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## High resolution solar spectrophotometry and narrow spectral range solar radiation measurements at the Hungarian Meteorological Service

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**Abstract**— Aims of the spectral radiation measurements can be devided to two wider areas: one is to get information about the radiation source, and the other is to get information about the properties of the space between the radiation source and the detector if output signal from the radiation source is known. In the latter case either the optical properties of the certain space or some optical parameter of an object placed in there is to be studied. The sun can be the object of the study or it can be used as natural radiation source to investigate some important properties of the atmosphere. The term 'solar spectrophotometry' refers to this.

Although detection of spectral distribution of the solar radiation is considered a special area that is relatively rarely used even today in atmospheric physical measurements, it still has big significance. In addition to the 'mere' knowledge of spectal solar irradiance, the measured data can be used in a considerably wide range. In special cases, the narrow spectral range informations about the radiation can be very useful. Typical example is the erythemally weighted UV radiation. Though it does not give spectral information, the spectral range that is characterized by it, is considerably narrower than that of the classical radiation components. So this type of measurements is also discussed here.

Main applied physical and technical principles of solar spectrophotometry, as well as spectrophotometers working in the UV, visible, and near infrared spectral range used at the Hungarian Meteorological Service (HMS), are shown in this paper. Measurement results and results from studies and researches using these data are also shown and analyzed. Also some special studies performed occasionally are shown.

Today the primary base for operation of high accuracy measurement systems is the calibration. Since we have reference instruments, QA/QC procedures are of crucial importance in our measuring practice. Our activitiy as we operate WMO Regional Center for Solar Radiation in Region VI gives even bigger emphasis to that.

*Key-words*: solar radiation, measurement technics, spectrophotometry, optical depth, total ozone, UV radiation, calibration

#### 1. Introduction

Solar radiation measurements in a station mean, in most cases of solar radiation practice of meterorological observations, the measurement of components of classical radiation budget of the atmosphere. Use of narrow spectral range or high resolution spectral solar radiation measurements can be considered relatively rare even today, despite the fact that it is not a brand new technique. The main reason is that it requires suitable special expertancy and special, rather expensive equipments that cannot be provided easily by institutions.

Narrow range and high resolution measurements of solar radiation have started at the Hungarian Meteorological Service (HMS) in the early nineties of the previous century by installation of ultraviolet detectors measuring erythemally weighted UV radiation and continued with installation of two high resolution spectrophotometers, then later by sunphotometers. It is important to note that primary acitvity of the HMS is to operate monitoring networks and establishing quality controlled databases. Consequently, the studies or researches concern different processing of data and analysis of processed data mostly and, only in a few case, some other 'more scientific' job in area of solar radiation measurements also.

The operation, and physical and technical background of the aforementioned systems, as well as some results from studies and researches based on their data are also shown in this paper. To detailedly describe the operation and technical background cannot be object of this paper, so they are concerned very briefly only.

## 2. Background physics

The background physics is not discussed in details because it can be found in numerous books and articles concerning theoretical and applied physics. The basic physics is the theoretical and experimental approaches of radiative transfer that can be performed in many ways depending on the type of the problem to be solved. It is, by all means, to be noted that the basic terms and definitions concern monochromatic radiation. It is, however, to be stressed that when monochromatic flux is mentioned in measurement technics, it means a flux belonging to very narrow, but finite range, because finite quantity cannot belong to a zero interval in reality. So the monochromatic quantities are only quasimonochromatic ones, actually. Consequently, if the wavelength of a spectral irradiance is given, the irradiance refers to a spectral band whose center is at the wavelength in question. Mathematically it can be expressed as follows:

$$I_{\lambda}^{q} = \int_{\lambda-d\lambda}^{\lambda+d\lambda} I_{\lambda}d\lambda , \qquad (1)$$

where

 $I^q_{\lambda}$  is the quasi-monochromatic flux,

- $I_{\lambda}$  is the theoretical monochromatic flux,
- $\lambda$  is the wavelength of the center of the spectral band,
- $\Delta \lambda$  is the half-width of the spectral band.

The narrower the band, the more accurate the flux referring to the given wavelength.

If the aim of the measurement is to get information about the space or, in atmospheric physics practically, a medium, or any component in the atmosphere, also the spectral flux at the top of the atmosphere (the so-called extraterrestrial flux) should be known. Considering the extraterrestrial flux and the flux measured at the surface, the total amount of a given component can be determined, obviously in the case when values of other physical quantities are known from measurement or calculation.

# 2.1. Determination of total columnar amount of atmospheric gaseous components

The calculation methods are well and fully described in several books and guidebooks, so they are mentioned here very briefly, and also some special physical quantities or methods are explained in more details. Determination of gaseous components using solar spectrophotometry is performed by the method of relative intensities (Dobson, 1957). The main principle is described here very briefly only, without quantification. This main principle is that the irradiance is measured at several wavelength pairs so that one of the pairs is at a wavelength where the absorption coefficient of the gas is considerably high and the other one is at a wavelength where the absorption coefficient is as low as it can be taken as zero. An important criterium is that the two wavelengths have to be very close to each other, so a spectral interval is needed where the absorption coefficient varies by high rate. The reason is that the properties of the aerosol being present in the atmospheric column between the solar disc and the detector at the time of the observation is not known. It is well-known, however, that the variation of the aerosol optical depth with wavelength can be described in each case by a smooth and non-rapidly changing function. Consequently, its effect on the calculated value of optical depth of the gaseous component, whose total columnar amount is to be determined, can be neglected (it means mathematically that the aerosol optical depth, as variable, is eliminated from the equation).

