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Continental ozone issues; monitoring of trace gases, data analysis and modelling of ozone over Europe

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Abstract

The poster and this paper consist of two parts: (1) Concentrations and internal production of ozone in Europe in relation to different concepts of the critical levels and (2) Exchange of ozone between the atmospheric boundary layer and the free troposphere. The first part describes the evaluation of 1989 ozone data from the EUROTRAC Tropospheric Ozone Research (TOR) project and the Co-operative Programme for Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe (EMEP). The data mostly from central and northwestern Europe, show that independent of ways of formulating the critical levels, there are exceedances over large parts of Europe.

The second part shows results from a Data Analysis Model that evaluates the transfer of air from the atmospheric boundary layer (ABL) to the free troposphere (FT) and vice versa. This quantity is of importance since it determines the effective emissions of shorter lived species to the global atmosphere. The model was run for the full year of 1989 to study the exchange processes at the location of Uccle (B). For the summer months June, July and August the results releal a net ABL to FT flux of O_3 with a magnitude of $0.43 * 10^{-3}$ mole m^{-2} day^{-1}. This number is in agreement with values derived from transport models.

1. Introduction

The Dutch part of the EUROTRAC-TOR project consists of contributions from TNO, KEMA and RIVM. The research activities concentrate on measurements at the high quality observatory 'Kollumerwaard', interpretation of the data, hosting the international TOR data base and chemistry and transport modelling. The interpretation activities centre around four basic tasks as they were posed in the NRP-TOR project proposal:

1. How much higher is the mean ozone concentration in the boundary layer over Europe than that averaged over northern mid-latitudes, and what is the seasonal, latitudinal and vertical variation of ozone within the adjacent free troposphere? Is there a secular trend in the concentrations of ozone and precursor molecules in the boundary layer or in the background atmosphere?;
2. What are the emissions and distribution of the precursors responsible for the particular excess of ozone?;
3. Can we measure how much of the excess ozone in the boundary layer spills over into the background atmosphere? Is it possible to quantify by co-measurements of ozone and other tracers the proportion of ozone produced in the troposphere to that transferred from the stratosphere to the troposphere at our location?;
4. How much ozone and how many precursors are transported across regional boundaries?

A full assessment of all the questions raised in the four tasks is beyond the scope of this extended abstract. In the following sections we discuss the internal production and the exceedance of critical levels of ozone in Europe and exchange processes between the boundary layer and the free troposphere. A more elaborate evaluation of the above mentioned questions is expected to be given in our final NRP report.

2. The internal production of ozone within Europe

The internal production of ozone within Europe was estimated by selecting monitoring stations from the TOR network which were found to be very little affected by local sinks as well by European ozone production and destruction. Emissions of NO_x from traffic in the vicinity of a monitoring station is an important sink for ozone, in particular during inversions when high NO concentrations may consume a substantial part of the ozone. Another sink is the deposition to vegetation during nighttime inversions. Both sinks may cause systematic diurnal variation in the concentration of O_3. In this evaluation the ratio maximum to minimum (O_3max / O_3min) hourly concentrations from the mean diurnal variation for the summer and winter half year respectively were used. The ratios varied considerably; from 5 to 8 in the areas most influenced by local sinks to 1.04 - 1.40 in the areas with the smallest influence from local sinks. The second criterion was that the stations should very seldom experience ozone episodes. Only four lowland (<500 m a.s.l) monitoring stations were found fulfilling these criteria. these were Jelöy and Svanvik in Norway, Mace Head in Ireland and Strath Vaich in Scotland. At these stations ozone episodes very seldom occurred.

The mean diurnal variation from the four background stations were assumed to represent the background ozone concentration in Europe. The summertime mean concentration at the four background sites was 32 ppb. The internal ozone production was then calculated as the difference between the diurnal curves from the actual monitoring station and the mean for the four background sites. At most sites this approach gives a production of ozone during daytime (mainly between 9:00 and 21:00h) and a destruction of ozone during the night. Since most vegetation effects are caused by stomatal uptake of ozone, it may be more appropriate to look only at daytime differences. Based on these assumptions the internal ozone production in Europe has been calculated to be of the order of 10-30 ppb at remote lowland sites in central Europe. In the south of Scandinavia the internal ozone production is of the order of 5-8 ppb. In the south of the UK, as well as at sites on the continent more influenced by local and mesoscale pollution, several sites show a mean ozone destruction, which at some sites may exceed 10 ppb. Table 1 summarises the analysis on internal ozone production in Europe.

