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ΠΡΟΕΔΡΙΑ ΝΙΚΟΛΑΟΥ ΜΑΤΣΑΝΙΩΤΗ

ΦΥΣΙΚΗ. — **Measurements of the background concentrations of air pollutants and aerosols (PAUR experiment)**, by *P. Theocaris, C. Zerefos, A. Hofzumahaus, A. Bais, I. Ziomas, D. Balis, K. Kourtidis, A. Papayannis, E. Kosmidis, C. Repapis, I. Isaksen, A. Krauss, M. Blumthaler, P. Suppan, P. Fabian, J. Pommereau**.

ABSTRACT

During the period 1.-20. June 1996, an extensive measurement campaign took place in Greece with the aim of studying the interrelationships between total ozone, UV-B radiation, aerosol load, air pollutants, photodissociation rates of NO_2 and O_3 and tropospheric ozone. This campaign was organized in the frame of the EU-funded Photochemical Activity and solar Ultraviolet Radiation (PAUR) project, which is part of the EU Environment and Climate Program. A wide range of atmospheric radiation, composition and meteorological parameters has been measured in a number of sites during the campaign. The present paper presents the main results from this extensive study of the atmosphere over Eastern Mediterranean.

INTRODUCTION

The main objective of the PAUR project was to investigate how increased penetration of UV-B solar radiation through the atmosphere, resulting from stratospheric ozone depletion, affects photochemical production and chemi-

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cal transformation of ozone and other photochemically active species in the lower atmospheric layers.

The background for the project followed from the WMO Ozone Assessments and other studies which have shown that northern middle and high latitude ozone has declined significantly during the last two decades (e.g. Stolarski, 1992). At that time the chlorine and bromine loading of the stratosphere had been increasing due to human activities, and there is increasing scientific evidence that the decrease in ozone is linked to enhanced halogen levels. The long term trends for the 1970's and 1980's during the winter and spring season were found to be $(-3.1 \pm 0.4)\%$ per decade over North America and Europe and slightly less over Asia. During the first years of the 1990's the ozone decline accelerated, reaching exceptionally low ozone column densities during winter/spring seasons of 1992, 1993, 1995, 1996 and 1997 (Bojkov et al., 1993; Gleason et al., 1993; Bojkov et al., 1995).

Observations of the UV-B (290-320 mm) in Thessaloniki in the past 4 years showed significant increases in UV-B solar radiation concurrent with the reduced columnar ozone densities (Zerefos et al., 1995 a, b). Model studies of UV-B changes in the troposphere for the same period, using observed reductions in ozone column densities predict significant increases in the photochemically active UV-B radiation (e.g. Madronich and Granier, 1992; Fuglestvedt et al., 1994). The enhanced UV-B solar radiation results to higher photodissociation rates of key trace species which control the chemical reactivity of the troposphere. Moreover, recent studies have shown that enhancement of UV-B radiation will have totally different impact on atmospheric chemistry whether they occur in polluted urban regions, or in prestine background areas (Tang and Madronich, 1994; Fuglestvedt et al., 1994). Indeed, observations and model studies show that in the less polluted background regions of the atmosphere, enhanced UV-B radiation levels will lead to enhanced photochemical activity which reduces ozone levels (e.g. Fuglestvedt et al., 1994), while enhancement of UV-B radiation in polluted urban and regional areas will lead to enhanced photochemical production of ozone and add to the burden of pollution in areas which already are strongly affected by photochemical pollution. Trends in photodissociation rate coefficients for tropospheric ozone have been estimated from satellite total ozone measurements to lie between about +0.3 and +0.4 percent per year (Tang and Madronich, 1994). These trends are expected to significantly alter the tropospheric chemical

composition particularly in the sunny regions of the earth. Of course penetration of UV-B to the lower atmosphere is extremely complicated and the ways that air pollution is interfering with the observed UV-B fluxes at ground level are not yet fully understood (Bais et al., 1993; Zerefos et al., 1995a) although tropospheric aerosols do attenuate solar irradiances (Seckmeyer and McKenzie, 1992; Blumthaler, 1993; Blumthaler et al., 1994). Modelling of UV-B fluxes through urban and rural conditions in variable environments (clouds, albedo, composition, aerosols etc.) is one of today's unresolved formidable tasks.

The overall PAUR project has been set up to study the different steps in a sequence going from reduced stratospheric ozone through increased photochemical activity in the lower atmosphere to the evaluation of the impact on regulations which aim at reducing ozone levels in polluted regions. The project aimed at increasing our understanding of how ozone and other photochemical pollutants are affected by variations and changes in UV-B fluxes.