## 2.2. Determination of aerosol optical depth

Calculation of aerosol optical depth (AOD) is also a well-known standard method and described in several publications (*Alföldy et al.*, 2007), so the reduction of the formula by which it is calculated is not shown here. The AOD is calculated by the equation as follows:

$$\delta_{A\lambda} = \frac{1}{M} ln \frac{I_{0\lambda}}{I_{\lambda}S} \left( \frac{P}{P_0} \delta_{R\lambda} + \delta_{O\lambda} \right), \tag{2}$$

where:

 $\delta_{A\lambda}$  is the aerosol optical depth at wavelength  $\lambda$ ;

 $I_{0\lambda}$  is the extraterrestrial irradiance;

- $I_{\lambda}$  is the irradiance at the observational point;
- *S* is the correction factor for the Earth-Sun distance (ratio of Earth-Sun distance at the time of the measurement to the mean value);
- *M* is the relative optical airmass;
- $\delta_{R\lambda}$  is the optical depth of Rayleigh scattering from atmospheric molecules;
- $\delta_{O\lambda}$  is the optical depth of ozone absorption:  $\delta_{O\lambda} = \alpha_{O\lambda}\eta$ ; where  $\alpha_{O\lambda}$  is the ozone absorption coefficient and  $\eta$  is the total ozone content of the air column between the solar disc and the detector;
- $P, P_0$  are the surface pressure and at the time of the observation and standard sea level pressures.

To obtain  $\delta_{A\lambda}$ , value of  $I_{\lambda}$  is measured by solar spectrophotometer and *P* is known by measurement also. Total ozone content  $\eta$  either can be observed at the given site or known from satellite observations. Values of both  $I_{0\lambda}$  and *S* are known.

Since high accuracy spectrophotometric total ozone content data are available at the Marczell György Main Observatory, accurate estimation of AOD is possible to perform. The standard wavelengths are used to estimate AOD are 368, 380, 412, 450, 500, 610, 675, 778, 862, and 1024 nm, and since the autocorrelation of AOD values estimated for these 10 wavelengths is very high, AOD for 500 nm is used generally for studies and in models.

## 2.3. Determination of graybody (broad band) optical depth

Graybody (broad band) optical depth (GBOD) can be determined if definition of monochromatic optical depth is extended to a wider spectral range if irradiances measured at the surface are available (*Németh et al.*, 1996). Consequently, the GBOD will then be determined in the following way. If  $I_{\lambda 0}$  is the irradiance coming onto the top of the atmosphere at wavelength  $\lambda$  and  $I_{\lambda}$  is the irradiance measured at the surface by a pyrheliometer in case of relative optical air mass *m*, then:

$$\int I_{\lambda} d\lambda = \left( \int I_{\lambda 0} d\lambda \right) e^{-m\delta GB} , \qquad (3)$$

where  $\delta_{GB}$  is the GBOD and  $S_{PYR}$  is the sensitivity range of the pyrheliometer.

Thus GBOD is given by the following equation if direct irradiance (denominator of the fractional) is measured:

$$\delta_{GB} = \frac{1}{m} ln \frac{\int I_{\lambda 0} d\lambda}{\int I_{\lambda} d\lambda}.$$
(4)

Though graybody optical depth is not a spectral quantity, as it is evident from its definition, results from studies concerning it are important to include here due to, on the one hand, its very close connection with spectral aerosol optical properties and on the other, its good usability to characterize short-wave radiation transmission of the atmosphere.

#### 2.4. Ångström exponent

The Ångström exponent  $\alpha$  characterizes the particle size distribution in the aerosol being present in the air column over the measuring site at the time of the observation. Value of  $\alpha$  is approximetaly 1.3 for average normal size distribution. Values higher than 1.3 are resulted in by the relatively higher frequency of smaller particles as compared with the large particles having a radius greater than 0.5 µm. Values lower than 1.3 mean the relatively higher frequency of large particles (*Ångström*, 1929).  $\alpha$  exceeds 2 considerably, or it does not reach 0.4 or 0.5 only in extreme situations.

 $\alpha$  is determined by the following equation (*Alföldy et al.*, 2007):

$$\ln \delta_{A\lambda} = \ln \beta - \alpha \ln \lambda, \qquad (5)$$

where:

- $\delta_{A\lambda}$  is the aerosol optical depth at the wavelength  $\lambda$ ,
- $\beta$  is the Ångström turbidity coefficient,
- $\alpha$  is the Ångström exponent.

This practically means that considering  $\ln \delta_{A\lambda}$  as a function of  $\ln \lambda$ ,  $\alpha$  is the steepness of the curve.  $\alpha$  can then be determined by a reasonable linear fitting in that regression.

#### 3. Narrow spectral range measurements

#### 3.1. Biologically effective UV radiaton

Due to the global ozone depletion, accurate monitoring of biologically effective UV radiation has become stressedly important in the last decades, and after the atmospheric ozone has started to recover, it has remained important due to the fact that UV irradiation is not decreasing despite the ozone increase.

If one wants to know the exact biological effect of an irradiation on a biological system, the response of the biological system in question to the irradation is needed to know. It means, practically, the spectral sensitivity of the biological system. The function describing the wavelength dependence of the sensitivity is called action spectrum and it is consequently a weighting function. The biological irradiance for a given wavelength can be obtained if the measured spectral irradiance is multiplied by the value of the action spectrum for the wavelength in question. The biological effective dose,  $I_{eff}$ , can be obtained by the following equation:

$$I_{eff} = \int_{\lambda_L}^{\lambda_U} I_{\lambda} A_{\lambda} d\lambda , \qquad (6)$$

where

- $\lambda_L$  and  $\lambda_U$  are the lower and upper limit of the spectral range where values of the action spectrum are used,
- $I_{\lambda}$  is the irradiance at the wavelength  $\lambda$ ,
- $A_{\lambda}$  is the value of the action spectrum for the wavelength  $\lambda$ .

