

Usage of the Polyphenylene Oxide Dosimeter to Measure Annual Solar Erythemat Exposures

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ABSTRACT

Poly (2, 6-dimethyl-1, 4-phenylene oxide) (PPO) film is a useful dosimetric tool for measuring solar UV in underwater and terrestrial environments. However, little is known about how the response of PPO changes with fluctuations in atmospheric ozone and also to seasonal variations. To resolve this issue this manuscript presents a series of long-term in-air solar erythemat response measurements made over a year from 2007 to 2008 with PPO. This data showed that the PPO dose response varies with modulations of the solar spectrum resulting from changes in season and atmospheric ozone. From this it was recommended that PPO only be calibrated in the season in which it is to be used at the same time as measurements were being made in the field. Extended solar UV measurements made by PPO with a neutral density filter (NDF) based on polyethylene are also detailed. These measurements showed that the lifetime of PPO could be extended by five days before saturation. As the dynamic range for PPO is known to be five days during summer at a sub-tropical location, the advantage of using the NDF is that half the number of dosimeters is needed to be fabricated and measured before and after exposure.

This is the authors' final corrected accepted version of:
Schouten, Peter and Parisi, Alfio and Turnbull, David (2010) *Usage of the polyphenylene oxide dosimeter to measure annual solar erythemat exposures*. *Photochemistry and Photobiology*, 86 (3). pp. 706-710. ISSN 0031-8655 Accessed from USQ ePrints <http://eprints.usq.edu.au/8338/>

INTRODUCTION

Previous studies have shown that the PPO dosimeter is capable of measuring long-term cumulative amounts of UV exposure (1, 2, 3, 4) with a dynamic range much greater than that of the more commonly used polysulphone dosimeter. Some of the tests performed in these investigations included cosine response, temperature sensitivity, interdosimeter variation, spectral response and dark reaction susceptibility. However the exposure lifetime of the PPO dosimeter can be extended drastically by the use of a neutral density filter (NDF). An NDF is simply a piece of material that is attached to the top side of a dosimeter that is only partially transparent to UV wavelengths. Parisi & Kimlin (5) have used a NDF based on developed black and white photographic film to extend the dynamic range of the polysulphone dosimeter by as much as six times beyond its usual limit. The first section of this manuscript continues this research and describes how a polyethylene NDF can be calibrated and used successfully with the PPO dosimeter in order to continuously measure in-air solar sun burning (erythemal) exposures over the space of a month in low SZA conditions without the need for replacement.

There have been very few studies carried out in the past that have extensively investigated the influence of seasonal change along with variable atmospheric constituents such as column ozone upon dosimetric calibration regimes. A study was performed where polysulphone was calibrated to solar exposures over the space of two years in Munich, Germany (6). From this work it was discovered that by factoring seasonal stratospheric ozone patterns along with local SZA variation into the measurement data, total exposure measurement errors could be reduced by as much as 23%. Kollias *et al* (7) has employed the polysulphone dosimeter to measure solar UVB variations over the time frame of several years in the northern hemisphere. The researchers found that there was a seasonal shift between the UVB data measured with the polysulphone dosimeter and synthetic UVB data derived from a model produced originally by Kollias *et al* (8). Fluctuations in total atmospheric column ozone thickness occurring during the measurement campaign were believed to have been one of the main contributing factors behind this seasonal shift along with variations in the rotation of the Earth around the sun and the rotation of the sun about its own axis. Casale *et al* (9) extensively calibrated the polysulphone dosimeter over each season at three different field locations throughout Italy. The results from this campaign showed that a strong correlation was present between variations in the measurement profiles and variations in total column ozone levels along with SZA. At this point in time no study has analysed the effect of seasonal change and atmospheric column ozone upon the response of the PPO dosimeter in-air. This manuscript will directly address this issue and will show that in-air measurements of the PPO dosimeter to solar exposures are susceptible to atmospheric and seasonal variability and change in a similar way compared to the polysulphone dosimeter.

