Proc. Estonian Acad. Sci. Biol. Ecol., 1998, 47, 3, 188–203 https://doi.org/10.3176/biol.ecol.1998.3.02

# FIRST FOUR YEARS OF ATMOSPHERIC TOTAL OZONE MEASUREMENTS IN ESTONIA

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Received 27 February 1998, in revised form 26 March 1998

**Abstract.** The measurements of atmospheric total ozone content were started in Tartu Observatory at Tõravere in 1993. A laboratory spectrometer with coelostat mirrors was used. The main results obtained in 1994–97 are presented. The reliability and the accuracy of the measurements and retrieval are discussed. Significant negative deviations from the long time mean have been recorded until the end of April due to the longitudinal peculiarities of the annual cycle. In winter the vertical amplitudes of the planetary scale Rossby waves govern the variations of total ozone. In the summer season a pattern of tropospheric highs is a key factor.

Key words: total ozone, spectrometer, solar ultraviolet, stratospheric pressure and temperature, Rossby waves.

#### **INTRODUCTION**

In 1993 the measurements of atmospheric total ozone (the effective thickness of the ozone layer) were started at Tõravere (58°16' N; 26°28' E) and since 1994 they are carried out regularly. This is a necessary stage in the preparation for the UV-B irradiance monitoring as the stratospheric ozone is the most important factor modulating the solar UV-B irradiance, and the atmospheric total ozone data are urgently needed for the interpretation of UV-B irradiance variations. Otherwise the variations of stratospheric and tropospheric origin in total ozone cannot be separated. Regular measurements of total ozone have never been carried out in Estonia before. Rough information about previous situations can be obtained using published global surveys, which are available since 1957 (London et al., 1976).

As there were no financial possibilities to obtain a Brewer spectrometer, the main instrument used in the World Ozone Network today, an existing laboratory double monochromator spectrometer SDL-1 was suited for these measurements. This looks like a step back to the very first observations of the atmospheric total ozone, including the first observations by Dobson, the initiator of global scale total ozone monitoring. An excellent review of the prehistory of total ozone studies has been written by Dobson himself (Dobson, 1968a). Already in 1926 he constructed a special double monochromator in optical wedge scheme for the total ozone monitoring. The major idea of the total ozone measurements proceeds from the differential absorption of solar radiation by ozone on selected wavelengths. There is strong absorption on one chosen wavelength and comparatively weak on another.

The main advance of the Dobson spectrometer was a possibility to carry out the total ozone measurements in optically unstable weather. Instead of photographing or serial recording of the full spectral interval under interest the ratios of irradiance or sky radiance on two selected wavelengths are recorded. This can be done much more quickly. The values of ozone absorption coefficients and atmospheric scattering coefficients on the Dobson wavelengths have been retrieved from laboratory measurements of the ozone absorption and theoretical calculations of atmospheric molecular scattering and then recommended for use in global total ozone network by World Meteorological Organization (WMO). The recommended values of ozone absorption coefficients were corrected twice during the operation of the world network. The new values were adopted for use since January 1992 (Komhyr et al., 1993), and all the previous total ozone data have been corrected. The results of the present work are based on these new values.

The major difference of the present work as compared to the measurements with the Dobson spectrometer is the continual recording of solar monochromatic irradiance values 300–340 nm. In 1994–97 about 500 spectra were recorded yearly on clear days since February to November. The total ozone values are expressed in Dobson units (DU), which correspond to millicentimetres. The slow and the rapid variations as well as different amplitudes of the total ozone variations were registered. A high variability was recorded every year until the end of April. In both odd years, 1995 and 1997, the winter–spring type anomalies lasted until the end of the first decade of May. In both even years the summer regime of the recorded total ozone behaviour started earlier. Both negative and positive total ozone anomalies occurred. The explanation requires understanding of atmospheric processes leading to the total ozone deviations from the seasonal mean value.

Among the reasons stimulating local measurements of atmospheric total ozone the study of long term trends is not the most important as the existing global data set containing both the ground based and space observations serves as a major source for this and the weight of observations from one location is small. Local observations are more valuable for the study of impacts and responses connected with irregularities of the ozone layer.

