rather close to the 15 ppbv measured on the island of Rügen in northern Germany in the 1950s [Feister and Warmbt, 1987]. On the other hand, emissions of some of the ozone precursors declined during the last approximately two decades of the 20th century. As a consequence, observed summer ozone concentrations have generally decreased after the mid 1980s [e.g. Roemer, 2001], while winter ozone levels have increased [e.g. Schuepbach et al., 2001]. Similar trends in the seasonal ozone concentrations are observed in the northern United States [NARSTO, 2000]. These observed trends in the seasonal variations of ozone are consistent with reductions in emissions of NO_x over the last few decades.

Free troposphere ozone

More important than the surface concentrations of ozone are the concentrations at higher levels, where changes lead to a stronger forcing of the climate. Current experimental work in Europe [e.g. Zanis et al., 2000] and the northern United States [e.g. Wang et al., 2002b] based on measurements from high elevation surface sites and aircraft campaign measurements, suggest that in the lower free troposphere over large continental regions NCP (see Section 4.1.2) is positive, especially during the transition from winter to spring. An analysis of ozone sonde data in the mid troposphere (around 5 km altitude) showed that northern hemispheric seasonal variations are quite similar spatially, with a maximum in ozone between April and September [Logan et al., 1999]. This maximum is caused by efficient photochemical ozone formation in summer, possibly also by strong STE during spring. NO_x emissions from lightning, which are most efficient over continents in summer, may contribute significantly to the chemical production.

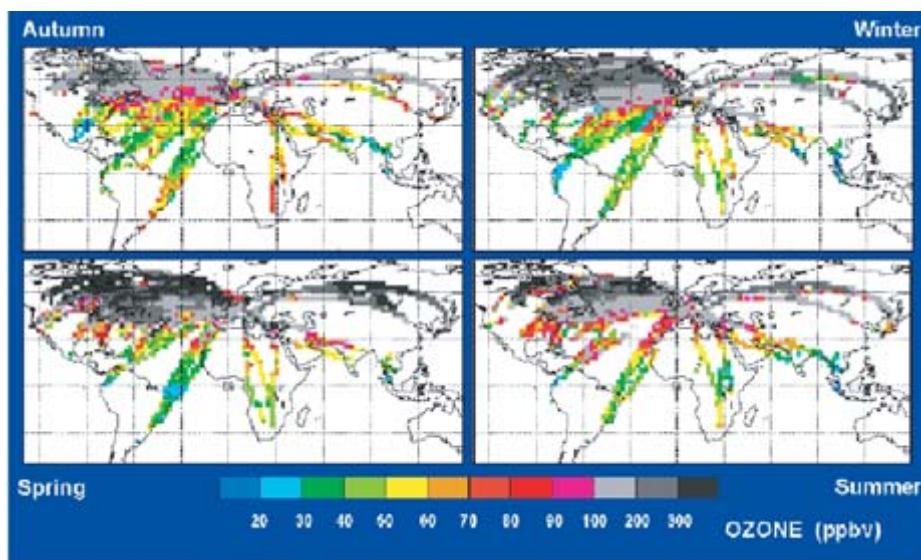


Figure 4.9 Seasonal climatologies of O_3 established from MOZAIC data collected in the altitude region 9-12 km between September 1994 and August 1996. The seasons specified in the panels refer to the northern hemisphere.

In the upper troposphere ozone observations of long-range Airbus A340 cruises from the long term program MOZAIC (Figure 4.9, from the 10-12 km altitude region) display a seasonal

variation in the northern hemispheric mid-latitudes with maximum concentrations in the late winter and spring. The peak concentrations over Europe are somewhat lower than over eastern north America and eastern Asia. However, Law et al. [2000] suggest that the magnitude of the maximum concentration depends on the position of the particular flight level where the measurements were collected relative to the height of the tropopause which is governed by the position of the large planetary scale ridge and trough pattern in the upper troposphere (ridge over Europe, trough over eastern north America and eastern Asia). In tropical areas, O₃ concentrations generally decrease toward the Equator and show less seasonal variation. At low latitudes the seasonal cycle is determined predominantly by the insolation since the direction of stratosphere-troposphere exchange is from the troposphere into the stratosphere. Hence, the influence of ozone originating from the stratosphere is very small throughout the year. The ozone concentration is thus mainly varying in line with photochemical destruction (which dominates the NCP) that minimizes on the winter and maximizes on the summer side of the Equator.

Outside the equatorial region, the seasonal variation in biomass burning emissions is an important factor. In the southern hemisphere, spring (northern hemisphere autumn) it is the dry season, when large-scale burning of biomass occurs in south America, Africa and Indonesia, which releases ozone precursors into the atmosphere. There is convective uplift of O₃ and its precursors from biomass burning regions into rather stable haze layers which can exist for many days and be transported large distances [Garstang et al., 1996; Diab. et al, 1996]. Due to the meteorological conditions, the biomass burning pollution is transported over large parts of the southern hemisphere. The influence by biomass burning between August and November has been observed in the seasonal cycle as seen in ozonesonde data at 500 hPa and 300 hPa [Logan et al., 1999].

Ozonesondes offer the longest record of free tropospheric ozone. Since 1970 observations are available weekly from a limited number of stations in a latitude range at mid to high latitudes in the northern hemisphere. Most stations show an increase in mid-tropospheric ozone from 1970 to 1980 [Logan, 1999]. Since then the changes are less clear. At some stations the increase continued in the period 1980-1996 (one in Europe, one in Japan), other stations had a significant decrease (all four Canadian stations), whereas other stations had no significant trend.

