Prediction of atmospheric chemical composition

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1. Introduction

In these notes I discuss briefly some issues that need consideration when moving from numerical weather prediction modelling (NWP) to chemical transport modelling (CTM). The main emphasis is on ozone, while the discussion is general and also applicable to a suite of other reactive atmospheric trace species including particulate matter. Below is discussed the main processes affecting reactive trace species in the atmospheric boundary layer; the linking of the local and global spatial scales; chemical transport models and their dependence on meteorological data; mesoscale modelling; improvements in numerical weather prediction; cloud cover prediction; boundary layer-free troposphere exchange rates over Europe and the North Atlantic; regional, intercontinental, hemispheric and interhemispheric chemistry and transport; and the capability to forecast ozone in the atmospheric boundary layer.

The formalism of setting up a chemical transport model (CTM) is not reviewed here. For the description of a state-of-the-art regional CTM, reference is given to the EMEP model (EMEP: European Monitoring and Evaluation Project) developed and in use at Norwegian Meteorological Institute (Meteorological Synthesizing Centre West in EMEP, MSC-W), for the analysis of transboundary air pollution in Europe and its abatement. The model is widely used within the framework of CLRTAP (Convention for Long Range Transmission of Air Pollution) and within the CEC (to support the development of directives related to air quality). See
http://www.emep.int/publ/reports/2003/emep_report_1_part1_2003.pdf
http://www.emep.int/publ/reports/2003/emep_report_1_part2_2003.pdf
http://www.emep.int/publ/reports/2003/emep_report_1_part3_2003_appB_p3-70.pdf

2. Main processes affecting reactive trace species in the atmospheric boundary layer

Changes in surface tracer emissions contribute to ‘global atmospheric change’. To assess the magnitude of global change, the handover of emissions and their transformation products from the source to the local, mesoscale, continental and global scales has to be followed, and the effects of changed emissions have to be quantified as they propagate through the same sequence of scales. The handover from the small to the larger spatial scales is a function of location and weather situation, and with very large differences from region to region.

As an example, ozone is handed over on all scales. It is formed in the atmospheric boundary layer and handed over to the larger scales, or the precursors are handed over to the larger scales and ozone is formed in the free troposphere. Dry deposition is the main removal mechanism for tropospheric ozone and takes place in the atmospheric boundary layer. Stratospheric-tropospheric exchange of ozone also occurs efficiently on the local scale in tropopause folds.

Time series of ozone concentrations can be separated into short-term, seasonal and long-term variations. Variations at different frequencies are caused by different processes. The synoptic-scale variability is short-term and can be attributed to weather fluctuations or changes in precursor emissions. Seasonal variations follow changes in the solar flux over the year, while the long-term variations are climate related or triggered by policy changes influencing the intensity of the precursor emissions (Rao et al., 1997). On the basis of statistical analysis of more than 10 years of ozone measurements at over 400 sites in the United States, Rao et al. (1997) concluded that baseline ozone (defined as the sum of the seasonal and long-term components) retains global information on the scale of more than 2 months in time and about 300 km in space. Short-term ozone is highly correlated in space, retaining 50% of short-term information at distances from 350-400 km. The correlation structure of short-term (weather related) ozone permits the prediction of ozone concentrations up to distances of 600 km from a monitor. In the high ozone cases, defined as exceedances of 120 ppbv, the information from a monitor extends only out to about 15 km, mainly because of the inhomogeneity in the NO_x levels in an urban area. Based on ozone data from the German measurement network, Tilmes and Zimmermann (1998) made an even stricter conclusion; they found that measurements at sites less than about four km apart, represent each other.

In situations with favourable meteorology and precursor emissions in the atmospheric boundary layer, ozone can be formed on the local and regional scales at a net rate of 10 or more ppb/h during the day, indicating that the ozone concentrations then can change significantly in the matter of a few hours. In the free troposhere the net formation or destruction rate of ozone is typically of the order of 0,1 ppb/h during the day, indicating a chemical lifetime of ozone there of the order of 50 days.