# THE INSTRUMENTATION

A schematic diagram of instrumentation is presented in Fig. 1. A double monochromator spectrometer SDL-1 (LOMO, St. Petersburg) is the major part of the instrumentation. The spectrometer weighs 600 kg and is installed in a laboratory on a ground floor having a window in the northern sector. The first problem is the guiding of the light beam (solar or the zenith sky) onto the entrance slit. In direct sun measurements it is realized using a heliostatic drive for solar tracking and two auxiliary mirrors. Quartz lens and a shutter are installed immediately in front of the entrance slit. In direct sun measurements the distance between the first mirror of the heliostat and the entrance slit of the spectrometer is about 20 m and the solar beam is reflected by three mirrors before reaching the



Fig. 1. Instrumentation for ozone measurements.

slit. The long distance makes the optical path very sensitive to the rotation errors of the heliostat as well as to the errors in the direction of the rotational axis. In zenith sky measurements only one mirror oriented under 45 degrees has been used. The universal spectrometer SDL-1 allows recording the spectra in the wavelength interval 200–500 nm with a grating 1200 grooves/mm and a photomultiplier (PM) FEU-39A (originally a detector). The instrumental dispersion in this region is 0.8 nm/mm. For ozone measurements an interval 300–340 nm was scanned regularly in both directions. This enables compensation for the errors caused by differences in solar elevation on the used Dobson wavelengths (305.5, 317.6, 325.4, and 339.8 nm). The time interval needed for each scan is approximately 2 min in one direction. Such a long time practically excludes the cloudy sky measurements due to rapid changes in absorptance and reflectance of clouds.

Using the originally installed PM FEU-39A, a signal to noise ratio occurring on the shortest wavelengths was much smaller than on longer wavelengths. This contrast was reduced by replacing this PM by a solar-blind FEU-142, whose response decreases rapidly with the increase of wavelength. Spectral responses of both multipliers are shown in Fig. 2, illustrating the way the signal on longer wavelengths is suppressed. The resulting PM instability does not exceed 0.05%. For recording photocurrent, a stepped current–voltage converter has been built using a special low-ohm voltage divider. An interface connecting the amplifier output with the PC has been built. The used analogue-digital converter (ADC)





ADC-6.5 has been developed by AS Enari Ltd, Tallinn (now Märtens & Trampärk Electronics, Tallinn). It is a 22-bit integrating ADC for digitizing 0–10 V voltage signals. The instrumental spectral response was calibrated using a certificated standard lamp.

# ACCURACY AND RELIABILITY OF THE MEASUREMENTS

The uncertainty of the ground based total ozone measurements using the Dobson's retrieval algorithm (Dobson, 1957) depends on the instrumentation errors, on solar elevation, and to some extent on the real vertical distribution of atmospheric ozone.

The instrumental uncertainty is dependent on the stray light level inside the spectrometer, the signal to noise ratios, the wavelength calibration, guiding errors, and the accuracy of the determination of spectral response. No imprints of a significant presence of stray light have been noticed. After the change of original PM to the solar-blind type the stray light safety increased. The wavelength scale has been checked by emission lines of a mercury lamp and by the positions of several evident details of the recorded spectrum during routine use of the instrument. The deviations of the positions of the mercury lines from regularly (after every 2.5 nm) recorded wavelength labels did not exceed 0.15 nm. The recorded spectra present a number of characteristic minima and maxima. whose centres have not floated more than 0.3 nm relative to the reference scale. The relative spectral response has been measured using a quartz halogen lamp Optronic DXW-1000, certificated traceable to NIST. The correction factor of the sensor varies between the shortest (305.5 nm) and longest (339.8 nm) Dobson's wavelength from 1.87 to 1.23 and it is measured with an accuracy of 2%. The guidance stability was checked carefully during the measurements to avoid errors by nonuniform illumination of the entrance slit. It was done by checking the light spot position on the last mirror and around the entrance slit.

One of the serious difficulties in the total ozone and the UV irradiance measurements is connected with the large diapason of the recorded spectral irradiation  $I_{\lambda}$  values. The total ozone retrieval has been performed using the Dobson wavelength pairs A (305.5 and 325.4 nm) and D (317.6 and 339.8 nm) as described in (Dobson, 1957) with the values of coefficients corrected in 1992 (Komhyr et al., 1993). The ratio of signal values on two A wavelengths is more than 20 and in the cases of low Sun the signal error on the shorter wavelength of the A pair is the most important factor limiting the accuracy.

