ANALYSIS OF A 7 YEAR TROPOSPHERIC OZONE VERTICAL DISTRIBUTION AT THE OBSERVATOIRE DE HAUTE PROVENCE

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Abstract

A seven year (1984-90) climatology of tropospheric vertical ozone soundings, performed by electrochemical sondes at the OHP (44°N, 6°E, 700m ASL) in Southern France, is presented. Its seasonal variation shows a broad spring/summer maximum in the troposphere. The contribution of photochemical ozone production and transport from the stratosphere to this seasonal variation are studied by a correlative analysis of ozone concentrations and meteorological variables, with emphasis on potential vorticity. This analysis shows the impact of dynamical and photochemical processes on the spatial and temporal ozone variability. In particular, a positive correlation (r = 0.40, significance >99.9%) of ozone with potential vorticity is observed in the middle troposphere, reflecting the impact of stratosphere-troposphere exchange on the vertical ozone distribution.

I. Introduction

The European project TOR (Tropospheric Ozone Research) aims at establishing the tropospheric ozone budget on a regional, European scale. The core of the project is a network of 39 surface stations measuring ozone, related trace constituents (NOx, hydrocarbons), and radiative and meteorological parameters, and of 7 stations for vertical ozone sounding. The goal of the surface measurements is to evaluate the impact of photochemical ozone production in the boundary layer on a regional scale. The vertical ozone soundings provide information on the photochemical ozone production in the free troposphere and the transport of ozone both from the stratosphere and the boundary layer. The Observatoire de Haute Provence (OHP, 44°N, 6°E, 700m ASL) in Southern France is part of the TOR network and vertical tropospheric ozone sounding is performed there both with a UV-DIAL lidar system and with electrochemical sondes. In this paper, the tropospheric ozone climatology obtained at OHP with Brewer-Mast sondes between 1984 and 1990, is presented. By relating the ozone data to meteorological variables, the relative importance of dynamical and photochemical processes for the spatial and temporal ozone variability will be studied.

II. The tropospheric ozone climatology at OHP between 1984 and 1990

Between 1984 and 1990, 140 ozone profiles up to approximatively 30 km altitude have been performed at OHP, using balloon-borne electrochemical Brewer-Mast sondes. The OHP is a rural site, the largest nearby urban agglomeration (Marseille) being located at 150 km distance in a south-west direction. From the instrument error analysis and two intercomparison campaigns involving several measuring techniques, it is concluded that our time series of ozone measurements is too short to derive an annual ozone trend with sufficient statistical significance [Beekmann, 1992], but that Brewer-Mast sondes are well suited to match the temporal variability of tropospheric ozone on a timescale of some days (20%) or of one year (40%). Furthermore, the spatial ozone variability (typically 20% in the Western Europe free troposphere) can be correctly assessed if stations using the same measurement technique (here Brewer-Mast sondes) are taken into account.

The ozone climatology at the OHP shows a broad spring and summer maximum in the troposphere (fig.1), as expected for a northern mid-latidude station. This seasonal variation is similar for the other stations in Western Europe. Figure 1 indicates however a meridional gradient of ozone in Western Europe, with larger values at the more northerly stations Uccle and Jülich (51°N), compared to the more southerly stations OHP (44°N) and Pic de Midi (43°N). These differences become more pronounced with increasing altitude. There might also be systematic differences depending on the continental character of a station: at Hohenpeissenberg (48°N, 11°E), located at about the same latitude as Payerne (47°N, 7°E), but more in the east, the larger ozone values are found. This analysis raises thus the question of whether first the spring-summer maximum and second spatial gradients are caused either by photochemical ozone production or by transport from the stratosphere.

