SOME OBSERVED RELATIONS BETWEEN OZONE AND ATMOSPHERIC FLOW FIELD

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Abstract

It is briefly review for the non specialist in meteorology the atmospheric ozone problem, along with some characteristic atmospheric flow field patterns including jet stream circulations, and illustration is made of their interaction with ozone mainly on observational grounds. Mention is made to the outside and inside cabin ozone content likely relation, according to research made elsewhere [5,17]. Ozone levels up to 300ppb in the troposphere and up to 1000ppb in the lower stratosphere have been measured which may concerne SST, but around 4 times higher values may be found in the middle stratosphere, which may concerne advanced SST.

1. Introduction

Mankind needs progressively larger knowledge of everincreasing regions of its environment, on which the earth's atmosphre plays a fundamental role from its upper top down to its lower terrestrial boundary layer. Thousands of persons of all ages and health conditions fly daily in the atmosphere sometimes for several hours, and this circunstance implies efforts to be made on a global basis of cooperation to offer them appropriate safeguard. In addition to weather forecasts, atmospheric composition needs to be considered in this specific context, embrassing its time and space more likely variations in relation to atmospheric circulations, and among the atmospheric components the ozone is relevant in many respects, and must be considered in the climatology of the atmosphere. On the other hand, the atmosphere has its mean circulations and composition, as for ozone, but flights are made in almost synoptic situations rather than in mean conditions, and this is a not the least reason to be aware of the relations between the perturbed field of motion and ozone distribution.

The importance of atmospheric ozone in the fields of Geophysics and Biology has long ago been recognized [6,10], including the potencial hazards of its presence inside the cabin of man-manned flying crafts [2,5,16]. This reason guide international efforts in the study of the ozone field on a cooperative basis mainly under IUGG and WMO coordination, strong support being given nowadays by UNEP [18] to extensively monitor and investigate the atmospheric ozone layer, a project to be implemented under the scientific guidance of the Commission for Atmospheric Sciences of WMO Working Group of Atmospheric Ozone.

The Instituto Nacional de Meteorologia e Geofísica of Portugal, formerly Serviço Meteorológico Nacional, initiated its cooperation with the International Ozone Comission more than two decades ago, and observes surface, total, and vertical ozone distribution in the atmosphere on a routine basis, as part of the WMO ozone network of Region VI (Europe), and has published a number of papers on this subject [12,13,14].

2. Ozone properties, sources and sinks

Pure ozone is the triatomic oxygen, O_3, it has the density 1,72 relative to air, is invisible under weak concentrations and ordinary temperatures, but it shows up blue under high concentrations due to its absorption of electromagnetic radiation in the yellow and orange bands. In addition, it has a characteristic strong smell, which permits the detetion of 100ppb by volume of the gas in air (0,2mg/m^3), which becomes lethal [10] with 4mg/m^3 ozone

content.

Ozone produces oxidations under ambient temperatures on a great number of products which otherwise would not be oxidize by oxygen, and this turns to be one of the major sinks of atmospheric ozone in the lower troposphere. A typical reaction takes place when only one atom of oxygen is fixed, namely

$$2\ IK + O_3 + OH_2 \rightarrow I_2 + 2\ KOH + O_2$$

This reaction is used to measure atmospheric ozone by the electrochemical method. In addition, the oxidation of organic compounds by ozone explains its biologic properties.

Given the above mentioned relative density of ozone to air, it would be expected a decreasing amount of ozone to be found in the earth's atmosphere with increasing hight, which observation proved not to be the case. In fact, ozone maximum is located within the layer 20 to 25km (50 to 20mb) of the stratosphere, and this is explained [7] by the photochemical theory, both for equilibrium and non-equilibrium conditions. There is however evident disagreament between theory and observation when seasonal and latitudinal variations of the gas are examined, which is due to the ozone transport by atmospheric circulations.

The photochemical theory in the assumption of equilibrium implies that atmospheric ozone is continuously being formed and destroyed in a dynamic process including both primary and secondary reactions. If mainly oxygen is involved, than the primary reaction is a photodissociation in the Herzberg continuum of the oxygen (1900Å to 2420Å) with the energy input of 128,000cal, namely

$$O_2 + h\nu \rightarrow O + O$$

Simultaneous conservation of energy and momentum during the ozone formation implies the presence of a third body (e. g. N_2), as

$$O_2 + O + M \rightarrow O_3 + M$$

A sink of ozone arises by its direct fotodissociation for $\lambda \leq$ 11,500Å, mainly by the UV Hartley bands (2320Å to 2930Å) in the upper stratosphere, and by the visible Chappuis bands (centered at 6000Å) at lower elevations. It absorves also in the UV-B band (2900Å - 3200Å). Another sink of the gas is the two-body collision

$$O^* + O_3 \rightarrow 2O_2$$

Computations based on this classical scheme have shown that the above reactions are the important ones under stratospheric conditions, although reactions with H - Compounds be of importance also below the stratopause (50 km).

