Intercontinental transport and the origins of the ozone observed at surface sites in Europe

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Abstract

A global three-dimensional Lagrangian chemistry-transport model is used to describe the formation, transport and destruction of tropospheric ozone using 1990s global emissions and 1998 meteorological archives. Using a labelling technique, the geographical origins of the ozone formed within the troposphere have been revealed, showing whether the ozone found at the surface in Europe has had its origins above the continents of North America, Europe or Asia or elsewhere in the world. In this way, contributions to the ozone found at 21 surface monitoring sites across Europe can be attributed to production over North America and Asia, demonstrating that intercontinental ozone transport is an efficient process. Sensitivity tests to the global man-made sources of NO\textsubscript{x} and carbon monoxide indicate that global ozone precursor emission controls may contribute towards reaching regional air quality policy goals for ozone in Europe.

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1. Introduction

Over the past few decades there have been conflicting explanations of the origins of surface ozone over Europe and of its spring-time maximum (Monks, 2000). Initially, stratosphere–troposphere exchange, an unmistakably natural process, had been proposed to explain the spring-time maximum in observed ozone mixing ratios at a mountain-top site at Arosa, Switzerland (Junge, 1963). However, more recently man-made processes are thought to have caused a doubling in surface ozone mixing ratios in Europe since pre-industrial times (Volz and Kley, 1988) based on observations at the Montsouris Observatory near Paris, France from 1861 to 1871. European Community emissions of NO\textsubscript{x} and VOCs have declined by 25–30\% between 1990 and 2000 (UNECE, 2002) leading to a decline in episodic peak ozone mixing ratios (Derwent et al., 2003) whilst mean ozone mixing ratios continue to rise at remote locations such as Mace Head, Ireland (Simmonds et al., 1997) and Strath Vaich, Scotland (Fowler et al., 2001). These apparent contradictions can be resolved if there are a number of sources of ozone in Europe and if a number of different mechanisms are operating to cause the observed seasonal cycle in ozone.

In this study, we use a global chemistry-transport model to study the likely origins of the ozone found at the surface across Europe. We use a labelling technique to elucidate the geographical origins (stratosphere, North America, Asia, Europe) of surface ozone and we identify intercontinental transport from North America and Asia as an important source of ozone within Europe. Finally, we show how global reductions in man-made emissions of NO\textsubscript{x} and carbon monoxide could reduce surface ozone levels over Europe.
2. The global 3-D chemistry-transport model STOCHEM

The model used in this study is the UK Meteorological Office global Lagrangian tropospheric 3-D chemistry transport model (STOCHEM). In the version employed here, the troposphere has been divided into 50,000 air parcels which are advected with a 3 h time step using archived winds from the Meteorological Office Unified Model. These winds are 6-hourly instantaneous values, taken from operational meteorological analyses at $0.83^\circ \times 1.25^\circ$ resolution for 1997 and 1998. The model calculations began at 1 July 1997 and run through to 31 December 1998. Results were taken from the last full year of the calculations with a 6-month period for initialisation. This model version is not the most recent (Collins et al., 2003) but is an earlier offline version which is faster and is run from operational meteorological analyses, full details of which are given elsewhere (Collins et al., 1997, 1999).