3. Linking the local and global scales

Both synoptic scale advection processes and convection play an important role in the vertical exchange in the troposphere. Observations have shown that trace gases are efficiently redistributed in deep convective clouds (Dickerson et al., 1987, Pickering et al., 1988), and model studies have indicated that the vertical redistribution of chemical trace species in rapid convective up- or downdraughts is important for their chemical processing (Chatfield and Crutzen, 1984, Pickering et al., 1992).

In some regions vertical exchange from the boundary layer to the free troposphere may be particularly important. One such region for the linking of the local emissions with global change may be over the Mediterranean, where the density of emissions is high at the same time as the land mass-ocean differences and the topography in the region give rise to circulations with a significant potential for rapid surface-mid troposphere exchange. This has been studied in some detail by the combined use of aircraft measurements of trace species and mesoscale meterologocal calculations.

Millán et al. (1996, 1997) summarize middle to upper troposphere injection mechanisms in the Mediterranean basin from available evidence and postulated processes, see Figure 1. In the lower to middle troposphere (2,5-3 km altitude) polluted layers are produced by thermally driven land-sea processes and the corresponding return flows. This occurs along south or east sloping coastal mountains. During the day the layer system circulates over the coastal areas in the cause of 2-3 days, and during the night these layers travel along the coast uncoupled from the surface until the next day. These processes are favoured along the east and south Spanish coasts, Italian coasts, south Turkish coasts, and the Lebanese and Israeli coasts, perhaps also over the northwestern African coasts.