Assuming that sources and sinks intensities tend to a state of equilibrium, then

$$\frac{\partial [O_3]}{\partial t} = \frac{\partial [O]}{\partial t} = 0$$

which allows equilibrium concentrations of O_3 and O to be known, provided the energy distribution in the incoming solar radiation, the absorption coefficients of oxygen and ozone, the reaction rates, and the thermal structure and density of the atmosphere are known. The vertical distribution of atmospheric ozone so computed agrees

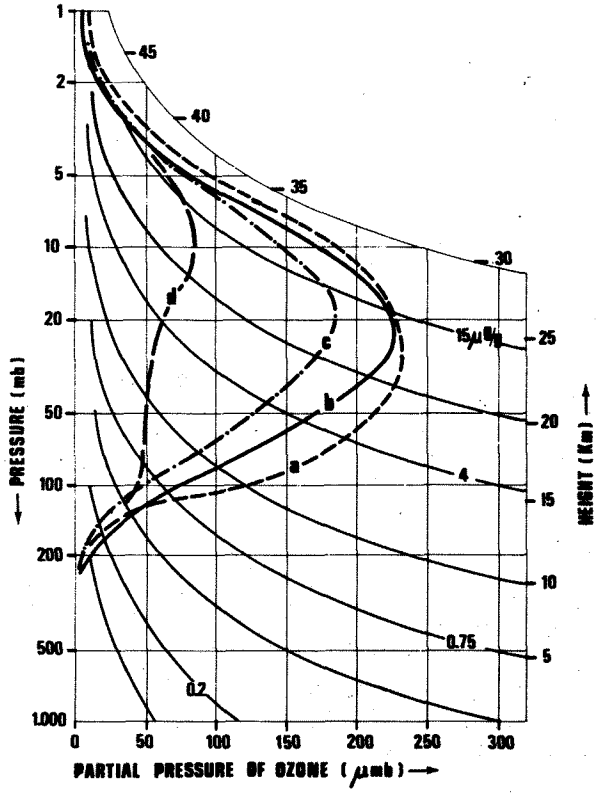


Fig. 1 – Vertical ozone distribution according to the photochemical equilibrium theory at different latitudes and seasons.
a = equator, June; b = 45°N, June; c = 60°N, June; d = 45°N, December (From Dütsch, 1969)

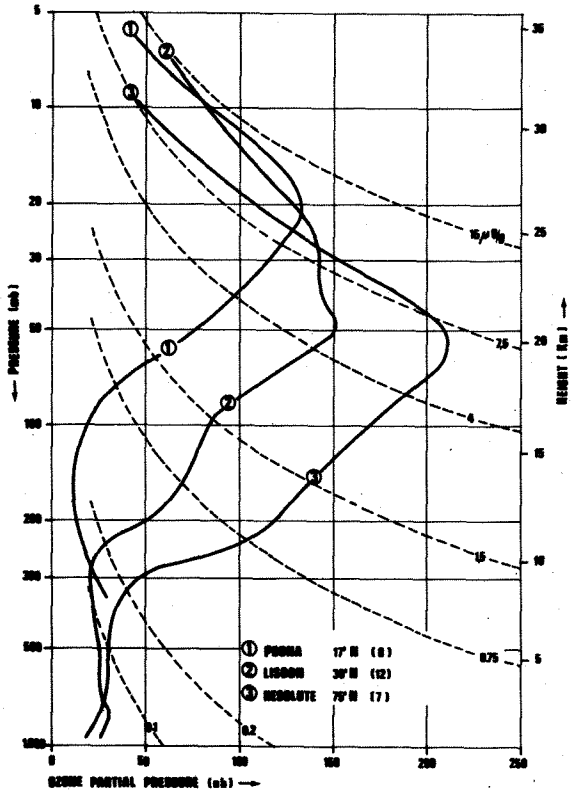


Fig. 2 – Vertical ozone distribution in spring from Umkehr (1) and ozonesonde observation (2;3)

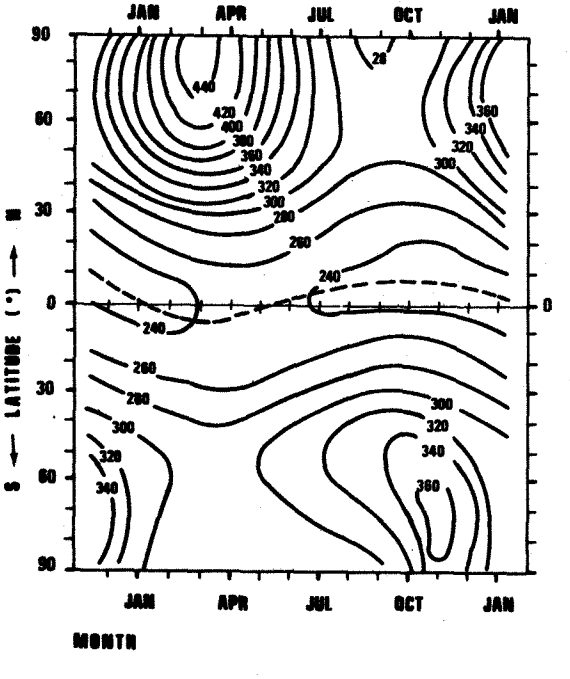


Fig. 3 – Average variation in total ozone (m Atm cm) by latitude and month, based on observations during 1957-1967. (From WMO, 1977)

reasonably well with observational results pertinent to a fixed location, but this is not so as regards seasonal and latitudinal distributions, as may be seen in figures 1, 2 and 3, the latter giving total ozone which is of course positivelly correlated with layered ozone, and this discovery gave a first hint, to be illustrated later, that the discrepancy may be due to the interaction between the ozone and the atmospheric flow field, which a great deal of research proved to be the case.

