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Recent developments of atmospheric research in Ukraine

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In recent years the Joint Laboratory of Atmospheric Optics and Aerosols of Ukraine has been carrying out atmospheric research in cooperation with international program of climate change studies. Our current research is aimed at studying aerosol and ozone in the Earth's atmosphere, because these constituents have a substantial influence on climate. In Ukraine, atmospheric aerosol remote sensing in the PHOTONS/AERONET network has been carried out since 2006 in Sevastopol and 2008 in Kyiv. For this research, sunphotometers CIMEL CE318 have been used. A mobile AERONET station has been developed, which consists of CE318N and portable Microtops II sunphotometers, as well as two self-designed experimental portable sunphotometer models and an ozonometer for aerosol and ozone study. The team's findings on aerosol spectral optical thickness, as well as optical and physical properties of aerosol particles (single-scattering albedo, distribution of particles by sizes, Ångström exponent), are discussed in the paper. In 2010, upon the establishment of the new regional atmospheric research station Nr. 498 Kyiv-Goloseyev, the team commenced with measurements of the total column density and vertical distribution of ozone, using the Dobson D040 spectrophotometer, in the framework of the Global Atmosphere Watch Program of WMO. The station has also been equipped with a Vaisala automatic weather station, a surface ozone 49i analyzer, and an experimental complex for monitoring secondary space rays. The aerosol and ozone measurements have been continuously submitted to data centres of AERONET (`http://aeronet.gsfc.nasa.gov/`) and WMO (`www.woudc.org/data/`). For aerosol and ozone research, the data from satellite sources (POLDER, MODIS, OMI, and SCIAMACHY) have also been analysed. The work on a proposal to design, build, and launch the space radiometer/polarimeter for global monitoring of atmospheric aerosols has commenced recently. This instrument should have the capability of investigating microphysical properties of aerosol particles. An overview of the devices, methods of data retrieval analysis of aerosol parameters and ozone dynamics, is presented in this paper.

Key words: aerosol, ozone, climate, remote sensing

INTRODUCTION

The main climate-influencing constituents in Earth's atmosphere (in addition to carbon dioxide CO_2 and methane CH_4) are aerosols, water vapour, and stratospheric ozone. Their impact on the transfer of solar radiation energy and the energy balance of the Earth has not yet been sufficiently examined [6]. Therefore the study of the influence of aerosols, water vapour, and ozone on climate change remains one of the fundamental problems of climatology. Atmospheric aerosols are produced by natural and anthropogenic sources, and vary greatly in particle size (from $0.001\,\mu$m in molecular clusters of gaseous contaminants, to $1000\,\mu$m in heavy dust) and chemical composition. Aerosols affect Earth's energy balance by scattering and absorbing radiation (direct effect) and by modifying the constituents and microphysical/radiative properties of clouds (indirect effects). The effect of aerosols on regional climate change is complex and varied: while weakly-absorptive aerosols, such as sulfates, can cool the atmosphere, carbon aerosols will have the opposite effect, increasing the temperature. Additionally, through interaction with water vapour, aerosols create an indirect climate effect by influencing the cloud cover and precipitation amounts. Therefore, the problem of quantitative estimation of the effect of aerosols, ozone, and water vapour on regional climate, is the basic scientific task of our current research [2].

AEROSOL MEASUREMENTS

The investigation of aerosols and their impact on climate change is based on measurements of ground-based networks and satellite radiometer/polarimeter observations [10, 12]. In April 2008, the monitoring of aerosol optical properties was commenced using

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the CIMEL CE318 sunphotometer at the Kyiv site, as a part of the PHOTONS/AERONET (AErosol RObotic NETwork) program with French support through PHOTONS (Service d'Observation from CNRS/INSU) [2, 7]. In November 2011, the mobile AERONET site was created, which consisted of sunphotometers CE318N and Microtops II, and was used for aerosol measurements in other regions of Ukraine. Additionally, the mobile equipment includes two experimental sunphotometers for aerosol and ozone observations, designed analogously to the Microtops II sunphotometer. Another, two-channel scanning sunphotometer, which will be used for aerosol transects from a moving car, is currently under development. For aerosol characteristic comparisons between the city and the outskirts (at the Kyiv site), an additional AERONET Kyiv-AO site for aerosol observations was established in the centre of city.

