

**ASSESSMENT OF THE EXPOSURE TO BIOLOGICALLY
EFFECTIVE UV RADIATION USING A DOSIMETRIC
TECHNIQUE TO EVALUATE THE SOLAR SPECTRUM**

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Abstract: A cost effective method employing polysulphone, nalidixic acid, 8-methoxypsoralen and phenothiazine as UV dosimeters is presented for evaluating the UV spectrum. The exposure measured by each dosimeter is a function of the source spectrum and the spectral response of the material. Each material has a different spectral response and records a different dose for the same exposure. A least squares method is employed to extract the source spectrum from the four dose measurements. A number of spectra have been evaluated and the differences between these spectra and the associated irradiances compared to the spectra and irradiances measured with a calibrated spectroradiometer is less than 20%. The technique allows simultaneous multi-site measurement at positions that may be inaccessible to sensitive and expensive equipment. The technique was employed to evaluate the spectrum on the chest and shoulder of four subjects. The erythema exposures were derived from the evaluated spectra with the chest exposures 0.7 to 0.8 those of the shoulder exposures.

1. Introduction

Studies by aeroplane, balloon, satellite and ground based measurements have confirmed that the Earth's protective layer of stratospheric ozone is being depleted due to human activities (Stolarski *et al.*, 1992). The reduction in the absorption of solar UV by ozone is of significant concern because of the concomitant increase in terrestrial UV. The waveband 280 nm to 320 nm (UVB) is that most strongly affected by changes in stratospheric ozone concentration (Frederick *et al.*, 1989). The irradiance at the earth's surface for wavelengths shorter than 280 nm (UVC) is insignificant because of the very high absorption not only by the remaining ozone but by other atmospheric constituents. Longer wavelengths of 320 to 400 nm (UVA) have relatively low absorption by ozone and are unaffected by any possible changes in ozone concentrations. Ambient solar spectra are subject to the influence of environmental factors such as cloud cover, aerosols and reflecting objects near the exposure site. As a result, the source spectrum of solar radiation and consequently the UV exposure is different at different exposure sites (Wong *et al.*, 1992).

Reviews are available in the literature of the potential biological impacts of these increases in solar UVB radiation on humans (Ambach and Blumthaler, 1993) and on terrestrial plants (Tevini and Teramura, 1989, Tevini, 1993). The relative biological effectiveness of radiation to produce a particular response is wavelength dependent. Hence the interfering effect of UV radiation on the natural function of important biological macromolecules is wavelength specific (Caldwell *et al.*, 1986) and a weighting function - the action spectrum - must be employed in determining the biological effective irradiance (UV_{BE}). For a selected biological process this is written as

$$UV_{BE} = \int_{UV} S(\lambda)A(\lambda)d\lambda \quad (1)$$

where $S(\lambda)$ is the source spectral irradiance in $W\ cm^{-2}\ nm^{-1}$ and $A(\lambda)$ is the relevant action spectrum determined for the particular process. Biologically effective irradiance must be employed in comparisons between studies where the source spectra are different and in predicting effects of changes in UV irradiance.

A knowledge of the UV source spectral irradiance, $S(\lambda)$, is therefore necessary in calculating the biologically effective irradiance. The UV source spectrum may be measured with a spectroradiometer, but such equipment is:

- Expensive, requires power and ancillary equipment such as a computer
- Impractical for simultaneous multi-site measurements (often inaccessible with equipment requiring careful handling)
- Impossible to attach to the body of the object under study to provide personal exposures as required in some studies. For example, in studies of the UV effects on humans, the influence of geometry, orientation, atmospheric conditions and optical properties of surroundings results in the ambient UV spectrum differing from that at various sites over a human body. Similarly, this applies in studies of the UV effects on plants, animals and materials.

Passive dosimeters such as Polysulphone have been used conveniently for assessing biologically effective exposure such as the effective exposure for erythema. The use of dosimeters requires the calibration against standard equipment such as a spectroradiometer (Diffey, 1989). The procedure often introduces errors which could be as high as 40% (Wong *et al.*, 1994). The calibration must be repeated for each type

of biological process or whenever there is a change in the source spectrum they are to measure. Each type of dosimeter has a sensitivity which is confined to a portion of the solar ultraviolet waveband, e.g. Polysulphone is sensitive to UVB (280-320 nm). There is not a single dosimeter which can be used effectively and conveniently for the entire solar ultraviolet waveband. This paper describes a technique employing four film dosimeters, which is suitable for multi-site simultaneous measurements of the entire solar UV spectrum over the object of study. The dosimeters are inexpensive, rugged and unaffected by rain or condensation and may be deployed in the open and at numerous sites over the objects of study. The technique is applicable where the source spectrum is known to be both relatively constant during the measurement period and a smooth function of wavelength.