If a high resolution spectrophotometer is available for the observations, the biologically effective radiation is determined by Eq. (6). However, there are broad band UV detectors, the so-called UV Biometers, whose output is the biologically effective dose. Spectral response of the human skin to the UV radiation is called Erythema, so the biologically effective radiation in case of human skin is called generally erythemally weighted radiation. The biophysical background of these processes is not object of this paper, so it is not discussed in details. Fig. 1 shows the meaning of the aforementioned facts in reality. A physical spectrum (red line) and the corresponding erythemally weighted spectrum (blue line) can be seen in the figure. The spectrum is from our database of high-resolution UV spectra recorded by Brewer spectrophotometer at the Marczell György Main Observatory of HMS in Budapest. The importance of use of the response function in case when biological effect of a radiation is studied is clear based on the figure. Due to the very rapid variation of the Erythema function, the biological spectrum considerably differs from the physical one.



*Fig. 1.* Physical UV spectrum measured by Brewer MKIII spectrophotometer and the calculated corresponding erythemally weighted (biological) spectrum.

## 3.2. Photosynthetically Active Radiation (PAR)

PAR is a special quantity that means the biological effectiveness of visible radiation on photosynthesis of plants. Consequently, nature of PAR is the same like any other action spectrum weighted radiations, but it is expressed in a special unit.

## 4. Measuring equipments at the HMS

The equipments that are used for high resolution spectral measurements and special narrow range measurements at the Hungarian Meteorological Service are shown very briefly in this section. To describe their technical details and specifications, as well as principles of their operation cannot be object of this paper. Their instrument manuals include detailed informations about them (see references).

## 4.1. Brewer MKIII double monochrometer spectrophotometer

This instrument is a high accuracy spectrophotometer that is the most accurate and most reliable spectral equipment in the UV region today. Its sensitivity range is from 286.5 nm to 363 nm with a spectral resolution of 0.5 nm. It is equipped with a double monochrometer to increase the effectiveness of filtering of stray light that is very important due to the low irradiances to detect. A photomultiplier is used as detector to produce suitably high signal-to-noise ratio. The equipment measures the total air columnar ozone and sulphur-dioxide content and records global UV spectrum in the range and with the parameters mentioned above (for any other details, see *Brewer MKIII Spectrophotometer Operator's Manual*, 1998).

In our operational measurement practice, it observes total ozone and sulphur-dioxide, as well as records UV spectra by approximately 15–20 minutes (the reason why the mesurement frequency varies a bit is that the observations are carried out at fixed solar zenith angles, and it, in addition to the observations in question, performs several tests concerning its optics, electronics, and mechanics to provide the possible highest quality data).

The Brewer spectrophotometer was installed in 1998, but the high accuracy total ozone observations have been started at the HMS in 1969 with the manually controlled Dobson spectrophotometer, that has been the highest quality instrument to observe total ozone then.

The two spectrophotometers worked simultaneously for one and a half year until terminating observations with Dobson spectrophotometer due to lack of manpower. These observations are carried out the Marczell György Main Obervatory in Budapest.

## 4.2. LI-1800 spectroradiometer / spectrophotometer

This high accuracy instrument is constructed to record electromagnetic spectra in the spectral range from 300 nm to 1100 nm with a spectral resolution of 1 nm. The diffraction spectra are produced by a monochrometer. The instrument is basically designed as spectroradiometer, namely to measure global (full-sky) irradiance, but, by using a suitably designed pipe with diaphragms, it has been made to be suitable to measure direct irradiance. It can thus work as both spectroradiometer and spectrophotometer.

In our operational measurement practice, LI-1800 records spectra by 15 minutes or 30 minutes normally according to the schedule set by us. Measurements are carried out only in cases when solar disc is not covered by cloud because the aim of the measurements with LI-1800 is to calculate aerosol optical depth and Ångström exponent characterizing size distribution of the aerosol. AOD values are archieved for all standard wavelenghts, though generally AOD at 500 nm is used for studies due to the very high autocorrelation of AOD values in a certain spectrum.

LI-1800 has options to measure PAR and illuminance, and very special quantities like leave transmissivity, leave reflectivity, etc. These quantities are not measured operationally but occasionally in special campaigns or for orders. LI-1800 is installed at the Budapest observatory also (for more technical details see: *LI-1800 Portable Spectroradiometer Instruction Manual*, 1989).

## 4.3. Sunphotometer SP02

Sunphotometers are special devices that are designed to measure one of the physical quantities characterizing the radiation transmissivity of the atmosphere.

The early sunphotometers measured turbidity, but the recent models masures rather aerosol optical depth. Filters are used in sunphotometers to select the required wavelengths, so their accuracy is considerably lower than that of the insturments having monochrometers. Their considerably less complicated construction and, as a consequence, their far lower prices makes them, however, practical to use in monitoring networks.

Sunphotometer SP02 is a relatively accuate and reliable device in its category. Two are operated by the HMS, one is at the Marczell György Main Observatory in Budapest and the other is at the Kékestető Observatory that is situated on the highest peak of Hungary called Kékesető.

SP02 has four channels to measure aerosol optical depth that are as follows: 412, 500, 675, and 862 nm. In case of SP02, the aerosol optical depth should not be calculated from measured irradiance, because its output is the aerosol optical depth itself (namely, the voltage output corresponds to aerosol optical depth value).

## 4.4. UV Biometer

UV Biometers are special broad band detectors and can be considered not so old type of detectors, since the first experimental copies started to work in the seventies, their use started to spread in monitoring networks in the late eighties and mainly in the early and mid-nineties, so most of the national networks are not older than 15 years. The UV Biometers output erythemally weighted irradiance.