Total ozone columns

Full global observations of tropospheric ozone can only be obtained using satellites. There are no direct satellite observations of ozone in the troposphere. However, the tropospheric ozone columns have been obtained indirectly, by subtracting the stratospheric burden from the total column [Fishman et al., 1990; Fishman and Brackett, 1997]. Nevertheless, these observations show some interesting features. The lowest ozone concentrations are found over the tropical Pacific Ocean, due to photochemical loss under low NO_x and high water vapour conditions. In contrast, a plume high in ozone rises from the continents to fill up the troposphere between south America and Africa over the latitude range 0°-20°S during austral spring. However, the largest ozone columns are clearly seen at mid-latitudes in the northern hemisphere, especially during spring and summer, when solar radiation stimulates production of ozone following the anthropogenic emissions of precursors.

- *Concentrations of tropospheric ozone vary strongly in space and with time of the year. Anthropogenic impact on tropospheric ozone has been possible to establish from long-term in situ ozone records and observation campaigns, when combined with observations of ozone precursors. For example, NO_x has been demonstrated to yield ozone production during summer and ozone loss during winter conditions.*

- *A strong increase in surface ozone has been observed over Europe since the late 19th century. At mid and high latitudes in the northern hemisphere there was an increase in mid tropospheric ozone in the 1970s, according to ozone sonde observations. Since then there has been a continued increase in some regions, but a decreasing or not significant trend in others.*

4.3.2 Modelling and interpretation

Over the last 10 years, there has been a considerable effort devoted to the calculation of pre-industrial tropospheric ozone distribution and associated radiative forcing of climate. For this purpose, studies have used global 3-D CTMs [e.g., Chalita et al., 1996; Berntsen et al., 1997; van Dorland et al., 1997; Stevenson et al., 1998; Hauglustaine and Brasseur, 2001]. These studies generally calculate more pre-industrial ozone than observed with the Schonbein technique. In order to match these sparse observations, several studies have forced ozone to match pre-industrial levels in their radiative forcing calculations [Kiehl et al., 1999; Mickley et al., 2001; Shindell and Faluvegi, 2002]. Under these assumptions, radiative forcings reaching values as high as 0.7 Wm^{-2} are calculated.

As an example, Figure 4.10 illustrates the pre-industrial and present-day ozone surface mixing ratios simulated for January and July conditions [Hauglustaine and Brasseur, 2001]. The distributions were calculated with the state of the art global 3-D CTM of the troposphere, MOZART. The model calculates the ozone distribution by taking into account emissions of ozone precursors (natural emissions in the pre-industrial and in addition anthropogenic emissions in the present day case). The chemical reactions impacting ozone as well as transport of chemical constituents are described. The meteorology that drives the transport of chemical species is assumed to be the same in both cases. Thus no interactions between the chemistry on one hand and the transport or climate change on the other is taken into account. The pre-industrial levels over the continents in the northern hemisphere range from a minimum value of 5-15 ppbv in January, associated with chemical loss and dry deposition, to a summer value reaching 10 ppbv in Europe and 20 ppbv in the central United States. In the southern hemisphere the pre-industrial mixing ratios over the ocean range from only 4-8 ppbv in January to values of 10-20 ppbv in July. The present day distributions clearly emphasise a strong impact of anthropogenic activities on ozone levels at a global scale. Maximum O₃ mixing ratios reaching 50-60 ppbv are calculated over polluted regions (i.e., northern America, Europe, and southeast Asia) during summer in the northern hemisphere. This corresponds to an increase of a factor of 3-5 from pre-industrial levels. The background ozone levels calculated over the ocean in the southern hemisphere have also increased by 5-10 ppbv in comparison to pre-industrial values.

The calculated pre-industrial ozone levels appear to be very sensitive to assumptions made for natural emissions of NO and hydrocarbons. The NO soil emissions represent a main surface source of NO_x at mid-latitudes in the northern hemisphere for pristine conditions, and assumptions on their pre-industrial level are crucial regarding simulated surface ozone. The situation is also complex in tropical regions where recorded pre-industrial ozone levels at the surface indicate very low values of only 5 ppbv. The calculated ozone levels in the tropics crucially depend on assumptions made for biomass burning emissions of ozone precursors by natural fires. The observed regional variability of O₃ trends is related to the transport of key precursors, particularly reactive nitrogen, CO and NMHCs. However, the chemistry of O₃ production is non-linear, so that increased emissions of NO_x do not necessarily lead to linear responses in O₃ concentrations over all ranges of likely values [e.g., Klonecki and Levy, 1997; Fiore et al., 2002]. Further, the relationship of precursor emissions to O₃ increase may also vary in time. The study by Wang and Jacob [1998] suggest that the O₃ production efficiency per mole of nitrogen oxide emitted has decreased globally by a factor of 2 since pre-industrial times.