Improvement of the photochemical theory of ozone sources and sinks advanced by [7] shows that on the assumption of non-equilibrium $[O_3]$ may still be used but representing the instantaneous, not the equilibrium value, thereby a relaxation time or time to reach the equilibrium being derived. This parameter is a minimum at the stratopause (50km) with values of the order of few hours, and increases downwards reaching values of the order of 10^5 days in the lower stratosphere (20 to 15km), wich means that photochemical reactions are too slow below 15-20mb to follow seasonal changes, and again the conclusion is that atmospheric transport processes play an important role in the distribution of ozone below about 25km.

It is known today [9,18] that the aforementioned is only the main part of the ozone chemistry in the stratosphere, for which other natural and man-made products are of importance. Keeping to the scope of this work, mention is only made to the hydrogen system involving the radicals OH and HO_2, in which case CH_4 diffusing upward may interfere; the nitrogen system, NO_x, injections of which in the lower stratosphere by stratosphere-flying aircrafts, both subsonic and supersonic [4] are under study as regards ozone layer reduction; the chlorine system, ClO_x, is of some concern because the chlorine included in man-made chlorofluoromethanes (CFMs) released into the atmosphere enters a catalytic chain and destroys ozone; the chlorine nitrate effect; and the bromine system, including reactions similar to the chlorine system

Beside the layered ozone, total ozone is commonly measured at ground based and aboard satellite spectrophotometers, and is the result of the integrated vertical distribution of the gas, usually referred to in Dobson Unit (DU) or mAtmcm. It cha-

racterizes the total height of pure ozone reduced to normal pressure and temperature, contained in a vertical column of unit section of the atmosphere above (or below) the observing site. A typical value of 300D.U. (300mAtmcm or 0,3cm) may be retained for reference.

3. The average atmospheric motion

The so-called mean general circulation of the atmosphere is made up of a large spectrum of motions averaged over large time (decades) and space (global) scales. The statistics so obtained obeys reasonably well to the hydrostatic equilibrium as a common qualitative property holding throughout the atmosphere, and to the geostrophic equilibrium which prevails in middle and higher latitudes. Hydrostatic equilibrium means approximate balance between gravity and the vertical pressure gradient forces, and geostrophic equilibrium means approximate balance between the horizontal Coriolis force $f = 2 \Omega \operatorname{sen} \varphi$ and the horizontal pressure gradient, respectivelly

$$\partial p = -\rho g dz$$

$$\bar{U} = (g/f)\, \bar{k} \times \operatorname{grad} z$$

where Ω is the magnitude of the angular velocity of the Earth ($7{,}292 \times 10^{-5}\ s^{-1}$), $\bar{U}$ is the horizontal wind velocity, ρ is air density, $\bar{k}$ is a unit vector positive upward, and φ is latitude measured northward. Combined they give the thermal wind equation

$$\frac{\partial \bar{U}}{\partial p} = -(R/fp)\bar{k} \times \operatorname{grad} T$$

which relates the vertical wind shear to the gradient of temperature along a constant pressure surface, R = Cp - Cv being the gas constant for air ($2{,}87 \times 10^{6}\ cm^2\ s^{-2}\ deg^{-1}$). The last equation is responsible for the local wind variation wind height as a consequence of the thermal structure of the atmosphere.

In addition, the atmosphere as a fluid system is governed by the basic hydrodinamic and thermodynamic laws, which includes the law of conservation of mass, Newton's second law of motion, and the second law of thermodynamics. Other set of laws includes radiation, where the interaction of electromagnetic radiation with the atmospheric composition must be known, ozone becoming very important in this context as referred to in section 2, the laws governing the evaporation and condensation of water, and others. Once written the laws, computational work would permit to visualize the actual state of the motion of the atmosphere and its future, a tremendous task indeed, and this explains why appeal to observation and analysis of the atmospheric conditions needs to be made on a routine basis, whereby synoptic and mean circulations are derived.

However, when the actual circulations are averaged, such entities as fronts and migratory cyclones may be filtered out, but this and other perturbations are very important features of the atmosphere for the users of the meteorological information. Many examples of both actual and averaged filds could be shown , but use will be made of a few readily available from previons work. Fig. 4 shows a schematically long term averaged vertical profile of temperature, and figure 5 is the time and longitude averaged zonal wind in January. On the other hand, figure 6 provides synoptic observed wind up to 100mb on the 21 st January 1959 with two very well defined tropospheric jet streams which must be compared to figure 5; figure 7 shows the 30mb stratospheric contours and their changes at five days' interval along 50ºN; and figure 8 give the evolution for three days (14 to 17 Jan 1962) of the wind field over Zaragoza between 500 mb and 5 mb, illustrating the ocurrence of well defined tropospheric and stratospheric westerly jet streams over Europe. The vertical motions associated to the jets provide a very efficient mechanism whereby mass exchange between the upper and lower layers of the atmosphere is achieved which is important in relation to the ozone redistribution.