Measurement of UV radiaton has not been an important task in meteorological and atmospheric physical observations in the previous decades, because its flux density is neglectably low as compared with that of the visible and infrared ranges, so consequently it has no important role in radiation budget of the atmosphere. Its crucial role in production of vitamin D in human body and its harmful effect on biological systems, however, has motivated some scientists or institutes to establish sporadic campaigns to measure UV spectrally or in broad band way. But no any long term UV monitoring network has been operated until discovering atmspheric ozone depletion. The number of national networks has increased rapidly since then. Due to their simple construction and, consequently, their considerably lower price, use of broad band UV detectors are far more practical in networks than use of higher accuracy spectral equipments.

UV monitoring network of the HMS includes five sites that are as follows: Budapest, Kékestető, Kecskemét, Sármellék, and Siófok. The former four stations have started to operate in 1994, while in Siófok, the measurements have been started in 2009.

## 5. Quality assurance and quality control

QA/QC procedures are of crucial importance nowadays in operating high accuracy networks. The calibrations, routine checkings and tests are performed in the measuring practice of the HMS in the ways and with the frequencies that are recommended by the manufacturers of the equipments. Each procedure for the different instruments are performed by following the working instructions of the HMS. To descript the details would not be reasonable in this paper, but it is still to be noted that to follow the given instructions is of stressed importance for us, we operate a regional center for solar radiation.

## 6. Operation of regional center for solar radiation, WMO Region VI, Budapest

The HMS has been operating the regional center for solar radiation since 1980. The solar radiation regional centers of WMO are operated to represent the World Radiation Reference (WRR) of the World Radiation Center, Davos, Switzerland, for the instruments of the countries of the given region. The absolute cavity pyrheliometer that represents the WRR in our regional center is the HF 19746 instrument. It participate in each international pyrheliometer comparisons held in the World Radiation Center in every fifth year. The imortance of our activity as regional center has decreased a bit during the decades since 1980, as more and more countries have started to operate absolute cavity pyrheliometers. The spectrum of our activity has, in the same time, widened by installing high resolution spectral instruments, narrow spectral range detectors, and sunphotometers. Currently we have reference to calibrate follows: spectrophotometers, instruments as spectroradiometers, filterradiometers, sunphotometers, UV Biometers, and special spectral devices including PAR meters, illuminance meters, personal UV dosimeters, filters, or other special radiometers.

As concerns the future of the operation of the regional center, it is to be noted that though its significance as representing world reference pyrheliometric scale had a bit decreased in the previous years, increasing interest appeared from other scientific areas, such as biology, agronomy, daylighting, etc. Particularly, the inclusion of UV radiation-related studies and experiments started to be more frequent in the last decade in the scientific areas mentoned above.

Another task that will probably be more and more important even in the near future is the testing of diode array spectroradiometers. This is a brand new technology and its three main advantages are the very high speed, the high spectral resolution, and the fact that the diode array spectrometers do not include moving parts. Nowadays, however, several problems are still unsolved, so the accuracy and reliability is considerably lower than that of traditional monochrometer spectrometers. Results of calibration of some of these instruments that we have made confirmed the fact mentioned above.

## 7. Selected results

Selected results from studies and researches based on the data coming from the activities shown in the previous sections are shown here. The results shown are a small part of the entire set of results. Detailed analyzis of the results is not discussed here, because it would be the aim of separate papers. Thus, in addition to very brief analyzes, the references where the details can be found are given instead.

## 7.1. Total ozone

Total ozone measurements have been carried out at the HMS since 1969. *Fig.* 2 shows the long term variation of total ozone for Budapest for the period 1969–2012. The percentage deviations of yearly means from the mean of many years are shown in the figure. The mean of many years have been calculated for the period 1969–1980 to eliminate the effect of the stronger ozone decrease of the later decades on the estimated mean that characterizes a quasi-unperturbated condition.



*Fig.* 2. Percentage deviations of yearly means of total ozone content from the mean of many years for Budapest, 1969–2012.

It is well-known that total ozone has a yearly course in the midlatitudes that is more and more stressed from the equator towards the poles. *Fig. 3* shows the calculated average (smoothed) yearly course for Budapest with the upper and lower limits of natural variability (defined as two times of the standard deviation).

It is to be noted that despite the ozone recovery, considerable ozone losses were found for most of the summers of the last decade. The summer ozone losses in the last decade were significant even considering all 43 summers despite the increasing trend in the yearly averages. It is difficult to answer the question today whether it is a consequence of some variations in the circumastances influencing ozone concentration that are of climatic scale or they were incident events.



Fig. 3. Average yearly course of total ozone for Budapest.

## 7.2. Short-wave radiation transmission of the atmosphere

## 7.2.1. Graybody optical depth and the scattering parameter

Some results concerning the behavior of transparency of the atmosphere are shown in this section.

*Fig.* 4 shows the long term variation of graybody optical depth for Budapest for the period 1967–2011 that is demonstrated by the yearly means. The values are increasing up to 1994 and decreasing from then (similar behavior was found for each monthly trends), thus a sectioned trend analysis was performed as an experiment for the period 1967–1994 and for 1995–2011. Considering the different months, based on the trend analysis for the period 1967–1994, the only month was December for which significant increase was not found. It is to be noted, that reason for no yearly averages are shown in the figure for 2008 and 2009 is that no sufficient number of measured data were available for the correct calculation for both years due to susbequent failures of the solar tracking system. It has unfortunately taken for a long time while the manufacturer's trouble-shooting trials has become succesful.

Yearly course of GBOD is shown in *Fig. 5*. It is clear, based on the figure, that transparency of the atmosphere is higher in winter than in summer, as it has been expected otherwise.



Fig. 4. Yearly means of graybody optical depth for Budapest for the period 1967–2011.



