

## STRATOSPHERIC OZONE FLUCTUATION AND ULTRAVIOLET RADIATION OVER SERBIA

by

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Increased ultraviolet radiation potentially detrimental to health is a direct effect of the reduced ozone layer of the Earth's atmosphere. Stratospheric ozone over the territory of Serbia and immediate surrounding depleted in the considered period of 1979-2008; its amount reduced by 7.6%. Solar and volcanic activities have significant effect on the interannual variability of the stratospheric ozone. Ozone layer over Serbia is 13.8% thicker than the planetary layer, with the ozone hole forming once in five years on average without serious health implications.

*Key words: stratospheric ozone, ultraviolet radiation, health, ozone hole, solar activity, volcanic activity*

### INTRODUCTION

About 90% of the Earth's atmospheric ozone is in the stratosphere, with the greatest concentration at altitudes between 22 and 25 km. Decreases of ozone concentration in the stratosphere, occurrence of ozone holes and increased ultraviolet radiation (UVR), on the Earth's surface have become focal problems of public interest. The ozone layer measured at the British Antarctic Survey (BAS) base has declined each September and October since 1977 at a rate of over 40% [1]. Scientific cause and consequence researches are becoming topical today.

A significant cause indicated by researchers in 1928 is chlorofluorocarbons (CFC). These compounds, chemically stable, not toxic or flammable, have found extensive application in industry, with the consequence of the increasing release of CFC in the atmosphere. While mixing with air molecules in general circulation of the atmosphere, CFC are gradually spreading over the globe [2] unchanged in the troposphere through an interval from 50 to 100 years [3].

CFC in the atmosphere slowly ascend upward in the stratosphere in which UVR disrupts chemical bonds of the compounds. Chlorine atoms released in the process destroy stratospheric ozone through a catalytic reaction; each released chlorine atom destroys about 100,000 ozone molecules.

BAS measurements indicated unexpected ozone depletion in the Antarctic springtime, so severe that computers receiving data from satellite Nimbus 7 often rejected the ozone amounts as the measurement errors. A particular concern of the scientific public is the annual depletion of ozone gradually approaching the southern tip of South America [4].

An international convention in 1985, known as the Vienna Convention, is aimed at protecting the ozone layer from harmful man-made substances. After the experimental evidence that CFC are great destroyers of stratospheric ozone molecule [5], the Montreal Protocol has been signed for restricted emission of substances harmful to ozone. These international motions stand for reduction of CFC emission which has been significantly reduced since the reference year 1989 [6].

Satellite measurements indubitably indicated interrelation between the state of the ozone layer and UVR. Increased solar UVR over the Earth's surface is a result of the stratospheric ozone depletion. The effects of UVR on health are variable. Exposure to UVR has a beneficial effect on human organism in that it activates skin protective mechanisms and provides for conversion of 7-dehydrocholesterol into vitamin D3. In contrast, UVR can be detrimental to human health in exposures to its cancerous, genotoxic, mutagenic, and immunotoxic effects. Summarizing the research results the British National Radiological Protection Board [7] report reads: "*there is convincing evidence*

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that UVR can cause damage to DNA and in animal experiments it has been shown to be a cause of cancer. Exposure to UVR also increases the risk of skin cancer in man and produces other undesirable health effects. The main tissues of the human body affected are those of the skin and the eye. There are also effects on the immune system, the significance of which for human health is not yet clear. Excess sun exposure can increase the risk of both non-melanoma and melanoma skin cancers, the latter being the main cause of skin cancer death. Skin cancers cause about 2000 deaths each year in Britain, which is about 1.4% of all cancer deaths" (DNA in the quotation denotes deoxyribonucleic acid).

Research data for the Republic of Serbia [8] include records of 13475 male and 12187 female new patients affected with cancer in central Serbia, where males were mainly affected by bronchopulmonary, colon, and rectum cancer, while females were mainly affected by breast and cervical cancer. Of the total number of patients with cancer for the observed year, skin cancer has affected 10.9% of male population, and 11.1% of female population. Skin cancer caused death of about 0.4% of the affected males and around 0.3% of the affected females out of all cancer deaths, or about 4.0% of males and around 2.7% of females affected by skin cancer.

In the context of the beneficial UVR, UVR is used in curing rickets, psoriasis, eczema, and some forms of jaundice. In small doses, it is necessary for vitamin D and calciferol synthesis [9].