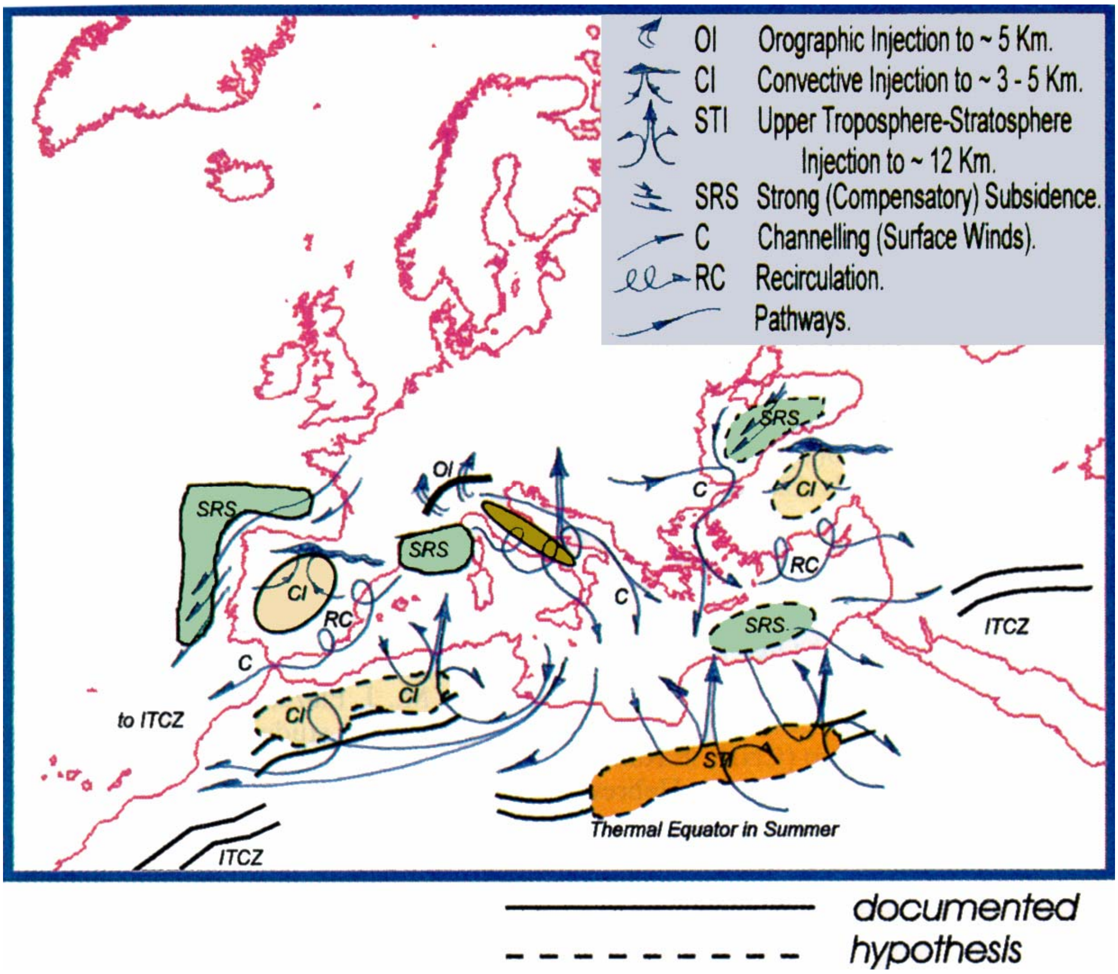

Figure 1 Schematic summary of observed and postulated circulations for the Mediterranean basin in summer (Millán et al., 1997)

Over the Alps, along the Apennines and over the Spanish and Turkish Plateaus and perhaps over the Atlas mountains in Marocco direct injection into the middle troposphere (up to 5 km) is documented to take place from diurnal convective cycles over large and dry land areas and from orographic injection. Condensation can occur. The stratified layers move away with the synoptic flow at their height, in particular during the night. The advection speed can be quite high (10-20 m/s). If the layers drift over the Mediterranean Ocean, large scale subsidence is likely.

Millán et al. (1997) also propose as a working hypothesis (Figure 1) that injection could occur up to 10-14 km as a result of convective pumping with cumulus congestus developing in the late afternoon. Chemical transformation and removal due to the formation of precipitation will be important. The injected pollution is advected by the upper level flow. Evening storms develop regularly in sea breeze or upslope wind systems in mountain ranges like the Apennines in Italy. Over North Africa sea breeze circulation systems may flow toward the Intertropical Convergence Zone and be pumped into the upper troposphere.

These flow regimes are likely to be very important for the exchange rate of pollutants between the boundary layer and the free troposphere throughout the Mediterranean Basin. Their dimension is limited, however, and to assess their impact on the global scale models need to resolve the most important features of each event. The local scales up to the global scales are interlinked, and the handover processes need to be treated in a consistent way. This is an important conclusion from the Mediterranean Basin studies. Another important conclusion is that the mesoscale circulations and convective events in the Mediterranean Basin may transport ozone and its precursors to the free troposphere so efficiently that the contribution to global change is significant. Some evidence for this is found in the tropospheric ozone residuals derived by Fishman et al., 1990 from satellite data. The averaged tropospheric ozone residual for June-August indicate a significant enhancement of the residual ozone column over the Mediterranean Basin 5-10 Dobson Units above the adjacent regions (40 ppb over a layer of 1 km near the surface corresponds to 4 Dobson Units).

4. Chemical transport models, dependence on meteorological data

The quality of chemical transport model calculations depends on the following main groups of data:

1. Model formulation (grid resolution, numerical methods, parameterizations)
2. The physical data for winds, clouds incl convective clouds, and precipitation, as calculated by some kind of state-of-the-art mesoscale, synoptic scale or global scale weather prediction or global circulation models
3. The quality of the boundary conditions for the chemical tracers (both lateral and upper boundary), as calculated by a chemical transport model which has a larger domain
4. The quality of the emission data and the description of the chemical transformation and removal processes

The importance of error propagation from group (2) into chemical tracer calculations is to a large extent overlooked in the literature, because it is generally believed that modern numerical weather prediction models has advanced far and that the trust placed in the quality of the weather analysis and forecast is ”inherited” when chemical tracer calculations are analysed. Also good observations for the validation of clouds and boundary layer mixing processes are not generally available, contributing to the weak emphasis on the validity of these parameters.