Table 1
Internal ozone production in Europe calculated from the TOR network as the difference in daytime ozone concentration between the actual monitoring station and the mean of the concentrations at four 'boundary layer background' sites.

Monitoring area	Number of stations	Internal ozone production (ppb)
Continental Europe; remote sites	6	5 ~ 30
Continental Europe; urban sites	8	-5 ~ +10
British Isles - south of 54°N	9	-13 ~ +8
British Isles - north of 54° N	6	0 ~ 3
Scandinavia - south of 62° N	12	5 ~ 8
Scandinavia - north of 62° N	6	-3 ~ +3

3. Ozone concentrations in Europe in relation to the critical level concept

In order to develop effect-oriented control strategies for regional air pollution the concept of critical loads and levels was developed. The concept was accepted in Europe for the assessment of the effects of acidic deposition and it will probably be the basis for the development of control strategies for ozone as well. It is now based on the exceedance of ozone concentrations over a threshold concentration and it is expressed in ppb.hours units.

One important issue is to what extent episodic ozone and background ozone contribute to the exceedance of the proposed 40 ppb base concentration. If episodes make the largest contribution, it seems reasonable to concentrate control strategies to eliminate episodic ozone but if the background ozone makes an important contribution it may be necessary to direct strategies to the free tropospheric background.

The importance of episodes for a selected number of TOR sites was evaluated. If we define episodes as hours when the ozone concentration exceeds 60 ppb, episodes contribute to the exceedances by more than 50% at most sites, where the exceedances are above 2000 ppb.hours per growing season. For sites having an exceedance of less than 2000 ppb.hours per growing season, the contribution from episodes is at most sites less than 50% and at remote sites less than 30%. So the central European oxidant problem is mostly an episode problem while episodes play a minor role for the exceedance of the critical levels at the outskirts of Europe.

It is obvious that the ways the critical levels are formulated may influence the control strategies for photochemical ozone in Europe. If critical levels are set to an exceedance of 50 ppb, the control of ozone will almost entirely be directed towards central Europe and one may assume that the exceedance may disappear at the outskirts by controlling the central European problem. If, on the other hand, the base level is set to 30 ppb, ozone becomes an all - European problem and control strategies may be directed towards all Europe. Since non-episodic conditions play an important role for the exceedance of the 30 ppb level it may be necessary to direct control measures to sources outside Europe and also to longer lived components like CH_4 and CO.

4. Exchange of ozone between the atmospheric boundary layer and the free troposphere

The ozone budget in the troposphere is composed of transport from the stratosphere, photochemical production, deposition at the earth's surface and photochemical destruction. The net photochemical production includes ozone formation both in the free troposphere (FT) and in the atmospheric boundary layer (ABL). There is transfer of air between the FT and the ABL; therefore a separate ozone budget of both layers may be drawn up. Despite the fact that the importance of transport processes on the FT and ABL ozone budgets has been known for long, there is still a large quantitative uncertainty about the fluxes involved.

Except for transfer of ozone between the ABL and the FT, the transport of VOC and NO_x is also of importance because vertical mixing of these components may, due to the non-linearity in ozone formation, cause more efficient ozone production from the same emissions in the FT than would occur directly in the ABL.

The influence of the diurnal cycle in the ABL depth, caused by convective activity, on exchange between the ABL and the FT was studied using a combination of the ground based TOR network, vertical profiles of ozone and meteorological parameters, synoptical information and land-cover data. These 'ingredients' were integrated with parameterisations of atmospheric processes in a data analysis model. The convective exchange processes and their influence on the ozone budget at the TOR station Uccle (B) were evaluated with this data analysis model for every hour covering the full year of 1989.

The analysis revealed a flux directed from the ABL to the FT at the Uccle (B) location averaged over June, July and August 1989 of 0.77×10^{-3} $mole.m^{-2}.day$. The downward FT to ABL flux was 0.33×10^{-3} $mole.m^{-2}.day$ in the same period. So the net flux is directed upward from the ABL to the FT and has a magnitude of 0.43×10^{-3} $mole.m^{-2}.day$. If we multiply this number with the surface of the countries in northwestern Europe ($2{,}442{,}510 \times 10^6$ m^2) it results in a net ABL to FT transport of 1.0 Gmole. day^{-1}. This corresponds to 4 ~ 6% of the daily cross tropopause flux of ozone in the northern hemisphere in summer.

Besides convective growth of the ABL depth, the exchange of air between the ABL and the FT takes place through several meteorological phenomena like clouds, frontal systems, heat island phenomena etc. The influence of these processes is not assessed in the present analysis.