2. Materials and Methods

Four types of materials which are sensitive to various wavebands of UV radiation and which undergo a change in optical absorbance upon exposure have been employed as dosimeters. The substances (supplied by A Davis, 3 Cumley Rd., Toothill, Ongar, Essex, CM5 9SJ, UK) employed in a thin film form are polysulphone, nalidixic acid (NDA), 8-methoxypsoralen (8-MOP) and phenothiazine. These materials have spectral responses which effectively cover different parts of the UV spectrum and with peaks in the spectral responses at different wavelengths. Hence the four materials provide measurements of the exposures in the respective UV wavebands. These four exposure measurements therefore allow broad scale reconstruction of the original source spectrum.

Polysulphone dosimeters have previously been employed in measuring human UV exposure (Diffey, 1989, Herlihy *et al.*, 1994) and have been investigated for use in measuring plant exposure (Parisi and Wong, 1994). The technique described here follows similar procedures: The dosimeters consist of a thin film of the material between 26 and 40 μm thick and approximately 1 cm^2 mounted in a cardboard holder which is attached at the point of interest. In this technique four dosimeters made from the different materials are fixed in close proximity to each other.

Measurements of UV exposure rely upon the deterioration of the dosimeter material due to the incidence of UV radiation. In order to standardise the effect of any dark reaction, it is necessary to employ a constant read-out time after exposure. In this case, the optical absorbance of the film was measured immediately prior to and after exposure to UV radiation (an alternate read-out time may be employed provided it is maintained as constant). The change in optical absorbance between these measurements, ΔA is related to the dose received.

The optical absorbance of the materials is measured using a spectrophotometer (Shimadzu Corp, Kyoto, Japan) at wavelengths 330 nm for polysulphone and NDA, 305 nm for 8-MOP and 280 nm for phenothiazine. These are the wavelengths at which the largest change in absorbance occurs for the particular materials. Four measurements of optical absorbance are taken at different locations on the film to avoid possible errors. In practice the variation in ΔA for dosimeters of the same type exposed to the same dose exceeds the variation indicated by the four measurements made on a single dosimeter. The larger variation between dosimeters is estimated to be of order 10% and this is the uncertainty ascribed to the measurements of ΔA .

The change in optical absorbance of a dosimeter exposed to a source spectrum of $S(\lambda, t)$ over a time interval T is given by the expression

$$\Delta A_i = \int_0^T \int_{UV} S(\lambda, t) R_i(\lambda) d\lambda dt \quad i = 1, \dots, 4 \quad (2)$$

where $R_i(\lambda)$ is the spectral response of the dosimeter and the subscript is used to designate one of the four different types of dosimeter material used. If $S(\lambda, t)$ is relatively constant over the exposure period, then Equation (2) becomes

$$\Delta A_i = T \int_{UV} S(\lambda) R_i(\lambda) d\lambda \quad (3)$$

where $S(\lambda)$ now represents the time averaged spectrum.

The spectral response functions $R_i(\lambda)$ give the change in optical absorbance for the different materials due to unit irradiance in the wavelength interval λ to $\lambda+d\lambda$. The relative spectral responses of NDA (Tate *et al.*, 1980), 8-MOP (Diffey and Davis, 1978) and polysulphone (CIE, 1992) are in the literature. Measurements to verify these results in the literature, calculate any normalisation factors and to determine the spectral response function of phenothiazine (Diffey *et al.*, 1977) were made as follows: Each dosimeter material was exposed through narrow band UV filters (Melles-Griot, Irvine, CA, USA) to radiation from a quartz tungsten halogen lamp for periods of time sufficient to cause significant degradation in each case. The lamp was powered by a current regulated power supply to ensure constant output during this time. A range of different filters was used; the full width at half maximum of each filter is approximately 10 nm and the peak transmission wavelengths are 286, 291, 311, 317, 339, 345 and 379 nm. A curved metal sheet with two rows of 12 mm diameter holes at the lamp level (Wong *et al.*, 1989) at a distance of 10 cm from the lamp allowed simultaneous exposure of a series of filter/dosimeter combinations. The spectral irradiance through each filter was measured with a spectroradiometer based on a double holographic grating (Jobin-Yvon, model DH10, France) and R212 photomultiplier tube (Hamamatsu Co., Hamamatsu City, Japan) with calibration traceable to the primary Australian standard lamp housed at the National Measurement Laboratory, Lindfield, NSW, Australia. The total irradiance rates through each filter were found by summation of the measured spectral irradiance in 1 nm steps over the waveband for which the transmission through the filter was non zero.

In this manner, each dosimeter was exposed to a known dose of UV radiation within a narrow bandwidth. For each filter/dosimeter material combination the resulting change in optical absorbance, ΔA_i , divided by the measured dose was taken as the average value for $R_i(\lambda)$ at the wavelength corresponding to the peak transmission wavelength of the filter. The method was applied to derive the spectral response functions for NDA, 8-MOP, phenothiazine and polysulphone.