Fig. 5. Average yearly course of graybody optical depth for Budapest.

These studies were performed for another special quantity called scattering parameter ( $\Theta$ ). It is an indicator of ratio of diffuse irradiance to the total global irradiance in the way that measured diffuse-to-global ratio is normalized by the diffuse-to-global ratio theoretically calculated for the Rayleigh-atmosphere, and it is in high correlation with turbidity (*Kaskaoutis et al.*, 2007) Thus it is expressed in the following way:

$$\Theta = \frac{D/G}{D_R/G_R}$$

where D and G are the diffuse and global irradiances, respectively, measured on a horizontal surface,  $D_R$  and  $G_R$  are the diffuse and global irradiances calculated for Rayleigh-atmosphere for the time of the observation, respectively.

Long term variation (1967–2002) and average yearly course were also determined for scattering parameter, and the results are very similar that those obtained for graybody optical depth.

Scattering parameter is a useful quantity to examine how close, in respect of transparency, the real atmosphere can be to the Rayleigh atmopshere in the clearest cases (least polluted cases, actually). In order to perform this, the lowest D/G values (occurred during the experimental period) were to determine. A reference can be the minimum value of a month (monthly absolute minimum value) from the long-term database. *Fig.* 6 shows the yearly course of absolute minimum values of D/G (solid line) and values of D/G calculated for Rayleigh-atmosphere ( $D_R/G_R$ ) for the middle of the hour intervals used for the study (dashed line). The important message of *Fig.* 6 is that in the clearest (least polluted) cases the atmosphere above the observatory almost equalled to the Rayleigh-atmosphere.



*Fig.* 6. Yearly course of monthly absolute minimum values of D/G and calculated values of D/G for Rayleigh-atmosphere.

#### 7.2.2. Optical properties of the aerosol

Optical properties of aerosols were studied by using AOD and Ångström exponent. Due to the consequentially very high autocorrelation between the AOD values calculated for the different standard wavelengths from the same spectrum, AOD for 500 nm (AOD<sub>500</sub>) was used for each study. To determine

long term variation of  $AOD_{500}$  was not possible because of the insufficient number of observations (that is a consequence of the fact that AOD, due to its definition, can be estimated when solar disc is not covered by any clouds).

However, a mean value (a quasi-mean of many years, actually) of AOD for Budapest was determined. The value obtained, of course, is not too reliable due to the abovementioned insufficiency. However its estimation, in contrast with the trend analysis, has reasonability due to the fact that for this estimation all data together are needed. The same can be stated for the mean of many years for the Ångström exponent.

Aerosol optical depth values are available for the period 1996–2011, so the mean of many years is valid for a period of 16 years. Since the results obtained for GBOD, that should relatively be in close connection with AOD, show that the transparency of the atmosphere has started to incease from the mid-nineties, the obtained mean for  $AOD_{500}$  will then underestimate a mean of many years concerning a longer period. A value of 0.28 was found as mean of many years for AOD<sub>500</sub>. A general experience for values of AOD at 500 nm is well-known as a result of numerous previous studies. It says that urban and industrial areas are characterized by values higher than 0.3, while rural areas are characterized by values lower than 0.2. Values between 0.2 and 0.3 characterize areas polluted on the average. Though the obtained value seems to be indicative of the suburban situation of the observatory, where there is no considerable industrial pollution, but traffic, it is to be noted that the correct value supposed to be some higher due to the expected underestimation of AOD<sub>500</sub> due to the tenth aforementioned reasons.

The obtained mean of many years for wavelength exponent is 1.31. Considering the facts about different values of  $\alpha$  written in 2.4., this value can be called usual value. It shows that the usual particle size is the dominant in the aerosols being present in the air colum above the observatory.

Dependence of Ångström exponent on  $AOD_{500}$  was also studied and the result is shown in *Fig.* 7. It was found that despite the considerably high dispersion of the set of dots, the two quantities are inversely proportional to each other, namely the dominant particle size increases with the increasing aerosol content of the air column. This fact has an important message to us: it means that there is a weak tendency showing bigger aerosol masses tend to be composed by larger particles with higher likelihood, statistically, and conversely: lower aerosol optical depths are, with higher statistical likelihood, produced by aerosols including smaller particles.

A parameterization technique was developed to estimate AOD data from GBOD data (*Tóth*, 2008). In principle, GBOD and AOD values calculated from irradiances that were measured at the same time, differ only due to GBOD, which is influenced by total vapor and ozone absorption (as main absorbers that can considerably vary), while AOD is not. Dependence of AOD at 500 nm on GBOD is shown in *Fig.* 8. If the aforementioned facts are accepted, the dispersion of GBOD values that belong to a given value of  $AOD_{500}$  is caused by effect of total water vapor content and total ozone content on GBOD. Consequently, if total water vapor content and total ozone content dependence

of the residuals are determined, a correction can be computed by which the relationship between the two optical depths can be improved. It means that AOD values can be computed from GBOD values by considering total precipitable water and ozone.



Fig. 7. Relationship between AOD<sub>500</sub> and Ångström exponent.



Fig. 8. Relationship between GBOD and AOD<sub>500</sub>.

Total precipitable water data calculated from rawinsound observations regularly performed in the observatory and total ozone data coming from ozone measurements carried out also operationally at the observatory by the Brewer spectrophotometer were used for the parametrization, so the parametrization equation was as follows:

$$\delta_{A\lambda} = C_{0\lambda}\delta_{GB} - C_{1\lambda}X_{TPW} - C_{2\lambda}X_{O3} + C_{3\lambda}, \tag{7}$$

where

 $\delta_{A\lambda}$  is the aerosol optical depth at wavelength  $\lambda$ ,

 $\delta GB$  is the graybody optical depth,

*XTPW* is the total precipitable water,

*XO3* is the total air columnar ozone content,

 $C_{0\lambda}$ ,  $C_{1\lambda}$ ,  $C_{2\lambda}$ ,  $C_{3\lambda}$  are the conctants to determine.