4. Atmospheric ozone and circulations

The observed fields of motion of the atmosphere as those shown in the well known weather maps and in satellite pictures are indeed the situations the aircrafts in flight are faced with on a routine basis, and the fields of atmospheric composition

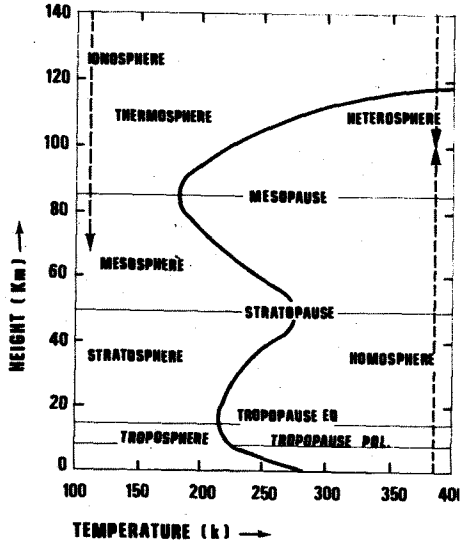


Fig. 4 – Vertical profile of atmospheric temperature and usual nomenclature

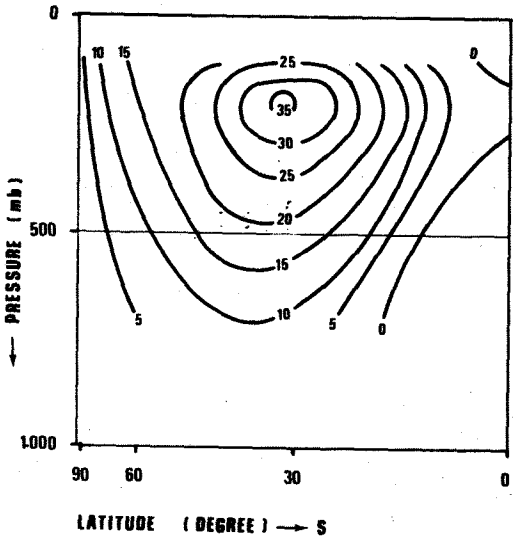


Fig. 5 – Time and longitude averaged zonal wind January, in m s^{-1} (From Lorentz, 1967)

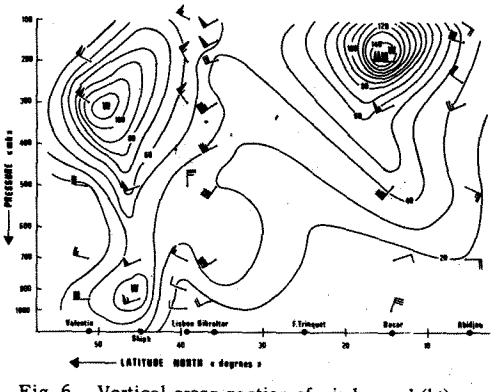
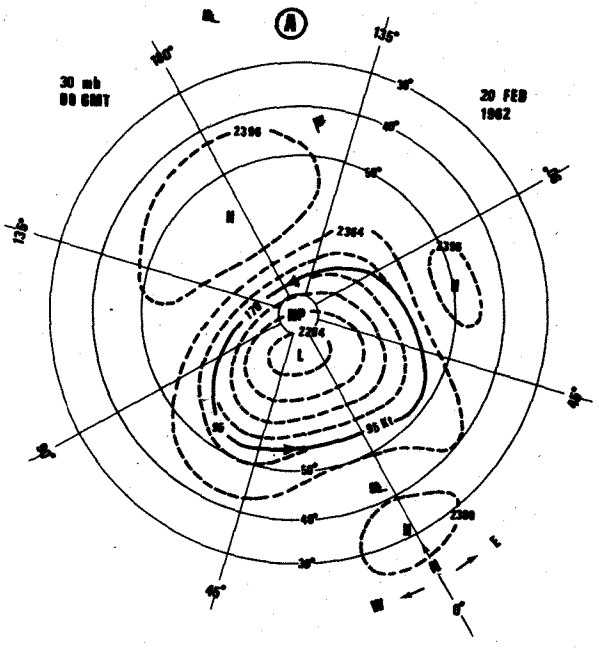


Fig. 6 – Vertical cross-section of wind speed (kt) near 10°W, 21 Jan 1959 (From Figueira, 1965)



