# VARIABILITY OF TOTAL COLUMN OZONE CONTENT MEASURED AT CHISINAU SITE, REPUBLIC OF MOLDOVA

**A. Aculinin**

*Institute of Applied Physics, Academy of Sciences of Moldova, 5 Academiei Str., Chisinau, MD-2028, Republic of Moldova; E-mail: akulinin@phys.asm.md*

## Abstract

Results of the total column ozone measurements at the ground-based solar radiation monitoring station in Chisinau, Moldova are presented. Column ozone content was derived from the ratios of direct solar ultraviolet radiances measured at 3 discrete wavelengths 305-, 312-, 320-nm within UV-B range. Ozone measurements are regularly carried out with hand-held MICROTOPS II Ozonometer, Solar Light Co. Monthly averages of total column ozone content measured with MICROTOPS II ozonometer at the Chisinau site are in close agreement with those retrieved from the multi-year (1957-2005) measurements carried out at number of ground stations and at the satellite platforms Nimbus -7, Meteor-3, and Earth Probe equipped with the Total Ozone Mapping Spectrometer (TOMS). Total ozone content reveals distinctive seasonal variation at the Chisinau site with maximum and minimum values in spring and fall, respectively. Climatic mean value of total column ozone content derived from TOMS data at the Chisinau site amounts to 334.7 DU.

## 1. Introduction

Ozone in atmosphere serves as a shield against the harmful erythemal UV radiation, which reaches the Earth's surface and exerts unfavorable influence on the human health and on other biological life. Distribution of the atmospheric ozone and ozone trends on regional and global scales are changing as a result of natural processes and increasing of anthropogenic activities, and complex dynamical processes taking place in the atmosphere. It was well established that total column ozone content at the middle latitudes ($35^oN$-$60^oN$) has been decreasing for several decades over 1978-2003 [1]. During these decades of observations a specific trend with ozone depletion was clearly observed. In the period from pre-1980 to 1997-2001 global ozone depleted on an average by about 3%, in the northern middle latitudes ($35^oN$-$60^oN$) total ozone decreased by about 3% and about 6% in southern middle latitudes ($35^oS$-$60^oS$). The value of ozone depletion also depends on season of the year. In the Northern Hemisphere (NH) ozone depletion amounted to 4% and to 2% in winter-spring and in summer-autumn, respectively. In the Southern Hemisphere ozone depletion was about 6% during all seasons [1]. Possible reasons of ozone trends, inter-annual and seasonal variability at the northern middle-latitudes are chemical and dynamical. These reasons are closely connected with each other through the complex processes such as heat and mass transfer and chemical reactions in atmosphere. Chemical cause is related with the increase of the anthropogenic halogen generation such as chlorine, bromine and chemical compounds on their basis. These chemicals induce ozone depletion through numerous and complex reactions both in gaseous phase and on the surface of the aerosol particles. Dynamical cause is connected with the atmospheric circulation processes such as Brewer-Dobson circulation, vertical and meridional motions near the tropopause and the lower stratosphere, planetary wave flux, specific North Atlantic Oscillation phenomenon, etc. [2].

In this paper there is investigated variation of the total column ozone content derived from the ratio of direct Sun radiances measured with hand-held sunphotometer at three wavelengths in the UV region of solar spectrum. Daily, monthly, seasonally and yearly mean values of the total column ozone retrieved at the Chisinau site are analyzed in comparison with the TOMS data. Trends in column ozone content are evaluated from the linear interpolation of gridded multi-annual datasets retrieved from the TOMS measurements at the satellite platforms during a period from 1978 to 2005.