The wavelength used for the study was 500 nm. This parameterization resulted in an approximately 30% increase in correlation coefficient. It means that a method is available to calculate aerosol optical depth from graybody optical depth if total precipitable water and ozone contents are known.

The discussion of the methods used for the study and detailed analysis of the results has been published in a conference proceedings (*Tóth*, 2008).

## 7.2.3. Relationship between aerosol size distributions and aerosol optical depth spectra

In a study, the size distribution of aerosol and its relationship with wavelength exponent was investigated. The size distribution was estimated by a mathematical inversion method developed by King et al. (*King et al.*, 1978). The size distribution can be deterined from aerosol optical depth spectrum by using this method.

It was found that the obtained size distributions are in good connection with the wavelength exponent. Though they are not independent from each other due to the fact that the aerosol optical depth spectrum is the base for both, to analyze their relationship is still reasonable, because they are very different quantities. While the wavelength exponent is one number that characterizes the fequency of larger particles, and the dominant particle size can be estimated from it by using an empirical equation determined by Ångström (*Ångström*, 1929), the size distribution determined by King's method is a function estimating a realistic size distribution of the particles being present in the aerosol at the time of the recording of the spectrum. So we had to find a property of the function that could be brought into relationship with a number. The shape of the size distribution to be suitable to compare. It was found that the shape of the size distribution changes with the value of  $\alpha$ . Results are shown in *Fig 9*. To make details more easily visualizable, size distributions are graphed for 5 different values of  $\alpha$ , but the effect found is the same if all size distributions is involved.



Fig. 9. Shapes of computed aerosol particle size distributions for different values of Ångström exponent.

The size distribution function for the lower values of  $\alpha$  has a typical shape that is decreasing slowly for the shortest particle radii (approx. 0.3 µm – 0.4 µm) and from that value it is decreasing by a bit higher rate. This shape tends to be quasi-linear and then tends to be inflectious from about 0.6 µm, but oppositely to the shortest radii: first it is decreasing a bit rapidly, then slowly, and the inflection point is at about 0.25 µm that remains almost unvariable along the studied radius range (a bit shifted to the longest radii, but even the maximum shift is only some hundredth µm). The another inflection point was found somewhere between 0.6 µm and 0.7 µm. *Fig. 10a, b,* and *c* show that differences between the shape of the size distributions are very small for a given value of  $\alpha$ .

## 7.2.4. Dependence of spectral diffuse-to-direct irradiance on the atmospheric turbidity

A special investigation was performed previously concerning dependence of spectral diffuse-to-direct beam irradiance ratio on the atmospheric turbidity and solar zenith angle for the wavelength range from 300 nm to 1100 nm (*Kaskaoutis et al*, 2007). It was performed by modeling based on measured data. Since the detailed analyzes cannot be discussed in this paper, only two of the several results are shown here. *Fig. 11* shows the spectral ratios for  $AOD_{500} = 0.3$ ,  $\alpha = 1.3$ , and solar zenith angle of 20°, for three different values of single cattering ratio.



*Fig. 10.* Computed aerosol size distributions for high (a), normal (b), and low (c) values of Ångström exponent.

*Fig. 12* shows the ratios for AOD = 0.3, single scattering ratio = 0.9, and solar zenith anlge =  $60^{\circ}$ , for three different values of  $\alpha$ . It can be seen from both figures that the ratio increases moderately towards the shorter wavelength in most part of the visible range, then this increase becomes very rapid in the shortest part of the visible range, and the rate of increase is considerably sharper

in the ultraviolet range. The increase rate increases with increasing single scattering albeldo, as it has been expected. At the same time, the increase rate increases as wavelength exponent increases (the dominant particle size decreases). Detailed analysis of all results can be found in the paper referred.



*Fig. 11.* Spectral diffuse-to-direct beam ratios for  $AOD_{500} = 0.3$ ,  $\alpha = 1.3$ , and solar zenith angle of 20°.



*Fig. 12.* Spectral diffuse-to-direct beam ratios for AOD = 0.3, single scattering ratio = 0.9, and solar zenith anlge =  $60^{\circ}$ .

## 7.3. UV radiation

Some results concerning UV radiation are shown in this section.

Fig. 13 shows the characteristic maximum spectra for the different months for Budapest. It is clear from the figure that no considerable UV irradiance is received by the Earth's surface below 296 nm in Budapest even in case of the summer months. Despite this fact, measuring biologically effective UV radiation at the shorter wavelengths is reasonable, because the biophysical effects of the shortest wavelengths that yet reaching the Earth's surface are not known exactly today.



*Fig. 13.* Characteristic maximum UV spectra for the different months for Budapest coming from observations by Brewer MKIII spectrophotometer

The yearly totals and summer season totals of the biologically effective UV dose are shown in *Fig. 14* and *Fig. 15*, respectively, for the period 1995–2012. It is to be noted that despite the UV monitoring network of HMS includes five stations, only four were used for studying long term variations of UV. The reason is that one of the stations, Siófok, has started to operate in 2009, as it was mentioned in Section 4.4, so its inclusion in this study was not reasonable. An increasing trend of 3-6%/10 years was found for the different stations for both quantities. The obtained value fits the world-wide tendencies and the values computed from models for these areas (*Lytinska et al.*, 2009). The reason for the UV increase is that the increase of atmospheric transparency presumably compensates the UV reducing effect of the total ozone increase.

## 7.4. Effects of atmospheric circumstances on UV radiation reaching the Earth's surface

It is very important to know how atmospheric circumstances affect UV irradiances reaching the Earth's surface. Some results concerning this are shown in this section.