Aerosol data processing from CE318 and CE318N sunphotometers is derived automatically using the AERONET algorithm. The measurement results are regularly uploaded to the AERONET[1] database. The aerosol parameters – spectral aerosol optical thickness (AOT) and Ångström exponent – are retrieved from direct solar irradiation measurements. AOT is computed using the Beer-Lambert-Bouguer law in consideration of Rayleigh scattering, ozone absorption and molecular absorption by water vapour and other absorbing gases. Water vapour content is determined from atmosphere total extinction measurements by CE318 and CE318N sunphotometers at 936 nm, using the filter-dependent constants of the sunphotometer. AOT is a basic characteristic of the total aerosol content in the atmospheric column. The error of AOT determination from CE318 measurements is approximately ± 0.01. The Ångström exponent $\alpha(\lambda_1, \lambda_2)$ is a measure of the wavelength (λ) dependence of the AOT and is computed here as

$$\alpha(\lambda_1, \lambda_2) = -\ln(AOT(\lambda_1)/AOT(\lambda_2))/\ln(\lambda_1/\lambda_2) \quad (1)$$

from AOT data at $\lambda_1 = 440$ nm and $\lambda_2 = 870$ nm using a linear fit of α. The Ångström exponent is a basic measure of the aerosol particle size distribution because AOT(λ) depends on the extinction process governed by the Mie ratio $2\pi a/\lambda$, where a is the radius of the spherical particle. For example, $\alpha \leq 1$ indicates large particle ($>0.5\,\mu$m) dominance in the atmospheric column, whereas $\alpha \geq 1.5$ corresponds to small particle ($<0.5\,\mu$m) dominance. The columnar properties of aerosol particles, notably single-scattering albedo, complex refractive index, and aerosol particle size distribution, are retrieved from sky radiation measurements using algorithm [5]. Microtops II measurements data give information about AOT and Ångström exponent. The results of aerosol observations and data analysis are discussed in the next section.

Satellite passive and active techniques enable the derivation of global aerosol distribution by scanning the atmosphere along and across the satellite ground track. Several global aerosol datasets are available from various satellite sensors, e. g., MODIS, AVHRR, POLDER [12]. Our team used polarization measurements provided by the POLDER (POLarization and Directionality of the Earth Reflectance) instrument [3] for aerosol properties analysis over land surface. The key advantage of polarized observations is their ability to systematically correct the ground reflectance contribution. The POLDER instrument, launched aboard French microsatellite PARASOL in December 2004, is still operational. The POLDER imaging spectroradiometer offered multispectral, multidirectional, and polarized measurements in 443, 490, 565, 670 (polar), 763, 765, 865 (polar), 910 and 1020 nm wavelengths. This data enables the derivation of aerosol particle size and scattering phase function, as well as aerosol optical thickness [4]. Aerosol parameters received from POLDER data have a horizontal spatial resolution of 18×18 km. The monthly average AOT at 865 nm, and Ångström exponent computed between 670 and 865 nm, are used in our analysis.

AEROSOL DYNAMICS

We study atmospheric aerosol dynamics over Ukraine, based on measurements obtained using the sunphotometer CE318. Over four years of observation, a set of aerosol particles parameters mainly over Kyiv site was obtained. Additionally, aerosol properties data over other Ukrainian regions were obtained using sunphotometer CE318 and Microtops II, and the results were compared using satellite data. Figure 1 shows the monthly mean spectral AOT values as obtained by the Kyiv site measurements in 2008–2011.

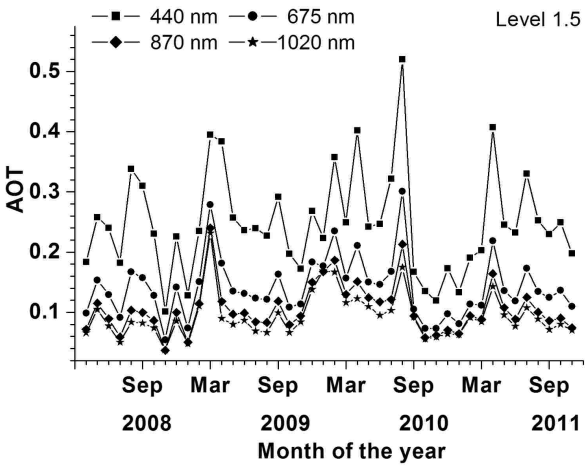

Fig. 1: The monthly means of spectral AOT at Kyiv site.