## 2. Equipment and measurement approach

Since July 2003 measurements of the total column ozone or total ozone content (TOC) in the column of atmosphere have been regularly carrying out with hand-held narrowband filter ozonometer MICROTOPS II (Solar Light Co) at the solar radiation monitoring station ($\varphi$=47.00$^0$N, $\lambda_o$=28.82$^0$E, h=205 m a.s.l.) at the Institute of Applied Physics in Chisinau [3]. The instrument is equipped with the highest grade and long stability filters with ion-beam assisted deposition centered at $\lambda$= 305.5, 312.5, 320, 936 and 1020 nm [4]. MICROTOPS II Ozonometer gives an accuracy of ~2% for total column ozone measurements made by much larger and more expensive instruments, such as Dobson and Brewer spectrophotometers [5]. Ozonometer operates as photometer and measures simultaneously direct solar ultraviolet radiance at 3 discrete wavelengths 305.5, 312.5, 320 nm within UV-B range. Filters for these channels have the full-width at half-maximum (FWHM) band pass of 2.4 nm. Registration of signals in all channels is made simultaneously. Single measurement random error for UV radiation is 0.3%. Total column ozone is deduced from the ratio of solar UV radiation measured at two wavelengths. Measurements of UV radiance at the third wavelength are used to account corrections due to aerosol and stray light. Additional channels with 936 nm and 1020 nm are used for measurements of direct solar radiances in the near IR wavelength region to derive precipitable water content and spectral aerosol optical depth in atmospheric column. These near-IR channels are equipped with filters having a FWHM band pass of 10 nm. The baffled collimators for each of channel have field of view of < 2.5$^o$. Detailed description of the instrument and measurement algorithm can be found elsewhere [4]. Due to portability and mobility of instrument, and opportunity to make fast (from a couple of seconds up to 30 sec, depending on the length of scan) measurements of the total column ozone, ozonometer can be successfully used for measurements in conditions of broken clouds. In this case column ozone content measurements are successfully carried out during short time interval of order ~ 10 sec by pointing instrument to the Sun in the clear sky gaps between the clouds. Principally, the measurements of the total column ozone are carried out in solar culmination during midday hours, when the small values of air mass *m* occur or during the hours with an appropriate weather conditions. MICROTOPS II ozonometer allows making reliable measurements of the total column ozone for air masses *m* up to values *m*= 3–3,5 (for AM and PM) [4].

## 3. Results of measurements

Regular total column ozone measurements at the ground station in Chisinau started from July 2003. Results of TOC measurements were processed and compiled into the database. Direct solar radiances measured in the UV-B range at $\lambda$= 305.5, 312.5 and 320 nm, aerosol optical depth at 1020 nm, precipitable water thickness, and atmospheric pressure have been added to this database. Multi-year TOC values acquired from measurements at number of ground stations over 1957-1975 [6] and at the satellite platforms such as Nimbus -7 (N7), Meteor-3 (M3), and Earth Probe (EP) with the TOMS during 1978-2005 [7] are utilized. Detailed information with description of the TOMS instrument, measuring and calibration techniques, and algorithm for retrieval of total ozone content can be found elsewhere [8]. TOMS Version 8.0 gridded datasets are presented in the NASA Goddard Space Flight Center archive with the typical resolution of $1^o$- latitude by $1.25^o$- longitude grid on the Earth's surface. Satellite datasets are used to compute respective interpolating values of the TOC specific to coordinates of the Chisinau site. Interpolated data will be utilized for comparison with the data collected directly from the ground observations at the Chisinau site.

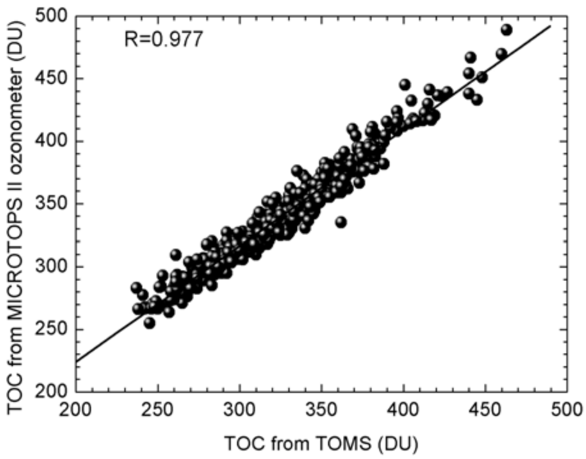


Figure 1. Comparison between MICROTOPS ozonometer data and EP TOMS data acquired over a period from July 2003 to August 2005. Correlation coefficient between datasets is equal to R = 0.977.

