Correlated Short Term Fluctuations in Aerosol Optical Thickness and Shortwave Radiative Quantities

S. E. Schwartz
Environmental Chemistry Division
Brookhaven National Laboratory
Upton, New York

Abstract

Close examination of direct normal solar irradiance (DNSI) and downwelling diffuse irradiance (DDI) on cloud-free days at the Atmospheric Radiation Measurement (ARM) Southern Great Plains (SGP) site reveals anticorrelated short-term (several minute) fluctuations, especially prominent in the hours around local solar noon; i.e., DDI increases as DNSI decreases. These fluctuations are correlated or anticorrelated with other direct and derived radiometric quantities, including aerosol optical thickness (AOT) from multifilter rotating shadowband radiometer (MFRSR) and Ångström exponent, evaluated as -d ln (AOT)/d ln wavelength. These fluctuations are attributed to increased relative humidity associated with intermittent rising air parcels. Examples of correlations will be given together with analysis of relative magnitudes of fluctuations in the several quantities establishing the consistency of this interpretation.

Motivation

- Aerosols are the greatest contributor to uncertainty and variability in the shortwave radiation budget for cloud-free skies. At 60° solar zenith angle, a change in AOT at 550 nm of 0.01, the present limit of accuracy in measurement, corresponds to a change in DNSI of 14 W m^{-2} (Halthore et al. 1997).

- Shortwave radiative forcing of climate change by anthropogenic aerosols is considered to be a potentially substantial offset of greenhouse gas warming over the industrial period, but is highly uncertain (IPCC 1996; Schwartz and Andreae 1996).

- Accurate characterization of aerosol scattering and absorption properties is required to constrain models of atmospheric absorption. An overestimate of 0.02 nm in AOT at 550 nm, compensated by a corresponding underestimate in direct beam absorption, leads to an underestimation of atmospheric absorption by 5% (Halthore et al. 1998).

Observations

The ARM complement of shortwave instrumentation provides a unique opportunity for characterization of aerosol influences on shortwave radiation. The following are the radiometric quantities used here and the instruments employed:

- DNSI: normal incidence pyrheliometer (NIP).

- Direct Beam Extinction: MFRSR.

- Total and DDI: Precision Spectral Pyranometers (unshaded, shaded).

Results are reported here for April 18, 1996, characterized by an absence of clouds throughout the day.

Comments on the Observations

Figure 1 shows time series of AOT and other radiometric quantities at the SGP site on April 18, 1996; local standard time = UT - 6 h; solar noon ca. 1830 UT.

AOT at a given wavelength, τ_λ, is from MFRSR. Path extinction is evaluated as -ln (V/V_0), where V is measured voltage and V_0 is voltage corresponding to top of atmosphere as inferred from Langley plots. Vertical extinction is path extinction divided by airmass. AOT is vertical extinction minus Rayleigh scattering optical thickness and ozone absorption optical thickness.

The points at 1730-1800 show DNSI calculated with the Moderate Resolution Atmospheric Radiance and Transmittance Model (MODTRAN)-3. The smooth black curve denoted “Modeled DNSI” passing through these points