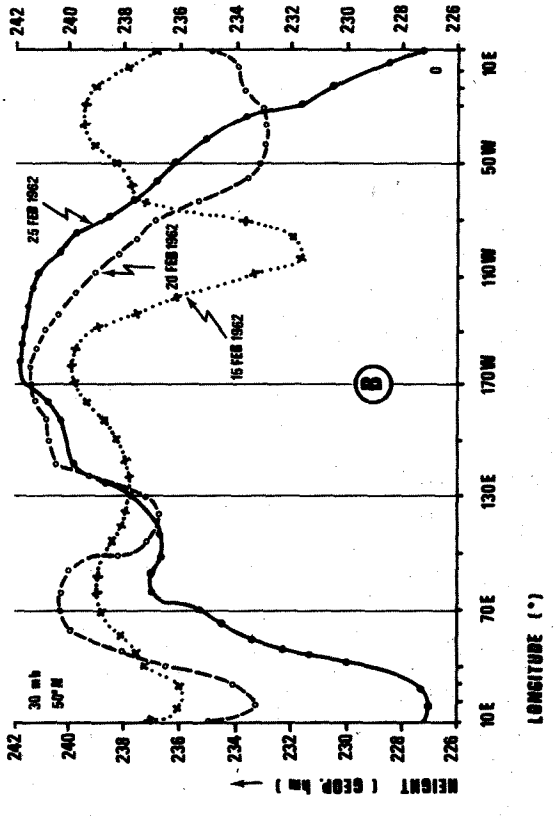


Fig. 7 – The polar vortex (from Labitzke) in geopotencial decameters, the associated stratospheric jet stream, (A), and the variation of the zonal index at the same level along 50°N, (B)

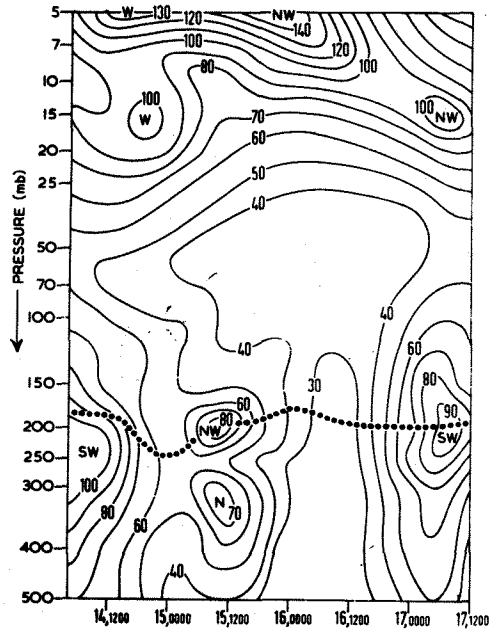


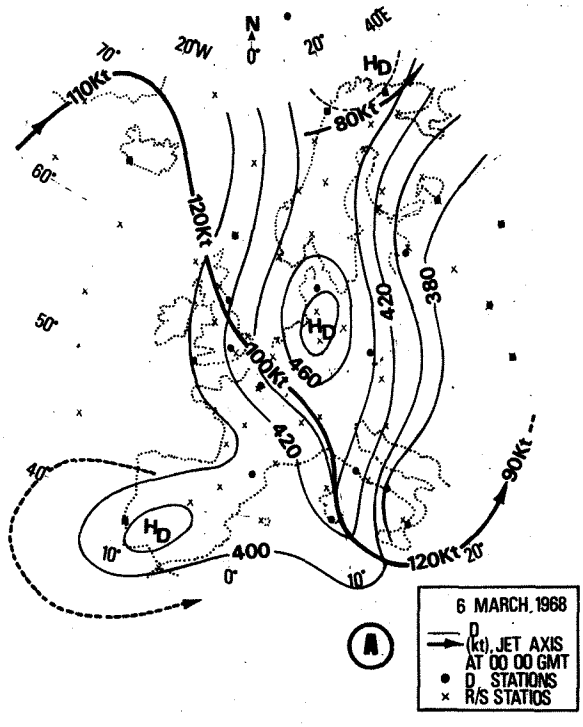
Fig. 8 – Time-height series of wind speed (kt) over Zaragoza in January, 1962 (From Figueira, 1965)

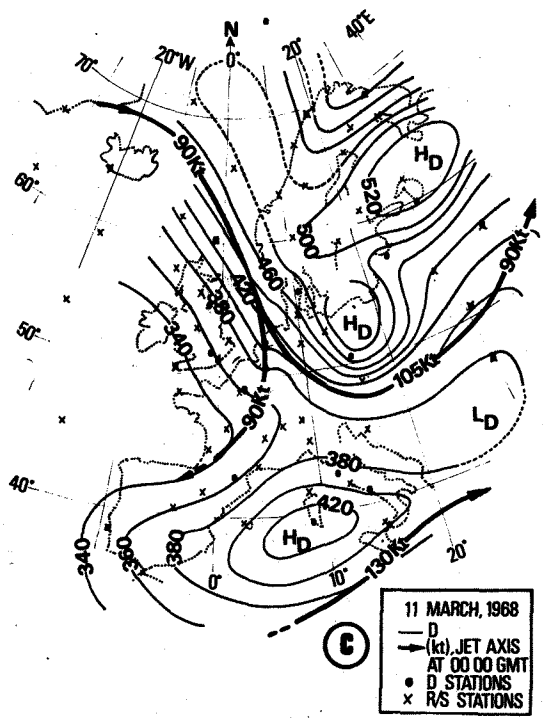
are obviously linked to them. Theoretically, one only needs to write down and, solve the appropriate balance equation for the componente X, which must include its substancial time variation, diffusion, and sources and sinks, that is, the properties of the environment and of the component itself. It seems correct to state that in a real atmosphere the circulation is composed by the motion field and the associated fields of the remaining meteorological variables always in a state of redistribuition, including of course natural and artificial composition.