The performance of the hand-held ozonometer in current use at the ground station at the Chisinau site was evaluated through the comparison between short time series of data obtained with MICROTOPS and EP TOMS data over a period from July 2003 to August 2005. Results obtained with ozonometer and TOMS instrument are found to be in close agreement with each other, and overall correlation coefficient between these datasets is equal to 0.977 (see Figure 1). Values of TOC obtained from measurements with MICROTOPS ozonometer were found to be systematically higher by about 24 DU in comparison with the EP TOMS data. This value is obtained from the interception of the fitted line with the y-axis, and it represents typical "offset". This "offset" may be connected with the instrument's errors and measurement techniques, which are used in TOMS and MICROTOPS. Differential optical absorption technique in the UV wavelength range is used in both instruments. TOMS measures backscattered UV solar radiances from the Earth's surface and from atop of clouds. Possible source of errors may be the following: errors in the measurement of the radiances, errors in the values of input physical quantities obtained from laboratory calibration measurements, errors in the parameterization of atmospheric properties used as input to the radiative transfer computations, errors due to degradation of optical elements, etc. [8]. MICROTOPS ozonometer utilizes classical optical scheme, which is widely used for direct solar radiance measurements [4,5]. In this case main errors are due to technique applied to direct solar radiance measurements, clearness of entrance window and experience of observer, which may play essential role in obtaining TOC data of high quality.

Total column ozone content over any of the observation sites is defined by concurrent complex dynamical processes in the atmosphere and photochemical processes. These processes result in diurnal, monthly, seasonal and yearly changes of TOC.

On the average total column ozone amounts observed at the Chisinau site show a little diurnal variation with an order of few DU. Some intense diurnal variation of TOC was observed on April 3 and April 29, 2004 (see Figure 2). Relative changes of TOC during these days amounted to ~ 15-20 DU. These diurnal changes of TOC may be attributed to air mass transport from the regions having high (for April 29) or low (for April 3) column ozone amount and to traveling of these air masses over the ground station at the Chisinau site.

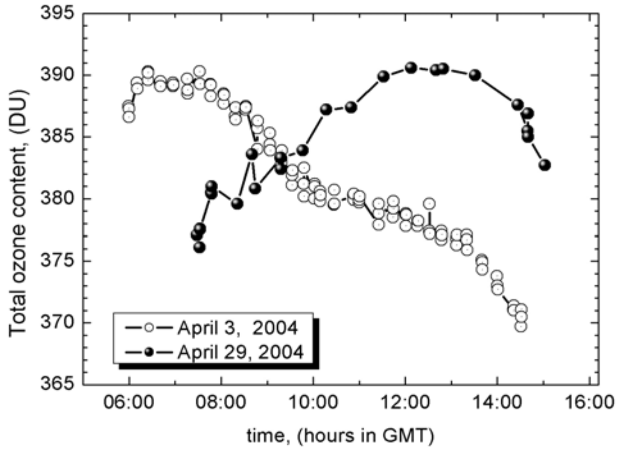


Figure 2. Diurnal variation of the TOC amounts measured with hand-held MICROTOPS ozonometer at the Chisinau site.

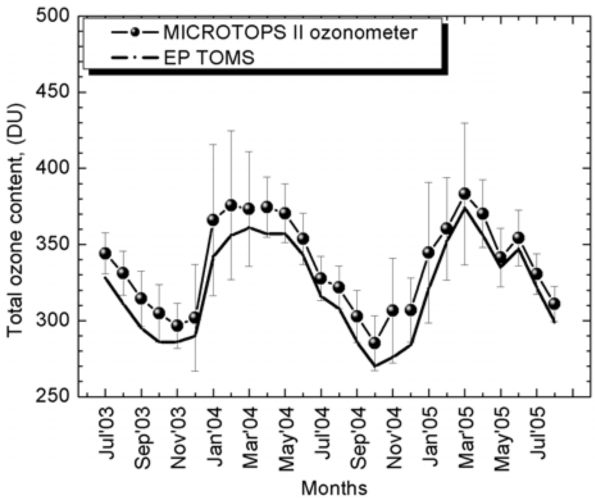


Figure 3. Time series of monthly mean values of the total column ozone content from MICROTOPS ozonometer and EP TOMS measurements during 2003-2005. Error bars show ±1σ values of standard deviation of monthly mean MICROTOPS data.