The response functions were then used to determine the source spectrum as follows: Four dosimeters were exposed to the source - in the examples shown here the quartz tungsten halogen lamp was used with an exposure time of 30 min at distances of 10 and 12 cm, along with 5 min exposure to summer sunshine in Brisbane, Australia on a horizontal plane at 09:18 and 11:39 EST (Eastern Standard Time) - The resulting change in optical absorbance for each dosimeter, ΔA_i , was measured. A predicted value for the change in absorbance for each dosimeter, $\Delta A_i'$, was calculated using an assumed

function, $S(\lambda)$, for the source spectrum based on a prior estimate of the source spectrum. The parameters of the assumed source spectrum were then iteratively varied until a minimum was found in χ^2 , defined by:

$$\chi^2 = \sum_{i=1}^N \frac{1}{\sigma_i^2} (\Delta A_i - \Delta A_i')^2 \quad (4)$$

where N is the number of dosimeters and σ_i is the error in ΔA_i . The iteration converged rapidly and was generally insensitive to the starting parameters. The iterative non-linear Marquardt method (description and Pascal code in Bevington and Robinson, 1992) was used to minimise χ^2 in the examples shown in this paper. This technique of assuming a function for an unknown and varying the parameters of the function to provide the optimum fit to the measured data has been applied by previous research (Rundel, 1983, de Gruijl *et al.*, 1993) to extract action spectra for particular biological processes. This paper employs the method to estimate an unknown source spectrum from a series of dose measurements made by dosimeters of different materials.

Minimisation of χ^2 , by the above or other techniques, in itself is insufficient to determine the source spectrum reliably. Additional information used to constrain the process is that the extracted function must have a root at the wavelength where the source spectrum has little or no irradiance, for example, at a wavelength of 250 nm for the quartz halogen lamp. Also choice of the starting function for $S(\lambda)$ must be based on a prior estimate of the expected spectrum. The latter is not an unreasonable requirement since the technique is intended to identify moderate variations in the terrestrial UV. Based on these constraints, a function with the following form was employed.

$$S(\lambda) = (\lambda - \lambda_0) \left(\sum_{i=1}^n a_i \lambda^{i-1} \right) \quad (5)$$

where λ_0 is the wavelength where the irradiance is zero and the first function fitted by minimising χ^2 is for $i=1$. The value of n is increased in steps of 1 with a function fitted in each case by minimising χ^2 . The value of n is limited by the number of dosimeters, in this case 4. The process is stopped when an additional term does not provide a significant improvement in the fitted function according to an F-test calculated from successive values of χ^2 .

Following estimation of the source spectrum, the UV_{BE} for a particular biological process can be calculated according to Equation (1) by employing the appropriate action spectrum or alternatively, the UVB, UVA or total UVA and UVB irradiance may be calculated by summation of the irradiance in the wavebands 280 to 320 nm, 320 to 400 nm and 280 to 400 nm respectively.

Each of the individual dosimeter materials has been calibrated for erythemally effective exposure for the summer solar spectra in order to compare the exposures derived from the spectra evaluated with the technique described in this paper with those obtained by employing each of the dosimeter materials individually. The dosimeters were exposed for a series of exposure times to summer solar UV between 08:22 and 13:53 EST under a clear sky and the solar spectrum measured with the calibrated spectroradiometer scanning at intervals over the waveband 280 to 400 nm. The erythemally effective exposure for each exposure period was calculated by employing the erythema action spectrum (CIE, 1987) in Equation (1) and multiplying the UV_{BE} by the exposure period,

T. The spectroradiometer measures the spectral irradiance in finite wavelength increments, $\Delta\lambda$, allowing the erythemally effective exposure to be calculated as:

$$\text{Exposure} = T \sum_{280}^{400} S(\lambda)A(\lambda)\Delta\lambda \quad \text{J cm}^{-2} \quad (6)$$

Measurement of the erythemally effective exposures in this manner along with the change in optical absorbance of each dosimeter material provides a calibration for the dosimeters.

As an example of application, the method of employing the four dosimeters to evaluate the spectrum was applied to measure human exposure to solar UV radiation during normal outdoor activities. The result was used to determine the erythemally effective dose. For this purpose, the system of four dosimeters has been miniaturised into one badge (3 cm x 3 cm) containing four holes of 0.6 cm diameter with one of the four different materials over each of the holes. This system of dosimeters has been employed to measure the personal exposure of four subjects for a 30 minute noon exposure in early winter in Toowoomba, Australia. During the exposure period, all of the subjects undertook outdoor activities with subject A jogging (31 May between 12:40 and 13:10 EST) and subjects B to D sitting and walking (2 June between 12:29 and 12:59 EST). A badge was attached to the top of the clothing and parallel to the body surface at two anatomical sites on each subject, namely, the center of the chest and the top of the left shoulder.