Each parcel holds the concentrations of 73 chemical species including NO$_x$, O$_3$, OH, HO$_2$, PAN, methane, ethane, ethylene, propane, propylene, butane, toluene, o-xylene, isoprene, formaldehyde, acetaldehyde, acetone, methyl glyoxal and methyl vinyl ketone. The chemical species react according to 174 photolytic and thermal reactions, with a 5-min chemistry time step. Surface uptake, dry deposition and wet removal are unchanged from Collins et al. (1997). Emissions into the model are implemented as an additional term in the production flux for each species during each integration time-step. The emissions compiled to represent the situation of the 1990s are listed in Table 1. Biomass burning, vegetation, soil, oceans and ‘other’ are all surface sources based on two-dimensional (latitude, longitude) distributions as in Collins et al. (1997). The aircraft and lightning NO$_x$ sources are three-dimensional. Methane sources from paddies, tundra, wetlands and animal sources (see Table 1) are held constant throughout the year at their yearly average values. Natural biogenic sources from soils, vegetation and the oceans, vary by calendar month. No seasonal nor diurnal dependences are assumed for any of the industrial emissions. The 6-hourly vertical wind fields were used to calculate a stratosphere–troposphere exchange ozone flux across 100 mbar surface on a $5^\circ \times 5^\circ$ grid using monthly ozone fields from Li and Shine (1995). The total annual injection of ozone through the 100 mbar surface amounted to 1216 Tg yr$^{-1}$, with a NO$_y$ flux of one thousandth of the ozone flux by mass (as N), based on the study of Murphy and Fahey (1994). Inter-parcel mixing was treated by relaxing the mixing ratios in each parcel towards the average

<table>
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<tr>
<th>Species</th>
<th>Industrial</th>
<th>Biomass burning</th>
<th>Vegetation</th>
<th>Soil</th>
<th>Oceans</th>
<th>Aircraft</th>
<th>Lightning</th>
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$^a$Emissions are in Tg yr$^{-1}$ except for NO$_x$, which are in Tg N yr$^{-1}$.
$^b$Emission totals and their spatial distributions were based on entries in the EDGAR database (Olivier et al., 1996).
$^c$Aircraft NO$_x$ emissions for 1992 were taken from IPCC (1999).
$^d$Includes paddies, tundra, wetlands, termites and animals.
$^e$Acetone source from vegetation represents production from terpene oxidation.
background mixing ratio as described by Collins et al. (1997).

We have participated in a number of global 3-D chemistry-transport model intercomparisons to establish the level of STOCHEM model performance. These include intercomparisons of fast photochemistry (Olson et al., 1997), tracer transport (Stevenson et al., 1998), ozone (Kanakidou et al., 1998), carbon monoxide (Kanakidou et al., 1999) and of past, present and future tropospheric ozone distributions (Gauss et al., 2003). In all of these intercomparisons, STOCHEM performs within the range of current models. However, in common with all other complex models, it is sometimes difficult to reconcile good model performance with one species in one location with poor model performance elsewhere or with other species. In previous work, Collins et al. (1997) have described comparisons of model results with observations for global mean OH, surface ozone for the Atlantic Ocean, for remote background sites, with ozonesondes and NOx measurements throughout the troposphere. From these comparisons it was concluded that the model was well able to provide a good approximation to the main controlling processes for OH, NOx and ozone in both the winter and the summer and that the model ozone distribution contains all the most important features of the observed distribution. In addition, Collins et al. (1999) presented comparisons between the model results and observations for hydrogen peroxide, formaldehyde, methyl hydroperoxide and acetone, all potentially important free radical sources in the troposphere. Furthermore, the methane lifetime due to OH removal was found to be 9.0 years in these model experiments, in agreement with Prinn et al. (1995) who reported a lifetime of 8.9 ± 0.6 years. The global ozone budgets, burdens and turnover times calculated here are well within the ranges in the literature as reviewed by Collins et al. (2000).

The mixing ratios of each of 73 model species, i, were calculated from:

$$\frac{dx_i}{dt} = P_i - L_i x_i + M_i.$$  

(1)

The production rate $P_i$ for each species was calculated at each time step from the local, instantaneous rates of emission and chemical production from the set of 174 reactions. The loss rate $L_i x_i$ for each species was calculated from the local, instantaneous destruction rate due to chemical reaction, dry and wet deposition and aerosol scavenging. $M_i$ represents the net production from interparcel mixing. In addition to ozone, the air parcels carried other forms of ozone, $O_3^{A}$, $O_3^{B}$, $O_3^{C}$, ..., in which ozone is labelled by the geographical region in which it was formed. Our approach adapts and extends that pioneered by Roelofs and Lelieveld (1997) for ozone of stratospheric origins. Each of the additional forms of ozone has the same loss coefficient, $L_i$, hence the sum of all of the mixing ratios of the different forms of ozone, $x_{O_3}^A$, ..., in each air parcel adds up to the mixing ratio of ozone, henceforward, ‘total ozone’, $x_{O_3}$, to distinguish it from the labelled forms.