MATERIALS AND METHODS

Neutral density filter properties. After an initial analysis, it was decided that polyethylene sourced from common waste bags would be used as the NDF material, primarily due to their availability in local Australian supermarkets and also because of their ability to withstand the elements of nature such as wind and rain without succumbing to critical amounts of damage. The waste bag chosen for use in this research was the Coles Savings brand bag (referred to as the NDF material from this point on). The transmission and absorbance spectra as measured for this NDF material by a spectrophotometer (model 1601, Shimadzu Co., Kyoto, Japan) are depicted in Figure 1. Figure 1 details the spectral transmission and absorbance of the NDF material after an equivalent solar erythemal dosage of 45 kJ m^{-2} , which equates to approximately 30 days solar exposure in mid autumn at the measurement location (the University of Southern Queensland campus in Toowoomba, Australia (27.5°S , 151.9°E , 693 m above sea level)). From this data it is clearly seen that the NDF material had high UV transmission and low UV absorbance making it very transparent to UV wavelengths, along with near linear characteristic transmission and absorbance spectra across the UV waveband for which the PPO dosimeter is most responsive (approximately 300 nm to 340 nm). This attribute of the NDF material was desirable as it allowed for equal amounts of solar energy to be filtered through to the PPO dosimeter at each wavelength, leading to the reduction of measurement uncertainty that could be brought on by changes in the incident solar UV spectra.

Also from Figure 1, it is clear that the optical properties of the NDF material do change slightly after exposure to a substantial amount of solar energy. For example, at 315 nm a difference of 3.5% is present between the pre and post exposure transmission spectra. Also at the 315 nm point, a difference of 0.08 between the pre and post exposure absorbance spectra was measured to occur. These changes are taken into account in the calibrations.

PPO dosimeter and neutral density filter measurements. The NDF and PPO dosimeter system was produced by cutting out a 1.3 cm x 1.7 cm piece of NDF material and attaching it to a PPO dosimeter by using standard electrical tape. Cumulative erythemal exposures were measured by a Solar Light UV broadband meter (model 501, Solar Light Co. PA, USA) calibrated to a Bentham spectroradiometer system (Bentham Instruments, Reading, UK) with a calibration regime traceable to the National Physical Laboratory (NPL). A full description of the complete specifications of the Bentham spectroradiometer system is available in a previous study (10). Complete operational and response specifications for the Solar Light UV broadband meter can be found at <http://www.solarlight.com/products/501.html>. At this website it has been claimed by the manufacturer that the Solar Light UV broadband meter has an angular response of within $\pm 5\%$ compared to the ideal cosine response. Also, the total uncertainty of the Solar Light UV broadband meter has been estimated to be approximately $\pm 11\%$ (11). The first NDF and PPO dosimeter measurement series (Measurement A) ran in early autumn from 5 March 2008 until 28 April 2008, while

the second series (Measurement B) ran in late autumn from 29 April 2008 to 27 May 2008. One dosimeter was used to obtain pre and post absorbance data for each time point in these trials. The dosimeters were left out for approximately seven hours each day. The response of the dosimeters in these series was calibrated to the erythemal solar UV dose (12) with respect to the horizontal plane. The total solar UV dose measured by the Solar Light UV broadband meter was obtained by measuring the amount of solar UV incident on the dosimeters each day. As soon as the dosimeters were exposed to the sunlight in the morning the Solar Light UV broadband meter was turned on. After the seven hour measurement session had been completed in the afternoon the Solar Light UV broadband meter was turned off. At the end of the measurement series, the seven hour daily total dosages were added together in order to produce the total erythemal solar UV dose. A single filtered dosimeter (with an attached NDF) was removed from solar exposure every second day and a single unfiltered (without an attached NDF) dosimeter was removed from solar exposure each day during the measurement campaign. For the early autumn readings, the last remaining filtered and unfiltered dosimeters were left out until they were dark orange in colour, which for PPO film is a visual indication that complete optical saturation is about to take place. For the late autumn readings, the final filtered and unfiltered dosimeters were removed before the beginning of winter. Eleven unfiltered and eight filtered dosimeters were used in the early autumn measurements. Ten unfiltered and eight filtered dosimeters were used in the late autumn measurements. A spectrophotometer (model 1601, Shimadzu Co., Kyoto, Japan) was used to measure pre and post exposure absorbance data for the dosimeters at an optimal wavelength of 320 nm as specified in previous studies (1, 2, 3, 4). Across both of the Measurement trials, local cloud coverage ranged from 0 to 8 oktas.