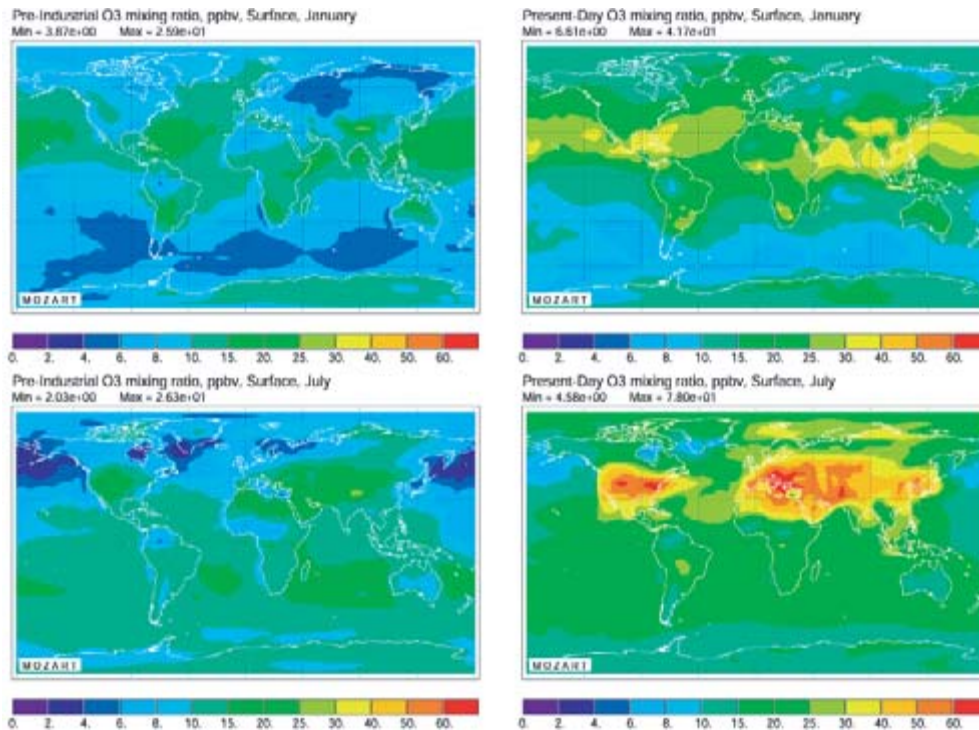


Figure 4.10 Surface distribution of ozone in January and July for pre-industrial and present day conditions in ppbv [Hauglustaine and Brasseur, 2001].

A recent survey of CTM studies provides global average increases in integrated tropospheric ozone ranging from 7 to 13 DU (Dobson Unit = 2.687×10^{16} molecules cm^{-2}) from pre-industrial to present [IPCC, 2001]. These results are summarised in Table 4.1. The spread among the various models clearly stresses the uncertainty on the pre-industrial ozone level estimates associated mainly with assumptions made on natural emission of precursors and on the magnitude of the ozone input from the stratospheric reservoir. The associated radiative forcings of climate range from 0.28 to 0.49 Wm^{-2} and reflects this uncertainty. The spread among the models is similar when normalised forcing (radiative forcing per ozone change, $\text{Wm}^{-2}\text{DU}^{-1}$) is considered, pointing to uncertainties also in the calculations of the radiative forcing for a given ozone change.

Several recent studies have also used global models and ozonesonde measurements to examine the ozone trend from pre-industrial to present based on historical reconstructions of ozone precursor surface emissions [Berntsen et al., 2000; Lelieveld and Dentener, 2000; Shindell and Faluvegi, 2002]. These studies emphasise the dominant contribution of stratosphere-troposphere exchanges on the ozone trend at high latitudes, of industrial and fossil fuel emissions at northern mid-latitudes, and the important role played by natural emissions in the tropics. All these various contributions and their time evolution have to be adequately considered in order to simulate the past evolution of tropospheric ozone. The adequacy of the meteorological fields used to transport ozone and its precursors is also essential in order to try to represent the inter-annual variability. The study by Shindell et al. [2001b] suggests, however, that the impact of climate change from pre-industrial to present is minor as far as the tropospheric ozone change is concerned.

Table 4.1 Global integrated tropospheric ozone increase from pre-industrial to present (DU), associated radiative forcing of climate (Wm^{-2}), and corresponding normalised forcing ($\text{Wm}^{-2}\text{DU}^{-1}$) based on global CTM studies. The right-hand column specifies the type of chemistry and

the type of model used. The term “intermediate” denotes a first generation CTM with lower degree of complexity in its representation of transport or physical processes. Note that Berntsen et al. [1997] reported two values, as two different radiative forcing calculations were performed.

Study	Tropospheric O3 change	Net radiative forcing	Normalised forcing	Note
Lelieveld and van Dorland [1995]	9.4	0.49	0.052	Intermediate CTM (NMHCs)
Chalita et al. [1996]	...	0.28	...	Intermediate CTM (NMHCs)
Levy et al. [1997]	9.5	Parameterised chemistry
Berntsen et al. [1997]	7.6	0.28	0.037	NMHCs included
Berntsen et al. [1997]	7.6	0.31	0.041	NMHCs included
Van Dorland et al. [1997]	8.1	0.38	0.047	Intermediate CTM (NMHCs)
Wang and Jacob [1998]	11.0	Intermediate CTM (NMHCs)
Brasseur et al. [1998a]	...	0.37	...	Intermediate CTM (NMHCs)
Stevenson et al. [1998]	8.7	0.29	0.033	NMHCs included
Haywood et al. [1998]	7.9	0.31	0.039	Parameterised chemistry
Roelofs et al. [1997]	7.2	0.42	0.058	NMHCs not included, GCM
Kiehl et al. [1999]	8.4	0.32	0.038	Based on observations
Mickley et al. [1999]	12.6	0.44	0.035	Some NMHCs included (GCM)
Berntsen et al. [2000]	9.6	0.34	0.036	NMHCs included
Lelieveld and Dentener [2000]	7.7	0.42	0.054	NMHCs included
Shindell et al. [2001b]	7.5	0.32	0.042	NMHCs not included, GCM
Hauglustaine and Brasseur [2001]	8.9	0.43	0.048	NMHCs included
Mean	8.9 ± 1.4	0.36 ± 0.06	0.043 ± 0.008	