SITES AND INSTRUMENTATION

As part of the PAUR project, an extensive measurement campaign termed «Olympian» took place in Greece during the period 1.-20. June 1996. During the campaign ambient gas analysers, gas chromatographs, DOAS systems, a LIDAR system, meteorological masts and balloon systems were deployed in several preselected locations. The main sites, where full sets of instrumentation have been deployed were the island of Aghios Efstratios and the suburban site of Tatoi in Athens. The existing Athens monitoring network of the Ministry of Environment, Land Use and Public Works provided also its data (air quality and meteorological parameters). Other sites included Varnavas (a small village 40 km north of Athens at a hill top close to the north coast of Attiki, where the composition of the air during Etesian wind episodes was expected to be characteristic of that of the Aegean sea), Faliro (on the shore line between the cities of Athens and Piraeus, where the air composition was expected to be characteristic of the plume of the city by Etesian wind and of the air returned by the sea breeze from the bay of Athens during the afternoon in case of light synoptic north wind associated with pollution episodes) and Pertouli (a forest site away from pollution sources at the Pindos mountain range in north-central Greece at 800 m ASL elevation that is often exposed in free tropospheric air).

Radiosonde data were provided by the Hellenic National Meteorological Service, together with a number of other meteorological parameters. Radiosondes were launched twice a day (00 and 12 GMT) from the Hellinikon Airport in Athens. Ozonesondes were further launched by LAP in Thessaloniki for a number of days.

The experimental approach was completed by four flights of an instrumented aircraft, flying over the Aegean at different altitudes in the range 0-12 km ASL and continuously measuring ozone and spectrally resolved radiation.

The following institutions participated in the experimental campaign:

- LAP-Laboratory of Atmospheric Physics, Aristotle University of Thessaloniki, (GR)
- UMUN-Lehrstuehl fuer Bioklimatologie und Immissionsforschung, University of Munich, (D)
- CNRS-Service d'Aéronomie du CNRS, (F)
- MetCon-Meteorologie Consult GmbH (D)
- UMS-University of Innsbruck (A)
- PERPA-Ministry of Environment, Land Use and PublicWorks, (GR)
- NTUA-National Technical University of Athens, (GR)
- AcA-Research Centre for Atmospheric Physics and Climatology, Academy of Athens, (GR)
- EMY-Hellenic National Meteorological Service, (GR)
- KFA-Kernforschungsanlage Juelich (D)

RESULTS AND DISCUSSION

The meteorological situation during the first 10 days of June was dominated by a high pressure system over Northern Europe with a ridge extending towards the Balkan. This gave a circulation pattern with air being transported from Scandinavia over eastern Europe, the Black Sea and finally over Turkey before entering the Aegean. Later the high pressure system moved towards the British isles and a more direct flow pattern from Eastern Europe towards the Aegean was established. The establishment of the Etesians during the campaign is also demonstrated in the calculated backtrajectories. Completely clear skies were observed during a major part of the experiment above the investigation area, and no precipitation events took place.

Total ozone exhibited little variation during the period of the campaign over Greece. Values ranged from around 310 D.U. to around 350 D.U. (Fig. 1). Figure 1 shows that a diurnal variation in total ozone was observed. At all three sites where total ozone measurements were made (Thessaloniki, Aghios Efstratios, Tatoi) this feature was evident, and was observable with both the Brewer and the Bentham instruments. This diurnal variation in total ozone, might induce an additional rhythm in UV-B fluxes. The possibility of a diurnal variation in total ozone caused by diurnal variations of the tropospheric ozone column is currently under investigation by LAP.

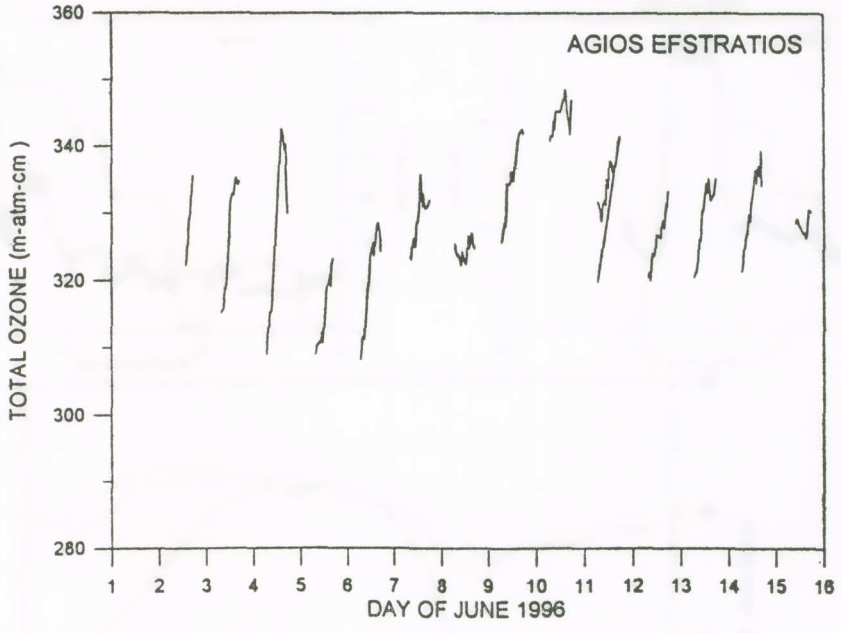

Fig. 1. Brewer total ozone measurements at Aghios Efstratios.