3. Results

Figure 1 shows the absolute spectral responses of NDA, 8-MOP, polysulphone and phenothiazine. Polysulphone exhibits a high response at UVB wavelengths with a rapid drop for wavelengths longer than 320 nm, NDA has a peak in response at approximately 325 nm and 8-MOP a peak at 300 nm. Phenothiazine responds to all UV wavelengths with a peak at about 320 nm.

Table 1 shows the measured changes in optical absorbance for the four dosimeter materials exposed to the quartz tungsten halogen lamp for a period of 30 min at a distance of 10 cm. Also shown are values predicted using the measured spectrum and the dosimeter response functions. The measured and predicted values agree within the typical variability of 10%. The source spectrum extracted using the measured values from Table 1 is shown in Figure 2 along with the spectrum measured using the spectroradiometer. The extracted spectrum has the form of Equation 5 with $n = 2$ and a value for χ^2 of 0.75 with two degrees of freedom. Increasing to $n = 3$ does not provide a significant improvement in fit according to an F-test. A more complicated UV spectrum may provide a significant improvement by increasing to $n = 3$. This is possible with the current system of four different dosimeter materials.

An ‘integrated difference’ between the evaluated and measured spectra is quantified by summing the absolute differences between the spectra at 1 nm intervals and dividing by the integrated spectral intensity of the measured spectrum; the result in this case being 8%.

Sensitivity of the method to the previously mentioned variability of the dosimeters was tested by repeating the procedure and varying the ΔA value $\pm 10\%$ from the measured values in Table 1. Each of the 16 possible combinations were examined; none of the

extracted spectra had integrated differences as described above of more than 23% in the range 250 nm to 400 nm. Table 2 presents the UVA, UVB and the total UVA and UVB, the erythema weighted (CIE, 1987) and the hazard function weighted (IRPA, 1989) (combined effects of skin and eye) irradiance for both the evaluated and measured spectra. The maximum discrepancy between the evaluated values and the measured values is 19% for the UVB waveband. The differences for the erythema weighted and the hazard function weighted irradiances are less than 10%.

Figure 3 plots the biologically effective UV for the quartz tungsten lamp at 10 cm calculated by weighting the evaluated source spectrum with the action spectra available in the literature for erythema (CIE, 1987) and the Hazard function (IRPA, 1989) for humans. For comparison the source spectrum measured with the spectroradiometer is similarly weighted and plotted in Figure 3. Figure 3 also displays the biologically effective UV calculated employing the generalised plant damage action spectrum (Caldwell, 1971) and the plant damage action spectrum derived for a variety of photoresponses in intact cucumber (Coohill, 1989). The comparison is reasonably good for the biologically effective UV in all cases.

The result of changing the source intensity by shifting the lamp to a distance of 12 cm and extracting this different spectrum is presented in Figure 4. The evaluated spectrum appears to deviate from the measured spectra for wavelengths shorter than 280 nm. This deviation is exaggerated by the logarithmic scale on the vertical axis of the graph and has no significant effect on the total UV irradiance as the lamp output below 280 nm is very small. Both the lamp spectra obtained at distances of 10 cm and 12 cm can be evaluated with the present method to an accuracy of about 10%. The results of evaluating the solar UV spectra produced 'integrated differences' between the evaluated and measured spectra of 17% or less. Table 3 lists the UV irradiances in the respective wavebands and the erythema and hazard function weighted irradiances for both the evaluated and measured spectra of solar exposures. For both of the measurement times of 09:18 and 11:39 EST, the differences are less than 16%.

The erythemal irradiances in Table 3 derived from the spectra measured with the spectroradiometer and the spectra evaluated with the technique described in this paper have been converted to the erythemal exposure for the five minute exposure periods and presented in Table 4. In the last four columns of this Table, these are compared to the erythemal exposures obtained by employing each of the dosimeter materials individually to derive an erythemal exposure from the measured ΔA of each material due to the irradiance. The discrepancies between the erythemal exposures obtained with the spectra measured with the spectroradiometer and the individual dosimeters were the smallest for the polysulphone dosimeters. However, if the source spectrum changed significantly from that for which the dosimeters were calibrated, the error may be as large as 40% (Wong *et al.*, 1994). In Table 4, the maximum discrepancies for NDA and 8MOP are 24 and 45% respectively. For phenothiazine, the erythemal exposure measured with phenothiazine is more than double that obtained with the spectroradiometer. These discrepancies are due to the differences between the spectral response of the dosimeters and the erythemal action spectrum. This is overcome with the technique described in this paper employing the combined system of four dosimeters to evaluate the spectrum. Knowledge of the UV spectrum allows the

biologically effective exposure to be derived for any action spectrum without the need of re-calibration for different source spectra.