Emissions of ozone precursors are expected to increase significantly in the future. This is particularly the case in regions where rapid economic growth or population increase are expected (i.e.,

south-east Asia, south and central America, Africa). Several studies have pointed out the potentially important impact of Asian emissions on the background level of pollutants over the Pacific and northern United States [Berntsen et al., 1996, 1999; Jaffe et al., 1999; Jacob et al., 1999; Mauzerall et al., 2000]. In the tropics, pollutants are rapidly transported to the upper troposphere where subsequent redistribution through long transport can take place [Grewe et al., 2001; Hauglustaine et al., 2001]. Other studies have also illustrated how background ozone levels over Europe are affected by long-range transport of pollution from Northern America [Li et al., 2002]. Estimates of future ozone levels have been performed with several models and under various emission scenarios [Chalita et al., 1996; van Dorland et al., 1997; Stevenson et al., 1998, 2000; Brasseur et al., 1998a; Hauglustaine and Brasseur, 2001; Grewe et al., 2001].

Recently, an intercomparison exercise of future ozone levels has also been performed by 11 global models [Prather et al., 2001]. The simulated surface ozone levels in 2100 under the IPCC SRES A2 scenario indicate large increases along the southern edge of Asia from Arabia to eastern China, in Africa and Central America [OxComp, Prather et al., 2003]. The changes in tropospheric column ozone from the OxComp study are shown in Figure 4.11. The largest increase in ozone abundance occurs near the tropopause at 40°N. Some models tend to concentrate this increase in the tropics and others push it to high latitudes. These changes are likely to affect significantly the background ozone concentrations in most pristine regions and to represent a sizeable increment towards violation of air quality standards over the next decades [Hauglustaine and Brasseur, 2001; Prather et al., 2003] and also to yield a significant climate impact (see Section 4.4.1). The globally averaged increases in ozone between 2000 and 2100 in the OxComp study was in the range 11-21 DU.

Aircraft emissions are believed to have changed the chemistry of the upper troposphere due to their emissions in this region (see Section 4.2.5). In the current atmosphere the ozone increase due to aircraft has been estimated to 0.4 DU, as a global average (results from the state of the art Oslo CTM2). This is modest compared to the ozone increase since pre-industrial times. However, aircraft impacts are restricted to certain latitude bands. The maximum change in zonally averaged ozone column is 1.4 DU, occurring in June. As aircraft emissions are anticipated to increase markedly in the future (see Section 4.2.6) their impacts on tropospheric ozone are expected to be significant.

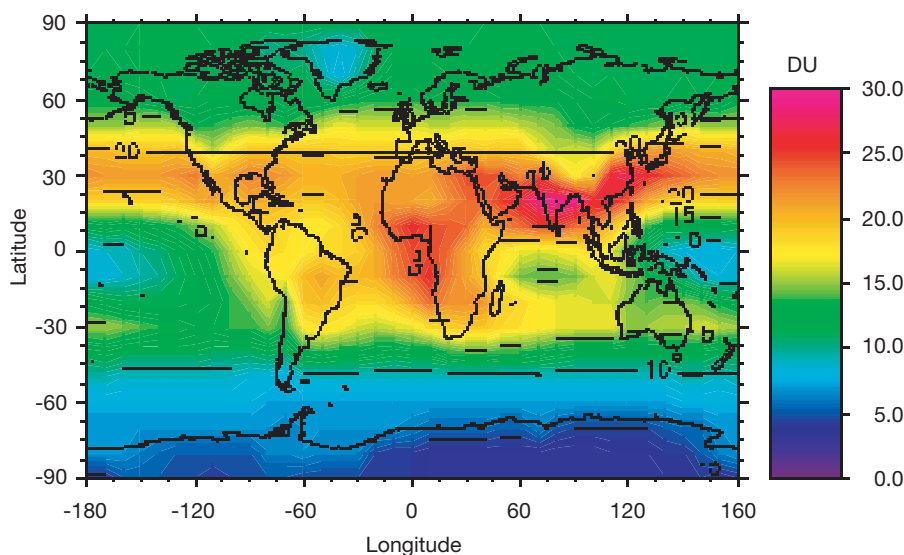


Figure 4.11 Changes in tropospheric ozone columns (DU) from year 2000 to 2100, as calculated by the 11 models in OxComp. The results shown are averages for all the models. (based on results in Gauss et al. [2003]).

- *Due to a lack of observational data for pre-industrial and even to some extent current ozone, models are used to establish changes in tropospheric ozone up till present time to assess the radiative forcing and the climate impact. A recent survey of CTM studies provides global average increases in integrated tropospheric ozone ranging from 7 to 13 DU.*
- *The ozone increase due to aircraft has been estimated to 0.4 DU, as a global average. This is modest compared to the ozone increase since pre-industrial times. However, aircraft impacts are restricted to certain latitude bands. The maximum change in zonally averaged ozone column is 1.4 DU.*
- *Future changes in tropospheric ozone have been calculated with several models and for several scenarios for emissions of ozone precursors. A recent model intercomparison study has applied the IPCC A2p scenario introduced above. The range of changes in global average ozone from 2000 to 2100 was estimated to 11-21 DU.*

4.4 Climate impact of current and future changes in tropospheric ozone

A first measure of the impact of ozone resulting from anthropogenic emissions of ozone precursors on climate is the associated change in the radiative energy flux through the tropopause (the boarder region between the troposphere and the stratosphere), called the radiative forcing. The concept of radiative forcing is introduced since as a first approximation, changes in surface temperature are proportional to changes in the radiative forcing (see Chapter 2).