Time series of the total column ozone content measured with hand-held ozonometer at the ground station in Chisinau represents a short series of observations lasted from July 2003 to August 2005. Ozonometer dataset and data retrieved from the EP TOMS measurements for this period of observations are shown in Figure 3. MICROTOPS ozonometer and TOMS data show similar annual and inter-annual variations during that period, although hand-held ozonometer records all the time slightly higher values of TOC. Analysis of discrepancy between MICROTOPS and TOMS daily mean values of TOC, $\delta=(X_{MT}-X_{TOMS})/X_{MT}$, gives on an average, $<\delta>=4.5\pm2.7\%$, where variables X are the values of total column ozone content and they are expressed in Dobson Units (DU); subscript 'MT' is referred to the measurements of TOC carried out with MICROTOPS ozonometer. Maximum variability of monthly means of TOC is observed during winter-early spring months. Daily mean values of TOC vary in a wide range during each month due to dynamical processes in the atmosphere. The largest amplitude of variation of daily mean values of TOC, defined as the difference between the maximum and minimum values of daily column ozone content observed during every month, is typical for winter-spring months and it reaches maximum value of ~ 155 DU on February 12, 2003. Monthly mean values of TOC show distinct seasonal variation with minimum and maximum values occurring in fall and winter-early spring, respectively. Various air mass circulation states specific to each season in the NH middle-latitudes are responsible for the variability of TOC. Variation of monthly averages of TOC retrieved from multi-year TOMS measurements at three satellite platforms such as N7, M3 and EP during period of 1978-2005 is shown in Figure 4. Data collected from the ground based measurements with MICROTOPS ozonometer at the Chisinau site from July 2003 to August 2005 are also presented in Figure 4. MICROTOPS data reveal very good resemblance with the multi-year monthly averages derived from TOMS data over a period of 1978-2005. Annual course of TOC from TOMS measurements shows smooth variation with respective minimum value in fall and maximum value in spring, which in general is typical for the NH. Some abnormal behavior of TOC was observed at the Chisinau site during summer-fall in 2003. MICROTOPS data were found to be higher relative to the multi-year TOMS overpass data and difference between them amounted to ~ 14 DU during the summer-fall period of observation. At the same time global maps of monthly means of TOC retrieved from EP TOMS measurements in the NH [7] show persistent and relatively high values of TOC for these months in 2003. This may be due to the lasting period of transportation of rich ozone air masses to the atmosphere of the middle latitudes in the NH and to the photochemical processes responsible for the partial restoration of ozone layer.

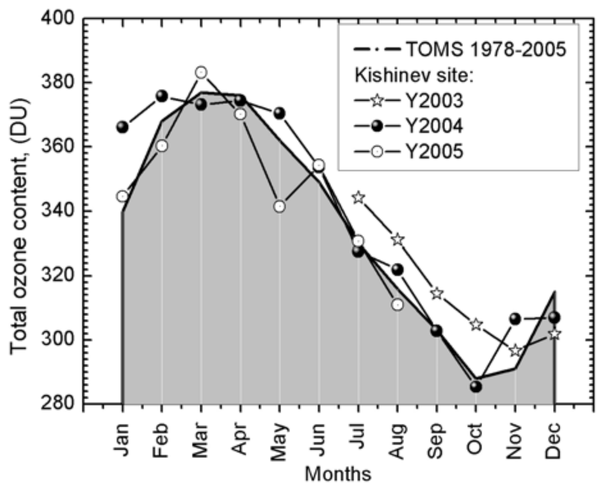


Figure 4. Variability of monthly means of TOC amounts retrieved from multi-year statistics of TOMS measurements from 1978 to 2005 and observed from ground based measurements with MICROTOPS ozonometer at the Chisinau site from July 2003 to August 2005.