In agreement with the Commission International de l'Eclairage definition (CIE), UVR spectrum is divided into three wavelength ranges [10]: UVC (100-280 nm), UVB (280-315 nm), and UVA (315-400 nm), where nm denotes nanometer ( $10^{-9}$ m). The limits between the divisions are empirical. Solar UVR of 280 nm and longer wavelengths (UVA and UVB) reaches the Earth's surface, but the rest of the UVR spectrum (UVC) is selectively absorbed by the atmosphere [11]. There is a small probability that wavelengths shorter than 280 nm reach the Earth's surface, because stratospheric ozone fully absorbs UVC and most of UVB component. Thus, UVR at the ground level consists of high UVA and very low UVB radiation which is biologically most effective. In divisions associated with biological effect, the limit is 290 nm, because wavelengths shorter than 290 nm are believed to cause sudden death of live cells [12]. The upper UVB boundary at 315 nm is rather arbitrary with relation to the microbiological effect of UVB radiation. Recent photomicrobiological researches show that radiation wavelengths in excess of 315 nm are photobiologically effective so that the UVB limit at 330 or 340 nm would be more adequate than the present limit. UVB radiation has highly deleterious effect. A large amount of it does not reach the Earth, which depends on the ozone depth.

This paper is concerned with the research, based on satellite measurements, of changes in stratospheric

ozone as an indirect cause of UVR variability – a significant factor of health risk over an arbitrary selected location which consists of Serbia and bordering areas of Southeast and Central Europe.

## METHODS OF RESEARCH

The first step of the research concerned the data base for ozone layer over the selected location. Satellite measurements from the last thirty years were used, made available to public by the National Aeronautics and Space Administration (NASA), the Goddard Space Flight Center – Distributed Active Archive Center. The processed data come from five satellites: Nimbus-7 – the Earth-observing satellites NASA and NOAA – The National Oceanic and Atmospheric Administration (1978-1993), Meteor-3 – Russian meteorological satellites (1991-1994), Japanese Earth resources satellites ADEOS – Advanced Earth Observing Satellite (1996-1997), Earth Probe (1996-2006), and OMI – the Ozone Monitoring Instrument (from 2004 to date). All above mentioned satellites used the same measurement technique – Backscattered UltraViolet (BUV) and instruments – Totale Ozone Mapping Spectrometers (TOMS).

The satellite data formed the main base and were most useful in the study of the phenomena characteristic of ozone in the atmosphere. Raw satellite data use an algorithm for processing in NASA/GSFC-DAAC before presentation [13]. Data used in this paper are the latest version 8 of an improved TOMS processing algorithm. The available database contains a textual file, the daily global records for each day of satellite measurements.

The problem first encountered in forming database was selection, for a territory over 10 GB in size, from over 50,000 daily records through thirty years of measurement. Another problem was the change of the satellite measurement resolution. Satellites Nimbus 7, Meteor 3, ADEOS, and Earth Probe made measurements at a predetermined network with resolution of  $1^\circ$  for geographic latitude and  $1.25^\circ$  for geographic longitude. Satellite OMI made measurements from 2004 at resolution of  $1^\circ$  for both latitude and longitude. The change in the network provided greater global data-coverage, but also shifted measuring sites of the new series of data in relation to the previous ones and additionally complicated the research.

This research covered Serbian domain, a rectangle including the territory of Serbia and respective extensions on the adjoining countries in agreement with the network of measurement sites. The two measurement networks are not essentially different for a small location as the Serbian domain. For both resolutions ( $1^\circ$   $1.25^\circ$  and  $1^\circ$   $1^\circ$ ) 24 sites were used for deduction of the amount of ozone over the selected site. The difference is only in the smaller number of network

points for Serbia at the resolution of  $1^\circ \times 1.25^\circ$  than at the resolution of  $1^\circ \times 1^\circ$  (due to the relative shift of the points with the change in the network resolution). The rest of measurement sites out of total 24 sites are located on the territory of neighboring countries in the designated domain, equally distributed on all sides.

A daily report on the measured amount of ozone is generated each day of the satellite observation. A section with the measurement information is in the beginning of the report. An example of the daily report section on ozone measurement for February 20, 1998, from the Earth Probe satellite is given in tab. 1. The same goes for the ozone-measuring satellites in function before it.