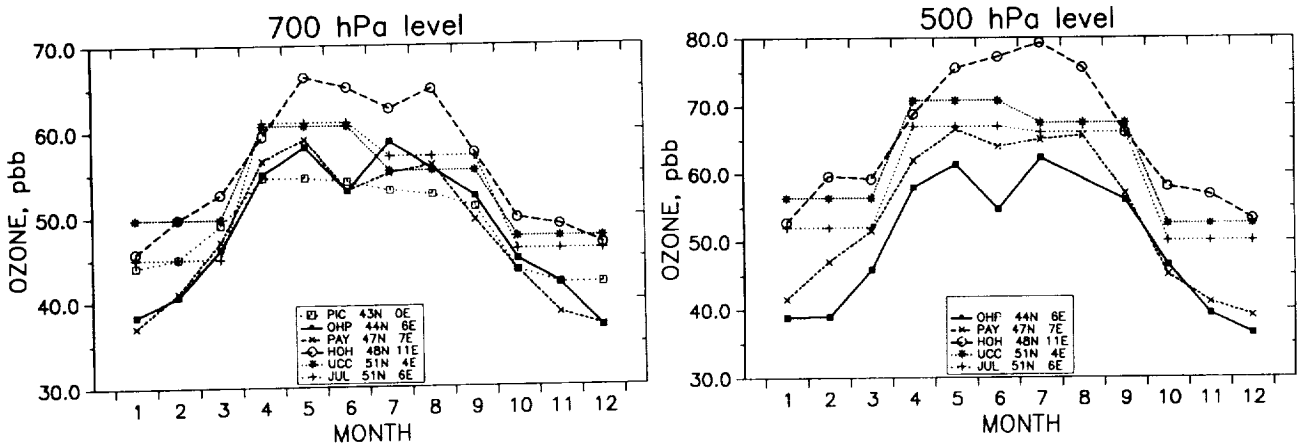

fig.1 : seasonal variation of tropospheric ozone at different Western European sounding stations,

	station	*location*	*period*	*instrument*	*reference*
PIC	Pic de Midi	43°N, 0°E, 3000m ASL	1982-84/90-91	UV-photometer	Nedelec, 1991
OHP	Obs. de Haute Provence	44°N, 6°E, 670m ASL	1984-90	Brewer-Mast	Beekmann, 1992
PAY	Payerne	47°N, 7°E, 490m ASL	1982-88	Brewer-Mast	Staehelin et al., 1991
HOH	Hohenpeissenberg	48°N,11°E, 980m ASL	1985-89	Brewer-Mast	Sonderbeobachtungen 86-90
UCC	Uccle	51°N, 4°E, 100m ASL	1984-88	Brewer-Mast	Bull. trimestr. 1985-89
JUL	Jülich	51°N, 6°E, 100m ASL	1989-91	ECC	Smit et al., 1991.

III. Analysis of ozone with respect to meteorological data

A first approach to relate the observed ozone variability at OHP to physico-chemical and dynamical processes, is to perform a statistical analysis of ozone concentrations with respect to meteorological data such as potential vorticity, humidity, geopotential height and trajectories. Contrary to concentrations of ozone precursors (NOx, hydrocarbons), these data are available in the free troposphere for each ozone profile, either directly from sonde measurements (pressure, temperature, humidity) or from meteorological models, providing objectively analysed temperature and wind fields for potential vorticity and trajectory calculations.

In this paper, we focus on the analysis of potential vorticity, which is a tracer of stratospheric air masses transported into the troposphere. In the absence of a vertical gradient of diabatic heating and in the absence of frictional forces, potential vorticity is a conserved quantity. Due to the similar vertical gradients of ozone mixing ratios and potential vorticity (PV) in the lower stratosphere, a positive correlation of ozone and PV in this region is expected. This is verified for the OHP ozone data base at the 225 hPa level, where a strong and significant correlation between both variables is found (r= 0.82, significance > 99.9%). At the 500 hPa level, a significant, but weaker, positive correlation between ozone concentrations and potential vorticity is observed (r= 0.40, significance > 99.9%). This positive correlation reflects the impact of stratosphere-troposphere exchange on the ozone variability in the middle troposphere. The correlation is weaker for the mid-troposphere than for the lower stratosphere, because in the troposphere ozone and PV are partially decorrelated by photochemical processes on one hand, acting on ozone, and by diabatic processes on the other hand, acting on PV (e.g. latent heat release, radiative processes including the interaction with clouds, sensible heat flux at ground). The ozone/PV correlation is most pronounced in the period spring/early summer (tab.1), indicating a larger importance of stratosphere-troposphere exchange during this period. The correlation coefficients are almost unchanged if one considers ozone concentrations and potential vorticity without their seasonal variation (residuals of monthly means), which means that the variability of both values is also correlated for timescales smaller than one month.

	r	p
225 hPa, all seasons	0.83	>99.9%
500 hPa, all seasons	0.40	>99.9%
mar. - july	0.49	>99.9%
aug. - oct.	0.27	93%
nov. -feb.	0.40	98%

tab.1 : correlation coefficients **r** between ozone and potential vorticity; 1 - **p** is the probability that a higher correlation coefficient than **r** would have been obtained if both data sets were completely uncorrelated (exactly valid only for binormal distributions).