Seasonal in-air erythemal measurements of the PPO dosimeter to deduce the influence of ozone and SZA upon responsivity. The measurements for this investigation were also made at the University of Southern Queensland campus in Toowoomba, Australia over 12 months from March 2007 through to February 2008 inclusive again using the Solar Light UV broadband meter calibrated to the Bentham spectroradiometer system. Only unfiltered dosimeters were employed for this trial. The SZA range for the autumn measurements was 20° to 70°. The SZA range for the winter measurements was 35° to 65°. The SZA range for the spring measurements was 8.4° to 53°. The SZA range for the summer measurements was 5.5° to 44°. Each measurement series ran for seven days in total. An average value across six dosimeters was used to obtain pre and post absorbance data for each time point in these trials. The dosimeters were exposed to the sun for approximately seven hours per day. Cloud coverage varied from 0 to 8 oktas over the year-long trial period. As explained in the introduction, the measurements were made for the PPO dosimeter over all four seasons so that the effect of different atmospheric column ozone levels and changing SZA on the response of the PPO dosimeter could be quantified. Four separate campaigns were carried out in each season in order to produce a substantial amount of data from which seasonal complete data sets could be formulated. As was

done for the early and later autumn NDF and PPO dosimeter readings, the change in optical absorbance for each of the dosimeters in the seasonal measurements was calibrated to the CIE (12) erythemal UV dose with respect to the horizontal plane. The erythemal action spectrum was chosen for this research as it is by far the most commonly used measure of the effect of damaging UV radiation upon the human population. The calibration curves were obtained by measuring the ambient erythemal effective UV dose (UV_{ERY}) with the Solar Light UV broadband meter over the required UV wavelength range and the corresponding change in PPO film absorbance (after and before exposure) which depends on the time-integrated erythemal effective UV irradiance incident on the dosimeter:

$$UV_{ERY} = \sum_{t_1}^{t_2} \sum_X^Y S(\lambda)A(\lambda)\Delta\lambda\Delta t$$

where $S(\lambda)$ is spectral irradiance in units of $W m^{-2} nm^{-1}$, $A(\lambda)$ is the erythemal action spectrum (12, 13, 14), t_1 and t_2 is the time interval over which the dosimeters were exposed to the sun and X and Y are the limits of the UV spectrum defined as 280 nm and 400 nm respectively. Lester et al (1) has shown that the PPO response spectrum is similar to the erythemal action spectrum predominantly within the waveband running from 280 nm to 340 nm. However, a comparison between the PPO and erythemal response spectra using two spectral scans measured across the terrestrial UV waveband at SZA of 20° and 60° on the same day under the same ozone level and cloud free conditions showed that a percentage difference of about 16% can occur between the two weighted spectra. This percentage difference may increase or decrease alongside changes in cloud cover, ozone and SZA.

Ozone monitoring. Over the duration of both the PPO and NDF measurements and the year-long measurement campaign, column ozone levels above Toowoomba were monitored by accessing OMI satellite information (http://jwocky.gsfc.nasa.gov/ozone/ozone_v8.html) each day. This data was used in order to deduce if ozone variations had any influence upon the measurement data from season to season. Figure 2 shows a time series of the ozone levels measured by the OMI satellite from March 2007 to May 2008 above Toowoomba. Aerosol levels were not analysed as Toowoomba is a high-altitude location with an atmosphere that is relatively low in aerosol concentrations due to minimal levels of anthropogenic emission output in the region.

RESULTS

PPO dosimeter and neutral density filter measurements

Figure 3 shows the measurement data sets measured for the filtered and unfiltered dosimeters during autumn 2008. The x-axis error bars represent the $\pm 7\%$ error inherent within each in-air PPO dosimeter measurement as estimated in previous works (1). These data sets obtained for each of the filtered and unfiltered trial

measurements were fitted with second-order polynomial equations passing through the origin. The equation obtained for the early autumn unfiltered series was:

$$UV_{ERY} = 2.85(\Delta A_{320})^2 + 14.10(\Delta A_{320})$$

with an R^2 value of 0.96 and with UV_{ERY} measured in kJ m^{-2} . For the early autumn filtered series the measurement equation was:

$$UV_{ERY} = -1.57(\Delta A_{320})^2 + 21.58(\Delta A_{320})$$

having an R^2 value of 0.93 and UV_{ERY} given in kJ m^{-2} . The measurement equation for the late autumn unfiltered series was given as:

$$UV_{ERY} = 8.5(\Delta A_{320})^2 - 5.86(\Delta A_{320})$$

with an R^2 value of 0.84 and UV_{ERY} once again measured in units of kJ m^{-2} . The final equation produced was for the late autumn filtered series and it was as follows:

$$UV_{ERY} = 15.11(\Delta A_{320})^2 - 9.99(\Delta A_{320})$$

where the R^2 for this fit was 0.93 and with UV_{ERY} provided again in units of kJ m^{-2} .