**Table 1. Daily report section on ozone measurements from Earth Probe satellite for February 20, 1998**

Day: 51 Feb. 20, 2998	Days in the year (51) and respective dates
EP/TOMS	Name of satellite (Earth Probe)/measuring instrument (TOMS)
NRT OZONE	Measured parameter (ozone)
GEN: 04.116 V8	Algorithm version of data generation (version 8)
ALECT: 12:00 AM	Time of measurement
Longitudes	288 bins centered on $179.375^\circ$ W to $179.375^\circ$ E (1.25 degree steps)
Latitudes	180 bins centered on $89.5^\circ$ S to $89.5^\circ$ N (1.00 degree steps)

The last two lines in the section are about the measurement sites of the satellite and data arrangement for subsequent measurement.

For the former network step  $1^\circ \times 1.25^\circ$ , the starting measurement site is at the latitude of  $-89.5^\circ$  (south) and the last at  $+89.5^\circ$  (north), or 180 measurement sites with half-degree centres on latitude. The data are divided in groups, with one group corresponding to a latitudinal zone, given in the end of the group (for example, lat.  $45.5$  corresponds to the measurements taken at latitude  $45.5^\circ$  north.). Data within each group correspond to  $1.25^\circ$  step site of the longitude, with the first site at  $-179.375^\circ$  (west) and last at  $+179.375^\circ$  (east), or a total of 288 longitudinal zones. Each group includes three-digit information on the amount of ozone in Dobson units (DU)<sup>1</sup>. The serial number of information is the longitude of the measurement site. The total number of sites in which the satellite scans the Earth's atmosphere with the given resolution and the total number of data in each daily report are obtained by multiplying the total longitudinal and

latitudinal measurement zones, or  $180 \times 288 = 51840$  measurements.

More recent measurements from OMI satellite have the resolution of  $1^\circ$  for latitude and  $1^\circ$  for longitude. Latitudinal measurement zones are the same as those of the previous satellites, between  $-89.5^\circ$  (south) and  $+89.5^\circ$  (north), or 180 zones centered at the half-degree of latitude. Data within each latitudinal zone correspond to the longitudinal zones which are subdivided by  $1^\circ$  step between  $-179.5^\circ$  (west) and  $+179.5^\circ$  (east), or a total of 360 longitudinal zones. A denser network provides more data. Daily reports from OMI satellite are similar to the reports from earlier satellite measurements. The total information on sites of the Earth's atmosphere scanned from OMI and the total number of data in each daily report from this satellite is obtained as a multiplication result of the total longitudinal and latitudinal measurement sites, or  $180 \times 360 = 64800$ .

The problem of data selection for the Serbian domain was solved by specific software that automatically selected relevant data from the textual files of both networks, and led to the final data base for the thirty-year measured ozone layer over Serbia. Manual selection of data from the daily reports was impossible, because it would take years. The database contains satellite measurements from November 1, 1998, to October 31, 2009, with a gap for 1995 and 1996.

Measurement sites for the Republic of Serbia domain are between latitudes  $41.5^\circ$  and  $46.5^\circ$  north and between longitudes  $18.125^\circ$  and  $23.125^\circ$  east, or a total of 24 measurement sites for the given resolution. The co-ordinates of the measurement sites (MS) for the measurement of ozone layer thickness from the Serbian domain for the resolutions of  $1^\circ \times 1.25^\circ$  and  $1^\circ \times 1^\circ$  are shown in tab. 2.

In order to obtain a sufficiently long series of data, the satellite records from resolution sites  $1^\circ \times 1.25^\circ$  in 1979-2004 were combined with the measurements from the sites  $1^\circ \times 1^\circ$  after 2004. There were minor discrepancies between the measurement sites of the two networks (fig. 1), but the continuity of the series was maintained. Net measurement sites with the resolution of  $1^\circ \times 1.25^\circ$  are shown in white color, while the measurement sites with the resolution of  $1^\circ \times 1^\circ$  are in black color. The error of combining two series was even negligible, because the distance between two sites was always less than 50 km (maximum 45 km for the easternmost sites beyond the Serbian territory).

The other step in the research is based on the deductions from the calculations of statistical parameters and statistical analysis.