For chemical tracers water vapour, shear in wind speed (giving very different transport times from a source region to a receptor volume in the free troposphere), convective activity and cloud cover and type are very important parameters. Water vapor is a key factor in the determination of the hydroxyl radical concentration, which determines the lifetime of most atmospheric trace species. Cloud cover and type influenced photolysis rates significantly.

Also data group (3) propagate serious problems into the interpretation of chemical transport model calculations in that ill defined upwind boundary conditions can be a dominant component in the distribution of long lived species like CO and NO_y far into the interior of the model domain. The method used is not really well defined if it turns out that factors outside of the domain can be dominant in explaining the concentration distributions of chemical tracers in the model. For data group (4) the surface emissions are the driving force for the concentration of chemical tracers.

It is difficult to balance a model development appropriately. The sophistication of the various model elements should approximately reflect the quality of the underlying physical or chemical data (resolution in time and space for instance). Also the model should resolve the spatial and temporal distribution of the phenomenon to be analysed. This means that the model results should not depend too much on ill defined initial or boundary conditions.

5. Mesoscale modelling

The modelling of ozone requires observational basis and theoretical skills in treating physical and chemical processes on all spatial scales, starting on the scale of the city plumes and convective cells, and ending with the global scale.

In the United States ozone plumes from major conurbanations have been observed with a lateral dimension of the order of 10 km (Imhoff et al., 1994, Chameides and Cowling, 1995). The rate of formation of ozone is a nonlinear function of the precursor concentrations, therefore this observation implies that a model calculation of ozone needs to resolve the 10 km scale, or at least allow testing and parameterization to assess the errors when a coarser resolution is used. If the objective is to make a careful assessment of the exposure of the population or crops to ozone levels above certain thresholds, a grid resolution of 10 km or finer in the horizontal can be required.

Such models, often denoted meso-scale chemical transport models, have been developed for many locations both in the United States and in Europe, and they depend on meso-scale hydrostatic or non-hydrostatic meteorological models to quantify the physical fields, and on synoptic scale weather prediction and chemical transport models for boundary values and initial concentrations.

Mesoscale nonhydrostatic models have to applied when the size of the topographic elements is typically one order of magnitude or less than the required horizontal grid resolution. To generate appropriate boundary conditions for a small grid domain with high grid resolution, the small domain can be nested once or several times in coarser models in succession.

The capability of mesoscale meteorological models to calculate the physical parameters which are essential for chemical transport models is generally believed to be good. However, in the German Tropospheric Research Programme (Schaller and Wenzel, 1999) extensive physical and chemical measurements are applied in model validation. Mixing height and specific humidity are two essential parameters in a chemical transport model calculation, but not so important in weather forecasting. In the German programme these mesoscale parameters show systematic and large biases with a too high mixing height in the model calculation, while the specific humidity is too low. When the mixing height is overestimated all precursors, notably CO and $NO_{x,}$ will be underestimated, which is confirmed by the chemical measurements, while the underestimated specific humidity will cause the calculated hydroxyl concentration to be on the low side, causing the calculated chemical lifetimes of trace species like the hydrocarbons, CO and NO_2 to be too long compared to what a measurement would have shown.

These findings are very important because they are somewhat unexpected. Their implication is that it is hard to validate the chemical aspects of a mesoscale chemical transport model, because the physical parameters are so important for the result of the chemical calculations. It is required to constrain the error bounds on parameters like mixing height and cloud cover before it is possible to state with confidence that discrepancies between for instance measured and modelled NO_x, CO or nonmethane hydrocarbons is due to a inferior emission inventory or not properly understood chemical transformation or removal processes.

6. Improvements in numerical weather prediction

Chemical transport model calculations depend critically on the validity of the weather prediction system. Numerical weather prediction has improved over the last one or two decades, at the same time there are aspects of weather prediction which are particularly important for chemical tracer calculations and not so much for the weather forecast. This is for example the distribution and optical thickness of clouds, the mixing properties of the atmospheric boundary layer including predictions of the mixing height, and the distribution of water vapor. These physical properties determine in turn the rate of photodissociation that drive atmospheric photochemistry and the concentration profile in the atmospheric boundary layer and hence the dry deposition rate as well as the rate of exchange between the atmospheric boundary layer and the free troposphere.