The average of the shoulder erythematol exposures for the thirty minute period is $7.6 \pm 0.5 \text{ mJ cm}^{-2}$ with the variation represented as one standard deviation. Similarly, the average of the chest exposures is $5.6 \pm 0.5 \text{ mJ cm}^{-2}$. Comparisons of the chest and shoulder exposures show that the chest exposures were 0.7 to 0.8 the shoulder exposures.

4. Conclusion

A method using a dosimetric technique has been developed for the evaluation of the UV spectrum. The method does not detect any fine structure in the spectrum; the evaluated function is a smoothed version of the measured spectral irradiance. The extracted spectrum is useful in the calculation of UVB, UVA and total UVA and UVB doses, and in calculating the biologically effective ultraviolet radiation for a particular biological process for studies of the effects of increased UV radiation. The method can be used to evaluate the UV spectra and biologically effective UV with a maximum difference of less than 20% compared to those measured with a calibrated spectroradiometer. The accuracy of the method is limited by the number of suitable materials available to use as dosimeters. The package used in the present study is four and it restricts the number of parameters in the fitted equation (curve) to four. The accuracy can be improved by increasing the number of materials used in the dosimetric package. The technique may readily be streamlined into a computer package with the user inputting the four changes in absorbances and the type of UV source, for example, solar, quartz tungsten lamp or any other lamp. The software would maintain a database of the constraints and the initial values of the parameters in the iteration for that particular UV source and output the values of the parameters of the evaluated spectrum. With this streamlined approach, the spectrum would be evaluated in several minutes.

The method provides the advantages over the conventional method of spectrum measurement employing spectroradiometers in that it is cost effective and the small passive detectors allow simultaneous evaluation of the UV spectrum at multiple sites and at any orientation. The system can be used to estimate the biologically effective exposure once the data of the spectral response functions and the action spectra for the biological process are entered into the computer program. There is no need to re-calibrate the system for each biological process or whenever the source spectrum changes. An example is the evaluation of the UV spectrum at various human body sites to determine the personal erythematol exposures during normal daily activities. The system of four dosimeters provides the ability to evaluate the spectrum for any orientation. It can be attached at any point over the object of study to evaluate the spectrum at that point. The system was applied to measure the erythematol exposures of subjects during outdoor activities with the chest exposure 0.7 to 0.8 those of the shoulder. Processing four dosimeters instead of one will increase slightly the experimental time however it is more than compensated for as the calibration procedure is greatly simplified. Due to the ruggedness, portability of the dosimeters and the absence of the requirement of power and ancillary equipment, the technique is of importance in field experiments measuring personal erythematol exposures where the usage of sensitive and expensive equipment is impractical.

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Table 1. Measured and predicted changes in the optical absorbance of each dosimeter exposed to the quartz tungsten halogen lamp for a period of 30 minutes at a distance of 10 cm.

Dosimeter	Measured ΔA	Predicted ΔA
Nalidixic acid	0.037 ± 0.004	0.034
Polysulphone	0.055 ± 0.006	0.049
8-methoxypsoralen	0.019 ± 0.002	0.017
Phenothiazine	0.43 ± 0.04	0.42

Table 2. Comparison of the measured and evaluated irradiances for the quartz halogen lamp at a distance of 10 cm in the UVA and UVB wavebands and for the erythema and Hazard function weighted irradiances.

Waveband	Measured ($\mu\text{W cm}^{-2}$)	Evaluated ($\mu\text{W cm}^{-2}$)
UVA and UVB	807	815
UVB	72	86
UVA	735	729
Erythema weighted	40	43
Hazard function weighted	26	27

Table 3. Comparison of the measured and evaluated irradiances for the solar summer UV spectrum at 09:18 and 11:39 in the UVA and UVB wavebands and for the erythema and Hazard function weighted irradiances.

Waveband	Solar at 09:18 EST		Solar at 11:39 EST	
	Measured ($\mu\text{W cm}^{-2}$)	Evaluated ($\mu\text{W cm}^{-2}$)	Measured ($\mu\text{W cm}^{-2}$)	Evaluated ($\mu\text{W cm}^{-2}$)
UVA and UVB	5278	4836	6458	5448
UVB	265	231	344	317
UVA	5013	4605	6114	5131
Erythema weighted	16	17	22	23
Hazard function weighted	4.0	3.5	5.7	4.8

Table 4. Comparison of the measured and evaluated solar erythemat exposures with the exposures derived for the same exposure period with the individual dosimeters.