[1] http://aeronet.gsfc.nasa.gov/

The AOT value experiences seasonal variations (e. g. from 0.1 to 0.4 at 440 nm) with increase in spring and late summer, and spontaneous spikes like those in August 2010, that correspond to period of wildfires in Russia. The AOT parameter is a characteristic of total amount of aerosol in the atmosphere over the site. The AOT over Kyiv compared with other planetary regions shows that atmosphere over Kyiv is relatively clean with low amounts of aerosol. However, AOT can vary significantly, at times increasing by a factor of 10 in a month or even a day. The results of Ångström exponent measurement in Fig. 2 show that particles ~0.1–0.3 μm dominate in the atmosphere over Kyiv ($\alpha \geq 1.5$). However, in some cases the coarse aerosol fraction prevails when $\alpha \to 0$. Usually the fine mode of particles represents aerosol of anthropogenic origin [8].

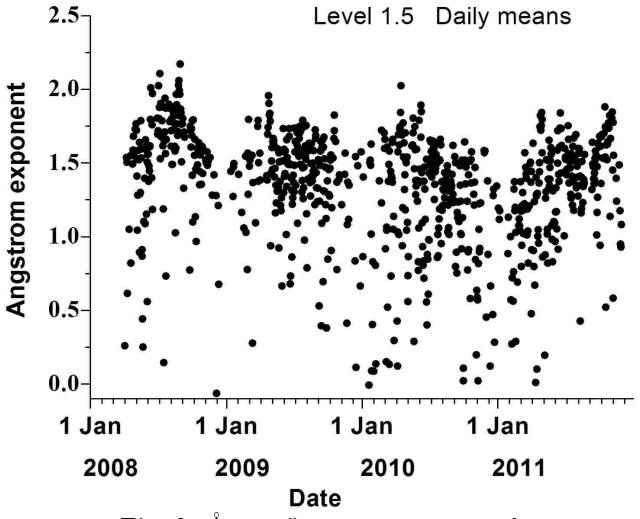

Fig. 2: Ångström parameter trend.

The comparison of aerosol properties over the Kyiv site versus other AERONET sites in Europe is shown in Fig. 3. Note the peaking aerosol pollution in Eastern Europe during strong wildfires in Russia in August 2010, when aerosol optical thickness increased by a factor of ten. The analysis shows that the atmosphere aerosol pollution over the Kyiv site is similar to other urban sites in Europe: with dominance of fine mode aerosol particles.

The aerosol particles size distribution is used in radiance flux and radiative forcing estimations. In the inversion problem solution, the aerosol size distribution is modelled by log-normal law that in most cases is bi-modal (see Fig. 4), but sometimes it is tri-modal. The minimum value of bi-modal distribution is considered as the limit for separation of the fine and coarse mode aerosol fractions. According to our data, the mean limit value for the Kyiv site is about 0.5–0.6 μm, however it could vary between 0.3–1.05 μm during shorter periods of averaging (from one to three days). For the Lugansk site, the mean limit value is about 0.4–0.8 μm (Fig. 4).

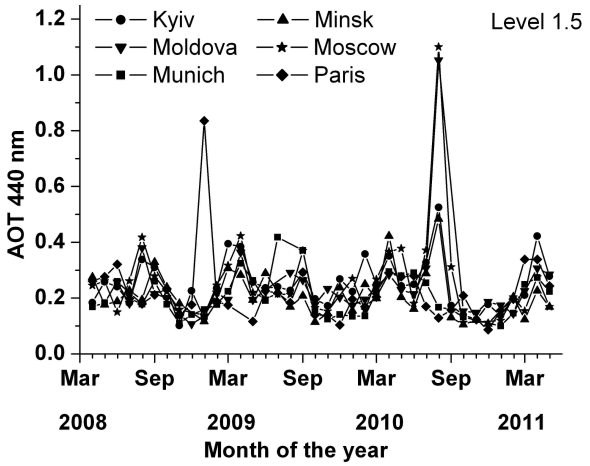

Fig. 3: Urban aerosol AOT over cities in Europe.