In this chapter the radiative forcing associated with changes in tropospheric ozone is discussed first in Section 4.4.1 followed by a discussion on the changes in temperatures associated with the radiative forcing, in Section 4.4.2.

4.4.1 Ozone changes and radiative forcing

Changes in tropospheric ozone impacts climate *directly* through the radiative forcing, as explained above. Also, ozone changes impacts climate *indirectly* through chemical reactions that changes the concentrations of other radiatively active gases, in particular methane. These direct and indirect forcings are subjects of the following two subsections.

4.4.1.1 Direct radiative forcing

The fact that ozone is radiatively active in the long-wave as well as the shortwave spectral region results in a different radiative forcing (RF) dependency on atmospheric and surface conditions as compared to pure greenhouse gases on one hand and aerosols on the other. Due to its long-wave characteristics, mainly through the 9.6 and 14 μm bands, the RF is critically dependent on the temperature contrast between the surface and the altitude of ozone change [Forster and Shine, 1997; Van Dorland et al., 1997]. This property is shared with the well-mixed greenhouse gases. The short-wave forcing is sensitive to surface albedo, cloud cover and solar zenith angle via both the amount of solar radiation on a horizontal plane and the path length through the atmosphere, as for aerosols. The role of clouds is twofold: it masks the greenhouse effect of tropospheric ozone, but it enhances the shortwave effect [Roelofs, 1999]. The net effect is that in the global and annual mean the RF is reduced by about 13% given the current distribution of clouds [Van Dorland et al., 1997].

Several studies of global tropospheric ozone change, and subsequent changes in the radiative forcing, since pre-industrial times have been performed. These studies give an increase in global yearly ozone mean of more than 30% (from 25 to 34 Dobson Units) with corresponding radiative forcings (RF) in the range from 0.2 to 0.5 Wm^{-2} (Table 4.1). Recent estimates of radiative forcing have been mostly based on output from CTMs [e.g. Hauglustaine et al., 1994; Chalita et

al., 1996; Forster et al., 1996; Berntsen et al., 1997; Van Dorland et al., 1997, Brasseur et al., 1998a, Haywood et al., 1998; Stevenson et al., 1998; Hauglustaine and Brasseur, 2001]. Roelofs et al. [1997] used the fully interactive tropospheric chemistry/climate European Centre Hamburg Model (ECHAM4) to compute the RF due to tropospheric ozone changes. Portmann et al. [1997] and Kiehl et al. [1999] combined an ozone climatology based upon satellite measurements of the tropospheric column content [from Fishman and Brackett, 1997] with a model calculation to derive estimates of the ozone RF for the tropics and for the globe, respectively. Mickley et al. [2001] found that the RF could be significantly larger due to uncertainty in estimates of pre industrial ozone levels. They obtain an instantaneous global yearly mean RF due to ozone changes of 0.72-0.80 Wm^{-2} and this would make ozone at least as important as methane as a greenhouse gas when considering increases by human activities.

The geographical distribution of the RF due to tropospheric ozone changes is much more inhomogeneous than that of the well-mixed greenhouse gases. Most studies show that the annual mean RF reaches a peak over north Africa due to a coincidence of relatively large ozone increases with the region where those changes are most effective due to a warm surface, high surface reflectance and cloudless skies [Van Dorland et al., 1997; Berntsen et al., 1997]. Many studies show distinct peaks in the seasonal cycle of the ozone forcing: most pronounced is the high RF at the northern hemisphere subtropical belt in July/August (Figure 4.12). A secondary maximum is found in the tropical region in autumn due the effects of biomass burning.

Tropospheric ozone levels are projected to increase further in future. The IPCC Special report on Scenarios developed 4 “marker scenarios” which represent different demographic, economical and technological developments. The so called A2 scenario represents the extreme range of emissions and results in the highest emissions of all 4 scenarios. The other scenarios are far less extreme. The older emission scenario IS92a results in emissions that are less than scenario A2 but more than scenario A1 with relatively low emission increases between 2000 and 2100. By using the IS92a scenario, Van Dorland et al. [1997], have calculated a 70% ozone change between 1990 and 2050 as compared with pre-industrial to 1990. This would result in a radiative forcing of 0.83 Wm^{-2} and 0.66 Wm^{-2} in 2050 (from pre-industrial time) for the annual mean northern hemispheric and global average respectively. Brasseur et al. [1998a] and Hauglustaine and Brasseur [2001] found for the same time horizon a comparable global mean RF of 0.63 Wm^{-2} and 0.69 Wm^{-2} , respectively, while the model of Stevenson et al. [1998] yields a lower RF of 0.48 Wm^{-2} in 2100 due to temperature and associated humidity increases resulting in lower projected ozone levels.

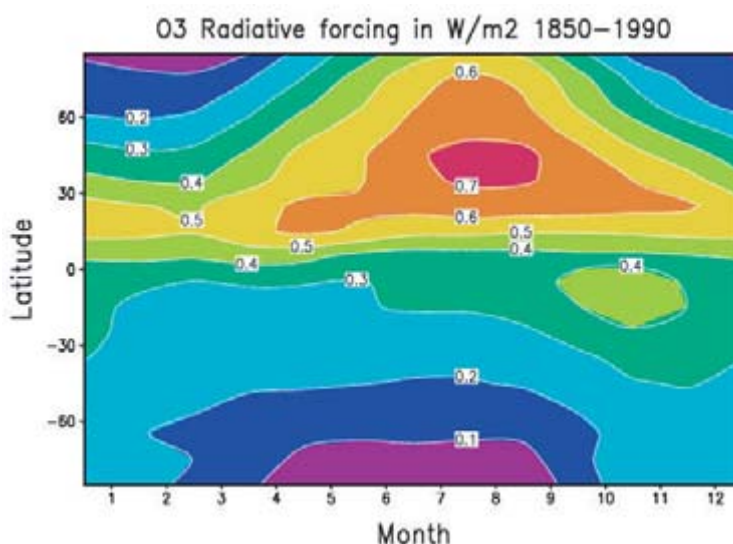


Figure 4.12 Radiative forcing due to changes in tropospheric ozone from pre-industrial to present conditions [Van Dorland et al., 1997].