Fig. 14. Yearly totals of erythemally weighted UV irradiances for 4 sites of Hungary.



Fig. 15. Summer season totals of erythemally weighted UV irradiances for 4 sites of Hungary.

*Fig. 16* shows the relationship between total ozone content and UV radiation reaching the Earth's surface. Theoretically well-known that, as a consequence of strong ozone absorption in the UV-B range, the UV irradiance measured at the surface should be in inverse relationship with total ozone: the higher the total ozone content, the lower the UV irradiance. This fact can be seen well in *Fig. 14*, where the daily course of erythemally weighted UV radiation is shown for two days for which total ozone has considerably differed.

This dependence well-traced exactly for our total ozone and UV radiation dataset.



*Fig. 16.* Daily course of erythemally weighted UV irradiance for two days with different total ozone content.

A relatively frequently used quantity is the RAF (Radiation Amplification Factor) that is a practical indicator of the principle mentioned above. RAF shows the percentage increase in erythemally weighted UV irradiance that is resulted in an effect of 1% decrease in total ozone content. Value of RAF was determined by us by using spectrophotometric total ozone data and measured broad band UV data. The RAF obtained by us was in Section 1.17 (Németh et al., 1996) that is in good agreement with values obtained by other authors. Though not being used generally, to determine a special spectral RAF (SRAF) seemed to be very informative (it can be called the 'monochromatic version' of the aforementioned broad band RAF). SRAF was determined from data coming from Brewer measurements. Fig. 17 shows the dependence of SRAF on the wavelength. The slope sharply incresases towards the shorter wavelengths. The very important medical conclusion from the figure is that relatively smaller ozone losses can have dangerous effects, mainly for persons having extremely sensitive skin, because the pattern suggests that considerable amount of photons can appear at the very short wavelengths where the human skin is not prepared for their effect. Though it is not clear if it can be more dangerous either on short time scale or on longer time scale, it seems to be sure that the effect is considerably dangerous on longer scale. Furthermore, considering the significant ozone deficits in the last 6–7 summers, this result warns us to be very careful with sunbathing.



Fig. 17. Dependence of SRAF on the wavelength.

The effect of cloudiness on the UV irradiance received by the Earth's surface is an important parameter. The results have been published by us previously (Németh et al., 1996), so they are discussed here only very briefly. Though radiative transfer is known exactly in theory and can be modeled with relatively good reliability, in very practical tasks, for example in case of UV forecast, a reliable empirical relationship can solely be used due to the fact that numerous parameters of the cloud influencing radiative transfer in the cloud is not known exactly at the time of the given observation. A non-heighdependence CMF (Clud Modification Factor) was determined by us, which means that only the percentage cloudiness was considered in the study. All data were used for the study with no any filtering that depends on atmospheric circumstances. The results are shown in *Figs. 18a* and *b* for solar elevations higher than 30° and lower than 30°, respectively. The non-linear dependence is clear from the figures, but a peculiar behaviour is revealed from Fig. 16b: UV irradiances can, with high statistical likeliohood, be higher in case of 10-40%cloud coverage than irradiances detected for clear sky conditions. The explanation for this unexpected behavior is the very effective radiation scattering towards the Earth's surface on the edges of the clouds. Cloud coverages between 10 and 40% can be produced by mainly cumuli with relatively high probability and, due to their ragged, fragmented structure, the scattering on the cloud edges can thus be considerable.



*Fig. 18.* Dependence of UV transmission on cloudiness in the atmosphere in Budapest of solar elevations higher (a) and lower (b) than  $30^{\circ}$ .

#### 7.5. Verification of surface solar radiation outputs predicted by ALADIN model

Verification of predicted solar radiation data from ALADIN weather forecast model was carried out in 2001 (*Tóth*, 2002). Not the results of the verification themselves are that are shown here, because on the one hand, it cannot be aim of this paper and on the other, several improvements have been made on the model since the time of the verification in question. The aim is to show how high resolution spectral data can be useful for and can contribute to the verification by pointing out the strongnesses and weaknesses of the concerned module of the model. The verification was performed for total global fluxes for three stations and direct as well as diffuse fluxes for one station, because direct and diffuse

radiation measurements have been carried out at only one station (Budapest) then. The predicted data for both the subsequent day and the day after subsequent day were verified.

Two main factors affect solar radiation flux reaching the Earth's surface in case of wider energy range radiation: total water vapor content and total aerosol content of the air column. A study was still invented to find out how ALADIN would be able to estimate surface solar radiaton fluxes in cloudless cases or in other words: how it would be able to estimate atmospheric radiation transmission. Implicitly, the data were used for this study that has been produced at cloudless situation. The detailed description of filtering that was made in order to select data meeting with the criteria given by us, see the paper referred. To represent radiation transmission condition of the atmosphere, aerosol optical depth at 500 nm (AOD<sub>500</sub>) was selected.

The aim of the study was to obtain any information on reliability of ALADIN-made surface solar radiation forecasts concerning different atmospheric radiation transmission conditions. It is very important, since considerable radiation extinction can in many cases occur even when sky is cloudless.

Relationship between  $AOD_{500}$  and difference of observed and predicted solar irradiances (DIF\_DIR, DIF\_DIF, and DIF\_GL in the figures) was studied for all three radiation parameters. Results are shown in *Figs. 19, 20,* and *21.*  $AOD_{500}$  is represented on horizontal axis and differences between observed and forecasted irradiances are represented on vertical axis in each figure.



*Fig. 19.* Dependence of DIF\_DIR on AOD<sub>500</sub>.