Ozonesonde ascents, ground-based and aircraft measurements show that high background ozone values observed at ground level during the period of the Etesian winds at some sites in Greece (Kourtidis et al., 1997) persist throughout the troposphere above the whole North-eastern Mediterranean (Fig. 2). The background concentrations of air pollutants (nitrogen oxides and hydrocarbons) is presented in Fig. 3 and shows that the air both over the Aegean and at Tatoi exhibited very low concentrations of the main pollutants.

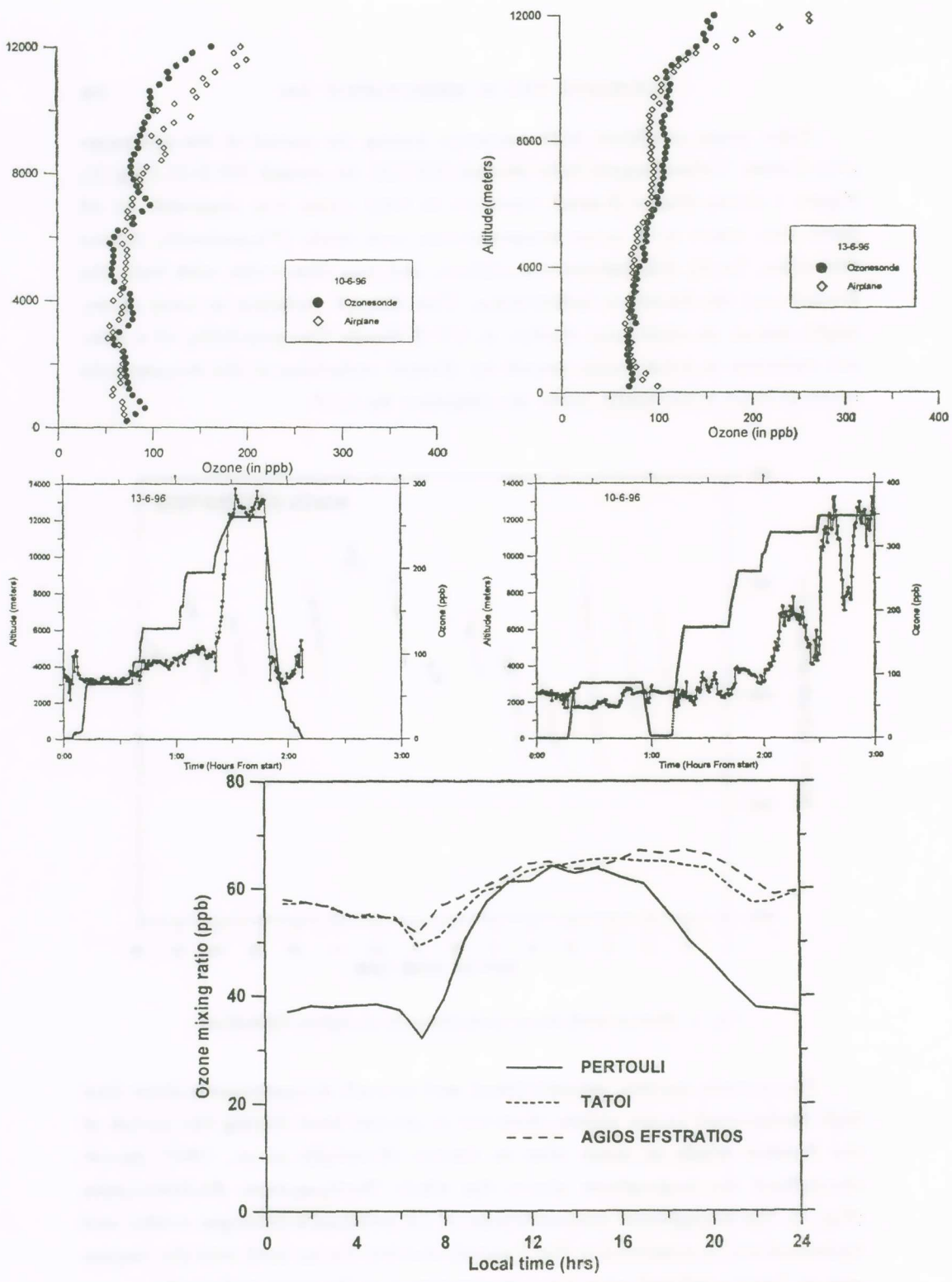

Fig. 2. Background ozone values over Greece. Upper panels: Measurements of tropospheric ozone with ozonesondes (Thessaloniki) and the FALCON aircraft (Athens-Thessaloniki flight path). Middle panels: Aircraft ozone measurements and their variability at different altitude levels of the flight path. Lower panel: Mean diurnal variation of ground ozone during PAUR at three sites.