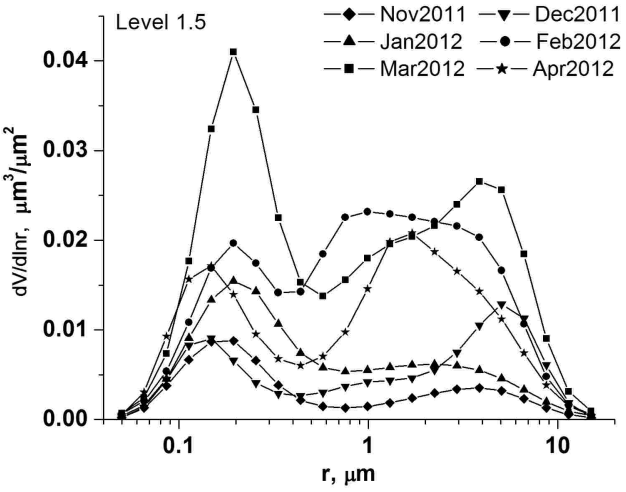

Fig. 4: Aerosol size distribution in Lugansk site.

The results of Microtops II sunphotometer measurements in several Ukraine regions show that AOT varies from 0.1 to 0.5 for the 440 nm spectral band for all periods except the summer period of wildfires in Russia in 2010, when AOT increased to 1.4. Fine fraction aerosol prevailed in Ukraine according to Ångström exponent retrievals from Microtops II data.

The satellite POLDER/PARASOL data were used to analyse aerosol atmosphere pollution over Ukraine together with our AERONET measurements. Some results of aerosol distribution and particle properties over Ukrainian regions are shown below. The primary area of our interest is Eastern Europe – a region that is considered to be the source of both natural and anthropogenic aerosols. We retrieved AOT satellite data maps over Ukraine for period March 2005 – May 2012 using information from ICARE, Cloud-Aerosol-Water-Radiation Interactions Data and Services Center archive[2].

[2] http://www.icare.univ-lille1.fr/archive/

The region is characterized by numerous agricultural, grass, shrubbery, forest and peat wildfires, as well as soil erosion in steppe regions. Saharan dust is transported to Eastern Europe over the Mediterranean and can increase AOT in the Eastern Europe region by 0.02–0.07, mostly in autumn and spring [9]. In spite of the large number of pollution sources, the investigated territory is characterized by a relatively low AOT (ranging between 0.05 and 0.15 at 870 nm) in comparison to that of areas in East Asia affected by industrial pollution, or areas in Southern Africa, caused by Sahara dust events and forest wildfires (where the AOT ranges from 0.2 to 0.6 at 870 nm). The monthly mean AOT at 865 nm for August 2010 and 2011 are presented in Fig. 5 and 6. Anomalously high surface temperatures (35–41°C) and low relative humidity (9–25%) from mid-June to mid-August 2010 created ideal conditions for forest and peat wildfires. Analysis showed that the region in Western Russia nearby Moscow (Fig. 5) was most severely impacted by wildfire emissions. Average values of AOT over Northern and Eastern Ukraine in August 2010 ranged from 0.2 to 0.4 at 870 nm due to wildfires in this area and transportation of biomass burning aerosols. The Ångström parameter computed during the wildfire period for 670 nm and 870 nm is between 1.8 and 2.0, and it corresponds to fine mode particles.

Fig. 5: Monthly mean AOT distribution at 870 nm in August 2010.

Fig. 6: Monthly mean AOT distribution at 870 nm in August 2011.

Summarising, it can be concluded that the atmospheric aerosol pollution over Ukraine is similar to that over Europe, with dominance of fine mode aerosol particles. It is not surprising that anthropogenic aerosols are detected over industrial areas. According to our analysis, the AOT values are higher over Eastern and central parts of Ukraine. Larger Ångström exponent values correspond to fine mode particles of anthropogenic origin, as seen over Eastern Ukraine where the industrial pollution is mainly concentrated.