The projected tropospheric ozone maximum for the 21st century shifts significantly southward as a result of increasing industrial activities in countries with emerging economies (see Section 4.3.2). Concurrently there is a shift in the time of the year of the ozone maximum from July/August for the period 1850-1990 to August/September for the period 1990-2050. As a follow up of the OxComp study for year 2100, radiative forcing has been estimated for the ozone distributions calculated in the intercomparison [Gauss et al., 2003]. The geographical distribution of the forcing reflects the estimated ozone changes, but is amplified over desert regions with high surface albedo (Figure 4.13). The range of the forcing as calculated in the Gauss et al. [2003] study is 0.40-0.78 Wm⁻², associated with the simulated tropospheric ozone increase from present day to 2100.

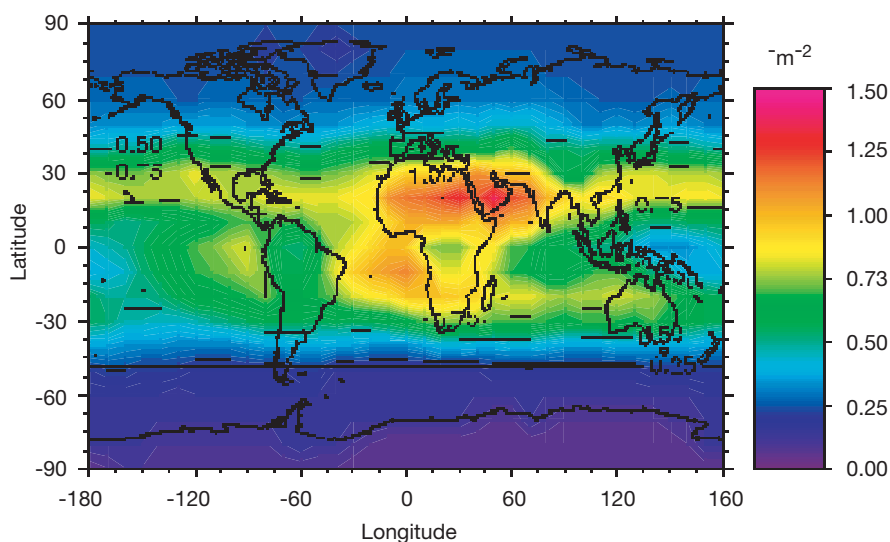


Figure 4.13 Radiative forcing due to changes in tropospheric ozone from 2000 to 2100 (average of 11 models in Gauss et al., [2003]).

- Several studies have yielded an increase in global yearly mean of tropospheric ozone since pre-industrial times of more than 30% (9 Dobson Units) with corresponding radiative forcings in the range from 0.2 to 0.5 Wm⁻². This ranks ozone as the third most important greenhouse gas in this time period after CO₂ and CH₄.
- A recent model intercomparison study which applied the IPCC A2p scenario calculated radiative forcing due to changes in ozone between 2000 and 2100 to be in the range 0.40 to 0.78 Wm⁻².
- The geographical distribution of the radiative forcing due to tropospheric ozone is much more inhomogeneous than that of the well-mixed greenhouse gases, more like the forcing due to aerosols.

4.4.1.2 Indirect radiative forcing

The hydroxyl radical, OH, which removes methane and other GHG from the atmosphere, is produced from ozone and water vapour in the troposphere, in the presence of solar radiation. The impact of the ozone precursors (CH₄, CO, VOC and NO_x) on climate thus has an indirect climate effect by their impact on OH, in addition to their impact through production of ozone [see e.g. Lelieveld et al., 1993, 1998; Johnson and Derwent, 1996; Fuglestedt et al., 1996, 1999]. This can partly be viewed as an indirect effect of changes in tropospheric ozone. It is important to notice, however, that the ozone precursors in addition influence the OH concentrations through reactions not involving ozone. Only part of their indirect climate effect therefore involves ozone in an explicit manner. Any regulatory action aimed at reducing tropospheric ozone must, however, take these processes into consideration.

Table 4.2 Impacts of ozone precursor gases on the radiative forcing. + denotes a positive RF, - a negative RF. * denotes low NO_x conditions, see text.

Source gas	Tropospheric Ozone	Methane and other gases removed by OH
CH ₄	+	+
CO	+ (- *)	+
NMVOC	+ (- *)	+
NO _x	+	-

Table 4.2 summarises the impact of the ozone precursors on the radiative forcing. As previously shown, CH₄, CO, VOC and NO_x lead to an increase in ozone and a positive (marked as + in Table 4.2) radiative forcing. The exceptions are effects of CO and VOC (perhaps also CH₄) in regions of extremely low NO_x, where they can result in ozone reductions and a negative (marked as -) radiative forcing (hence the * in Table 4.2). This effect has only a weak impact on the global radiative forcing. CH₄, CO and VOCs react with OH and therefore reduce OH and impose a positive radiative forcing. An opposing effect is the production of OH from the increase in ozone. This is usually a somewhat weaker effect, yielding an overall positive radiative forcing due to the impact on OH. The role of NO_x is different since it increases OH also in the first place (by shifting the HO₂/OH balance towards higher values of OH), yielding a negative radiative forcing from NO_x. Wild et al. (2001) found that this effect dominated over the positive forcing due to production of ozone, yielding an overall slightly negative radiative forcing for NO_x when integrated over 100 years.