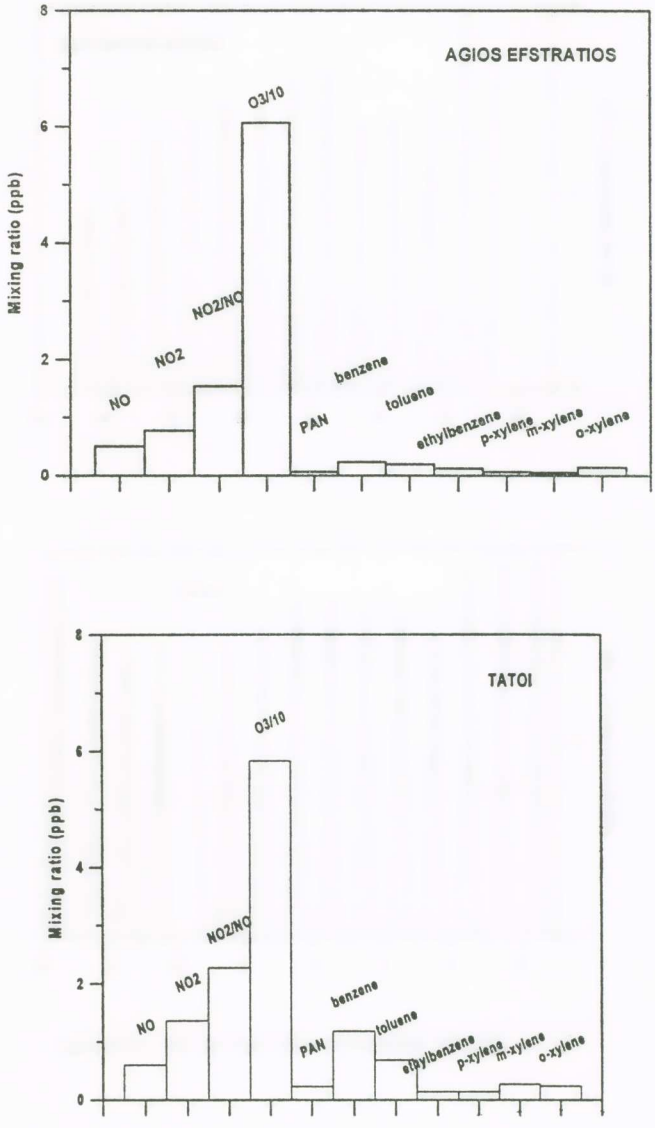

Fig. 3. Campaign mean ozone and precursor gases fingerprints at Aghios Efstratios and Tatoi. Note that ozone values are divided by 10 to fit in the range of the figure.

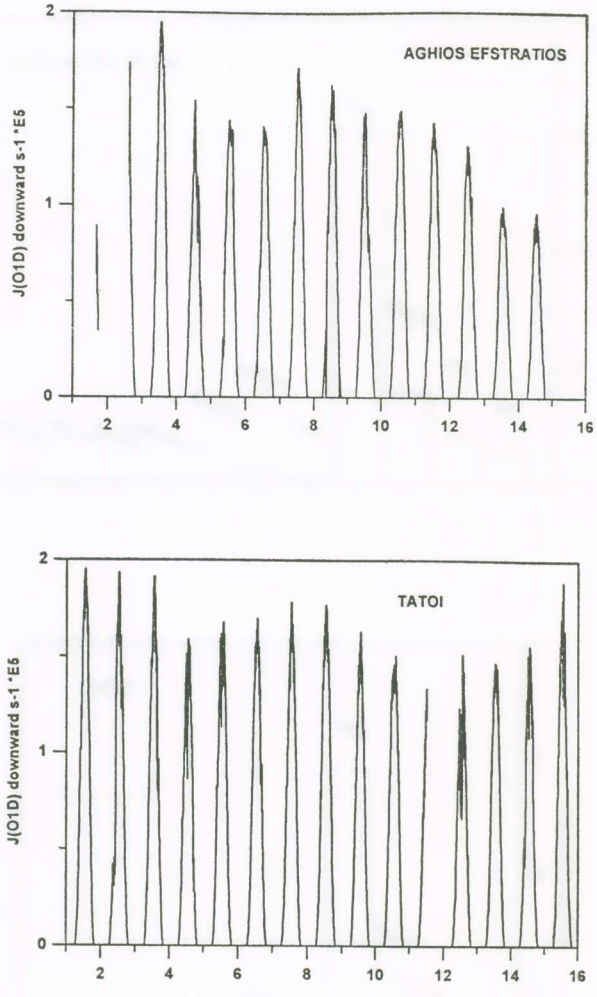

Fig. 4. J(O1D) measurements during the campaign.