Time (EST)	Erythemat Exposures (mJ cm^{-2})					
	Measured	Evaluated	NDA	Polysulphone	8MOP	Phen
09:18	4.9	5.1	3.7	5.3	7.1	11.7
11:39	6.6	7.0	5.8	7.4	8.7	17.9

- Figure 1. Spectral responses of nalidixic acid (Tate *et al.*, 1980) (1), 8-methoxypsoralen (Diffey and Davis, 1978) (2) and polysulphone (CIE, 1992) (3) with the normalization factors applied as determined in this research and the spectral response of phenothiazine (4) as determined in this research.
- Figure 2. Comparison of the spectral irradiance for the evaluated spectrum (1) and the spectrum measured with a spectroradiometer (2).
- Figure 3. Biologically effective UV for the quartz tungsten lamp at 10 cm for the erythral action spectrum (CIE, 1987) with the calculated (1) and the measured spectrum (2), the hazard function (IRPA, 1989) with the calculated (3) and the measured spectrum (4), generalised plant damage (Caldwell, 1971) with the calculated (5) and the measured spectrum (6) and for plant damage for a variety of photoresponses in intact cucumber (Coohill, 1989) using the calculated (7) and the measured spectrum (8).
- Figure 4. Spectral irradiances for a lamp distance of 12 cm showing the evaluated spectrum (1) and the spectrum measured with a spectroradiometer (2) compared to the evaluated spectrum (3) and the measured spectrum (4) for a lamp distance of 10 cm.

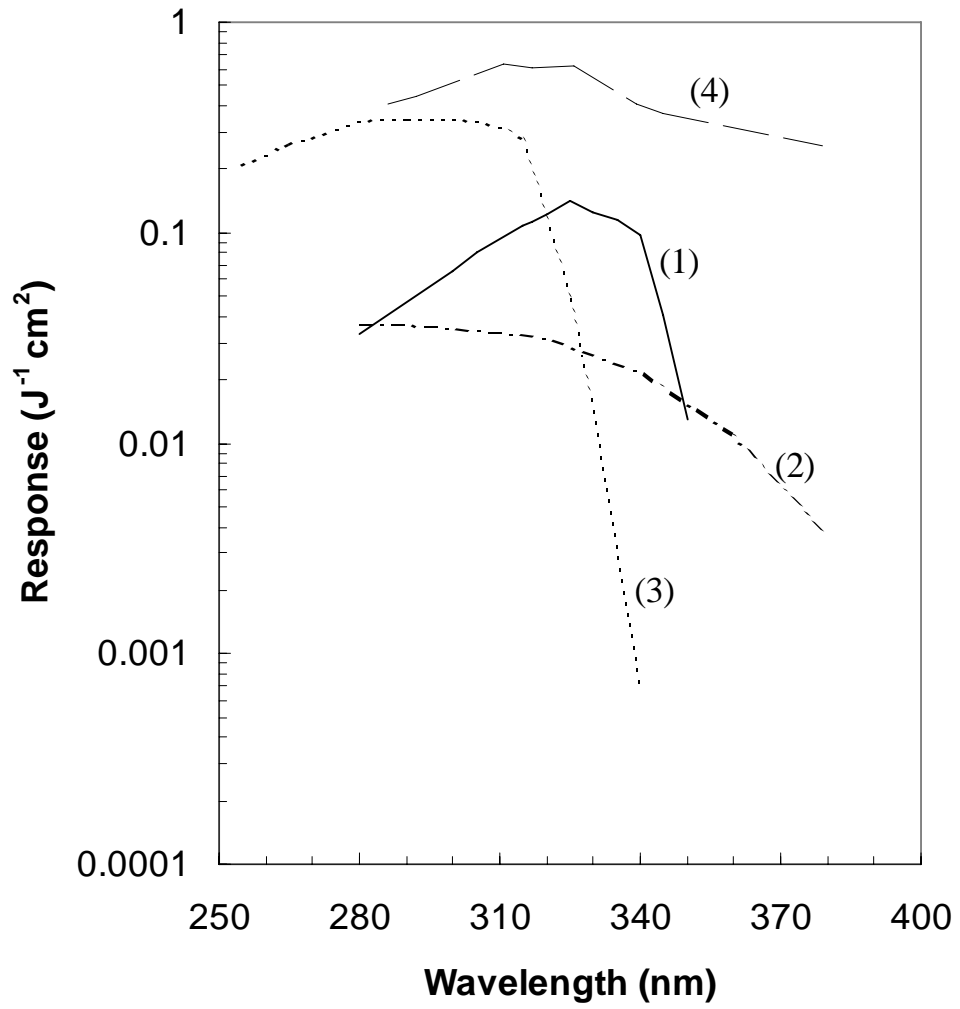


Figure 1.