The uncertainty of measurements with Dobson instruments has been discussed in several publications. For example, Basher (1982) and Degorska &

Rajewska-Wiech (1989) underlined that the total ozone values measured at high atmospheric masses are dependent on solar elevation and have significant day-to-day variations explained with the variable contribution of atmospheric scattered radiation from solar aureole. The so-called atmospheric stray light problem in our measurements is similar to that for the original Dobson instrument. The measurements were carried out at as small as possible solar zenith angles to suppress this uncertainty.

In the retrieval algorithms it is expected that the vertical distribution of ozone in the stratosphere corresponds to the yearly averaged one. Often the deviations of total ozone from the seasonal mean occur due to the anomalous shape of its vertical profile. In pressure-created anomalies over the tropospheric highs the layers above are lifted up and the ozone becomes diluted. The total ozone increase happens over the tropospheric lows. In the case of horizontal displacements of the air from the tropics or from the polar latitudes ozone rich or ozone poor air appears only in thin layers called the laminae. Usually the upper part of the vertical profile of ozone remains undisturbed. As a result, both the effective height and the effective thickness of the disturbed layer can be changed. In Estonian latitude the maximum of the stratospheric ozone layer is located in the interval of the geometric height 20-22 km. When the real height of the ozone maximum differs from the supposed mean 2 km, the calculated total values at low solar positions will contain significant uncertainty (16 DU at  $z = 75^{\circ}$ ). At  $z < 60^{\circ}$  the uncertainty is less than 3 DU (z designates solar zenith angle).

The uncertainty of the measured values of total ozone depends significantly on the solar zenith angle and increases rapidly at  $z > 60^{\circ}$ . If the measurement is carefully checked and the weather is stable, the error at  $z < 60^{\circ}$  will not exceed 5 DU, but on solar zenith angles  $z > 75^{\circ}$  it reaches 20 DU. Consequently, the measurements of atmospheric total ozone at high UV irradiation values in the summer season (since the middle of March up to the beginning of October) have higher accuracy than those made in winter. For measurements at  $z < 60^{\circ}$  the mean standard deviation of our measurements is slightly more than 1% of the value or less than 5 DU.

In the ozonometric stations that use the Dobson and at present mostly the Brewer instruments most of the measurements are carried out recording the direct solar irradiance or the zenith sky radiance. The clear zenith sky measurements are calibrated using statistical regression with the quasisimultaneous direct sunlight measurements. The derived empirical relationships are presented in the form of the so-called zenith sky charts. We have checked the correspondence of our chart compiled using the simultaneous direct sun and zenith sky measurements data 1994–95 to the widely used Canadian and European (for Uccle) charts (Komhyr, 1960; De Muer & De Backer, 1992). The coincidence



Fig. 3. Correspondence of the Tõravere  $N_{AD}$  values to the widely used zenith sky charts for total ozone values 300, 350, and 400 DU.

illustrated in Fig. 3 is good for the interval of total ozone values of 300–400 DU. The measured values of the combined ratio

$$N_{AD} = \log I_{317.6} / I_{339.8} - \log I_{305.5} / I_{325.4} - 0.020$$

are compared to those of the above-mentioned charts. In most cases when clear sky measurements were carried out direct sun measurements were available and the present paper deals only with the latter as these are more precise. The comparison here illustrates the fact that our results, collected using a nonstandard instrument, correspond fairly well to those collected using standard instruments.

### MAIN FEATURES OF THE TOTAL OZONE ANNUAL CYCLE

The results of the total ozone measurements made at Tõravere in 1994–97 are presented in Fig. 4. In January–February as well as in November–December the weather conditions and the high uncertainty of measurements do not allow regular data collecting and their detailed analysis.











Figure 5 presents the longitudinally averaged annual mean curve of the total ozone for the northern latitude 58° and the mean curve for the same latitude interpolated for the Estonian longitude (25° E). Both curves were constructed using the published monthly mean total ozone maps and corresponding gridded data for 1957–67 (London et al., 1976). The third curve in Fig. 5 presents the observed mean at Tõravere in 1994–97. The current monthly mean values in winter tend to be systematically lower than those for the past chemically undisturbed atmosphere with the smaller content of greenhouse gases. The current seasonal curve shows bite-outs from the seasonal mean ozone curve of 1957–67 since January to the middle of April or even the first decade of May.