OZONE OBSERVATIONS: VARIABILITY AND CLIMATOLOGY

Historically, the total ozone column (TOC) observations in the Kyiv region were commenced in 1973 using filter ozonometers. These data were archived in the World Ozone and Ultraviolet Radiation Data Centre (WOUDC[3]) database and are available for the observation period 1973–1997. In the same region, analysis was performed using the filter ozonometer M-124 in Lisnyky station near Kyiv for the time period of 1997–2002. Since 2010, observations with the Dobson spectrophotometer have been carried out at the Kyiv-Goloseyev station. A joint team of observers is currently carrying out the TOC and Umkehr height profile measurements at a new site, registered in WOUDC as "Kyiv-Goloseyev STN498" station. In 2010, the station was included in the Global Atmosphere Watch (GAW) Program of the WMO, as the Regional GAW Station, with GAWID 'KGV'. The D040 Dobson spectrophotometer allows researchers to calculate the total ozone column from estimations of solar radiation absorption in near-ultraviolet range (305–340 nm), using the well-known Beer-Lambert-Bouguer law. The TOC values are also determined from zenith sky measurements using statistical dependencies.

To estimate the quality of the ground-based Dobson D040 measurements, the measurements were compared to satellite data (Fig. 7, the range of two standard deviations from the mean difference value is indicated with a dotted line). Satellite overpass data obtained with OMI/Aura and SCIAMACHY/Envisat instruments[4] have been considered. This comparison indicates a high quality of the Direct Sun (DS) and Zenith Blue (ZB) Dobson observations, as mean satellite vs. ground-based differences do not exceed 1.5–2.0% and standard deviation is equal to 3–4% from annual mean ozone values (about 320 DU for Kyiv-Goloseyev station). It should be noted that an ozone station is typically evaluated as "good" if the above-mentioned difference is $< 3\%$ with a standard deviation of $< 4.5\%$. The dispersion increases in the case of the Zenith Cloud measurements (the standard deviation is nearly 6%) due to the non-uniform structure of the clouds and the difficulty of modelling their structure under varying conditions using the same algorithm. The sea-

[3] http://www.woudc.org
[4] http://www.temis.nl

sonal cycle significantly affects the total ozone differences (Fig. 7). The seasonal cycle is caused by low efficiency of Dobson algorithm at solar zenith angles exceeding 70° and by dependence of the total ozone difference on zenith angle. Observations under these conditions are predominantly carried out near winter solstice, when solar zenith angles reach 73° at noon. Influence of scattered light increases at high solar zenith angles causing total ozone underestimation with standard algorithm considering only direct solar radiation [1].

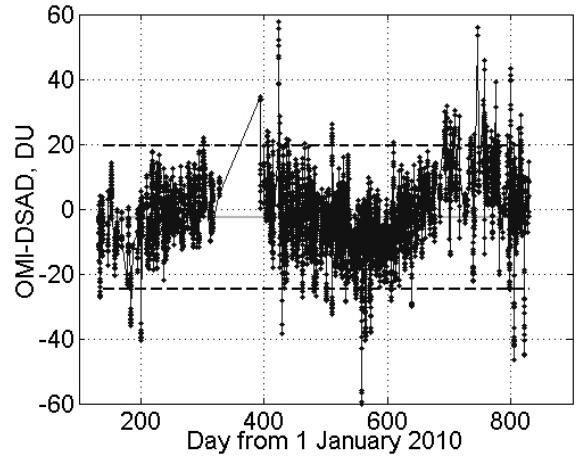

Fig. 7: Differences between OMI satellite measurements over Kyiv-Goloseyev and Direct Sun Dobson data.