The main results from these measurements can be summarised as follows:

Measurements of photolysis rates of ozone, J-O^1D are presented in Fig. 4. During the campaign period, the day-to-day variability of the J-O^1D daily maxima was around 45% while the one of J-NO_2 daily maxima was 8%. The

variability in the solar direct UV-B irradiance was strongly correlated with the total ozone variability, while the variability in global UV-B irradiance seemed to be more affected by changes in the aerosol loading. Tropospheric vertical aerosol optical property measurements were performed with a LIDAR system at the island of Aghios Efstratios. Total aerosol optical depth was measured at Tatoi, Thessaloniki and Aghios Efstratios with two Brewer and one Bentham instrument. During the period with intensive Etesian flow, aerosol optical depth was around 0.2. During periods with not very intensive Etesian flow, it increased up to 1. The day-to-day variability of aerosol optical depth was an order of magnitude, while the daily variability as determined from multiple measurements during one day, was up to a factor of 2.5.

As mentioned above, the approach of PAUR was complemented by aircraft measurements of $J(O^1D)$ and spectral radiation as part of a joint PAUR - ATOP venture. The aircraft performed 4 flights (on the 10, 11 and 13.6.1998) where $J(O^1D)$ and UV spectral solar radiation was measured together with tropospheric ozone. These measurements showed that the actinic flux increases by a factor of 1.3-2.2 between 0-12 km. Further, multiple scattering and

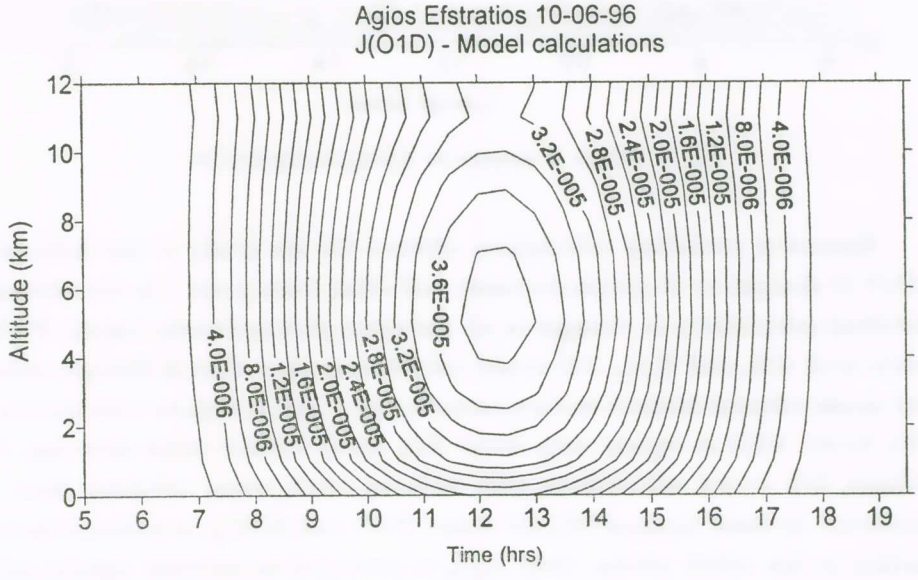

Fig. 5. Model calculations of the diurnal variation of $J(O^1D)$ in the troposphere for the 10th of June 1996, Aghios Efstratios.

absorption by ozone lead to significant differences in the shape of UV-A and UV-B vertical profiles. Aerosols influence considerably the vertical profile of ozone and NO2 photolysis frequencies (Figs. 5 and 6).

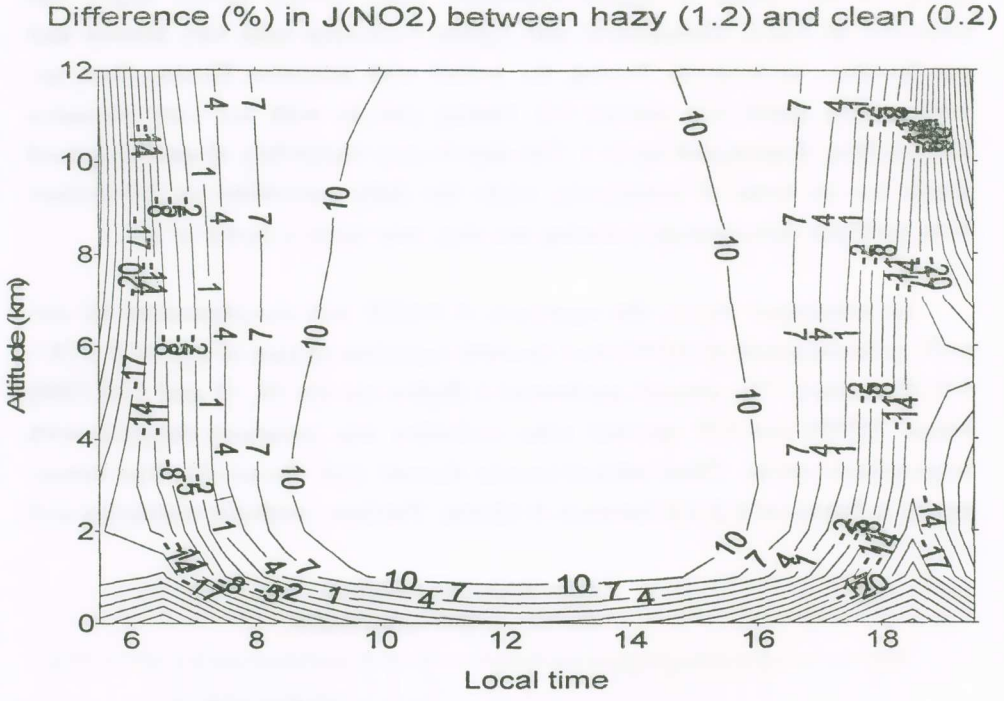

Fig. 6. Effects of aerosols on $J(NO_2)$ during PAUR.