The improvement in numerical weather prediction over the last decade is in parts due to better observations, fewer simplifying assumptions are invoked when solving the fundamental equations, numerical solution techniques have been advanced, model resolution has increased, physical parameterizations have been advanced and data assimilation improves model initialization.

Bengtsson (1999) discussed the importance of these factors, and concluded that useful predictive skills over most of the northern hemisphere extratropics has been extended to more than a week at present from about three days through the 1960s and 1970s. Improved observation systems was the main factor until the late 1970s, since then improved forecasting systems including model physics, dynamics and numerical methods have been the main factors. Data assimilation has been of fundamental importance in order to realize fully the model improvements. The combination of an accurate model and an advanced data-assimilation scheme makes it possible to reduce the error of the initial state significantly. Bengtsson (1999) further concluded on the basis of results from ECMWF that using today's models and data-assimilation systems, past forecasts are significantly improved even by only using observations available at the time. This finding justifies the ongoing reanalysis at major weather prediction centres of past observations by today's forecasting systems. In that way the full value of past global observations is exploited.

The forecast skill varies considerably from day to day, and according to Bengtsson (1999) the length of useful forecasts can vary by more than 6 days for 90% of the predictions. The remaining 10% have an even larger span. Ensemble prediction studies (ECMWF) suggest that the major part of this variability is related to the inaccuracy of the initial state and the way these inaccuracies are exposed to rapidly growing perturbations. The growth of errors is for example much faster in active baroclinic zones than in the inner regions of large anticyclones.

Chemical tracer calculations can be very sensitive to errors in advection and physical processes which affect the transformation and hence the lifetime of chemical trace species. Forecast errors can propagate into the chemical tracer calculation and in some cases amplify errors that arise due to for example poorly known emissions or chemical transformation mechanisms and rates.

7. Cloud cover prediction

The calculation of photolysis rates requires an accurate description of the distribution and optical thickness of clouds. When deep convective clouds occur, they affect the vertical transport of trace species in a dramatic way. Cloud sensors on satellites are providing datasets that are gradually been used for model validation. In chemical transport models cloud cover and type of cloud are important parameters because of their direct influence on the photolysis rates and convective exchange. Lin et al. (1994) developed a parameterization of subgrid scale convective cloud transport in a mesoscale regional chemistry model for Eastern and Central North America, and showed that the model calculations of ozone and precursors with the cloud transport parameterizations were in much better agreement with aircraft observations than those without. Flatøy et al. (1995) showed how convective processes parameterized in a mesoscale chemical transport model over Europe significantly reduced the maximum daytime concentrations of ozone in the atmospheric boundary layer in anticyclonic weather over polluted continental areas.

Discrepancies in cloud cover and type of cloud propagate into the chemical transport model results and bias the calculated distribution of the chemical trace species in quite unpredictable ways. If a convective event over a region with high emissions was calculated to take place in a neighbouring grid square where the emissions are much lower, a significant bias is introduced in the calculation of the formation of ozone in the atmospheric boundary layer. Summer time boundary layer formation of ozone usually takes place in weather situations which are also conducive to the formation of thunderstorms. Convective events occur over continents where also anthropogenic emissions can be significant. Events that can lead to photo-oxidant episodes in the boundary layer may therefore coincide much more frequently with convective activity than the frequency of convective activity itself indicates. In convection over polluted continents, the boundary layer is often a net source to the free troposphere both of ozone and precursors. Convection therefore leads to fewer episodic peak concentrations of ozone near the surface, but the precursors vented into the free troposphere cause formation of ozone there, and in addition more ozone is formed in the free troposphere, for the same amount of precursors, than in the boundary layer. The ozone lifetime in the free troposphere is also much longer than in the atmospheric boundary layer.