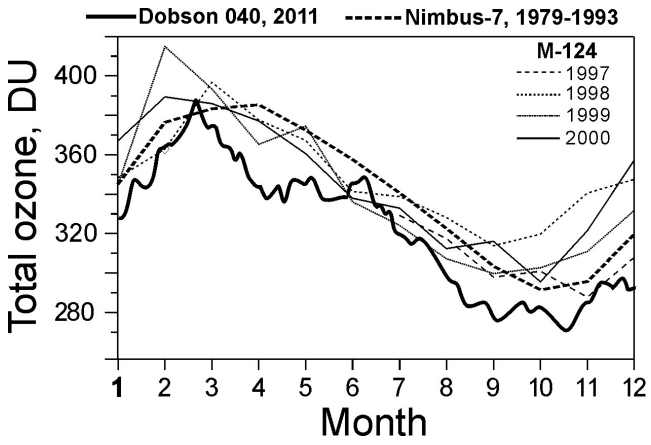

Fig. 8: Seasonal change of the total ozone over Kyiv region by the ground-based and satellite measurements in 1979-2011.

The total ozone content varies seasonally according to both historical ozonometer data and recent Kyiv-Goloseyev measurements. Fig. 8 shows averaged seasonal variation of the total ozone over the Kyiv region from three different instruments: the Nimbus-7 satellite spectrometer for observations in the time period from 1979 to 1993 (archive overpass monthly means, thick dashed curve), the ground-based filter ozonometer M-124 for the 1997–2000 period (Lisnyky station monthly means, thin curves), and the Dobson spectrophotometer for 2011 observations (30-day running means, thick solid curve). The seasonal ozone variation is typical for the Northern Hemisphere middle latitudes, with the maximum ozone content in winter-spring, and the minimum in autumn.

More detailed seasonal evolution of the TOC levels over Ukraine can be seen from Fig. 10, where the continuous satellite measurements over nearly 15 years (1979–1993) are presented. An archive of daily ozone measurement for four Ukrainian ground stations shows a partial TOC decrease in late March-early April, which interrupts the mean monotonic TOC level variation near the seasonal maximum (as marked by arrows in Fig. 10a). This feature is not seen in the monthly mean data (Fig. 10b) as the temporary ozone minimum falls to the month transition interval. The causes of this ozone decrease should be investigated in the future work.

Seasonal climatology (Fig. 10b) gives a reference annual TOC cycle for revealing the anomalous TOC levels over the station. In particular, according to recent Kyiv-Goloseyev data, daily TOC values exceeded 400 DU (Dobson units) in February-April and dropped to 250–260 DU in October-November (compare this to the climatological maximum ∼380 DU and minimum ∼300 DU, depicted with rectangles in Fig. 10b). Using this method, several specific events with an anomalous TOC decrease were observed. For example, lowered TOC values were recorded in August 2010, which are most likely related to atmospheric pollution due to forest/peatbog wildfires in Russia. In another case, very low values of ∼220 DU were observed in October 2011, which can be correlated with the ozone mini-hole event.

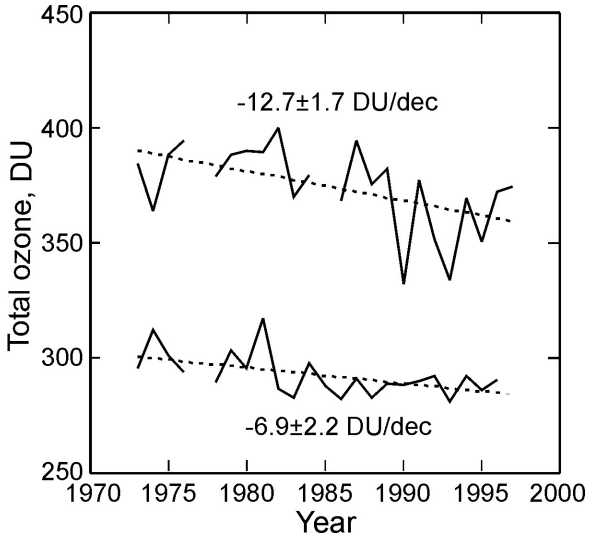

Fig. 9: Long-term trend in the total ozone change over Kyiv.