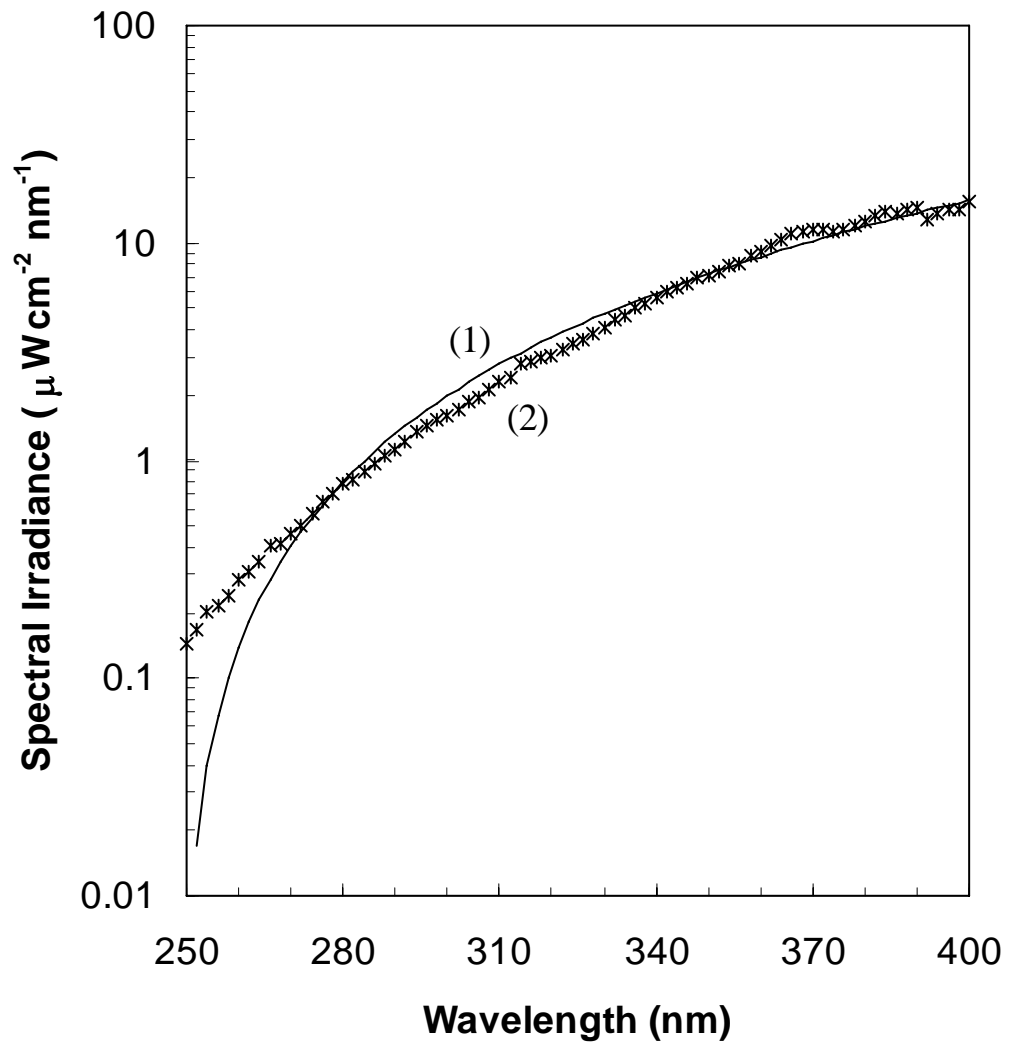


Figure 2.