In conclusion, when comparing the calculated distribution of clouds from numerical weather prediction models or in global circulation models, the agreement with measurements is fair at best. The discrepancies can cause significant errors in chemical transport model calculations. Often errors in calculated chemical tracer fields as evaluated by measurements, are thought to be due to erros in the emissions estimates or in the parameterization of chemical transformation or removal. It turns out, however, that errors in calculated chemistry can often be due to errors in the data for cloud cover, advection or mixing properties in the atmospheric boundary layer.

8. Boundary layer-free troposphere exchange rates over Europe and the North Atlantic

Precursor emissions of ozone are mainly removed in the atmospheric boundary layer and do not enter the free troposphere. The fraction of NO_x entering the free troposphere is essential for the formation of long lived ozone there. A 3-dimensional chemical transport model where a numerical weather prediction model was used to generate the meteorological data needed to run a comprehensive chemical dispersion model was described for Europe and the North Atlantic by Flatøy and Hov (1995). Several such models exist (e.g. Atherton et al., 1996, Lin et al., 1998). Some results will be shown from a calculation by Hov and Flatøy (1997) of the rate of exchange of species between the atmospheric boundary layer and the free troposphere in comparison with other processes, as indicated by the continuity equation which for each species can be expressed as

$\delta[c]/\delta t$ = *emission + advection + diffusion - dry deposition + convection + chemical production – chemical loss*

Here $\delta[c]/\delta t$ is the local rate of change in the concentration of compound c. The terms for advection, diffusion and convection on the right hand side can be both positive and negative. The difference (chemical production – chemical loss) is the net rate of formation of the species c.

The model domain covered Europe, the North Atlantic and parts of Russia. Results are shown here for two sub regions for a 10 day period in July 1991 when there was a weak anticyclonic circulation over north Europe, northwesterly flow off the continent over the British Isles and an easterly flow over central and southeast Europe. A maximum in the ozone concentration in the atmospheric boundary layer was calculated over northern Italy with a ridge of high ozone all the way to Iceland caused by European emissions. Results for two sub regions, one covering 10x15 grid cells in the Eastern Atlantic west of the British Isles, and one covering 5x5 grid cells over Germany, The Low Countries, The Alps and Northern Italy (one grid cell covers approximately 150x150 km^2) are shown in Table 1 for the atmospheric boundary layer defined as the three lowest layers in the model (confined below 865 hPa).

Table 1 shows the mean tendency terms over 10 days in July 1991 for chemistry, (dry+wet) deposition, vertical advection, convection and emissions of NO_x, NO_y and ozone. Over the European continent 8% of the NO_x emissions are brought to the free troposphere as NO_x , while 16% of the NO_x emissions are brought to the free troposphere as NO_y-NO_x, i.e. as PAN or HNO_3. A similar calculation was made for a part of October 1994 when Europe was exposed to westerly winds. For this period the numbers were not significantly different from the July period numbers

Table 1: Mean concentration of NO_x *,* NO_y *(sum of* NO_x *and species generated when* NO_x *is oxidized, except that nitrate aerosol was not included in* NO_y*) and ozone for two sub regions in a 3d chemical transport model calculationd for 10 days in July 1991 by Hov and Flatøy 1997. The average of the tendency terms for chemistry (chem) in the atmospheric boundary layer (ABL), vertical advection (v.adv) and convection (conv) out of the ABL, average emissions, ratio between the rate of vertical transport of* NO_x *and* NO_y *out of the ABL and the rate of emissions into the ABL (v.adv+conv)/emis. The numbers in paranthesis are for a similar calculation for October 1994 with westerly winds over Europe and frequent precipitation in North Europe and shorter residence times of the air masses than in July 1991*