The knowledge of potential vorticity offers the possibility to sort the ozone profiles into two classes: one with air masses of larger PV values, affected by recent stratospheric ozone intrusions, the other with air masses of

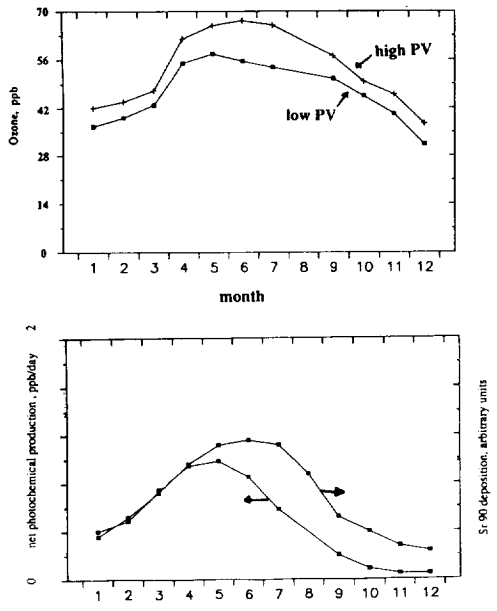

fig.2 : above: seasonal variation of ozone profiles of the high and the low potential vorticity class at 500 hPa,
below: seasonal variation of the net photochemical ozone production at 50°N and 4 km height [Isakson, 1988], Sr 90 surface deposition in Southern France [Marenco and Fontan, 1974].

smaller PV values, which are then, in the absence of recent stratosphere-troposphere exchange, representative of the photochemical ozone source. PV values, which are more than 0.1 PV-units above their monthly mean at the 500 hPa level and which are larger than 0.8 PV-units at the 350 hPa level (i.e. near the dynamical tropopause of 1.6 PV-units), belong to the high PV-class (1 PV unit = 1.10-6 K m²/ kg s). Figure 2 shows a similar seasonal variation of ozone for both classes at the 500 hPa level. The seasonal variation of the high PV-class compares well with that of the deposition of Sr 90 in Southern France at the end of the sixties [Marenco et al., 1974]. Sr 90 is a fission product released during atmospheric nuclear bomb tests and was widely used as a tracer of stratospheric air. Furthermore, the seasonal variation is in good agreement with that of the frequency of occurence of cut-off lows, which is maximum in June [United Kingdom Photochemical Oxidants Review Group, 1987]. The seasonal variation of the low PV-class corresponds well to that of the seasonal variation of the net photochemical ozone production at 50°N and at 4 km height, as computed by a 2D-model [Isaksen, 1988]. The larger amplitude in the seasonal variation of the ozone sources than in the seasonal variation of the ozone concentrations is explained by the larger deposition rates during the period spring/summer [Liu et al., 1987]. Thus, allthough a quantitative determination of the respective contribution of the two ozone sources is not possible by this analysis, it supports nevertheless the seasonal variation of the ozone sources, derived by photochemical modeling and by other tracers of stratosphere-troposphere exchange (Sr 90 deposition, cut-off lows).

The simultaneous knowledge of ozone and PV values allows to determine the ozone/PV ratio and its seasonal and altitude dependence at the OHP. The exact knowledge of this ratio allows the initialisation of ozone fields of models from PV fields and to calculate ozone fluxes from PV fluxes. At the OHP, the values of the ozone/PV ratio are much higher in the mid-troposphere than in the lower stratosphere (fig.3). They show a spring maximum at the levels 500 hPa and at 225 hPa and an early summer maximum at 350 hPa. The higher tropospheric values of the ozone/PV ratio are explained by a decrease of the potential vorticity in the troposphere due to diabatic processes and an increase of ozone due to photochemical production, particularly in the period spring/summer.