- *The ozone precursors (CH₄, CO, VOC and NO_x) have an indirect climate effect by their impact on OH, in addition to their impact through production of ozone. For CH₄, CO and VOCs this forcing is positive, working in the same direction as the ozone forcing. For NO_x the indirect forcing opposes the ozone forcing.*

4.4.2 Climate impact of tropospheric ozone change on temperature

Different climate models have different values for the global mean climate sensitivity parameter λ (see Chapter 2). The current range for λ is 0.4 to 1.2 K (Wm⁻²)⁻¹ [Ramaswamy, 2001]. The sensitivity represents the amalgamation of all feedback processes in the model, and the wide range reflects differences in model parameterisations, mainly relating to clouds and sea-ice. A further concern is that within a given model, whilst λ is approximately constant for the well-mixed greenhouse gases, it does vary somewhat for more spatially heterogeneous forcings, such as changes in ozone and aerosols.

Because ozone has a relatively short lifetime (from a few days at the surface in the tropics to several months in the upper troposphere), its distribution has changed due to anthropogenic activities in a spatially heterogeneous way. For ozone changes, the largest radiative forcings are produced for changes at tropopause levels, because of the low temperatures in this region [Lacis et al., 1990; Brasseur et al., 1998b]. For a given change in ozone, the largest forcings are seen over hot, cloud-free high albedo land, or areas of low, warm cloud with a high, cold tropopause (i.e. sub-tropical deserts and oceanic stratocumulus regions) [Gauss et al., 2003].

There have been only a few studies concerning the impact of tropospheric ozone changes on climate using GCMs [Hansen et al., 1997; Bintanja et al., 1997; Christiansen, 1999; Ponater et al., 1999; Stuber et al., 2001a, b; Grewe et al., 2002; Sausen et al., 2002; Joshi et al., 2003]. The GCMs used were of varying complexity, and the experiments generally considered only highly idealised ozone changes [apart from Ponater et al., 1999; Stuber et al., 2001a]. Hansen et al. [1997] found that λ was strongly influenced by cloud feedbacks, and was most sensitive to lower tropospheric ozone changes. Bintanja et al. [1997] found that λ depended on the latitudinal distribution of the

radiative forcing, mainly due the ice-albedo feedback. Consequently, climate was more sensitive to ozone changes at high latitudes. Christiansen [1999] also looked at climate sensitivity and the vertical structure of ozone changes, but found much less variation with height than Hansen et al. [1997]. Differences were attributed to the different treatment of cloud feedbacks in the two models. Stuber et al. [2001a, b] found that for ozone changes in the lower and mid-troposphere λ was similar to the value found for CO_2 , but a lower sensitivity was found in the upper troposphere. In contrast, a higher sensitivity was found for ozone changes in the lower stratosphere: this was related to a strong feedback with lower stratospheric water vapour. However, the feedback from H_2O on ozone through chemistry was no included. Joshi et al. [2003] have attempted to build a consensus view, by comparing results from the same experiments performed by 3 GCMs: the University of Reading, ECHAM4/DLR, and LMD models. All of these models found that for global ozone changes just below the tropopause, λ was reduced by 16-37% compared to the equivalent forcing from CO_2 , whereas λ was increased by 23-80% for ozone changes in the lower stratosphere (see Figure 4.14). There was a lack of consensus between the models about the sensitivity of the extra-tropics compared to the tropics. Overall, this study suggested that for the same magnitude radiative forcing, tropospheric ozone was $\sim 25\%$ less effective in generating a climate response, compared to CO_2 . The reason for this appears to be that increases in upper tropospheric ozone cause reduced dynamic coupling between the surface and the upper troposphere. Furthermore this ozone increase appears to reduce the cold point temperature at the tropical tropopause and hence reduces the water vapour transport to the stratosphere. However, in one of the experiments reported by Sausen et al. [2002] (experiment ON-LT), ozone changes in the boundary layer generate a 60% larger climate response than for an equivalent CO_2 forcing. Clearly, this could be important, as some of the largest observed increases in tropospheric ozone have been observed in this region.

Folland et al. [1998] used the Met Office Hadley Centre GCM to consider the individual impacts of a variety of anthropogenic forcings over the second half of the twentieth century, including the simulated change in tropospheric ozone. These authors found that the increase in tropospheric ozone generated a distinctive signature (tropospheric warming and lower stratospheric cooling), as compared to the response for the well-mixed greenhouse gases (tropospheric warming and upper stratospheric cooling).

Clearly, studies of the climate change in response to a given radiative forcing resulting from changes in tropospheric ozone are only in their beginning, representing a future research challenge. Potentially, even surprises may arise from fully interactively coupled CTMs due to new modes of the system.