The ‘total ozone’ production rate is given by:

$$\frac{dx_{O_3}}{dt} = E_{STE} + k_1[HO_2][NO] + k_2[RO_2][NO] - k_3[O^3D][H_2O]$$

$$- \text{other O}_3 \text{ loss terms} - \text{other NO}_2 \text{ loss terms},$$

(2)

where $E_{STE}$ is the local, instantaneous ozone production rate from stratosphere-troposphere exchange, $k_1$ represents the rate coefficient for the $HO_2 + NO = OH + NO_2$ reaction, $k_2$ for $RO_2 + NO = NO_2 + RO$, $k_3$ for $O^3D + H_2O = OH + OH$, the other loss terms for ozone involve loss through its reactions with OH, HO2, NO2 and its ozonolysis reactions with ethylene, propylene and isoprene and the other NO2 loss terms include its reactions with OH, peroxy acyl radicals, NO3 radicals and ozone. The two terms $k_1[HO_2][NO]$ and $k_2[RO_2][NO]$ were then used to drive the rate of ozone production of the ozone tracer labels, depending where the air parcel was located with respect to the Northern Hemisphere continental land masses, North America (defined arbitrarily as 140°W–20°W), Asia (60°E–140°W) and Europe (20°W–60°E), or elsewhere in the world southwards of 23°N. Since the selected regions fill the entire model domain, both horizontally and vertically up to the model top at 100 mb, then the sum of the tracer mixing ratios satisfies Eq. (1) and the mixing ratios of the ozone tracers could be determined along with the other 73 model species. Tracer conservation is assured in the formulation adopted for the Lagrangian transport scheme in STOCHEM.

3. Intercontinental sources of ozone in Europe

3.1. Comparison of model with observed seasonal cycles in Europe

Because this study focuses on the origins of ozone observed at the surface in Europe, attention is first given to how well the STOCHEM model describes the seasonal cycle of ozone across Europe. Here, the model results for ozone of all origins are used, the results for the so-called ‘total ozone’. A record is kept of the air parcels which are in the vicinity (within 1°) of particular locations and their ozone mixing ratios are noted at each time step. For the chosen locations, model points were found for 87% of the days in 1998. The model mixing ratios are plotted out as individual points in Fig. 1 and as monthly ranges, shown as monthly mean ± 1 standard deviation, as broken lines. Fig. 1 also presents the observed monthly mean maximum daily ozone mixing ratios in ppb (1 part in 10^9 parts by volume) during 1998.
Fig. 1. Observed mean monthly daily maximum ozone mixing ratios in ppb (solid lines), model calculated ozone mixing ratios (dots) and monthly model ranges as mean ± 1 standard deviation (broken lines) for 1998 at selected EMEP ozone monitoring network locations in Europe, country locations are given in Table 3.
at each of 10 selected remote, rural site locations in the EMEP ozone monitoring network (Hjellbrekke, 2000), as solid stepped lines. The observed monthly means of the maximum daily (i.e. mid-afternoon) ozone mixing ratios were plotted instead of the monthly mean values because of the marked diurnal cycles found at these continental rural sites due to surface uptake of ozone under shallow nocturnal stable layers which are not simulated in the global model. During the mid-afternoon, surface observations reflect ozone levels in a deeper atmospheric layer and provide a better comparison with model results that are for the lowest model layer at 950 mb. Details of the latitudes, longitudes, siting criteria and measurement techniques for the EMEP network are given in Hjellbrekke (2000) and the country locations are given in Table 3.