	Eastern North Atlantic			**Continental Europe**		
	NO_x	NO_y	ozone	NO_x	NO_y	ozone
[c], ppt	56	474	31.4	1391	3373	47.6
$(\delta c/\delta t)_{chem}$, ppt/h	-2.6	small	-36	-214	small	1075
$(\delta c/\delta t)_{dep}$, ppt/h	small	-7.4	-29	small	-113.5	-351
$(\delta c/\delta t)_{v.adv}$, ppt/h	-0.0	-2.0	-57	-5.3	-23.4	-194
$(\delta c/\delta t)_{conv}$, ppt/h	-0.0	0.1	3.5	-18	-47	-128
emission, ppt/h	0.7	0.7		290	290	
(v.adv+conv)/emis	0.09	2.9		0.08	0.24	
	(0.32)	(0.15)		(0.07)	(0.11)	

The convective exchange of ozone is not soimportant for the redistribution of ozone. It takes 44 h to generate the concentration of ozone chemically (47.6 ppb/1.075 ppb/h), while the the lifetime due to dry deposition which is 136 h (47.6 ppb/0.351 ppb/h). Convection is a significant factor in reducing the maximum ozone concentration in episodes, however, and this is important from a policy point of view (Flatøy et al., 1995). Vertical advection is on the average more important in the redistribution of ozone than convection. The term $(\delta[c]/\delta t)_{conv}$ for the ABL can be both negative and positive in a convective event, depending on the concentration of ozone in the ABL compared to the mid or upper troposphere, and when averaging over several days and large areas the individual terms tend to cancel.

In Table 2 is shown the average ratio $(\delta[O_3]/\delta t)_{chem}/(\delta[NO_x]/\delta t)_{emis+v.adv+h.adv+conv}$ over Continental Europe for the July 1991 calculation as a function of height. This ratio is one way of expressing the ozone producing efficiency of NO_x, an alternative ratio would be to put the transformation rate of NO_x to NO_y-NO_x in the denominator. The ratio used here expresses the net rate of chemical formation of ozone per NO_x molecule delivered by a physical process (emission, advection or convection) to the volume considered. This ratio in general increases with height and is typically 15-20 in the upper half of the troposphere over Continental Europe and around 5 in the atmospheric boundary layer (ABL). As an average for the total domain there is a net chemical removal of ozone at some altitudes where the concentration of NO_x is so low that the flux through chemical loss reactions of ozone (notably the O_3+HO_2 and O^1D+H_2O reactions) is larger than the flux through ozone production reactions like NO+HO_2 -> NO_2+OH followed by NO_2 photolysis. The ozone producing efficiency increases with altitude because the lifetime of NO_x is longer in the free troposphere. The conversion to HNO_3 and removal of NO_y in precipitation or by dry deposition are slower there than in the ABL.

Table 2: Ozone forming efficiency as a function of the local source strength of NO_x *given as the ratio between the local net chemical formation rate of ozone and the local net change with time of the local concentration of* NO_x *due to emissions, advection and convection for eight pressure heights for the sub region of 5x5 grid cells covering Continental Europe* $(\delta[O_3]/\delta t)_{chem}/(\delta NO_x]/\delta t)_{emis+v.adv+h.adv+conv}$

Pressure height (hPa)	**Continental Europe**
209	21.7
319	15.0
428	15.7
537	11.2
646	6.4
756	9.8
865	5.4
960	3.6

Liu et al. (1987) estimated that around 10 molecules of ozone is formed per NO_x molecule emitted in polluted air masses and around 100 for very clean conditions. See also discussion in Hov (1997).

9. Regional, intercontinental, hemispheric and interhemispheric chemistry and transport

There is a strong hemispheric component in the tropospheric distribution of ozone. This means that emissions over one continent causes ozone formation which appears as background ozone over other continents or as ozone in the free troposphere. Several measurement campaigns in the northern hemisphere have provided data on how anthropogenic and biogenic emissions over distant upwind continents have affected the chemical composition of the troposphere over the Pacific and the Atlantic. A summary of the major findings of a number of campaigns is given in the 1998 WMO assessment of ozone (WMO, 1999, Table 8-4 therein). The field experiments have been accompanied by a number of model developments set up to calculate the regional and global budgets of tropospheric ozone. These models are usually driven by meteorological data generated by a global circulation model, which means that the weather data used do not correspond to the weather situation for a particular date. The grid resolution is coarse which means that the description of the processes for the handover of precursors and secondary species from the sources to the global troposphere is heavily parameterized.