In order to estimate mean values of TOC variability at the Chisinau site TOMS instrument dataset over 1978-2005 was used. These values are defined as the difference between maximum and minimum of monthly averages of TOC, which were observed during every year in the time series of TOMS data. Maximum and minimum values of TOC variability were found to be ~136 DU (in 1985) and ~83 DU (in 1992). For comparison of TOC variation observed at the Chisinau site with the average amounts of column ozone in latitudinal belt from 45$^o$N to 50$^o$N multi-year dataset collected at number of ground based stations spread in this belt and dataset retrieved from TOMS measurements are used. Mean-latitudinal distribution of monthly and multi-year averages of the total ozone content in column of atmosphere retrieved from the combined TOMS overpass data acquired at the N7, M3 and EP satellite platforms over a period 1978-2005 and at number of ground stations during 1957-1975 are presented in Table 1.

Table 1. Mean-latitudinal (45$^o$–50$^o$) distribution of monthly and multi-year averages of the total ozone content (in DU) in column of atmosphere measured at number of ground stations and at the Nimbus-7, Meteor-3, and Earth Probe satellite platforms equipped with the TOMS. Total column ozone amounts measured with the MICROTOPS II Ozonometer at the ground-based station at the Chisinau site during 2003-2005 are presented in the last row.

| | Jan | Feb | Mar | Apr | May | Jun | Jul | Aug | Sep | Oct | Nov | Dec | Multi year |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| ground 1957-1975 | 359 | 386 | 389 | 392 | 358 | 335 | 317 | 303 | 293 | 290 | 302 | 329 | 339 |
| TOMS 1978-2005 | 340 | 368 | 377 | 376 | 362 | 348 | 330 | 315 | 302 | 289 | 291 | 314 | 335 |
| Chisinau2003-2005 | 355 | 368 | 378 | 372 | 356 | 354 | 334 | 321 | 309 | 295 | 302 | 304 | 337 |

One can clearly see a close agreement between monthly means of TOC observed at the Chisinau site during 2003-2005 and zonal monthly averages of TOC retrieved from multi-year measurements carried out at the ground stations and satellite platforms in the NH middle-latitudes over a period of 1957-2005. In the last column of Table 1 zonal multi-yearly averages of TOC are presented. Yearly mean value of TOC at the Chisinau site was deduced from two-year-series of ozone observations. Yearly mean values of TOC at the Chisinau site show very good coincidence with the multi-year averages of TOC for mean-latitudinal datasets from ground stations and satellite platforms.

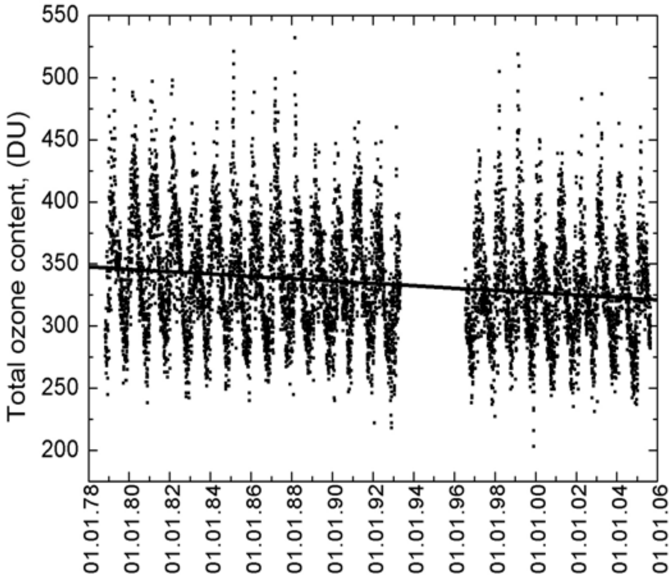


Figure 5. Time-series of daily mean values of TOMS gridded data of the total ozone content versus years. TOC data were retrieved and interpolated for the Chisinau site over the period of observation from 1979 to 2004. Linear regression over whole dataset gives the value of ozone trend of – 10.5 DU per decade or -3.1% per decade.