Results for direct irradiance is shown in *Fig. 19*. Based on our five-year-long aerosol optical depth data series, it was known that average value for the

Marcell György Main Observatory in Budapest was approximately 0.3 for 500 nm. It is clear from Fig. 19 that for the most part, overestimations are found for values of AOD<sub>500</sub> lower than 0.3 and underestimations are found for  $\tau_{500}$ values higher than 0.3. It means that the model underestimates direct irradiance reaching the surface in cases when atmospheric radiation transmission is high, namely when very small amount of pollutant is present and overestimates it in cases when atmosphere is considerably polluted. Based on it one can conclude, that atmospheric radiation conditions predicted by the model are generally approaching average conditions due to that the model draftly handles parameters influencing radiation transmission, namely its variability is lower than that of the corresponding true parameters. The model predicts more polluted atmosphere compared with reality in very clear cases being close to Rayleigh atmosphere. These findings are confirmed by results of the same study made for diffuse irradiance (Fig. 20). Pollutants and water vapor intensively scatter radiation, consequently diffuse (scattered) irradiance observed at Earth's surface increases as larger amount of them is present in the atmosphere. It can be seen in Fig. 20 that in case of diffuse irradiance, inverse effect was found as compared to that found for direct component: the model overestimates diffuse irradiance in cases of high transmission (AOD<sub>500</sub> < 0.3), namely it forecasts more polluted conditions than the true state of atmosphere and, at the same time, it underestimates in cases of lower transmission, namely it forecasts lower pollution than that appearing in true atmosphere. The same effect lies behind this phenomena like in case of direct fluxes so the explanation is the same: the model tends to produce such physical conditions of atmosphere that result in more average atmospheric transmission than those occurring in reality. Those mentioned above are valid for global radiation also (see Fig. 21), but the effect is relatively less evident due to the fact that global irradiance is the sum of the previous two parameters so the effect in queston decreases.



Fig. 20. Dependence of DIF\_DIF on AOD<sub>500</sub>.



*Fig. 21.* Dependence of DIF\_GL on AOD<sub>500</sub>.

#### 8. Other special studies

Special individual studies occur with variable frequency in our activities and concern wide area in solar radiation measurements. They concern radiation transmission of given materials, or variation of their radiation transmission due to longer exposure to solar radiation (mainly UV), or spatial distribution of radiation source's emitted radiation, etc. Amongst these studies, results for only one are shown that are very useful and instructive. Several experiments concerning UV radiation transmission change of UV blocking plastic foils used in agriculture and special seed holding materials used in agriculture. The plastic foils have been exposed to UV radiation in the whole vegetation period (from May to October), and the change both in their special mechanical parameters and in their radiation transmission was investigated. The latter was performed by us, but measured data were provided to the former also by the Hungarian Meteorological Service. The experiment were supported by Hungarian Institute for Agrucultural Engineering (for other details, see *Csatár* and *Fenyvesi*, 2008).

Five kinds of foils were examined: 1) a plastic foil that included no UV filtering material was for control, 2) white-colored foil including 5% UV filtering metarial, 3) white-colored foil including 20% UV filtering material, 4) violet-colored foil including 5% UV filtering material, 5) violet-colored foil including 20% UV filtering material. Only a brief summary is discussed here to concern the most important results that are shown in *Figs. 22a, b, c*.



*Fig.* 22. UV transmission spectra of the examined plastics for three different sampling dates during the experiment (unexposed, Aug 15, Sep 5)

The plastics have been installed at the end of May in the garden of the Marczell György Main Observatory of the HMS and have thus been exposed to solar radiation until the beginning of October covering the entire vegetation period. Samples have been taken out from them by two weeks and their mechanical parameters and UV radiation transmission spectra have been examined to study variations in them. UV transmission spectra for three selected sampling dates for the spectral range from 286.5 nm to 363 nm for five selected sampling dates are shown in the figure. The behavior of the plastics are very similar and difference was found only quantitatively. It is evident that the UV blocking effect starts to loose partly soon after the installation, namely their increasing UV transmission increases and become considerable for the white foils and increases by a low rate for the violet foils for the second half of the examined period. It is also clear that at the end of the period, a chemical recovery starts up because the UV trasmission decreases a bit. Also a smoothing effect can be seen in the fine structure of each of the transmission spectra, but one type (white 20 %) retrieves partly its original shape. One can conclude that the destructive effect of UV radiation on the UV blocking material is not entirely irreversible. These results have stressed importance for the agriculture in application of the foils in UV protection of plants.

## 9. Conclusions

Due to the nature of this paper, conclusions cannot be made in the usual way. A special measurement technique and its theoretical background, covering, at the same time, a wide scale of measurement types were shown that is operationally run at the Hungarian Meteorological Service. Results coming from studies and investigations concerning very considerably different areas were shown, so their detailed analyzes were not possible, though some conclusions was made straight after showing the given results.

It is, however, still to be noted that use of high resolution spectrophotometry and spectroradiometry in atmospheric physical observations and investigations is very useful for numerous purposes, and it has reason for existence both in operational measurements and special experiments. It should be clear based on the studies and results showed that modern solar radiation measurements require its inclusion to an increasing degree.

Acknowledgements—Since high number of very different studies was shown in this paper, numerous domestic and foreign colleagues have contributed to them, so to list them is on the one hand, unreasonable and on the other, impossible due to the fact that contribution or significance of contribution of given persons cannot be estimated correctly. Still, one person is to be mentioned by name, because he is the author's long term colleague in the solar radiation area and his work on most part of the aforementioned studies is of primary importance. He is *Zoltan Nagy*, who is currently the head of the Atmospheric Physics and Measurement Technics Division of the HMS where all activities on solar radiation belong to. Nevertheless, the author thankfully acknowledges every international and domestic project that has founded or contributes by any way to the tasks shown, as well as also thankfully acknowledges all persons who have contributed to them in any degree.

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