Proc. Estonian Acad. Sci. Biol. Ecol., 2007, **56**, 1, 69–76 https://doi.org/10.3176/eco.2007.1.06

# Changes in the spectral aerosol optical thickness in Estonia (1951–2004)

Viivi Russak<sup>a\*</sup>, Ain Kallis<sup>a,b</sup>, Anne Jõeveer<sup>b</sup>, Hanno Ohvril<sup>c</sup>, and Hilda Teral<sup>c</sup>

<sup>a</sup> Tartu Observatory, 61602 Tõravere, Estonia

<sup>b</sup> Estonian Meteorological and Hydrological Institute, Rävala 8, 10143 Tallinn, Estonia

<sup>c</sup> Institute of Environmental Physics, University of Tartu, Tähe 4, 50090 Tartu, Estonia

Received 27 January 2006, in revised form 4 July 2006

Abstract. Broadband solar radiation has been measured during several decades, but the history of spectral measurements is relatively short. Several models exist for the calculation of the spectral atmospheric aerosol thickness from broadband solar radiation data. Using the method elaborated at Moscow University, aerosol optical thickness at 550 nm was calculated for Tartu–Tõravere Meteorological Station (Estonia) for the period 1951–2004. A significant increase in the spectral as well as broadband optical thickness is characteristic until the late 1980s, while during the last 15–20 years a sharp decrease was observed. Besides changes in the aerosol burden, changes in the features of dominating particles, as well as in the seasonal cycle, were found during the period under study.

Key words: solar radiation, aerosol, optical thickness, multi-annual changes.

#### **INTRODUCTION**

Atmospheric aerosol plays an essential role in the Earth's radiation budget. First, it directly affects climate by scattering and absorbing solar and terrestrial radiation. Secondly, it has an indirect impact on climate through the influence on cloud parameters. A method for the remote sensing of the columnar aerosol content and its properties is based on the measurements of solar radiation in cloud-free conditions with the data from narrow spectral bands preferred as the most informative. Long-term monitoring of aerosol is necessary for understanding the changes taking place in radiative forcing and climate parameters.

<sup>\*</sup> Corresponding author, russak@aai.ee

During several decades broadband solar radiation has been measured at hundreds of meteorological stations throughout the world, while the history of continuous spectral radiation measurements is relatively short. Several models have been created for transition from broadband irradiance to spectral aerosol optical thickness (AOT $\lambda$ ). These models enable us to study aerosol parameters retrospectively during long periods.

### **DATA AND METHODS**

In Estonia routine broadband direct solar radiation measurements with the Yanishevsky thermoelectric actinometer started at Tartu–Tõravere Meteorological Station (58.25° N, 26.47° E) already in 1950. The receivers and the measurement methods have not changed since then. Spectral measurements of direct radiation by an automatic Cimel Electronique 318A Sun and sky spectral photometer of Aerosol Robotic Network (AERONET) at Tartu–Tõravere Meteorological Station began in June 2002 (AERONET, 2005).

In the present study, the method elaborated in Moscow University (Tarasova & Yarkho, 1991) for the calculation of aerosol optical thickness at 550 nm (AOT550) was used. This model contains more than ten formulas. The input parameters are broadband direct irradiance, solar elevation, the amount of precipitable water W, and the Ångström wavelength exponent  $\alpha$ . The postulated columnar ozone (O<sub>3</sub>) content of 0.3 cm agreed well with its values measured at Tõravere (Eerme et al., 2002), the nitrogen dioxide (NO<sub>2</sub>) content was not considered. The amount of precipitable water in our calculations was determined using its linear dependence on water vapour pressure on the ground. This linear connection was first determined by Okulov (2003) on the basis of radiosonde observations at Tallinn Aerological Station (59.48° N, 24.60° E) and adjusted by using photometer measurements made at Tõravere in 2002–2003 (Fig. 1).

The Ångström wavelength exponent  $\alpha$ , estimated from photometer measurements in the spectral range of 440–870 nm (AERONET, 2005), varied from 0.6 to 2.3 at Tõravere in 2002–2004 (Fig. 2). In the present calculations the value  $\alpha = 1.5$  was used.

The aerosol optical thickness at 500 nm measured by the photometer was transformed to 550 nm by using the Ångström classical formula. The correlation between the values of AOT550 calculated by the Moscow model and those from photometer measurements at Tõravere in 2002–2003 (405 cases) was relatively high, R = 0.99 (Fig. 3).

On average, the model yielded 8% smaller values than those obtained from photometer measurements in 2002–2003. As an exception, on days with extensive forest and peat bog fires in Estonia and nearby Russian areas in the summer of 2002, the difference reached 20%. This can be explained by the peculiarities of the aerosol originating from biomass burning. In 2003, when no large forest fires were recorded, the model overestimated the photometer data on average by about 1%.



Fig. 1. Dependence of the daily averaged precipitable water W on water vapour pressure on the ground e at Tõravere (2002–2003).