The model points scatter around the stepped solid lines at most locations during much of the year and the observed monthly mean daily maximum ozone values (solid lines) fall within the ranges (broken lines) of the model results. Exceptions were found during some winter months at central European locations, where the model results overestimate the observations. This is caused by the coarse scale of the NOx emission inventories used, leading to an underestimation of wintertime ozone depletion. At the remote Scandinavian locations, the model tends to overestimate the observations during some summer months because of its coarse resolution which means that too many regional pollution episodes have reached these remote locations. The issues of too coarse model resolution have been minimised by using ozone observations at regionally representative rural locations during the mid-afternoon when boundary layer depths are at their maximum, minimising the effects of local NOx emissions and surface uptake on ozone levels. No attempt has been made to assess the impact on model ozone of individual power station or urban plumes.

Seasonal cycles are clearly evident in both observations and model results in Fig. 1. At most sites, summertime ozone peaks dominate the seasonal patterns. However at some of the more remote sites, secondary maxima are apparent during the springtime in both observations and model. Such maxima are evident at Mace Head, Tortosa, Kollumerwaard, Birkenes and Asprevaten. At Waldhof, Montilibretti, and Iskrba, the observations reveal a summertime maximum that is also reproduced faithfully by the model. On this basis, it is concluded that the model exhibits some skill in reproducing qualitatively the observed seasonal cycles at a range of selected site locations in Europe. A clear geographical pattern has emerged from both the observations and the model calculations, with summertime maxima dominating in the Mediterranean region and central Europe and with some evidence of secondary springtime maxima in Scandinavia, and in the Atlantic Ocean fringes of north and west Europe.

In Table 2, the seasonal cycles in the observations and in the model calculations are compared in a more rigorous manner for 19 locations across Europe. The biases, fractional biases, root mean square errors (RMSE) and the fraction of the monthly model results within ±20% of the observations (i.e. about ±7 ppb) were calculated as described in the footnotes to Table 2. Taking the locations with springtime maxima, mean biases fell in the range −2.06 to 3.34 ppb, −5% to 8.5%, with an average of 1.3 ppb. They showed a wide range in performance as indicated by the fraction of monthly means within ±20%, from 0.11 to 0.92 and the RMS errors from 5.2 to 13.8 ppb. For the locations with summertime maxima, mean biases fell in the range −3.5 to 2.2 ppb, −7.5% to 5.9%, with an average of −1.1 ppb. They showed a much narrower range in performance compared to the locations with spring-time maxima, as indicated by the fraction of monthly means within ±20% of 0.42–0.67, and RMSE errors of 7.1–10.8 ppb.

Taking all 19 locations together, it appears that the model is capable of reproducing observed monthly mean daily maximum surface ozone mixing ratios with an accuracy of ±8.7 ppb, based on RMSE errors, during 1998 and with about two-thirds of individual monthly mean values within ±20% of the observations. This is a workable level of model performance that gives some confidence in the model-derived continental attributions which follow.

3.2. Intercontinental attribution of the origins of surface ozone in Europe

By using the tracer-labelling technique, the model is able to give an indication of the geographical origins where the ozone was produced in each air parcel arriving at each location. By averaging overall the air parcels arriving during 1998, a picture has been built up of the geographical origins of the ozone calculated for selected locations and this is presented in Table 3. Broadly speaking, adding up the columns across Table 3 gives the annual mean ‘total’ model ozone mixing ratio and these fall in the range 30–45 ppb. As the previous section shows, these ‘total’ values were directly comparable with the surface observations. The columns in Table 3, present estimates of the ozone mixing ratios that could be attributed to the particular continental source regions.