Seasonal in-air erythemal measurements of the PPO dosimeter to deduce the influence of ozone and season upon responsivity

A complete measurement series developed from weekly data sets obtained in each season from March 2007 to February 2008 is provided in Figure 4, with the x-axis error bars showing the standard deviation for each data point. To produce these complete seasonal measurements, data sets were recorded for four weeks per season and averaged to generate the four separate data sets which were then directly compared against each other in order to find out if they were dependent on season or atmospheric column ozone levels. These campaigns were run at staggered intervals throughout each season so that a broad cross section of varying atmospheric conditions was encountered each time. A second-order polynomial equation that went through the origin was used to describe the trend of each erythemal measurement data set:

$$UV_{ERY} = -\nu(\Delta A_{320})^2 + \kappa(\Delta A_{320})$$

where UV_{ERY} is the erythemal effective UV dose measured in units of kJ m^{-2} . In autumn this equation was:

$$UV_{ERY} = -3.1(\Delta A_{320})^2 + 11.2(\Delta A_{320})$$

with an R^2 value of 0.99 where UV_{ERY} is the erythemal dosage expressed as kJ m^{-2} . For winter the equation was:

$$UV_{ERY} = -3.2(\Delta A_{320})^2 + 8.9(\Delta A_{320})$$

with an R^2 value of 0.99 where again UV_{ERY} is the erythemal dosage in units of kJ m^{-2} . The equation for spring was:

$$UV_{ERY} = -3.4(\Delta A_{320})^2 + 14.3(\Delta A_{320})$$

with an R^2 value of 0.99 where UV_{ERY} is also measured in units of kJ m^{-2} . In summer the equation was:

$$UV_{ERY} = -3.22(\Delta A_{320})^2 + 17.5(\Delta A_{320})$$

with an R^2 value of 0.99 with the UV_{ERY} dosage given once more in kJ m^{-2} .

DISCUSSION

From Figure 3 it is clearly seen that from the measurement data sets that the filtered dosimeters were capable of measuring far greater amounts of solar erythemal exposure in comparison to the unfiltered dosimeters, thus extending the effective life time of the PPO dosimeter. In Measurement A (early autumn, 5 March 2008 until 28 April 2008) the filtered dosimeters measured an extra 14 kJ m^{-2} in comparison to the unfiltered dosimeters before the earliest beginnings of optical saturation. This equated to an extra five days of exposure time. In Measurement B (late autumn, 29 April 2008 to 27 May 2008) the difference was not as sizeable, with the filtered dosimeters measuring 2 kJ m^{-2} more erythemal exposure than the unfiltered dosimeters, which was an extra two days worth of solar exposure for late autumn. The changes occurring in the optical properties of the NDF material after solar exposure as displayed in Figure 1 suggests that it may have to be replaced after extended periods in the field. From this, it is recommended that the NDF material is substituted once a month in order to limit the effect that these inherent and unavoidable optical property changes have upon the responsivity of the PPO dosimeter. As the dynamic range for PPO has been measured to be approximately five days during summer at a sub-tropical location, the advantage of using an NDF is that as much as half the number of dosimeters is needed to be fabricated and measured before and after exposure saving the researcher a substantial amount of time and effort.

Again, from Figure 1 it can be seen that the spectrophotometer measured a 20% transmittance on average across the 300 nm to 340 nm waveband for the NDF material. This suggests that the NDF and PPO dosimeter system could theoretically be used to measure up to five times more solar exposure at any given time in comparison to an unfiltered PPO dosimeter. However, as described in the previous paragraph, the actual observed increase in the measured exposures was much less.