Fig. 2. Frequency of the values of the daily mean Ångström wavelength exponent  $\alpha$  (in the spectral area 440–870 nm) at Tartu–Tõravere in 2002–2004.



**Fig. 3.** The relationship between aerosol optical thickness at 550 nm estimated from photometer measurements and that calculated by the Moscow model.

#### RESULTS

The high correlation between the results of the Moscow model and the photometer measurements encouraged us to use this model for the calculations of AOT550 at Tartu–Tõravere for earlier years (Fig. 4). For this purpose we used the data of routine solar radiation and water vapour measurements at Tartu–Tõravere Meteorological Station covering the period from 1951 to 2004. As we lacked information on the possible values of the Ångström exponent  $\alpha$  for the earlier years, the exponent used in this study  $\alpha = 1.5$  may increase the inaccuracy of our results to some extent. Comparative calculations have shown that replacing  $\alpha = 1.5$  by  $\alpha = 1.3$ , the annual values of AOT550 will be by about 5% lower.

In addition to spectral aerosol optical thickness, we calculated the broadband (averaged over the entire solar spectrum) aerosol optical thickness (BAOT) at Tartu–Tõravere for the same period. The value of BAOT can be considered as a sum of optical thicknesses of an ideal (clean and dry) atmosphere, water vapour, and aerosol. The optical thickness of an ideal atmosphere was calculated by Okulov (2003) by the method proposed in (Gueymard, 1998). In these calculations absorption in  $O_3$  and  $NO_2$  was included besides the Rayleigh scattering. The optical thickness of water vapour was evaluated using the method of Zvereva (1969). To obtain the aerosol optical thickness, the calculated optical thickness of the atmosphere. The residual optical thickness is that of the aerosol.



**Fig. 4.** Time series of annual mean broadband and spectral (550 nm) aerosol optical thicknesses at Tartu–Tõravere. The arrows mark eruptions of El Chichón (1) and Mt. Pinatubo (2).

The time series of the annual mean values of AOT550 and BAOT were quite similar in the period under study (Fig. 4.). A significant steady increase is characteristic until the middle of the 1980s (significance p < 0.01 in 1951–1982), while during the last 15–20 years a rapid decrease was observed. The increase in the aerosol loading during the first decades of the period under study coincides with the period of extensive growth of industry and transport and should be considered as resulting mostly from human activities. Numerous volcanic eruptions also influenced the optical state of the atmosphere in these years. In the early 1980s successive eruptions took place (Soufrière in 1979, Mount St. Helens in 1980, Alaid in 1981, El Chichón in 1982), and the self-cleaning ability of the atmosphere diminished, leading to the accumulation of volcanic products in the atmospheric aerosol burden has been observed in the years after the El Chichón (1982) and Mt. Pinatubo (1991) eruptions. In these years increased values have been observed in the time series of BAOT as well as of AOT550.

The middle of the 1980s should be considered as a break point in the time series of the aerosol optical thickness in Estonia. Then the slow but steady increase was replaced by a relatively rapid decrease, which may be related to the economic decline of the socialist countries at the turn of the 1980s–1990s. For example, in the Czech Republic the emission of SO<sub>2</sub> decreased by 86% and that of NO<sub>x</sub> by 55% during 1989–2000 (Hejkrlik, 2002). During the period from 1990 to 1995 the emission of SO<sub>2</sub> from oil-shale-fired power plants in North-East Estonia diminished by about 60% (Eesti keskkonnaseisund, 2000).

Another reason for the observed cleaning of the atmosphere may be linked to the nature protection measures applied in Estonia, as well as throughout Europe. The aerosol originating from distant sources plays an essential role in Estonia. Rough estimates demonstrate that about half of the sulphur compounds precipitated in Estonia have been transported over long distances (Punning & Karindi, 1996). The increased transparency of the atmosphere may partly be due to the absence of major volcanic eruptions in these years. At present, the values of AOT550 and BAOT are comparable to those in the early 1950s.

The annual mean aerosol optical thickness at 550 nm mostly exceeded the respective broadband value. It is characteristic that changes in AOT550 were more rapid than in BAOT (Fig. 5.). This evidently indicates that besides changes in the aerosol loading also changes in its physical and chemical properties (size distribution, chemical composition, hygroscopicity, etc.) have occurred during the last half-century. Similarly to the time series of AOT550 and BAOT, also a break point in the time series of their observed ratio is obvious in the late 1980s.

In the annual course of the ratio AOT550/BAOT the highest values occur in February, followed by a continuously decreasing trend up to November (Fig. 6.). This feature apparently points to possible changes in the dominating aerosol sources. For example, in January–February cloudless days are often frosty and numerous ice crystals floating in the air may increase air turbidity. In March–April, after the snow has melted, the dust from the soil without fresh vegetation can be taken up by turbulence and convection. Bog and forest fires can essentially change the aerosol characteristics and loading in summer months. Long-range transport of aerosol depends mostly on the seasonal features of atmospheric circulation. Most frequently a high content of aerosol is connected with air



**Fig. 5.** Time series of the mean annual ratio of spectral to broadband aerosol optical thickness at Tartu–Tõravere (solid line) and its moving average (period 5 years) (broken line).