Time-series of daily mean values of TOC from TOMS instrument's measurements is shown in Figure 5. Total column ozone amounts were retrieved and interpolated for the Chisinau site over the period of observations from 1979 to 2004. Dataset, which is under analysis, consists of full year observations with the gap lasting from 1993 to 1996. TOMS data during that time gap were missing or incomplete. Linear regression applied to the TOMS overpass dataset at the Chisinau site gives the value of ozone trend of –10.5 DU per decade or -3.1% per decade, which is consistent with the value of ozone depletion specific to the NH middle-latitude belt from 35$^o$N to 60$^o$N [1]. Seasonal trend of TOC in column of the atmosphere is defined by transport of ozone within the low stratosphere, i.e. through advection of air masses and their upwelling, and photochemical reactions. TOC decline was found to be statistically significant for all seasons. Winter-spring trend is the highest one and it is higher than the fall trend by ~ 3.5 times. Seasonal trends of ozone depletion at the Chisinau site amount to ~3.5%, ~ 2.8% and ~1% for winter-spring, summer and fall, respectively. These values are in close agreement with the respective ones retrieved for the mid-latitudes in the NH, where seasonal trends of TOC amounted to 4% and to 2% in winter-spring and summer-autumn, respectively [1].

Seasonal values of zonal averages of TOC were deduced from measurements carried out at number ground stations and at the satellite platforms with the TOMS instruments during 1957-2005 (see Table 2). Seasonal mean values of TOC obtained at the Chisinau site during short period of observations from 2003 to 2005 are presented in the last row of Table 2. Statistically averaged TOC data for the longitudinal zone from 45$^o$N to 50$^o$N and Chisinau site are in close agreement with each other. Maximum and minimum of seasonal mean values of TOC observed at the Chisinau site amount to ~369 DU and ~302 DU in spring and fall, respectively and these extreme values are consistent with the ones deduced from the multi-year (1978-2005) TOMS data, thereby confirming the character of the seasonal variation of TOC in middle-latitudes in the NH.

Table 2. Seasonal values of zonal averages of TOC (in DU) retrieved from measurements at number of ground stations and at the satellite platforms with the TOMS over a period of 1957-2005 and seasonal averages of TOC observed at the Chisinau site during 2003-2005.

| | Winter | Spring | Summer | Fall |
|---|---|---|---|---|
| Ground +TOMS 1957-2005 (zonal averages) | 352 | 374 | 326 | 299 |
| TOMS gridded data during 1978-2005 | 342 | 373 | 332 | 295 |
| Chisinau site data during 2003-2005 | 341 | 369 | 336 | 302 |

Frequency of occurrences of seasonal variation of TOC retrieved from the TOMS instrument measurements at the Chisinau site during 1978-2005 is shown in Figure 6. The winter and