The annual mean ozone mixing ratios across Europe due to stratosphere–troposphere exchange fell in the range from 5.7 to 10.6 ppb, with Iskrba (Slovenia) at the lowest end of the range and Mace Head (Ireland) at the highest. The mean value and seasonal cycle found here for Mace Head for ozone of stratospheric origins is
in excellent agreement with that already reported by Collins et al. (2003). Mace Head (Ireland) being the westerly most location is most exposed to the weather systems travelling across the North Atlantic Ocean. These weather systems are thought to account for much of the stratosphere–troposphere exchange found in northerly midlatitudes (Stohl, 2001). Iskrba (Slovenia), on the other hand, is one of the easterly most locations in Table 3 and is thus farthest removed from the influence of the weather systems travelling across the North Atlantic Ocean. Averaged over the 21 locations in 1998, stratosphere–troposphere exchange contributed about 7.3 ppb (18%) to surface ozone mixing ratios over Europe.

Table 3 indicates that the bulk of the ozone calculated in air parcels arriving at the 21 selected locations carried the ‘European’ label. European contributions covered the range from 9.5 to 31.5 ppb, with Mace Head (Ireland) at the lowest end of the range and Montelibretti (Italy) at the highest. Mace Head (Ireland) is the location in Table 4 with the least exposure to European regionally polluted air masses, receiving European air on only 32% of occasions during the 1990–1994 period (Simmonds et al., 1997). Montelibretti (Italy), on the other hand, is surrounded by centres of European population and industry in all directions and so is greatly influenced by European ozone production. Averaged over the 21 locations in 1998, European ozone production contributed about 20 ppb to surface ozone mixing ratios over Europe and provided the largest continental contribution of about 50%.

The North American region contributed surface ozone mixing ratios over Europe in the range 5.8–12.2 ppb. Virolahti (Finland) and Laheema (Estonia), both Baltic Sea locations, received the least North American contribution and Mace Head (Ireland) the highest. As we have remarked above, Mace Head is the location most exposed to the weather systems moving across the North Atlantic Ocean and these systems appear to provide a mechanism for the intercontinental transport of ozone as well as stratosphere–troposphere exchange. The behaviour seen for stratosphere–troposphere exchange and North American intercontinental transport appear somewhat different pointing perhaps...
to different geographical patterns of influence from these two mechanisms across Europe as shown in Fig. 2. Averaged over the 21 locations in 1998, North American continental ozone production contributed about 8.2 ppb (20%) to surface ozone mixing ratios over Europe. 

The Asian region contributed surface ozone mixing ratios in the range 3.5–6.6 ppb. Montelibretti (Italy) and Iskrba (Slovenia) received the least Asian contribution and Mace Head (Ireland) the highest. The spatial pattern of the contributions from Asian sources show similarities with that from stratosphere–troposphere exchange (see Fig. 2), but differences from North American sources. Averaged over the 21 locations in 1998, Asian continental ozone formation contributed about 4.5 ppb (11%) to surface ozone mixing ratios over Europe. 

The mixing ratio of ozone with the stratospheric label reached a maximum of 11.2 ppb during March 1998, with a secondary maximum of 10.6 ppb during that December. The European labelled ozone reached its maximum of over 30 ppb during May–August 1998, with its peak of 38 ppb during July. North American ozone reached a peak of 12.7 ppb during August 1998 and exceeded 10 ppb for the period July–September. Asian ozone reached its maximum of over 4 ppb during the period August–December, with a peak of 6.2 ppb during November. On this basis, intercontinental transport from North America and Asia combined, was a significantly more important source of surface ozone in Europe compared with the stratosphere in all months, except December and February. Broadly speaking, the stratospheric contribution peaked in the spring, the local European production peaked during the middle of summer and the intercontinental transport contributions from North America and Asia, peaked during the late summer and autumn, respectively.