Extensive modelling calculations allowed for the study of the influence of UV-B changes on tropospheric ozone and other trace gases. The simulations involved calculations in changes in all the main photooxidants: Ozone, PAN, nitric acid, OH, and H_2O_2. 3-D model calculations show that in Europe, reduced ozone columns increase surface ozone by up 4 ppb in regions with elevated NO_x levels, while in regions with lower NO_x levels surface ozone decreases. In Athens, 3-D model calculations with reduced total ozone columns show a moderate increase (around 19% for ozone, PAN and HNO_3) in photooxidants, mainly in the urban plume. Only H_2O_2 is predicted to increase significantly (up to 40%). The predicted significant increases in H_2O_2 in the urban plume of Athens imply that there will be an enhancement in SO_2 to H_2SO_4 conversion,

and hence precipitation acidity, due to decreasing ozone columns. This effect deserves some further study. Increases in precipitation acidity due to UV-B induced increases in atmospheric H_2O_2 have been postulated earlier (Sakugawa et al., 1990) and recent data suggest that total ozone decline might have already affected H_2O_2 concentrations (Anklin and Bales, 1997). Box model calculations show that less stratospheric ozone leads in most cases to a higher abundance of oxidant species like ozone, PAN, HNO_3 and H_2O_2 in the boundary layer. Only under very clean conditions ozone might be reduced.

CONCLUSIONS

A comprehensive picture of the background composition of the summertime atmosphere in the Mediterranean region has been obtained. Of particular importance is the establishment of background boundary layer summertime NO_x and VOC levels over the Aegean due to their critical influence on ozone modelling results. Equally important is the confirmation of high summertime background ozone values in the region, throughout the troposphere. Further, changes in the chemical composition of air during travelling from the Varnavas site at the Northern part of Athens basin to the Faliro site at the coast have been quantified. Additionally, measured and modelled contribution to the NMHC burden from biogenic hydrocarbons indicate that the topic of biogenic NMHC emissions deserves further attention.

Further, effects of different aerosol properties in UV-transmittance have been clarified. For the campaign period changes in the aerosol loading is found to cause larger variations in the UV irradiance than changes in the total ozone. Compared to an aerosol free sky, the aerosol loading during PAUR reduced the UVB irradiance by 0.65-0.95. The spectral measurements of global UV irradiance show agreement with model calculations within $\pm 8\%$. The agreement between modelled and measured $J(NO_2)$ values is very good, within $\pm 5\%$, while the agreement between modelled and measured $J(O^1D)$ values is at the moment less satisfactory, pointing to misrepresentation of atmospheric properties in the spectral region of the photodissociation of ozone. The aerosol single scattering albedo was found to have a large effect on the model / measurement ratios. As it appears from the modelling studies, information on the aerosol composition is of great importance for the impact studies of aerosol to the UV transmittance. The actinic flux increases by a factor of 1.3-2.2

between 0-12 km, depending on the wavelength. Multiple scattering and absorption by ozone lead to significant differences in the shape of UV-A and UV-B vertical profiles. The shape of the $J(O^1D)$ profile, which was also reproduced by modelling, is strongly affected by the temperature dependence of the $O(^1D)$ quantum yield $\varphi_{O(1D)}$.

Measurements showed a diurnal variation in total ozone column. If ongoing investigations confirm the observed diurnal variation, this could potentially be due to the buildup of tropospheric ozone during the day and present a negative feedback of the troposphere to increased UV penetration that is presently unquantified. This diurnal variation could also induce an additional rhythm in processes controlled by UV-B fluxes.

In Europe, reduced ozone columns increase surface ozone by up 4 ppb in regions with elevated NO_x levels, while in regions with lower NO_x levels surface ozone decreases. In Athens, model calculations with reduced total ozone columns show a moderate increase in photooxidants. Only H_2O_2 is predicted to increase significantly (up to 40%). Presently, it appears that the continued total ozone decline will enhance, in cases significantly, ozone pollution episodes. However, for these results to have validity and applicability to abatement policies, more modelling should be performed that would also study combined effects of $J(NO_2)$ changes, $J(O^1D)$ changes, future projected changes in the fractionation of NMHCs in traffic emissions and future projected temperature changes due to greenhouse warming and their effects on water vapour and biogenic NMHC emissions. The effects of predicted H_2O_2 increases on the acidity of precipitation need some further study.