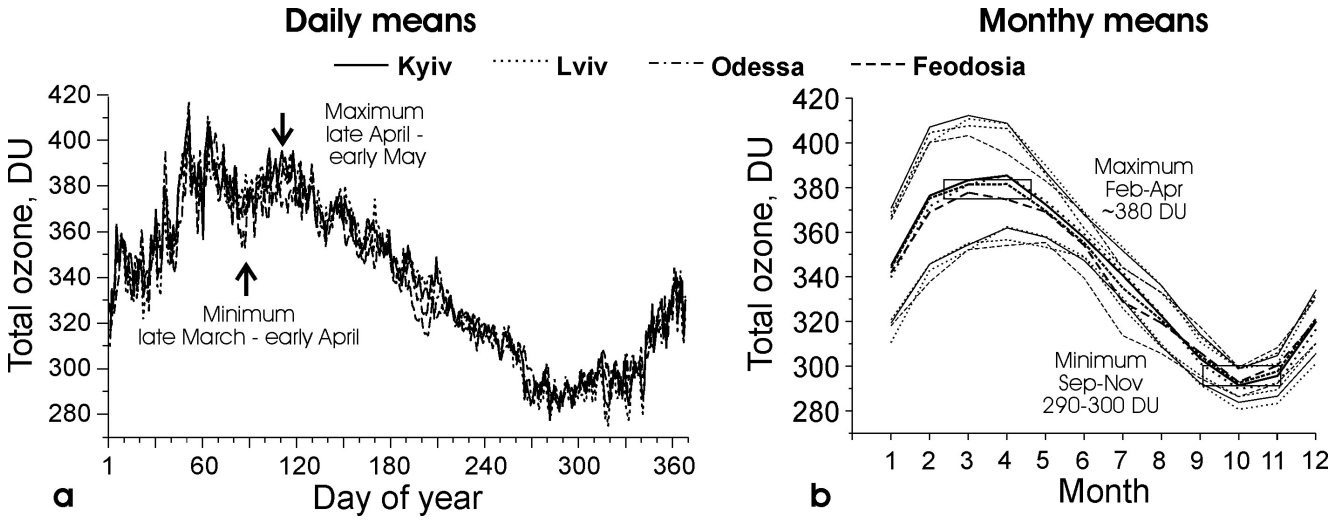

Fig. 10: Total ozone climatology for four Ukrainian stations by the Nimbus-7 TOMS V.8 Archive Overpass, 1979–1993: (a) daily mean values and (b) monthly mean values.

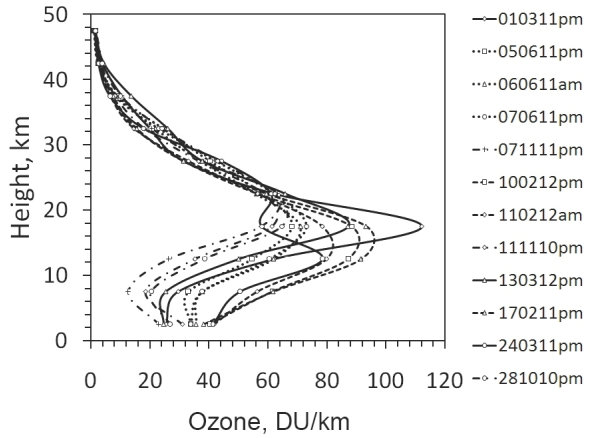

Fig. 11: Altitude ozone profile over Kyiv-Goloseyev station.

Recent TOC data from the Dobson measurements at the Kyiv-Goloseyev site in 2010-2012 (for example, thick solid curve in Fig. 8 for 2011) demonstrate generally lower TOC values than in the 1980s–1990s (by about 20 DU). This is evidence of the long-term ozone decrease observed in global ozone data. Fig. 9 illustrates decadal decrease of the total ozone over Kyiv in 1973–1997, estimated for the seasons of TOC maximum and minimum. The months of the seasonal TOC maximum (February-March, upper curve) and minimum (October-November, lower curve) are presented in Fig. 9. The figures show that historical and continued daily observations of the TOC variations over Ukraine can be important for analysis of the regional ozone distribution change.

VERTICAL OZONE DISTRIBUTION

Altitude ozone profile measurements serve as a data source for the investigation of atmospheric dynamics, as well as vertical ozone transport associated with Brewer-Dobson circulation. A study of altitude variations of ozone concentration above Kyiv was carried out in 2005–2008 using ground-based Fourier Transform InfraRed (FTIR) spectrometric observations [13].