<sup>1</sup> The Dobson unit [DU] is the unit measure for total ozone. A Dobson unit is defined as the ozone column  $10 \mu\text{m}$  thick at standard air temperature [ $0^\circ\text{C}$ ] and atmospheric pressure (1013.25 hPa). If you were to take all the ozone in a column of air stretching from the surface of the Earth to space, and bring all that ozone to standard temperature ( $0^\circ\text{C}$ ), and pressure (1013.25 hPa, or one atmosphere, or "atm"), the column would be about 0.3 centimeters thick. Thus, the total ozone would be 0.3 atm-cm. To make the units easier to work with, the "Dobson unit" is defined to be 0.001 atm-cm. Our 0.3 atm-cm would be 300 DU.

**Table 2. Coordinates of measurement sites for measurement of ozone layer thickness from Serbian domain**

MS <sup>1</sup>	Net resolution 1° 1.25		Net resolution 1° 1°	
	Geographical latitude	Geographical longitude	Geographical latitude	Geographical longitude
1	41°30'N <sup>2</sup>	20°37'30" E <sup>3</sup>	41°30'N	20°30'E
2	41°30'N	21°52'30" E	41°30'N	21°30'E
3	41°30'N	23°07'30" E	41°30'N	22°30'E
4	42°30'N	19°22'30" E	42°30'N	19°30'E
5	42°30'N	20°37'30" E	42°30'N	20°30'E
6	42°30'N	21°52'30" E	42°30'N	21°30'E
7	42°30'N	23°07'30" E	42°30'N	22°30'E
8	43°30'N	18°07'30" E	43°30'N	18°30'E
9	43°30'N	19°22'30" E	43°30'N	19°30'E
10	43°30'N	20°37'30" E	43°30'N	20°30'E
11	43°30'N	21°52'30" E	43°30'N	21°30'E
12	43°30'N	23°07'30" E	43°30'N	22°30'E
13	44°30'N	18°07'30" E	44°30'N	18°30'E
14	44°30'N	19°22'30" E	44°30'N	19°30'E
15	44°30'N	20°37'30" E	44°30'N	20°30'E
16	44°30'N	21°52'30" E	44°30'N	21°30'E
17	44°30'N	23°07'30" E	44°30'N	22°30'E
18	45°30'N	18°07'30" E	45°30'N	18°30'E
19	45°30'N	19°22'30" E	45°30'N	19°30'E
20	45°30'N	20°37'30" E	45°30'N	20°30'E
21	45°30'N	21°52'30" E	45°30'N	21°30'E
22	46°30'N	18°07'30" E	46°30'N	18°30'E
23	46°30'N	19°22'30" E	46°30'N	19°30'E
24	46°30'N	20°37'30" E	46°30'N	20°30'E

<sup>1</sup> Measurement site<sup>2</sup> Northern geographical latitude<sup>3</sup> Eastern geographical longitude

## RESULTS AND DISCUSSION

The average thickness of ozone measured in 1979-2008 at 24 sites over the research domain is 330 DU. The standard deviation in the same measuring period is 29.7 DU, maximum 528 DU, minimum 204 DU, and median 327.9 DU. Median thickness somewhat below the mean value indicates greater frequency of low values, but also more extreme high than low stratospheric ozone thickness, which is significant for UVR intensity on the ground level.

The mentioned statistical parameters are given in tab. 3.

**Figure 1. Ozone measurement sites over Serbian domain for both resolutions 1° 1.25° and 1° 1°**

Stratospheric ozone changed thickness over the territory of Serbia and immediate bordering area from 1979 to 2008 in a regular annual cycle. The minimum average thickness (287.7 BU) is in October. The mean value gradually increases to 371 DU in April and then decreases to October.

Pierson's correlation coefficient is high, mostly around 0.9, between the measurement sites. Correlation is the highest between two nearest sites, and the lowest between most distant longitudinal sites. This has been expected and agrees with the prevailing theory on the ozone latitudinal distribution [14]. The sample considered is representative, because the correlation on daily basis is calculated for a measurement period of 30 years. It leads to a confident conclusion that the amounts of ozone in all sites over Serbia are strongly interdependent. The high correlation between the measurement sites was expected for a small location like Serbia. The covariance of all sites is positive, very high, which corroborates the assumed phase of all