ΠΕΡΙΛΗΨΗ

Προκαταρτικὰ Ἀποτελέσματα τῶν Ἀεροπορικῶν καὶ Ἐπίγειων Μετρήσεων Πεδίου στὴν Ἀτμόσφαιρα τοῦ Ἑλλαδικοῦ Χώρου

Τὴν περίοδο ἀπὸ 1 ἕως καὶ 20 Ἰουνίου 1996, πραγματοποιήθηκε ἕνα ἐξαιρετικὰ ἐκτεταμένο πείραμα ποὺ κάλυψε ὅλη τὴν ἀτμόσφαιρα τῆς χώρας μας μὲ μετρήσεις τῆς σύστασής της, σὲ ὅλη της πρακτικὰ τὴν ἔκταση ἀπὸ τὸ ἔδαφος μέχρι τὴ στρατόσφαιρα. Οἱ μετρήσεις αὐτὲς πραγματοποιήθηκαν ἀπὸ ἕνα ἐπιτελεῖο περίπου 60 ἐπι-

στημόνων οἱ ὁποῖοι ἀνέλυσαν τὴν ποιότητα τῆς ἀτμόσφαιρας τόσο σὲ κατοικημένες περιοχὲς ὅσο καὶ σὲ περιοχὲς ἀπομακρυσμένες.

Οἱ μετρήσεις ἔγιναν τόσο στὸ ἔδαφος ὅσο καὶ στὴν ἐλεύθερη ἀτμόσφαιρα. Στὴν ἐλεύθερη ἀτμόσφαιρα οἱ μετρήσεις πραγματοποιήθηκαν μὲ ἀερόστατα, μὲ εἰδικὰ ἐξοπλισμένο ἀεροσκάφος, μὲ Laser Radar καθὼς καὶ μὲ εἰδικὰ κατασκευασμένους διπλοὺς μονοχρωμάτορες. Ἡ ἀναλυτικὴ περιγραφὴ τῶν ἐπιστημονικῶν ὀργάνων γίνεται στὸ κείμενο τῆς ἐργασίας.

Σχεδὸν σὲ ὁλόκληρη τὴν περίοδο πραγματοποιήσεως τοῦ ἐκτεταμένου αὐτοῦ πειράματος, οἱ μετεωρολογικὲς συνθῆκες ποὺ ἐπικράτησαν ἦταν ἰδιαίτερα εὐνοϊκὲς γιὰ τὸν καθορισμὸ τῶν συγκεντρώσεων τοῦ ὑποβάθρου στὴν ἀτμόσφαιρα τοῦ Ἑλλαδικοῦ χώρου. Πράγματι, ἐπικράτησαν κατὰ τὸ μεγαλύτερο διάστημα ἐτησίες ἄνεμοι οἱ ὁποῖοι μεταφέρουν, ὅπως ἀπεδείχθη, ἀκόμη καὶ στὴν Ἀθήνα ἀέρα ἀπὸ τὸ Αἰγαῖο πέλαγος ἐξαιρετικῶν χαρακτηριστικῶν ἀπὸ πλευρᾶς περιβαλλοντικῆς ποιότητας. Τὰ συμπεράσματα αὐτῆς τῆς μελέτης τοῦ ὑποβάθρου, ἡ ὁποία ἂς σημειωθεῖ εἶναι ἡ πρώτη ἐκτενὴς μελέτη καθορισμοῦ τοῦ ὑποβάθρου στὴν ἀτμόσφαιρα ὄχι μόνο τῆς Ἑλλάδας, ἀλλὰ καὶ διεθνῶς, εἶναι μοναδικά. Αὐτὴ ἡ μοναδικότητα ὀφείλεται στὴ μεγάλη λεπτομέρεια καταγραφῆς καὶ στὴν ἔκταση (ἀπὸ τὸ ἔδαφος μέχρι τὴ στρατόσφαιρα) στὴν ὁποία πραγματοποιήθηκαν αὐτὲς οἱ ἐξειδικευμένες μετρήσεις.