Increasing emissions over the continents augment the likelihood of pollution episodes in the atmospheric boundary layer, and may also cause an increase in surface ozone concentrations over other continents. From a global change point of view, however, the free tropospheric impact of increasing emissions is of particular importance since as already discussed, the ozone producing efficiency per molecule of NO_x is higher in the free troposphere than in the boundary layer. In addition ozone in the free troposphere has a longer lifetime than closer to the ground, and its radiative forcing is higher per unit change than nearer to the surface. Using a global 3-d chemical transport model driven by meteorological data from the GISS model, Berntsen et al. (1996) calculated that a doubling of the NO_x emissions over Asia gives ozone increases of up to 30% in the upper troposphere and with a resulting radiative forcing change of up to 0.5 W/m^2 which is 30-50% of the negative radiative forcing due to sulphate aerosols in the region.

At the UK Meteorological Office a global three-dimensional lagrangian tropospheric chemistry model has been developed where 50000 air parcels are followed. The meteorological data including the wind field is calculated in the UK Met Office UM global ciurculation model, and to make long calculations practical the meteorological data are smoothed as 18 days' means in the chemical transport calculations. In Collins et al. (1997) is reported a global calculation of tropospheric ozone, and the chemical production and loss processes are quantified and aggregated on a global basis. For the month of August it was calculated a net photochemical production rate of ozone of 5.7×10^{34} molecules/day, compared with a stratospheric input at 100 hPa of 8×10^{33} molecules/day, or less than 20% of the in situ net production. If the European NO_x emissions were reduced by 50% it was found that ozone over Europe in summer dropped by 10-20% while in the winter ozone went up by 40% in the same area. This illustrates how NO_x reduces surface near ozone in the winter over polluted continents, while in the summer decreasing NO_x in general reduces ozone over the continents.

The model objectives determine its formulation. Coarse grid hemispheric or global models of the troposphere which are based on global circulation model data are suited for climatological investigations rather than event studies. Processes on horizontal scales shorter than about two grid elements are parameterized, which means that the dynamical influence of land-ocean interfaces, topography and convection is indirect and of a diagnostic character in the calculation. On the other hand, such models can be run for time periods of sufficient length to estimate the global impact of slowly changing parameters like emissions.

10. The capability to forecast ozone in the atmospheric boundary layer

Modelling of atmospheric chemistry has been advancing with the progress in numerical weather prediction. One of the objectives of this chapter has been to discuss how errors in weather prediction may propagate into a chemical transport model calculation. In particular parameters like mixing efficiency in the atmospheric boundary layer, cloud cover and water vapour concentrations may not attract so much attention in the weather forecast, while they are essential for a reliable chemical transport calculation. Also more measurement data are becoming available for CTM validation, in particular from instrumented aircraft in the northern hemisphere. 3-d CTM forecasts are still in their infancy, not least because the initialization possibilities are still quite poor. Almost no chemical measurement data are available online to improve the determination of good initial conditions. Data assimilation has been a major break through for the skills in numerical weather prediction (Bengtsson, 1999), while the theoretical formulation has started in chemical transport calculations (Elbern and Schmidt, 1999). Due to the lack of an appropriate infrastructure which can provide the necessary measurements in near true time, the implementation of routine chemical assimilation cannot be realized. Ozone sonde observations are slowly becoming available with only one hour or so time delay, and ozone is used as a proxy for potential vorticity in the upper troposphere and lower stratosphere and in that way used to improve weather prediction model assimilation (ECMWF activity). The infrastructure which is set up in that way may become a channel for other chemical measurements as well.

The increasing awareness of the negative impact of ozone on the population and for crops has created a demand for online information on expected ozone levels near the ground for the coming days. A legal infrastructure is being implemented in Europe which requires such information to be available to the public. See Flatøy et al., 2000.

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