If a erosol-like behaviour may be assumed, as for ozone, than in regions far enough of the main formation and destrution regions, the fluxes of the entity may be computed with reasonable confidence provided the wind field and the concentration of the component are known. Given the difficulties to directly measure such fields as that of the vertical velocities and of the concentrations of the products in the free atmosphere namely on a routine basis, it is very useful to turn now the reader's attention to some case-studies where observed atmospheric ozone is related to observed atmospheric flow field features, namely the tropospheric jet stream location and the baroclinic zones associated to it.

In an attempt to synoptically relate the tropospheric jet complex to the field of total ozone observed from ground based equipment (14) found the ocurrence of very significant gradients of total ozone along and across the more likely location of the jet axis as illustrated in figure 9, A to C, where the main centers of high total ozone are located within a few hundreds of kilometers to the left of cyclonically curved jets, looking downstream. Tacking into account that the main source of atmospheric ozone is located at layers above the tropospheric jet complex, this suggests the ocurrence of important downwards atmospheric transport in those areas, which this analysis could not prove, but its consistence with the results from (1) is reasonable. In addition, previous work based on potential temperature, vorticity, and artificial radioactivity pointed toward the existence of gaps in the propopause layer to the left of the jet axis, linked to a baroclinic zone slopping downwards to the lower troposphere beneath the jet core, so that it would be expected to find ozone rich air, that is, ozone content with stratospheric concencrations, inside such baroclinic layers. This guessing could be consistently proved by direct observations, and two of this case-studies will be now briefly referred to.

During the Rockwell Polar Flight (16) with departure from Honolulu at 15 05 54 G.C.T. Nov 1965 via North Pole, London, Lisbon, Buenos Aires, South Pole, Christchurch, and arrival at Honolulu at 17 20 22 G.C.T. Nov 1965, direct measurements of atmospheric ozone were made based on the IK reaction principle. The tropopause layer was crossed four times according to flight reports, and this provided opportunity for almost synoptically direct observations of wind field and ozone concentration to be made in the troposphere and in the lower stratosphere, including in this case three times the jet stream region, but never the baroclinic zone beneath the jet core. Figures 10, A and B show the vertical profiles of the indicated parameters, and it is worth noting that figure 10A refers to North Hemisphere autumn when ozone is a minimum (see figure 3), and figure 10B refers to





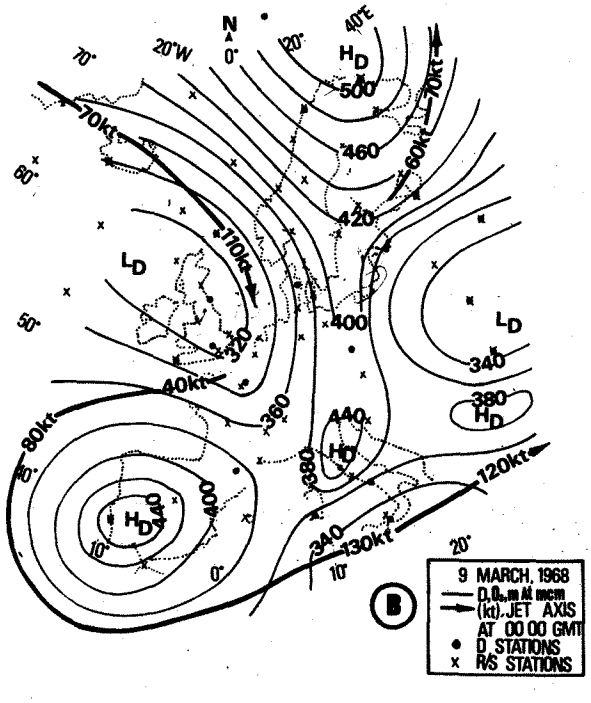


Fig. 9A, B and C – Total ozone in relation to jets (From Figueira, 1973)

South Hemisphere spring when it is a maximum, however lower than the North Hemisphere spring maximum. It is evident from figure 10A that ozone concentrations in the autumn stratosphere were 4 to 6 times higher than in the troposphere , and the highest value of around 110pphm by volume was observed in the spring stratosphere some 500km to the left of the main tropospheric jet axis located around 45ºS (see figure 10B). The results do not permit to read ozone gradients, neither across nor along the jets, not even for the jet located around 50ºN in figure 10A because ozone sampling was partly made in the troposphere and partly made in the stratosphere, and this leaves no possibilities of comparaison with figure 9 were total, not layered ozone is shown. However, latitudinal ozone gradients do exist.