In addition to the measurements obtained between both the filtered and unfiltered PPO dosimeters in the Measurement A and Measurement B campaigns having dissimilar magnitudes of total exposure, Figure 3 shows that they also have slightly different characteristic regimes. This was to be expected, as with the use of any UV NDF the incoming UV wavelengths are attenuated which alters their spectral energy distribution that in turn influences the response of the dosimeter underneath. So the PPO dosimeter must be calibrated for extended field use with an NDF attached to it. Additionally, this will also take into account any short and long-term variability in the absorption characteristics for the NDF material. The use of an unfiltered measurement regime to calibrate extended PPO and NDF dosimeter based exposures will most certainly lead to substantial errors and hence will not suffice.

Also from Figure 3, a substantial difference was also seen between the Measurement A (early autumn, 5 March 2008 until 28 April 2008) filtered and unfiltered data sets and the Measurement B (late autumn, 29 April 2008 to 27 May 2008) filtered and unfiltered data sets. The Measurement A campaign ran at an earlier time in autumn compared to Measurement B, which meant that the Measurement A dosimeters received on average a daily erythemal exposure of 1.6 kJ m^{-2} in contrast to the Measurement B dosimeters that were exposed to a sizeably lower average daily erythemal exposure of 1.03 kJ m^{-2} . This difference in average erythemal exposures caused by the gradual increase in peak SZA over autumn, means that the average solar erythemal spectra intercepted daily by the Measurement A dosimeters would have had different characteristics such as energy distribution and cut-off wavelength when compared to the average erythemal spectra intercepted daily by the Measurement B dosimeters, leading to the significant dissimilarities seen between the two data sets. Interestingly, ozone levels varied only slightly between the two campaigns with an average column ozone level of 262 DU measured during Measurement A and 261 DU measured during Measurement B. So column ozone was not believed to have played any part in influencing the deviations seen between the data sets gathered for the two Measurement series. In addition, the sensitivity of the PPO dosimeter to fluctuating fluence rates has already been analysed previously by both Lester et al (2003) and Schouten et al (2007). In these two studies the PPO dosimeter was measured to have a sound level of dose-rate independence in both terrestrial and underwater environments over a substantial amount of time. So the disparity between the Measurement A data and the Measurement B data could not have resulted from sensitivity to different fluence rates. Also, there does appear to be an offset in the small dosage response between the Measurement A data series and the Measurement B data series that did not occur in the data presented in Figure 4. One explanation that can be given for this is that during the Measurement B data collection phase a higher amount of cloud coverage was present in the local Toowoomba area which could have drastically influenced the composition of the incoming solar spectral irradiance, and as result changed the dose response of both the PPO dosimeter and the NDF.

Figure 4 shows that there were differences between the complete erythemal doses for the PPO dosimeter derived from the data sets measured over autumn, winter, spring and summer. The most substantial difference between all of the measurement regimes was measured to occur between the summer and winter measurements. For example, in winter a total solar erythemal exposure of 4.9 kJ m^{-2} resulted in a change of optical absorbance of approximately 0.75 in the PPO dosimeter. In comparison, the same amount of exposure received in summer resulted in a change in optical absorbency of close to 0.3. This equated to a substantial overall difference in optical absorbency of 0.45.

The coefficients calculated in the measurement trend equations derived for each season provide a picture of what is potentially causing the variability between the

seasonal erythemal measurement regimes. The ν coefficient given for each of the complete equations show only low differences with changes in season, with the combined average value measured for the autumn, winter, spring and summer data sets being 3.23 kJ m^{-2} with a standard deviation of $\pm 0.13 \text{ kJ m}^{-2}$, resulting in a level of variation equal to approximately 4%. Conversely, the κ coefficient appears to be the main factor behind the extensive variability between the seasonal complete measurement regimes. A combined average value of the κ coefficient for the autumn, winter, spring and summer data sets was calculated as 12.98 kJ m^{-2} with a standard deviation of $\pm 3.74 \text{ kJ m}^{-2}$ which equated to a 30% level of variation, over seven times as large as the level of variation estimated for the ν coefficient data. The primary cause behind the apparent inconsistency in the κ coefficient and in turn the fluctuations seen to occur between the complete erythemal measurements from season to season could be the disparity between the spectral response of the PPO dosimeter and the response of the Solar Light UB broadband meter.