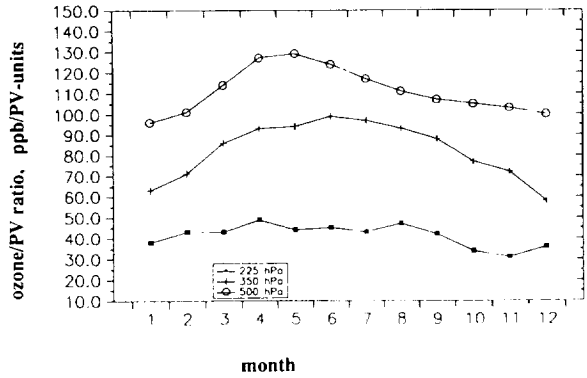

fig.3 : ozone / potential vorticity ratio at OHP for 500 hPa (upper curve), 350 hPa and 225 hPa.

A comparison of the ozone/PV ratios for a northern and a southern station (Uccle, 51°N, OHP, 44°N), shows only small differences compared to the error bars of the ratios (tab.2). As a consequence, the meridional ozone gradient in Western Europe is related to a similar gradient of potential vorticity, which shows the importance of dynamical effects (e.g. lower tropopause height at Uccle) for the spatial distribution of ozone in the upper and middle troposphere.

level	500 hPa		300 hPa	
station	OHP 44°N	Uccle 51°N	OHP 44°N	Uccle 51°N
ozone [ppb]	49.6 ± 4.6	61.8 ± 4.6	58.3 ± 7.7	99 ± 7.7
potential vorticity (PV-units)	0.45 ± 0.02	0.61 ± 0.05	0.95 ± 0.05	1.8 ± 0.2
ozone/PV ratio [ppb/PV-unit]	110 ± 15	101 ±15	61 ± 11	55 ± 11

tab.2: comparison of the ozone / potential vorticity ratio at Uccle (51°N) and at OHP (44°N)

We now make use of the established correlations between ozone and potential vorticity and also between ozone and relative humidity at the 850 hPa level (r = - 0.38, significance > 99.9%), in order to determine the part of a potential ozone trend due to dynamical processes. For the OHP data set between 1984 and 1990, a considerable variability of relative humidity and potential vorticity has been observed which would yield ozone trends between - 2.7% and + 1.2% for different pressure levels (tab.3). It is clear that these dynamical induced ozone changes have to be taken into account, if one wants to deduce accurately the ozone trend due to anthropogenic activities.

level	850 hPa	500 hPa	350 hPa
annual trend of relative humidity (84-89)	-4.5 ±1.4%		
annual trend of potential vorticity (84-89)		-2.0 ±2.2%	-11.8 ±5.2%
annual ozone trend induced by meteorological variables	+1.2 ±0.4%	-0.5 ±0.7%	-2.7 ±1.2%

tab.3 : the impact of the interannual variability of potential vorticity and relative humidity on a possible ozone trend.

4. Summary and future work

The tropospheric ozone climatology obtained at the OHP in Southern France between 1984 and 1990 shows a seasonal variation characterized by a clear spring/summer maximum of ozone concentrations. A statistical analysis of the ozone profiles together with meteorological variables, especially with potential vorticity, shows that this maximum is both due to transport from the stratosphere and to photochemical production. This is in good agreement with the ozone sources derived from model calculations (photochemical production) and from independent tracers of stratosphere-troposphere exchange (Sr 90 deposition, cut-off lows). We can also show the notable impact of dynamical processes on the meridional ozone gradient in Western Europe and on an interannual ozone trend.

The future work has to be devoted to a more particular analysis of the photochemical source of ozone, using the surface data of ozone precursors (NOx, hydrocarbons), which are now available in the TOR-database. Trajectory analysis, taking into account the photochemical transformation of air masses (Lagrangian modeling), could be used to relate the surface emissions of ozone precursors to the vertical ozone profiles, in order to establish the impact of the enhanced emissions of ozone precursors over Europe on the strengh of the photochemical ozone production in the free troposphere.

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