In May 2010 our team started an Umkehr-method observation of vertical ozone distribution in the stratosphere over Kyiv, using the Dobson spectrophotometer, to explain the origin of the ozone anomalies (see e. g. Fig. 10). The Umkehr method is based on the phenomenon of changes in ultraviolet radiation absorption by stratospheric ozone, varying with wavelength and zenith angle. The absorption ratio between weakly- and strongly-absorbed wavelengths is used as an indication parameter. Observations at large zenith angles show that this value first increases and then starts decreasing, which is caused by the Earth's sphericity and the irregularity of vertical ozone distribution. This technique involves observations according to a specific schedule of various Sun zenith angles during sunset/sunrise. TOC values from standard Direct Sun measurements were used for ozone profile calculation. Observation data were processed using the WOUDC software, with the assumption that the Earth's atmosphere is divided into 10 homogeneous layers, using the Dobson C-Umkehr retrieval algorithm [11]. The software outputs a set of values depicting the ozone distribution by height, averaged through the period of observation. The vertical profiles of atmospheric ozone for four seasons of Umkehr observations are shown in Fig. 11. The observation dates are listed on the right-hand side of the plot in Fig. 11. The winter profiles (February) have the lowest height of ozone maximum, at about 15 km, while spring profiles (March) exhibit stronger variations in ozone

density and height (solid lines in Fig. 11). An interesting feature of the spring vertical ozone profile is the double maximum on March 24, 2012. The maximum ozone densities of the summer and autumn profiles (at approximately 18–19 km) are nearly at half the value as for the other two seasons. It should be noted that total ozone retrieval at Kyiv-Goloseyev is carried out with a mean ozone layer height of 21 km using the corresponding algorithm.

The properties of vertical ozone distribution are influenced by seasonal variations of meridional ozone circulation (descending branch of Brewer-Dobson circulation), while horizontal ozone transport is mainly influenced by planetary waves and stratospheric polar jets. Local factors, such as synoptic vortices in the troposphere reaching the tropopause level, as well as global tendencies, can influence ozone distribution as well. All of these factors constitute the objective for our investigations.

CONCLUSION

The analysis shows that the atmospheric aerosol pollution over Kyiv/Lugansk sites is typical for urban sites in Europe, with dominance of the fine mode aerosol particles. The territory of Ukraine is characterized by aerosol optical thickness ranging from 0.05 to 0.15 at wavelength 870 nm. The transportation of aerosol from strong forest and peat fires in Western Russia and Northern Ukraine in mid-July – August 2010 caused the AOT to increase to 0.2–0.4 and up to 0.7 in the Lugansk region. Seasonal peaks of AOT in April-May and August-September are explained by infrequent agricultural fires, transportation of Saharan dust over Eastern Europe, harvest time, and forest/peat wildfires. Maxima of AOT value over investigated areas was observed in May 2006, August 2007, April 2009, and August 2010. More results of aerosol data retrievals are available at the project's web-page[5].

The total ozone measurements at the Kyiv-Goloseyev station give reliable data for Direct Sun and Zenith Blue observation cases. The seasonal variations between satellite and ground-based TOC values have been noted. The largest differences are observed during winter period, which are caused in particular by Dobson total ozone underestimation at large zenith angles. The mean TOC values over Ukraine in 2010–2011 (from satellite and Dobson spectrophotometer measurements) are lower than the mean data for 1973–2002 by about 20 DU (filter ozonometer measurements). In October 2011 very low TOC values close to 220 DU were observed, that shows the ozone mini-hole condition over Ukraine. Ozone height profiles derived from Umkehr observations using the standard WOUDC software show the mean maximum value of ozone layer at about 18 km, that is approximately 3 km below of the height of ozone layer currently used for TOC calculations. The resulting aerosol parameters and observations of ozone behaviour will be used for evaluation of radiative forcing, and to study the impact of atmospheric aerosol on regional climate variations. The experience and knowledge obtained in the recent atmosphere research is currently used for development of an aerosol-related space mission that has been started in Ukraine.

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