Fig. 5. Comparison of the Tõravere total ozone mean annual cycle for 1994–97 to that for 1957–67. The latitudinal mean for 58° N as well as the mean for the Estonian area are presented.

The large-scale features of the annual cycle of total ozone are dependent on the meridional transport intensity from equatorial to polar latitudes. This downward-poleward transport from the tropics occurs on heights above the

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subtropical barrier (Plumb, 1996). In the winter-spring season higher values of total ozone are observed because the ozone rich air is transported at lower heights. At the same time the winter-spring season is connected with the presence and breaking of the polar vortex – a system of strong circumpolar cyclonic winds. In the air exchange between the vortex and its surroundings the direction out of the vortex prevails. The filaments of the ozone poor (if the air is chemically treated in the cold vortex) or ozone rich air are reaching lower latitudes through the barrier of strong winds (Varotsos et al., 1994; Newman et al., 1996; Mlch & Laštovicka, 1996).

The longitudinal distribution of the total ozone around the winter vortex is believed to be governed by the vertical amplitudes of the planetary waves with wavenumbers 1-3 (Randel, 1993; Wirth, 1993), determing the preferred flow regimes of the stratosphere at the same time (Pawson & Kubitz, 1996). As a result, the longitudinal distribution of atmospheric total ozone shows significant asymmetry. The region of higher values of the column ozone is concentrated in the American-Pacific sector and the region of lower values in the European-Atlantic sector. In the relatively stable summer stratosphere the amplitudes of total ozone deviations from the long-term seasonal mean are smaller than in winter and spring when large contrasts have been recorded every year. In the summer season several events of extraordinarily low total ozone (deviation about 20%) over Estonia were recorded in 1995 and two such events in the summer of 1997. In both years the equatorial stratospheric winds were westerlies. All the 1995 summer events of low total ozone occurred dynamically in a similar way. In the even years 1994 and 1996, when easterlies blew in the tropical stratosphere, no significant negative deviations from the seasonal mean were noticed. All the deviations from the mean are created by the movement of stratospheric air of different ozone content or are pressure modulated. More than a half of the moderate variations of total ozone in summer are usually believed to be pressure modulated (Dobson, 1968b; Salby & Callaghan, 1993). It means that vertical displacements of stratospheric air are responsible for the variations of total ozone.

Due to frequent deviations towards lower total ozone values the contemporary annual curve of the total ozone over the Baltic longitudes differs from the latitudinal seasonal mean. The weighted maximum value is lower and has shifted closer to summer. The mean value of the total ozone by our measurements for every April was lower than that for May. This difference was less than 15 DU in 1994 and 1995 and a little over 30 DU in 1996 and 1997. In 1957–67 the annual maximum occurred at the end of March or at the beginning of April. Our measurements show that this maximum has shifted to May. Looking on the NOAA TOVS (National Oceanic and Atmospheric Administration TIROS Operational Vertical Sounder) atmospheric total ozone distributions available now through www (http://nic.fb4.noaa.gov/products/stratosphere/tovsto), it can be easily noticed that the longitudinal distribution is highly asymmetric especially in winter and spring.

Extremely low total ozone values in northern middle and high latitudes were recorded before our measurement cycle in 1992 and 1993 due to high stratospheric aerosol burden after the Mt. Pinatubo eruption (Bojkov et al., 1993; Kerr et al., 1993; Komhyr et al., 1994). By 1994 the excess aerosol content of this "century eruption" had lowered and since 1995 the stratospheric aerosol burden can be considered as corresponding to the current background aerosol. The stratospheric winters 1994/95 and 1995/96 have been classified to the extremely cold ones (Naujokat & Pawson, 1996). However, the long-term cooling trend of the stratosphere permits regarding these winters as normal ones before the warm 1997/98 winter due to the forcing by the El Niño event.