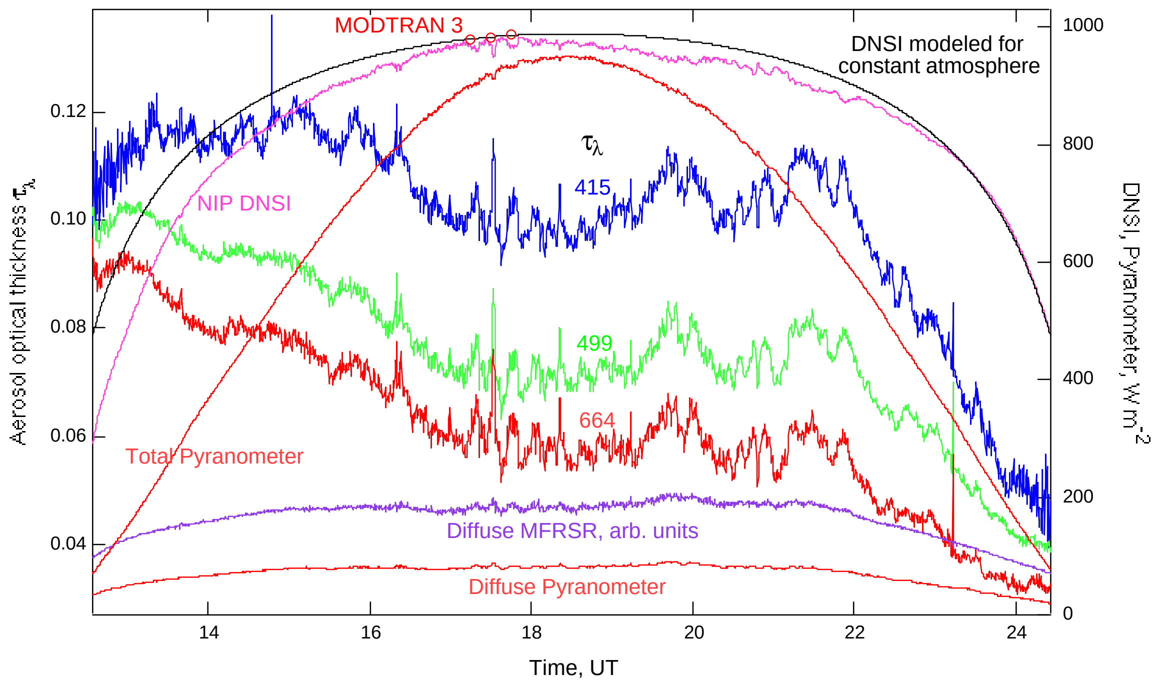

Figure 1. Shortwave radiometric quantities observed at the ARM SGP Central Facility in north central Oklahoma on April 18, 1996. See text for further explanation. (For a color version of this figure, please see *http://www.arm.gov/docs/documents/technical/conf_9803/schwartz(3)-98.pdf*.)

provides an approximation to DNSI for constant atmospheric conditions and may be compared to value determined with NIP.

Also shown are pyranometer measurements of total and diffuse irradiance and diffuse irradiance from the MFRSR.

Attention is called to fluctuations in the several quantities especially in the midday hours: decrease in DNSI and total downwelling corresponding to increase in AOT, and perhaps a hint of increase in diffuse downwelling from the pyranometer and MFRSR.

Comments on the Correlations

Figure 2 focuses on the hours around local noon. In addition to the traces for AOT and DDI, there is shown a quantity of DDNSI evaluated as the difference between the smooth black curve and the measured DNSI, taken such that a decrease in DNSI corresponds to an increase in DDNSI. Note the correlation of fluctuations in DDNSI with those in AOT.

Also shown is the Ångström exponent of the AOT, the negative logarithmic slope of AOT with wavelength evaluated as

$$\mathbf{å}_{415/664} = -\frac{\ln \tau_{415} - \ln \tau_{664}}{\ln 415 - \ln 664}$$

and plotted such that a decrease in $å_{415/664}$ is upward on the graph. Again note the close correlation of $å_{415/664}$ with AOT and DDNSI.

Causes of the Correlations

The sensitivity of DNSI to the several controlling variables virtually precludes the fluctuations being due to anything else but fluctuations in AOT.

Further the slope of the correlation between AOT and DNSI is entirely consistent with the model calculations, 11.6 W m^{-2} per 0.01 AOT (499 nm). So a decrease in DNSI with increase in AOT is expected.