3.3. Sensitivity of the intercontinental attribution of ozone in Europe to global emission source strengths

In this section, the sensitivities of the model surface ozone mixing ratios in Europe to the global man-made emissions of NOx and carbon monoxide are examined. They were studied by reducing by one half the global man-made source strengths in two separate model experiments. The 50% reductions were applied across-the-board, with no geographical or seasonal dependence. Table 4 presents the impact of a 50% reduction in NOx emissions on the ozone mixing ratios attributed to each continental source region at each of the 21 selected locations. No entries are provided for stratosphere–troposphere exchange because the ozone mixing ratios from this source were unaffected by the reductions emissions. The reductions in annual mean ozone mixing

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<th>Location</th>
<th>Stratosphere–troposphere</th>
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<td>25.2</td>
<td>7.2</td>
<td>3.8</td>
</tr>
<tr>
<td>Average</td>
<td>7.3</td>
<td>20.0</td>
<td>8.2</td>
<td>4.5</td>
</tr>
</tbody>
</table>
ratios due to the 50% reduction in global NO\textsubscript{x} emissions were largest for the European source region, 0.9–4.3 ppb, least for the Asian source region, 0.5–0.7 ppb, with North America 0.8–1.9 ppb. On a percentage change basis, the reductions in surface ozone attributed to the European source region were broadly similar.

The reductions in surface ozone attributed to the European source region, when averaged over the 21 locations, exceeded 5 ppb during the summer months of May to August and were close to zero during the winter months of December to March. In contrast, the reductions due to the North American source region showed much reduced seasonal variation, lying between 1.0 and 1.5 ppb, with a summer minimum, an autumn maximum and a subsidiary maximum during spring. The reductions due to the Asian source region followed an exactly similar seasonal pattern as those due to the North American region but covered a significantly wider range from 0.3 to 0.8 ppb.

On a percentage change basis, when averaged over all 21 locations, the reduction in surface ozone due to a 50% reduction in man-made NO\textsubscript{x} emissions attributed to the North American source region, varied from 10% to 25% depending on the time-of-year, and from 6% to 20% for the Asian source region, showing their minimum values during summer and their maximum values during winter. The seasonal variations in these percentage changes are characteristically different from that shown in the base case attributions to each source region shown in Table 3. Ozone attributed to the European source region showed little or no sensitivity to the reduction of man-made NO\textsubscript{x} emissions in wintertime and showed its greatest sensitivity of about 15% during summertime.

The impact of an across-the-board 50% reduction in global man-made carbon monoxide emissions on the surface ozone attributed to each continental source region are presented in Table 4. The reductions in ozone mixing ratios were again largest for the ozone attributed to the European source region, 0.4–0.8 ppb, least for the Asian source region, 0.1–0.2 ppb, with North American ozone reduced by 0.1–0.3 ppb. On a percentage basis, a 50% reduction in carbon monoxide emissions reduced surface ozone from Europe by 2.3–3.2%, from North America by 1.7–3.0% and from Asia by 1.8–3.0%. These responses to a 50% reduction in CO emissions appear to be largely independent of ozone production region, on a percentage basis. However, they are close to
the levels of noise in our 3-D chemistry-transport model and so it is difficult to be sure of their significance for air quality policy.

It is instructive to compare the results from the present study for a 50% reduction in global man-made NO\textsubscript{x} emissions with those of Wild and Akimoto (2001) for a 10% increase in surface fossil fuel NO\textsubscript{x}, CO and VOC emissions. Scaling the results from Wild and Akimoto (2001) gives a 0.5 ppb decrease in surface ozone found in Europe but formed over Asia, compared with 0.5–0.7 reported here, 1.0 ppb decrease in ozone formed over North America, compared with 0.8–1.9 ppb reported here and a 2.0 ppb decrease in ozone formed over Europe, compared with 0.9–4.3 ppb in this study. The estimates from Wild and Akimoto (2001), though for NO\textsubscript{x}, CO and VOC emission changes, are similar to those reported in this study and provide confirmation of our tracer modelling technique, bearing in mind that our calculations are for a larger NO\textsubscript{x} reduction and probably take the model into a region of greater \( \Delta \text{O}_3/\Delta \text{NO}_x \) slope compared with Wild and Akimoto (2001).