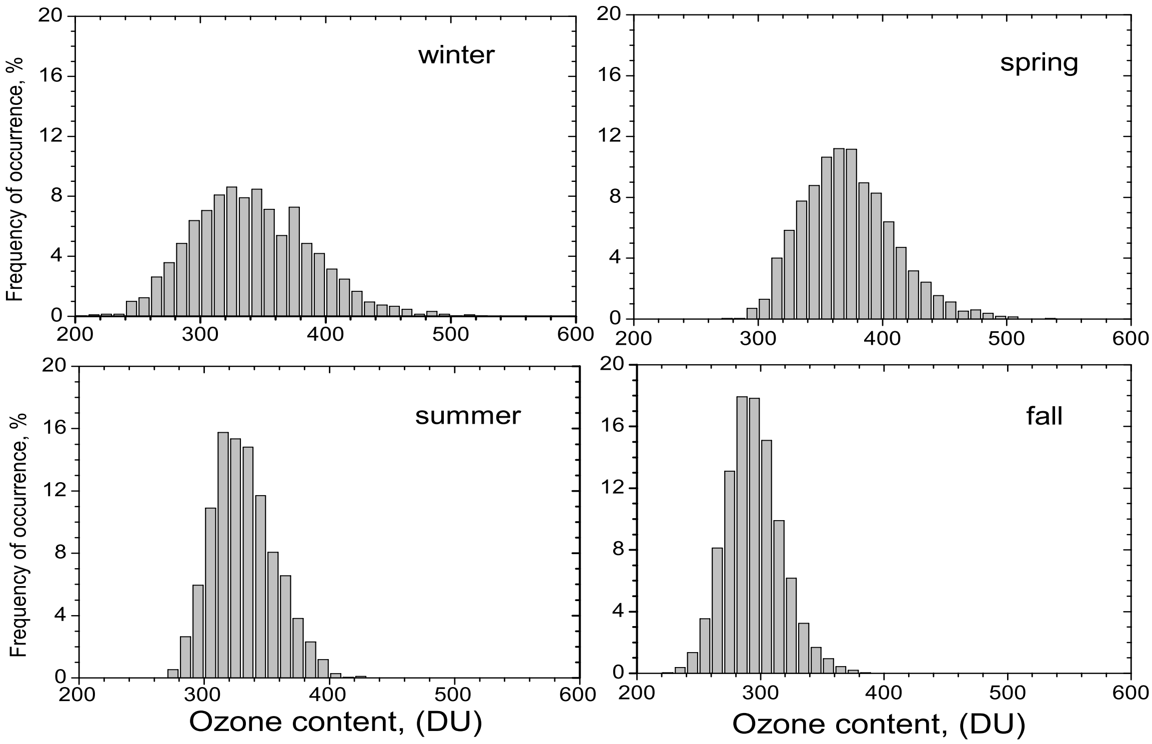


Figure 6. Seasonal variation of TOC retrieved from the TOMS gridded data at the Chisinau site during 1978-2005.

spring are characterized by a wider distributions of TOC's values with modal values at ~340 DU and ~370 DU, respectively. These distributions span a wide range of TOC values from 240 DU to 500 DU. This fact confirms that such extent of TOC variation observed during winter-spring is controlled by intense dynamical processes occurring in the atmosphere. The summer and fall have narrow probability distributions with modal values at ~330 DU and ~290 DU, respectively. The fall is characterized by the narrowest distribution and by larger shift of the maximum toward the lower values of TOC less than 300 DU.

Deviations in seasonal averaged values of column ozone content from TOMS data were calculated for winter (D-J-F), spring (M-A-M), summer (J-J-A) and fall (S-O-N) over period 1979-2004 at the Chisinau site (see Figure 7). These deviations indicate departures (in %) of seasonal averages of TOC in time series from the climatic mean value retrieved from TOMS overpass data for that period of observations. The highest deviations are observed in spring and winter. In spring months deviation is fully positive and ranges from 5.2% to 21.9%. These seasons are characterized as ones having higher values of TOC and such deviations may be due to influence of dynamical processes in the atmosphere. Maximum of long-term mean value of TOC from TOMS overpass data is peculiar to March and it amounts to ~377 DU (see Table 1). A full negative deviation is found in fall and it ranges from -17.4% to -8.7% during 1979-2004. This result may be also explained by dynamical reasons: air mass transportation from regions with low ozone content and upwelling of air within the low stratosphere. The fall is characterized as a season with low TOC (see Figure 4). The lowest monthly mean value of TOC ~ 289 DU was recorded in October (see Table 1).

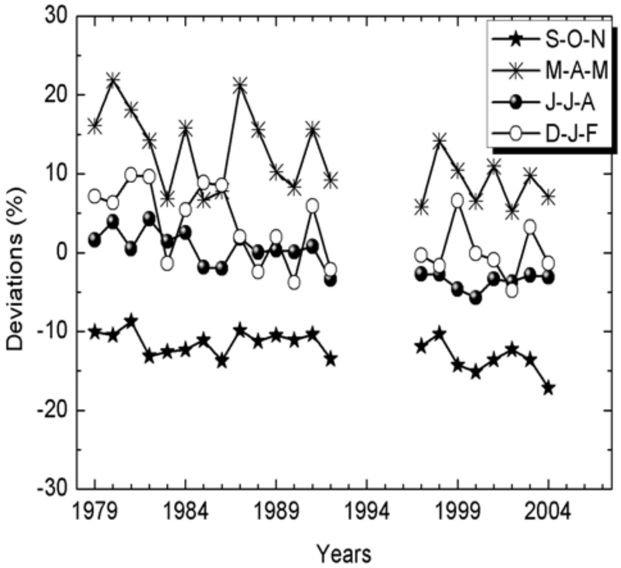


Figure 7. Deviations (in %) of seasonal mean values of TOC from climatic mean in winter (D-J-F), spring (M-A-M), summer (J-J-A), and fall (S-O-N).