**Table 3. Statistical parameters of ozone layer thickness over the selected location (1979-2008)**

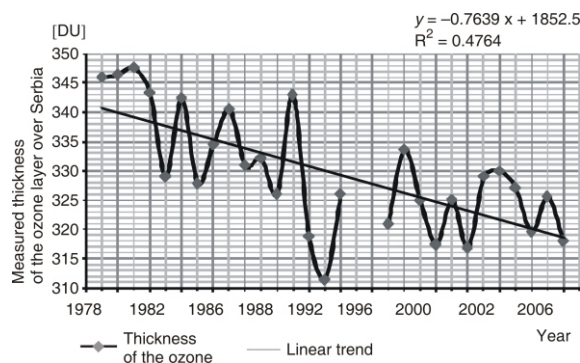
Months:	Jan.	Feb.	Mar.	Apr.	May	June	July	Avg.	Sept.	Okt.	Nov.	Dec.
Mean [DU]	334.6	357.3	365.6	371	356.9	342.8	325.1	312.4	303.3	287.7	291.6	311.8
Median [DU]	331	359	362	366	356	341	323	310	302	286	290	309
Minimum [DU]	204	217	240	263	263	277	273	266	243	232	209	214
Maximum [DU]	479	516	528	519	480	455	425	402	384	381	413	426
Standard dev. [DU]	42.5	46.5	41.9	35.8	30.1	24	21.7	18.4	19	19.1	25.5	32.1
Lower limit [DU]	292.1	310.8	323.7	335.2	326.8	318.8	303.4	294	284.2	268.6	266.1	279.7

the measurement sites. The maximum daily variance in ozone amount between sites is around 7%, and the average daily difference in relation to mean daily amount is about 3%. The difference in the ozone amount is the highest between the southern- and northern-most sites. It is comparatively small, and in relation to the mean daily thickness it is at the measurement error level.

In view of the fact that the correlations of the daily amounts of ozone over Serbian domain are high among all measurement sites and that the changes in all sites are in-phase among themselves, the Serbian domain may be taken to be the site of the satisfactory measurement accuracy. This approach for the considered series led to average annual amounts of ozone over Serbia (tab. 4).

The mean annual ozone concentration over Serbia of 330 DU is higher than the planetary concentration by as much as 40 units (13.8%). In relation to the mean annual ozone concentration for northern hemisphere, the amount of ozone over Serbia is higher by 23 DU (7.9%) [15]. The average annual ozone thickness at the latitude of Serbia is comparatively high, which implies minor health risk from UVR.

The ozone amount registered over Serbia was the highest (347.6 DU) in the first year (1981) of the time sequence and the lowest (311.36 DU) in 1993. The mean value of the three highest ozone depths in the time series (1979-1981) is 346.6 DU, in agreement with the highest three-year value of Wolf number (150.1) for the given period. This is consistent with the observed dominant contribution of solar radiation to the production of ozone [16-19]. The lowest ozone amount of 311.36 DU was registered two years after the Mt. Pinatubo eruption, which was the greatest volcanic eruption in the latter half of the 20<sup>th</sup> century [20]. The largest decline of 24.3 DU among the successive years was registered in 1992, a year after the strongest eruption. Great decrease in ozone concentration was



**Figure 2. Trends in stratospheric ozone decline over Serbia**

observed in 1983, a year after the eruption of El Chichon volcano. This is also consistent with the fact that sulphur and chlorine released in the form of volcanic aerosol are detrimental to stratospheric ozone [21].

Trends in stratospheric ozone decline over Serbia are shown in fig. 2.

The cumulative decline of the annual atmospheric ozone over Serbia for 1979-2008 was 7.6%, or 2.5% per decade. The decrease in stratospheric ozone agrees with the changes over the neighbor countries. The cumulative decline for Bucharest (1980-2003) is 8.1% and similar percents are obtained for other locations in mid-latitudes [22-24].

Studies of the solar activity effects on ozone revealed a connection between the changes in polar faculae and ozone [25]. The polar faculae show anti-phase synchronous with stratospheric ozone above Serbia, which shows that they contribute to the ozone depletion. Polar faculae are local magnetic fields in high heliographic latitudes, up to about 2300 km in diameter [26]. Polar faculae are in anti-phase with sunspots faculae, while the intensity of a local magnetic field can exceed 4 mT [27]. The connection mechanism of polar faculae and stratospheric ozone could operate through coronally holes round the Sun. Substance in coronally holes, under certain conditions, can overpower the pull of gravitational and magnetic forces of the Sun and leave it in the form of solar wind. Particles of the solar wind enhance formation of nitrogen oxides whose multiplied concentration decomposes ozone [25].