Fig. 6. Mean annual course of the ratio of spectral to broadband aerosol optical thickness at Tartu-Tõravere (1951–2004).

masses crossing Estonia from the southwestern and southern directions. Some amount of aerosol from the Arctic regions can reach Estonia in spring. A number of authors, e.g. Barrie & Bottenheim (1991), Rodionov & Marshunova (1992), and Vinogradova (2000), showed that in winter atmospheric circulation leads to the advection of aerosol from moderate latitudes into the Arctic, whereas in spring an essential part of it may return to middle latitudes.

#### **CONCLUSIONS**

Comparison of the values of spectral aerosol optical thickness calculated by the method elaborated at Moscow University with the corresponding values measured by a spectrophotometer showed a good coincidence. Annual mean values of aerosol optical thickness calculated by the model at 550 nm (1951–2004) increased steadily until the late 1980s, followed by a decreasing trend up to now. Besides changes in the aerosol burden, changes were detected in aerosol properties.

#### ACKNOWLEDGEMENTS

This research was supported by grant No. 5857 of the Estonian Science Foundation. The AERONET team and the Estonian Principal Investigator Dr. O. Kärner together with Dr. M. Sulev are appreciated for making accessible the spectral observation data. The data of broadband solar radiation were provided by the Estonian Meteorological and Hydrological Institute.

#### REFERENCES

AERONET, 2005. http://aeronet.gsfc.nasa.gov/

- Barrie, L. A. & Bottenheim, J. W. 1991. Sulphur and nitrogen pollution in the Arctic atmosphere. In *The Pollution of the Arctic Atmosphere*, pp. 155–183. Elsevier Science Publishers.
- Eerme, K., Veismann, U. & Koppel, R. 2002. Estonian total ozone climatology. Ann. Geophys., 20, 247–255.
- *Eesti keskkonnaseisund XXI sajandi lävel.* 2000. Keskkonnaministeeriumi Info- ja Tehnokeskus, Tallinn.
- Gueymard, C. 1998. Turbidity determination from broadband irradiance measurements: a detailed multicoefficient approach. J. Appl. Meteorol., 37, 414–435.
- Hejkrlik, L. 2002. Recent changes in air pollution in Czech Republic. In *Fourth European* Conference on Applied Climatology, ECAC 2000, Brussels, 12.11.2002–15.11.2002. Abstract Volume.
- Okulov, O. 2003. Variability of atmospheric transparency and precipitable water in Estonia during last decades. *Diss. Geophys. Univ. Tartu.*, 18.
- Punning, J.-M. & Karindi, A. 1996. Composition of Estonian atmosphere. In *Estonia in the System of Global Change* (Punning, J.-M., ed.), pp. 26–34. *Inst. Ecol. Publ.*, 4, Tallinn.
- Rodionov, V. F. & Marshunova, M. S. 1992. Long-term variations in the turbidity of the Arctic atmosphere in Russia. Atmos. Ocean, 30, 531–549.
- Tarasova, T. A. & Yarkho, E. V. 1991. Determination of atmospheric aerosol optical thickness from land-based measurements of integral direct solar radiation. *Soviet Meteorol. Hydrol.*, 12, 53–58.
- Vinogradova, A. A. 2000. Anthropogenic pollutants in the Russian Arctic atmosphere: sources and sinks in spring and summer. *Atmos. Environ.*, 34, 5151–5160.
- Zvereva, S. I. 1969. On attenuation of solar radiation in polar region. *Tr. AANII*, **287**, 171–187 (in Russian).

## Aerosooli spektraalse optilise paksuse muutused Eestis aastatel 1951–2004

Viivi Russak, Ain Kallis, Anne Jõeveer, Hanno Ohvril ja Hilda Teral

Atmosfääri sondeerimine päikesekiirguse abil on seal leiduva aerosooli kirjeldamiseks üks levinumaid viise. Detailsema teabe annavad päikesekiirguse spektraalsed mõõtmised. Eestis on integraalset päikesekiirgust mõõdetud rohkem kui poole sajandi jooksul, selle regulaarsed spektraalsed mõõtmised algasid aga alles 2002. a suvel. Päikesekiirguse integraalsetest andmetest aerosooli spektraalsete optiliste paksuste arvutamiseks on mitu meetodit. Artiklis kasutatud Moskva ülikoolis välja töötatud metoodika sobivust Eesti tingimustega on testitud AERONETsüsteemi fotomeetri mõõtmisandmete abil. Tartu-Tõravere meteoroloogiajaama päikesekiirguse andmetest arvutatud aerosooli spektraalsete optiliste paksuste (550 nm) aegrida kirjeldab muutusi nii atmosfäärse aerosooli hulgas kui ka optilistes omadustes aastatel 1951–2004.