Total column ozone content anomalies having extremely large or low values of total column ozone amounts are created by atmosphere anomalous circulation patterns. Climatic mean value of TOC derived from the historical series of TOMS data at the Chisinau site is equal to 334.7 DU with the maximum value of ~532 DU (March 3, 1988) and minimum value of ~203 DU (December 1, 1999). These mean and extreme values of total column ozone amount were retrieved from multi-year TOMS measurements during 1978-2005. Mean value of TOC derived from MICROTOPS ozonometer measurements during short series of observations at the Chisinau ground station amounts to 342.8 DU. Maximum and minimum values of TOC are found to be equal to ~491 DU (February 12, 2004) and ~252 DU (October 31, 2004), respectively. Anomaly ozone events, which are characterized with the lowest values of TOC, represent special interest due to increase of UV solar radiation having a harmful influence on the biological life. Such localized and transient events with substantial ozone depletion over area up to several thousand kilometers during a short period (few days) are known as ozone mini-holes. Ozone mini-holes occur in middle-latitudes, typically in autumn and winter [9]. These anomaly events are caused by dynamical processes such as advection of warm subtropical air with low ozone content, upwelling of air masses due to wave-breaking in low stratosphere [10-12], poleward transport [9], North Atlantic Oscillation [2,13]. Especially, the event with extremely low value of TOC ~165 DU was recorded over the North Sea on November 30, 1999. This event was extremely low one ever registered in TOMS measurements in the NH over period 1978-2005. It should be noted that extremely low TOC ever retrieved from TOMS records for Moldova amounted to ~203 DU on December 1, 1999 which was the day of the culmination of the ozone hole. This day with ozone anomaly was due to the December ozone hole, which stretched and covered the most part of the Europe, reaching the Mediterranean and Black seas. Next days ozone hole became thin in meridional direction and began to migrate to the east direction, and finally, on December 4, this ozone anomaly disappeared.

## 4. Summary and conclusions

Results of TOC measurements with MICROTOPS and TOMS instruments are found to be in close agreement with each other, and overall correlation coefficient between their datasets amounts to 0.977. Climatic mean value of TOC retrieved from TOMS overpass data at the Chisinau site during 1978-2005 is equal to 334.7 DU with the maximum value of ~532 DU (March 3, 1988) and minimum value of ~203 DU (December 1, 1999). Mean value of TOC derived from MICROTOPS ozonometer measurements at the Chisinau ground station during 2003-2005 amounts to 342.8 DU. Maximum and minimum values of TOC are found to be equal to ~491 DU (February 12, 2004) and ~252 DU (October 31, 2004), respectively. Monthly means of TOC from TOMS overpass data show distinct seasonal variation with minimum ~ 289 DU (in October) and maximum ~ 377 DU (in March). Extremely low TOC ever retrieved from multiyear TOMS records for Moldova amounted to 203 DU (in December 1, 1999) during the localized and short-term ozone mini-hole event observed over Europe on November 29-December 3, 1999. Seasonal trend of TOC is defined by transport of ozone within the low stratosphere through advection of air masses and their upwelling, and photochemical reactions. TOC decline was found to be statistically significant for all seasons. Seasonal trends of ozone depletion from multiyear TOMS data at the Chisinau site amount to ~3.5%, ~ 2.8% and ~1% for winter-spring, summer and fall, respectively. Linear regression of TOMS dataset at the Chisinau site gives value of ozone trend of –10.5 DU per decade or -3.1% per decade which is consistent with the value of ozone depletion observed in the NH middle-latitudes belt from $35^{o}$N to $60^{o}$N.

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