4. Discussion and conclusions

Evidence for the intercontinental transport of ozone and particles into Europe from North America and Asia based on ground-level, aircraft and satellite observations has been reviewed by Volz-Thomas (2003). The impact of man-made emissions from North America and Asia on surface ozone in Europe has been investigated using the global 3-D GEOS-CHEM model by performing sensitivity studies in which regional emissions were turned off (Li et al., 2002). According to these sensitivity studies, surface ozone in Europe decreased by 2–4 ppb when man-made emissions in North America were shut off and by slightly less for Asian emissions.

Shutting off North American man-made emissions in the GEOS-CHEM model (Li et al., 2002) decreased mean surface ozone mixing ratios in Europe by 2–4 ppb. The STOCHEM ozone tracer labelling technique indicated a somewhat larger contribution of 8.2 ppb from North America in Table 3. However, using the emission sensitivity approach in Table 4, a much smaller North American contribution of 1.2 ppb was identified.
by using a 50% reduction in man-made NO$_x$ emissions. Tracer labelling and emission sensitivity techniques appear to give different estimates of the magnitudes of the intercontinental transport of ozone because of important non-linearities in the fast photochemistry of ozone and its precursors. It may well be that the tracer labelling technique appears to give higher estimates and a more accurate quantification of intercontinental transport whereas the sensitivity approach provides a somewhat lower estimate but a more policy-relevant quantification by describing responses to emission controls.

In European air quality policy terms, the focus of concern is not so much with annual mean ozone concentrations (see Tables 3 and 4), as with ozone levels during the spring-time growing season for the protection of crops and vegetation as quantified by the accumulated exposure over a threshold of 40 ppb, the AOT$_{40}$ concept (Fuhrer et al., 1997). An empirical relationship has been constructed between mean monthly daily maximum ozone mixing ratios and the AOT$_{40}$ values within the EMEP database for 1998 (Hjellbrekke, 2000) for the 19 selected ozone monitoring sites in Table 2, as follows:

$$\text{AOT}_{40} \text{(in ppb h)} = 804 \times \text{ammdm} - 29,000,$$

where ammdm is the average monthly mean daily maximum ozone mixing ratio in ppb between April and July. Using this relationship, it has been possible to estimate the changes in AOT$_{40}$ exposure levels across Europe resulting from a 50% reduction in man-made NO$_x$ emissions. Reductions in North American NO$_x$ emissions reduced AOT$_{40}$ exposure levels by 620–1480 ppb h, 3.6–36%, with an average of 970 ppb h, 13%, across Europe. Reductions in Asian NO$_x$ emissions reduced average AOT$_{40}$ exposure levels by 410 ppb h, 5%, and in European NO$_x$ emissions by 4300 ppb h, 43%. Emission sensitivity studies on the hemispheric scale show that NO$_x$ emission controls in North America and Asia may produce significant benefits within Europe in terms of reducing future ozone exposures and protecting crops and natural vegetation.

On this basis, we conclude that the ozone tracer labelling technique can provide an account of the influence of intercontinental transport on surface ozone levels within Europe. On a year-round basis, the stratosphere appears to account for about 7 ppb, whereas intercontinental transport from North America, 8 ppb, and from Asia, 5 ppb. These intercontinental sources influence the different parts of Europe to different extents both spatially and seasonally and these variations help to explain the different seasonal cycles observed across Europe. Furthermore, because NO$_x$ emissions from North America, Asia and Europe have exhibited different trends over the 1970s–1990s (Naja et al., 2003), it may be possible to explain why there have been different observed trends in surface ozone in different parts of Europe during the 1970s–1990s. Background ozone levels may well be rising along the Atlantic fringes of Europe, influenced strongly by the steadily increasing North American NO$_x$ emissions and the rapidly rising Asian emissions. Ozone levels in central Europe may well remain roughly level because the increasing influence of North American and Asian emissions is counterbalanced by the influence of decreasing NO$_x$ emissions from central Europe itself. What could happen to ozone levels in Europe in the future could depend on future the NO$_x$ emissions from the different continents and this will be the subject of further studies with the ozone labelling technique.

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