Another aspect of the results of figure 10 discussed by the authors [16] is concerned with the relation between the inside and outside cabin ozone concentration, when they stressed that the level of 30pphm in the ventilating air during nearly all the

time flight in the stratosphere "represents a level which may concerne health officials if observed in surface air". However, they were sampling air in the lower stratosphere (14 km), and it is known from observation that the ozone maximum at around 20 to 25km may be several times its value in the lower stratosphere, depending on latitude and season (see figure 2), which may rise an interesting problem as regards advanced SST.

Another well-documented case of direct measurement of both tropospheric and stratospheric ozone in relation to jet stream location and tropopause folding is provided by [5]. The measurements were mada in April 1975 within the objectives of Project DUSTORM aboard the Electra aircraft and a commercial aircraft, flying perpendicular to the wind from Dever toward Oklahoma City. Observed winds and ozone are shown in figure 11 A for the tropospheric flight, revealing the hight ozone content inside the baroclinic boundary layer between the cold and the warm air which slopes downard beneath the jet core, and in figure 11 B the tropopause folding

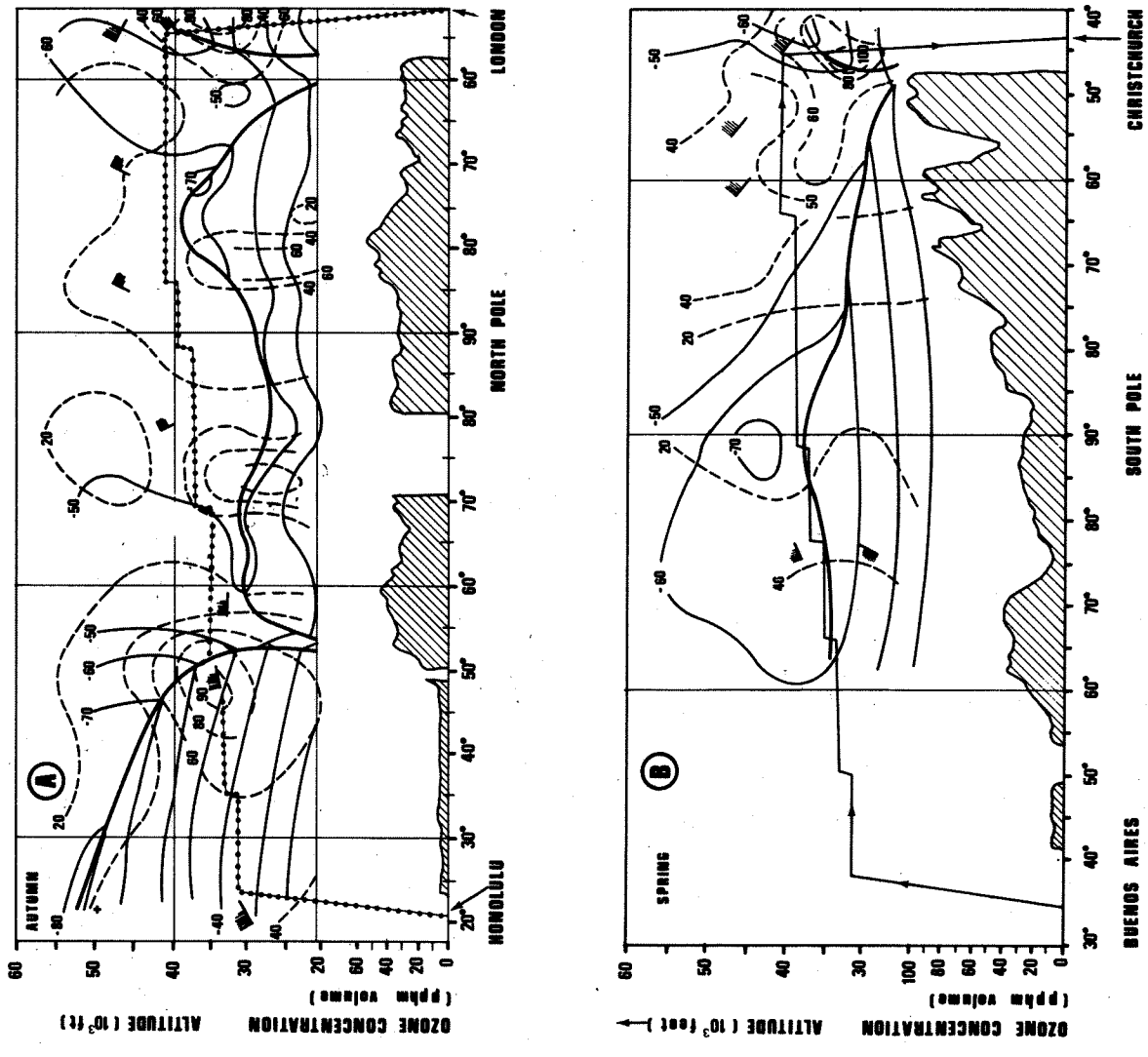


Fig. 10A and B – Meteorological and ozone cross-sections, including isotachs (---kt), isotherms (—°C), tropopause and frontal surfaces (⟶), and ozone in the lower part (simplified from Machta and others, 1969).