The most stable total ozone values occurred in August. The differences of yearly August mean from the 4-year August mean did not exceed 6 DU. The total number of recorded values was 10 in 1994, 17 in 1995, 15 in 1996, and 23 in 1997. In other summer season months the monthly mean values in 1994 and 1995 were systematically lower than in 1996 and 1997. The highest monthly mean values since April to July were recorded in 1997. The variance of recorded total ozone in summer months since May to the autumn equinox was smaller in the even years 1994 and 1996 when no large deviations were noticed. In 1994 three longer and one short period of negative deviations were recorded during March and April, and after that a deep anomaly was noticed only in October. All summer deviations from the smoothed mean were moderate, not exceeding a few per cent.

Since 1995 the TOVS total ozone global distributions have been available and studied in periods of the recorded anomalies. In 1995 a deep anomaly was recorded in March and then a deep short anomaly on 24 and 25 April. Four more deep anomalies were recorded on 14 June, 8 July, 1 September, and around 20 September. In 1995 the atmospheric activity during the summer period seemed to be higher than in other years. Small shallow holes often formed in the ozone layer along the northward directed low ozone waves over high pressure ridges. The April low ozone event was created by one such "hole" located over the Baltic and generated by two low ozone strips extending northward from the Mediterranean Sea and from the Caspian Sea. A similar ozone hole was formed on 13 June over the Gulf of Finland. The value of the total ozone decreased about 90 DU on 12 to 14 June. At that time a very stationary long strip of low total ozone in the Indian longitude and an extended area of high total ozone over the Northern Atlantic were present. The Atlantic high pressure wave could not extend directly towards higher latitudes and turned steeply eastward forming an elongated area of low ozone from central Norway to St. Petersburg. The anomaly recorded on 8 July was formed in guite a similar way. In the summer of 1995 those situations were highly stationary. At the same time three such holes were

present in different regions for example on 12 July. The hole observed over Estonia was caused by the powerful low between Iceland and Ireland forcing the Atlantic high pressure ridge into the east while another high pressure ridge blocked its further extension. We failed to record at least one similar anomaly in August due to cloudy weather. In September the eastward movement of the Atlantic high pressure air towards the Baltic was blocked by the powerful high pressure wave in the Caucasian longitude. This air had partly turned towards the Baltic (another branch towards the Taimyr) and was responsible for very low ozone over Estonia on 1 September. The low ozone event since 19 September can be explained by three ozone poor air strips that reached simultaneously the high latitudes (the first in the Atlantic, the second in the Baikal and the third in the Alaska longitude). These strips formed two low ozone areas. One of them extended from Scandinavia to the Taimyr and included Estonia. In October the accumulation of ozone rich air over the middle and high latitudes of the western hemisphere began and lasted until the breaking of the polar vortex in April.

In 1996 deep ozone anomalies were recorded in early March and in the first half of April. After that no deep anomalies have been noticed before October. In June and July the weather was often cloudy and the measurements could be carried out only on a small number of days. In 1997 a long period of low ozone occurred in February and March and a shorter one in the middle of April. Comparatively low values were recorded during the first half of May. After that high values were recorded until the end of August. One short negative deviation (-18%) occurred on 9 June. It was formed by the strengthening of the high pressure ridge from the Mediterranean Sea towards Scandinavia and the deepened low ozone area moved slowly over Estonia.

The short variations of total ozone of smaller amplitudes are believed to be pressure dominated like most of the large variations. It has been known for a long time that the total ozone maxima tend to occur at the passage of a cold front of a cyclone and the ozone minima can be found in the south-west sector of an anticyclone (Götz, 1951). The influence of tropospheric pressure changes on the stratospheric column ozone are dependent on the vertical extent of the pressure anomalies. Correspondingly the stratospheric column ozone changes must be more sensitive to the pressure contrasts in the upper troposphere than to those on the ground level.

We have studied the correlation of the measured column ozone changes with the ground level pressure changes using the pressure data recorded in the actinometric station of the Estonian Institute of Meteorology and Hydrology located at Tõravere. The correlation was calculated separately for every summer period since late April until the autumn equinox. The correlation of pressure changes with the column ozone value changes was searched in time intervals of 1–2 days if the pressure had decreased or increased monotonously. During fine weather periods both the pressure changes and the total ozone changes were small. The best correlation (correlation coefficient -0.44) was found for the summer of 1997 and the second best (-0.30) for the summer of 1994. For the following two summers the correlation coefficient was lower than -0.20. For 1997 a higher correlation was consistent with the low activity of planetary waves. The stratospheric air was not significantly disturbed by waves and so the pressure changes on smaller scales could modulate the total ozone.