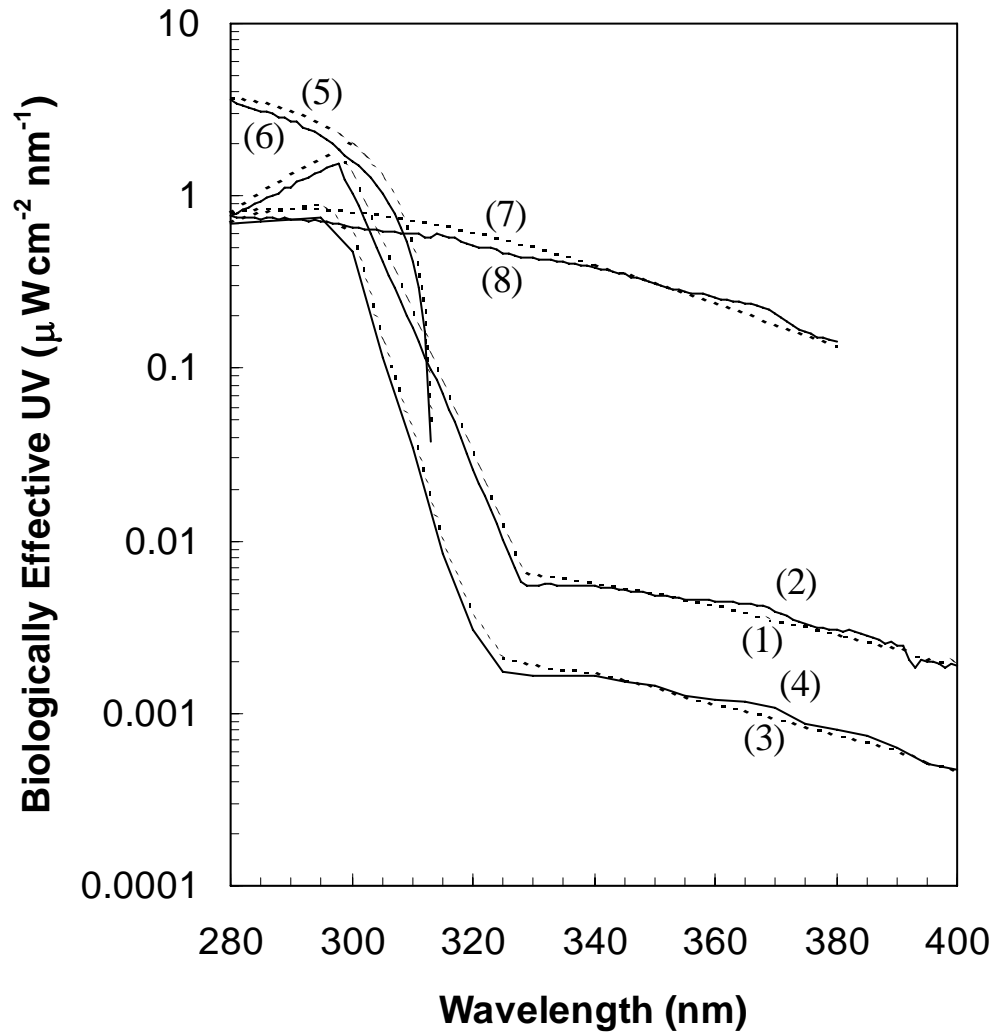


Figure 3.

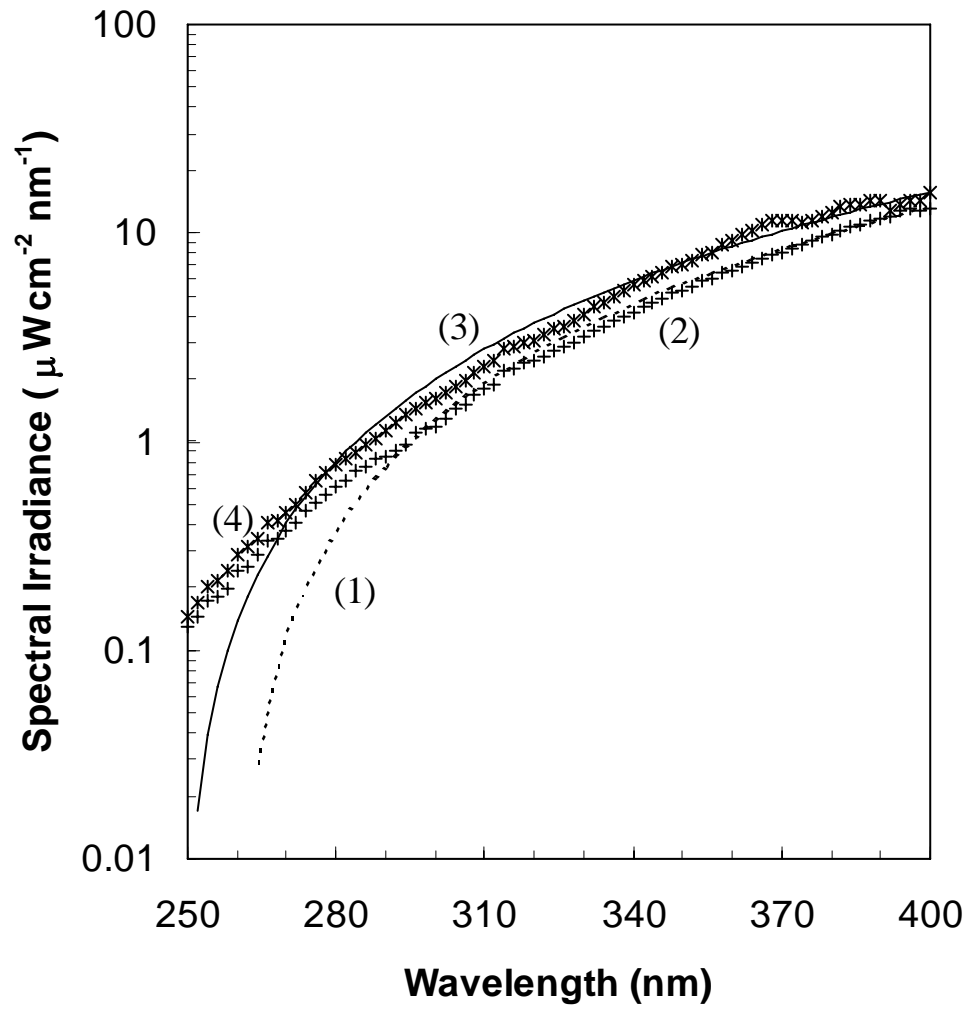


Figure 4.