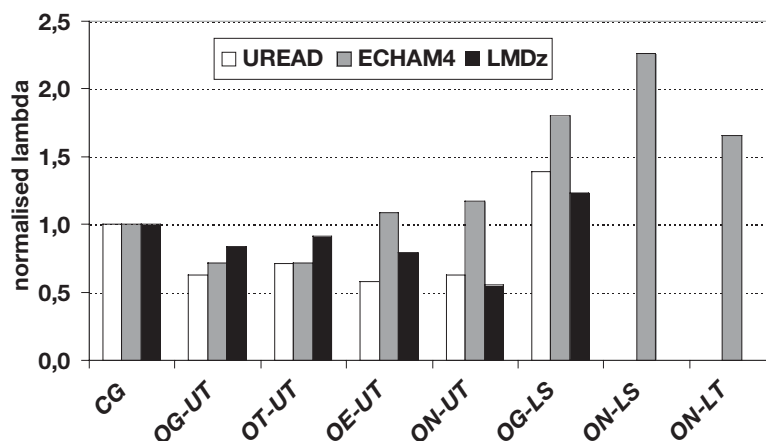


Figure 4.14 Normalised climate sensitivity parameters $\lambda/\lambda_{\text{CO}_2}$ as simulated by 3 different models (UREAD, ECHAM4, LMDz) for various experiments (CG: CO_2 added globally, vertically homogeneous; OG-UT: ozone added globally in the upper troposphere, i.e. in the levels just below the thermal tropopause; OT-UT: ozone added equator-ward of 30° in the upper troposphere; OE-UT: ozone added pole-ward of 30° in the upper troposphere; ON-UT: ozone added northwards of 30°N

in the upper troposphere; *OG-LS*: ozone added globally in the lower stratosphere (in levels that are everywhere higher than the tropical tropopause); *ON-LS*: ozone added northwards of 30°N in the lower stratosphere; *ON-LT*: ozone added northward of 30°N in the planetary boundary layer). The figure is adopted from Sausen et al. [2002] and from Joshi et al. [2003].

- For ozone changes, the largest radiative forcings are produced for changes at tropopause levels, because of the low temperatures in this region. In addition, the climate response to a given radiative forcing depends on the altitude of the ozone change.
- There have been only a few studies concerning the impact of tropospheric ozone changes on climate using GCMs. A recent study build on results from three different models suggested that for the same magnitude radiative forcing, tropospheric ozone was in general ~25% less effective in generating a climate response, compared to CO₂, with a range from ~50% less effective to ~50% more effective than CO₂ depending on altitude and latitude region of ozone change.

4.5 Conclusions

Tropospheric ozone influences the radiative balance of our planet, both by absorbing solar radiation and, more importantly, by acting as a greenhouse gas (GHG) through absorption and re-emission of infrared radiation from the Earth's surface. Thus ozone plays an important role in the climate system. It is now well recognised that the large increase in the background tropospheric ozone concentrations during the previous century is mainly due to anthropogenic emissions of ozone precursors. The ozone increase in the troposphere has lead to trapping of radiation in the troposphere, yielding a positive radiative forcing. Recent calculations show that the increase in tropospheric ozone represents the third largest positive radiative forcing since pre-industrial time (within the uncertainty rage 0.2–0.5 Wm⁻²). In this way the ozone increase has had a significant climate warming impact.

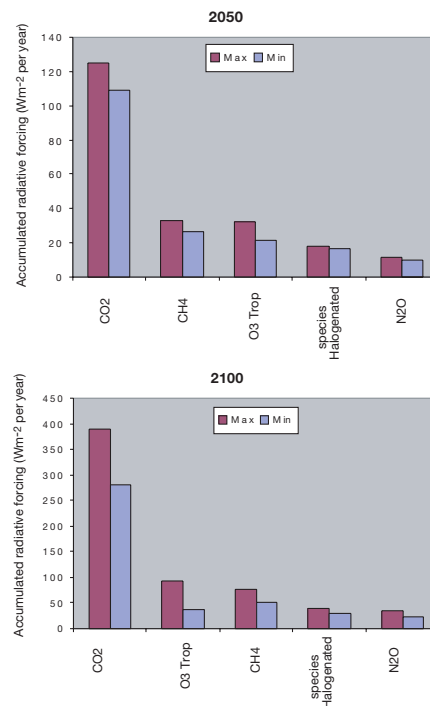


Figure 4.15 Accumulated radiative forcing (in Wm⁻² per year) due to greenhouse gases from 2000 to 2050 (upper panel) and 2100 (lower panel). The results are based on instantaneous radiative forcing scenarios (in Wm⁻²) for each 10 years between 2000 and 2100 estimated in IPCC [2001] for tropospheric ozone, CO₂, CH₄, N₂O, and a range of halogenated greenhouse gases (CFCs, HCFCs, HFCs, CF₄, C₂F₆ and SF₆). The numbers have been accumulated for this report.

Emissions of ozone precursors are expected to increase significantly in the future. This is particularly the case in regions where rapid economic growth or population increase is expected (i.e., south-east Asia, South and Central America, Africa). Thus tropospheric ozone caused by anthropogenic emissions will continue to contribute to climate warming during this century. According to scenarios established in IPCC [2001] the global radiative forcing due to ozone will range from 0.4 to 1.0 Wm^{-2} in 2050 and from 0.2 to 1.3 Wm^{-2} in 2100. Climate change is as a first approximation proportional to the accumulated forcing up to a given point in time. The accumulated forcing due to tropospheric ozone is given in Figure 4.15, up till 2050 as well as 2100. It is compared to the accumulated forcing due to other greenhouse gases, again based on scenarios for radiative forcing given by IPCC [2001]. The maximum as well as the minimum values are given for each greenhouse gas, representing an uncertainty range. The results show that tropospheric ozone will continue to be an important climate gas in the next hundred years, and perhaps be the second most important contributor to climate warming after CO_2 .

- *The increase in tropospheric ozone represents the third largest positive radiative forcing since pre-industrial time. Tropospheric ozone will continue to be an important climate gas in the next hundred years, and perhaps be the second most important contributor to climate warming after CO_2 .*