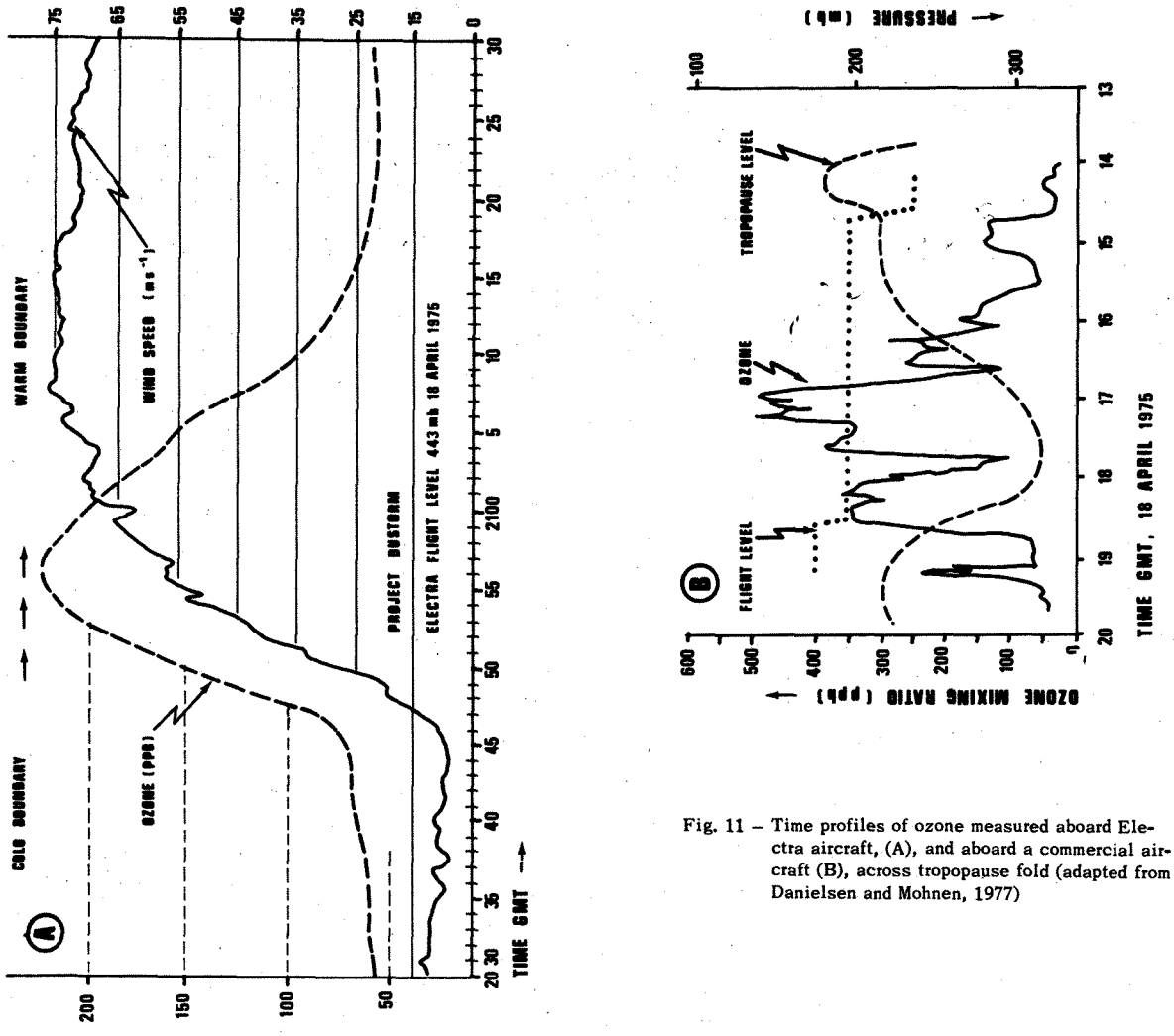


Fig. 11 – Time profiles of ozone measured aboard Electra aircraft, (A), and aboard a commercial aircraft (B), across tropopause fold (adapted from Danielsen and Mohnen, 1977)

and the stratospheric ozone mixing ratio are indicated for the stratospheric flight within the same weather system. It is pointed out that these observations were made during the North Hemisphere spring when the ozone is a maximum, in contrast to figure 10 A, and it is remarkable that stratospheric ozone levels are comparable in both cases, against the expectations, which may be explained, at least in part, to conditions above the tropopause and in part to instrumental differences. In both cases however the highest ozone mixing ratio appeared linked to the jet stream layer and attained values significantly higher than the 80ppb specified by the Environmental Protection Agency as an upper limit in air if it extends over an hour, thus the main conclusion seems to be that potential hazard to humans may exist at occasions due to atmospheric ozone. Finally a remark is made concerning the controversy between the likely relation between inside and outside cabin ozone content. Some workers [2,3] advanced that around 75% of the atmospheric ozone may be destroyed on its passage the through the aircraft compressors and ventilation system, but this bears a certain degree of uncertainity, as for the advanced 50% destruction indicated by [16]. On the other hand, almost synoptic comparaison [8] with ozonesonde observations led to the conclusion that no appreciable reduction is observed, the main sink of inside cabin ozone being due to smoke from cigarettes.

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