Τὸ πρῶτο οὐσιαστικὰ συμπέρασμα τῆς μελέτης αὐτῆς ἦταν ὅτι ἡ καθ' ὕψος μεταβολὴ τῶν ρυθμῶν φωτολύσεως, ἀπὸ τὸ ἔδαφος μέχρι τὴ στρατόσφαιρα, παρουσιάζει μιὰ ἀπότομη μεταβολὴ στὸ πρῶτο χιλιόμετρο ἀνόδου μέσα στὴν ἀτμόσφαιρα, ἡ ὁποία ἔχει σὰν συνέπεια τὴν ἐνίσχυση τῶν φωτοχημικῶν διεργασιῶν στὴν ἐλεύθερη τροπόσφαιρα. Τὸ Σχῆμα 5 δείχνει ἀκριβῶς τοὺς ὑπολογιζόμενους μὲ θεωρητικὸ πρότυπο ρυθμοὺς φωτόλυσης καὶ ἐνδεικτικὰ τὶς ἀντίστοιχες ἀεροπορικὲς μετρήσεις στὸ Αἰγαῖο, ἀπὸ τὸ ἔδαφος μέχρι τὴ στρατόσφαιρα γιὰ τὶς 10/06/1996. Ἀντίστοιχα, στὸ Σχῆμα 2, φαίνονται οἱ ὑψηλὲς συγκεντρώσεις ὄζοντος ὅπως μετρήθηκαν στὸ ἔδαφος καὶ στὴν ἐλεύθερη ἀτμόσφαιρα μὲ εἰδικὸ ἀερόστατο (ὀζοντοβολίδα) καὶ εἰδικὸ ἀεροσκάφος στὸ βόρειο Αἰγαῖο. Εἶναι ἄξιο παρατηρήσεως ὅτι οἱ τιμὲς τῶν 55-65 μερῶν στὸ δισεκατομμύριο ὄζοντος εἶναι φαίνεται κοινὸ χαρακτηριστικὸ γιὰ τὸ ὑπόβαθρο αὐτοῦ τοῦ ἀερίου καὶ ὁπωσδήποτε εἶναι αὐξημένο σημαντικὰ σὲ σχέση μὲ ἄλλες περιοχὲς τοῦ βορείου ἡμισφαιρίου ὅπως προκύπτει ἀπὸ τὶς θεωρητικὲς ἐκτιμήσεις τοῦ μοντέλου τῆς Νορβηγικῆς Ἀκαδημίας Ἐπιστημῶν.

Ἐκτὸς ὅμως ἀπὸ ὁλόκληρη τὴ σειρὰ τῶν μετρήσεων τοῦ ὑποβάθρου τῶν ἄλλων ἀερίων ρύπων (π.χ. ὀξειδίων τοῦ ἀζώτου καὶ ὑδρογονανθράκων), οἱ ὁποῖοι παρουσιάζονται στὸ Σχῆμα 3, ἕνα ἀκόμη ἐνδιαφέρον συμπέρασμα αὐτῆς τῆς μελέτης ἦταν ἡ ἐξαιρετικὴ ποιότητα τῆς ἀτμόσφαιρας στὴ βόρεια Ἀττικὴ (περιοχὴ Τατοΐου καὶ

Βαρνάβα) σὲ συνθῆκες ἐπικράτησης βορείων ἀνέμων. Ὅπως φαίνεται καὶ στὸ Σχῆμα 2, ἡ σύγκριση μεταξὺ τῶν τιμῶν ὄζοντος στὴ βόρεια Ἀττικὴ καὶ στὸν Ἅγιο Εὐστράτιο σὲ ὅλη τὴ διάρκεια τοῦ πειράματος δείχνει μιὰ ἐκπληκτικὴ σύμπτωση. Ἀντίστοιχη σύμπτωση στὴν ποιότητα τοῦ ἀέρα προκύπτει καὶ ἔμμεσα μὲ τὴ σύγκριση τῆς ποιότητας τῆς ἀτμόσφαιρας σὲ ὁλόκληρη τὴ φασματικὴ περιοχὴ τῆς ὑπεριώδους ἡλιακῆς ἀκτινοβολίας. Ἡ σύγκριση αὐτὴ δείχει ὅτι τὰ ἀτμοσφαιρικὰ αἰωρήματα στὴ βόρεια Ἀττικὴ ἔχουν περίπου τὶς ἴδιες ὀπτικὲς ἰδιότητες μὲ ἐκεῖνα στὸν Ἅγιο Εὐστράτιο καὶ στὴν οὐσία σὲ ὅλη τὴ διάρκεια τοῦ πειράματος, ἡ ἴδια θόλωση στὴν ὑπεριώδη φασματικὴ περιοχὴ ἐπικρατοῦσε τόσο στὴ βόρεια Ἀττικὴ ὅσο καὶ στὸ βόρειο Αἰγαῖο. Τὸ γενικὸ συμπέρασμα ἀπὸ τὸ α′ μέρος αὐτῆς τῆς μελέτης εἶναι ὅτι ἐπικρατεῖ στὴν Ἑλλάδα κατὰ μέσον ὅρο τὸ ἴδιο ὑπόβαθρο ποιότητας τοῦ ἀτμοσφαιρικοῦ περιβάλλοντος σὲ μετεωρολογικὲς συνθῆκες μέτριων βορείων ἀνέμων στὸ Αἰγαῖο ποὺ ἐπικράτησαν σχεδὸν ἐπὶ τρεῖς συνεχεῖς ἑβδομάδες. Στὸ β′ μέρος γίνεται ἡ περιγραφὴ τῆς καταστάσεως στὴν Ἀττικὴ σὲ περίοδο ἐξασθένησης τῶν βορείων ἀνέμων (περιπτώσεις ἐμφανίσεως φωτοχημικοῦ «νέφους»).

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