For day-to-day changes in the total ozone values exceeding 20 DU the correlation with the 50 hPa temperature has been studied using the stratospheric data base of the Free University of Berlin available through www. In the summer period when the ozone content in the stratospheric air can be considered conserved, the descending and ascending of air takes place fairly adiabatically. So the temperature of the ascending air is decreasing and that of the descending air is increasing (Stanford & Ziemke, 1996). Possibly due to the relatively high generalization of the stratospheric temperature maps, no day-to-day temperature deviations exceeding 1°C were found in the summer stratosphere over Estonia in the studied cases.

### STRATOSPHERIC OZONE MODULATION OF UV-B IRRADIATION

There is a worldwide need not only for continuous measurements but also for the prediction of UV-B irradiance. Because the UV-B irradiance is most strongly affected by the stratospheric ozone, understanding of the mechanisms of stratospheric ozone variations in every specific geographical region is of utmost importance. The ground level UV irradiance consists of direct solar irradiance and of scattered irradiance. The direct irradiation is attenuated by the stratospheric ozone and by molecular scattering enhanced by aerosol scattering. In the UV-B region (280–320 nm) ozone is the dominant attenuator. The threshold above which molecular scattering dominates is roughly on the wavelength 310 nm. It is determined by strong absorption in the Hartley band (200–310 nm with maximum absorption at 255 nm).

The recorded spectra for the ozone retrieval correspond to the solar UV irradiance in arbitrary units and can be used for studying the dependence of direct irradiance on columnar ozone. As a first step of such a study the ratios of irradiance on different wavelengths were calculated. The dependence of the ratio  $I_{305}/I_{320}$  on the solar zenith angle for three different total ozone values are presented in Fig. 6. For all three nominal values the real values in the interval  $\pm$  15 DU were included. The dependence of the ratio on the solar zenith angle is close to linear. The higher values correspond to the lower total ozone. In June



Fig. 6. Dependence of the direct solar irradiation ratio  $I_{305}/I_{320}$  on the solar zenith angle, z, for total ozone values 280, 350, and 420 DU.

values of the total ozone from 285 to 410 DU were recorded in the course of our measurement cycle. This means that the values of direct  $I_{305}$  can differ about 2 times.

#### ACKNOWLEDGEMENTS

This study was supported by the Estonian Science Foundation (grants Nos. 177 and 2008). Appreciation is expressed to NOAA for the availability of TOVS total ozone global distributions and to the Free University of Berlin for the permission to use their stratospheric database.

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# ATMOSFÄÄRIOSOONI KOGUHULGA MÕÕTMISE ESIMESED NELI AASTAT EESTIS

# Kalju EERME, Uno VEISMANN, Rutt KOPPEL ja Matti PEHK

On antud ülevaade aastatel 1994–97 Tõraveres (58°16' N; 26°28' E) tehtud atmosfääriosooni koguhulga mõõtmisest ja saadud tulemustest. On analüüsitud mõõtmise täpsust, selgitatud muutlikkuse üldtendentside põhjusi ja välja toodud suuremate registreeritud anomaaliate põhjused.

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#### INTRODUCTION

The matural distribution area of Actimizita Valoraile? (MaXimPole Repri-Maxim 18 the for fast (Rasson Primor's fruit Protth Robeit/Sikhafh) China (Reltar, 1949). In Estonia it has been cultivated as a decorative jithing but it deserves attention as a berry plant as well (Elliku, 1985). However, its use for the initiar purpose is inindered by its unisexuality. In 1972 more than 200 two to three year old plants of A koloratiza were brought to Estonia from the Far East (from the victarity of the Bounical Garden of Madiyostok). One of the plants furned out to be bisexual. According to the present localization of the plant in north eartern Estonia it has got the victetal name. Jiseku', Beginning from 1976 the plant has invetified richly every year including the augments after, the victor of the the transfer of 1978/79 and 1986/87 when the famerature in the north east of