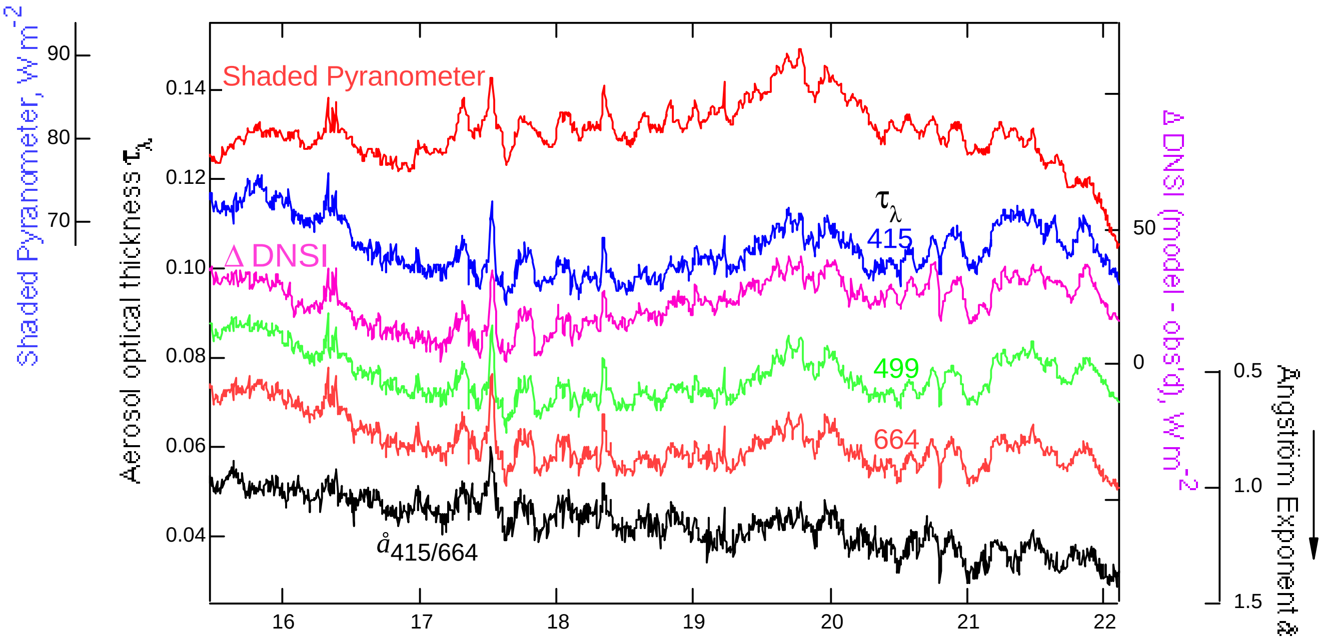

Figure 2. Correlations of rapid fluctuations in shortwave radiometric quantities observed at the ARM SGP Central Facility in north central Oklahoma on April 18, 1996. See text for further explanation. (For a color version of this figure, please see *http://www.arm.gov/docs/documents/technical/conf_9803/schwartz(3)-98.pdf*.)

Why then is there an increase in DDI with an increase in AOT? Assuming that AOT is due to light scattering, most of the light scattered out of the direct beam, especially at high sun elevation, is scattered in the downward direction.

Thus, an increase in aerosol light scattering should lead to a decrease in downwelling diffuse radiation.

Causes of the Fluctuations

What is responsible for these fluctuations? We propose that the fluctuations are due to turbulent convection.

Consider a parcel rising convectively due to localized surface heating. As the parcel rises it expands and therefore cools by doing adiabatic work on its surroundings. This cooling leads to an increase in relative humidity; an increase in relative humidity leads to growth of hygroscopic aerosol particles, thereby increasing their light-scattering coefficient.

Notice that an increase in AOT corresponds to a decrease in the Ångström exponent. The Ångström exponent is a rough measure of the size of the particles responsible for light scattering; a decrease in the Ångström exponent (i.e., flatter spectral dependence of light scattering) corresponds to larger particles. Thus, when AOT is increasing, particle size is increasing.

Now consider a parcel rising under turbulent convection. We have already suggested that the increase in AOT arises from accretion of additional water by hygroscopic particles, increasing their size.

The increase in Ångström exponent is entirely consistent with this, supporting the plausibility of hygroscopic growth induced by convection as the cause of the fluctuations in AOT and the resulting fluctuations in the several other radiometric quantities.

A manuscript is in preparation. For further information contact ses@bnl.gov.

References

Halthore R. N., S. E. Schwartz, J. J. Michalsky, G. P. Anderson, R. A. Ferrare, B. N. Holben, and H. M. ten Brink, 1997: Comparison of model estimated and measured direct-normal solar irradiance. *J. Geophys. Res.*, **102**, 29,991-30,002.

Halthore R. N., S. Nemesure, S. E. Schwartz, D. G. Imre, A. Berk, E. G. Dutton, and M. H. Bergin, 1998: Diffuse-sky downward irradiance (DFDI) at surface in cloud free atmospheres - a closure experiment. This proceedings.

IPCC (Intergovernmental Panel on Climate Change), 1996: Climate change 1995 - the science of climate change. contribution of WGI to the second assessment report of the Intergovernmental Panel on Climate Change. J. T. Houghton, L. G. Meira Filho, B. A. Callander, N. Harris, A. Kattenberg, and K. Maskell, Eds. Cambridge University Press, Cambridge, United Kingdom, 572 pp.

Schwartz S. E., and M. O. Andreae, 1996: Uncertainty in climate change caused by anthropogenic aerosols. *Science*, **272**, 1121-1122.