Ozone holes over Serbian territory and immediate surrounding, defined by the ozone thickness (1979-2008) of 220 DU or less, occur once in five years on average. Only once, in one measurement site, an ozone hole appeared for two consecutive days (December 1 and 2, 1999, tab. 5). All others were one-day occurrences registered in the November-February interval when the Sun is low on the horizon and the level of UV radiation index is also low. Ozone holes over the given domain were most frequent in November and December, and never covered more than half the Serbian territory.

**Table 4. Average ozone thickness over Serbia**

Year	Mean value [DU]	Maximum error [DU]	Year	Mean value [DU]	Maximum error [DU]
1979	345.97	0.03	1994	326.07	0.03
1980	346.39	0.03	1995		
1981	347.60	0.03	1996		
1982	343.35	0.02	1997	320.92	0.02
1983	328.98	0.02	1998	333.65	0.02
1984	342.42	0.03	1999	324.95	0.02
1985	327.86	0.04	2000	317.40	0.02
1986	334.56	0.03	2001	324.99	0.03
1987	340.51	0.02	2002	316.83	0.01
1988	330.88	0.03	2003	329.03	0.02
1989	332.20	0.02	2004	329.91	0.03
1990	326.06	0.02	2005	327.09	0.02
1991	343.03	0.02	2006	319.54	0.05
1992	318.75	0.02	2007	325.61	0.02
1993	311.36	0.02	2008	318.02	0.02

**Table 5. Ozone hole occurrences in network of ozone monitoring sites from the Serbian domain (1979-2008)**

Dates of ozone hole occurrences 220 DU	02. 02. 1988	03. 12. 1992	15. 11. 1994	01. 01. 1998	01. 12. 1999	02. 12. 1999	30. 11. 2000
MS01	217						
MS02	220						
MS03							
MS04	219						217
MS05	217						
MS06	220						
MS07	220						
MS08							211
MS09	220	218					215
MS10	220	219					
MS11	220						
MS12							
MS13		220					209
MS14		219		220			218
MS15				220			
MS16				219			
MS17							
MS18		220		210			218
MS19				209			220
MS20				213			
MS21				215		219	
MS22		218	219	209			
MS23		214		204			
MS24		218		209	219	216	

## CONCLUSIONS

The research of changes in stratospheric ozone over Serbia and immediate surrounding (1979-2008) for 24 measurement sites indicated the most likely average ozone depth of 330 DU. The mean local ozone is 40 DU greater than the planetary thickness. The annual cycle of change is regular, with minimum (287.7 DU) in October and maximum (371 DU) in April. The cumulative decrease of stratospheric ozone for the same period is 7.6%, or 2.5% per decade. The maximum ozone depths at the beginning of the time-series agree with the solar activity maxima. The research results are consistent with the hypothetical effect of Sun's polar faculae on the ozone decomposition. The lowest amount of ozone (311.4 DU) was recorded two years after the Mt. Pinatubo eruption, which was the greatest volcanic eruption in the latter half of the 20<sup>th</sup> century.

Ozone holes over the Serbian territory in the observation period (1979-2008) occurred once in five years on average. There was a single occurrence of one ozone hole remaining for two consecutive days in this period at one measurement site. All other occurrences were registered in the period November-February when UVR index is low.

The annual amount of ozone layer over the latitudes of Serbia is comparatively high. Consequently, the health risk is smaller from the observed trend of decreasing stratospheric ozone concentration. Ozone holes and the associated increased UVR over Serbian

location are not a serious threat to the health of the population.

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### **ДИНАМИКА СТРАТОСФЕРСКОГ ОЗОНА И УЛТРАВИОЛЕТНО ЗРАЧЕЊЕ ИЗНАД СРБИЈЕ**

Смањење дебљине озонског омотача Земље директно утиче на повећање ултравиолетног зрачења са потенцијалним ефектима на здравље људи. У раду је испитивана динамика стратосферског озона над територијама Србије и непосредног окружења у периоду 1979-2008. године. Констатован је кумулативни пад дебљине озона за 7.6%. Указано је на утицај Сунчеве и вулканске активности на међугодишњу варијабилност дебљине стратосферског озона. Дебљина озонског омотача над територијом Србије је за 13.8% већа од планетарне, а озонске “рупе” се јављају просечно једном у 5 година и не представљају озбиљнију претњу по здравље становништва.

*Кључне речи: стратосферски озон, ултравиолетно зрачење, здравље становништва, озонска рупа, Сунчева активност, вулканска активност*