Alongside the dissimilarity existing between the response spectra for the PPO dosimeter and the erythemal effect, inconsistent levels of atmospheric column ozone may have also modulated the seasonal measurement regimes. The column ozone trend over the year-long measurement campaign given in Figure 2 shows that ozone levels were at their lowest during the months of autumn and summer and at their highest during the months of winter and spring. As ozone is only capable of absorbing energy from the solar wavebands with wavelengths shorter than those in the UVA, erythemal UVB wavelengths incident upon the PPO dosimeters during autumn and summer would have been attenuated to a lesser extent in comparison to the erythemal UVB wavelengths incident in winter and spring leading to changes in the features of the solar erythemal spectra from season to season. To eliminate the introduction of any major errors in field exposure measurements made in-air brought on by seasonal changes in SZA coupled with varying column ozone trends, calibrations are required to be made with respect to the source spectrum that they will be measuring. For example, if field measurements are to be made in early winter, it is highly recommended that a calibration be performed simultaneously in the same location in order to factor in current trends in the atmospheric and geometric parameters directly affecting the response of the PPO dosimeter. It is also important to note that the PPO dosimeter should only be used under real daylight, as after exposure to simulated UV it may provide erroneous results unless the measurements are constrained and calibrated to that particular UV source.

This manuscript has shown that the PPO dosimeter can be used for the specific measurement of long-term solar erythemal UV at a substantial level of reliability as long as calibration campaigns are carried out under the necessary conditions. Additionally the employment of a polyethylene NDF has been detailed and proven to be a useful tool for application in extended measurements of the solar erythemal UV with the PPO dosimeter

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FIGURE CAPTIONS

Figure 1. Transmission and absorbance spectra across the waveband running from 300 nm to 340 nm for the NDF material after a solar erythemal exposure of 45 kJ m^{-2} .

Figure 2. Ozone over the Toowoomba region from March 2007 to May 2008.

Figure 3. Total solar erythemal doses measured by the Solar Light UV broadband meter for the filtered and unfiltered PPO dosimeters over the months of autumn 2008.

Figure 4. Calibration curves obtained in this study: Complete solar erythemal doses for winter, spring and summer and their second order fitting curves.

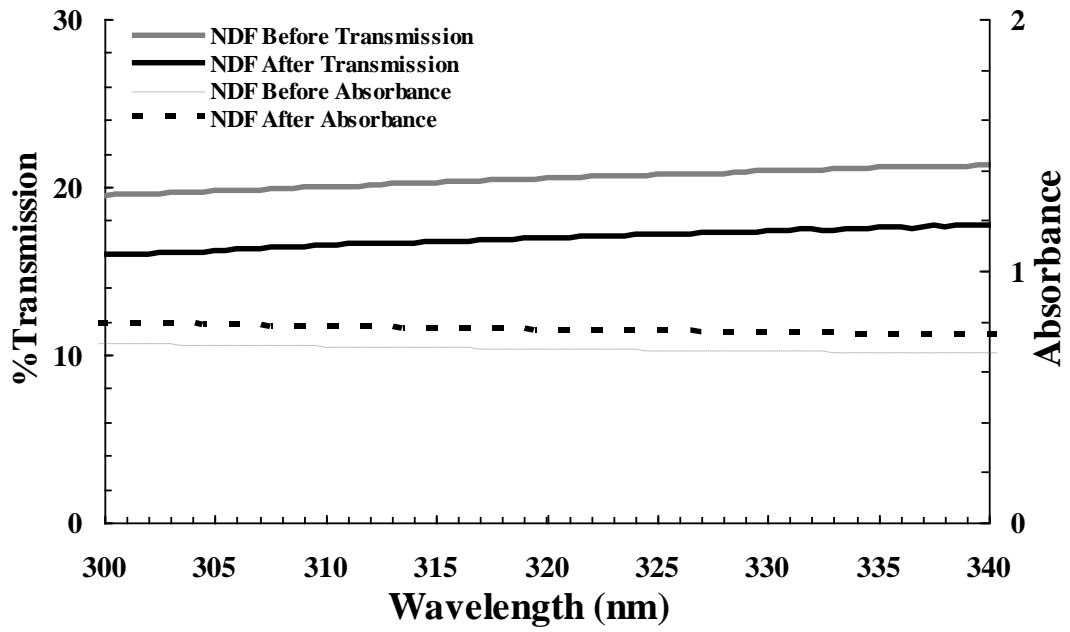


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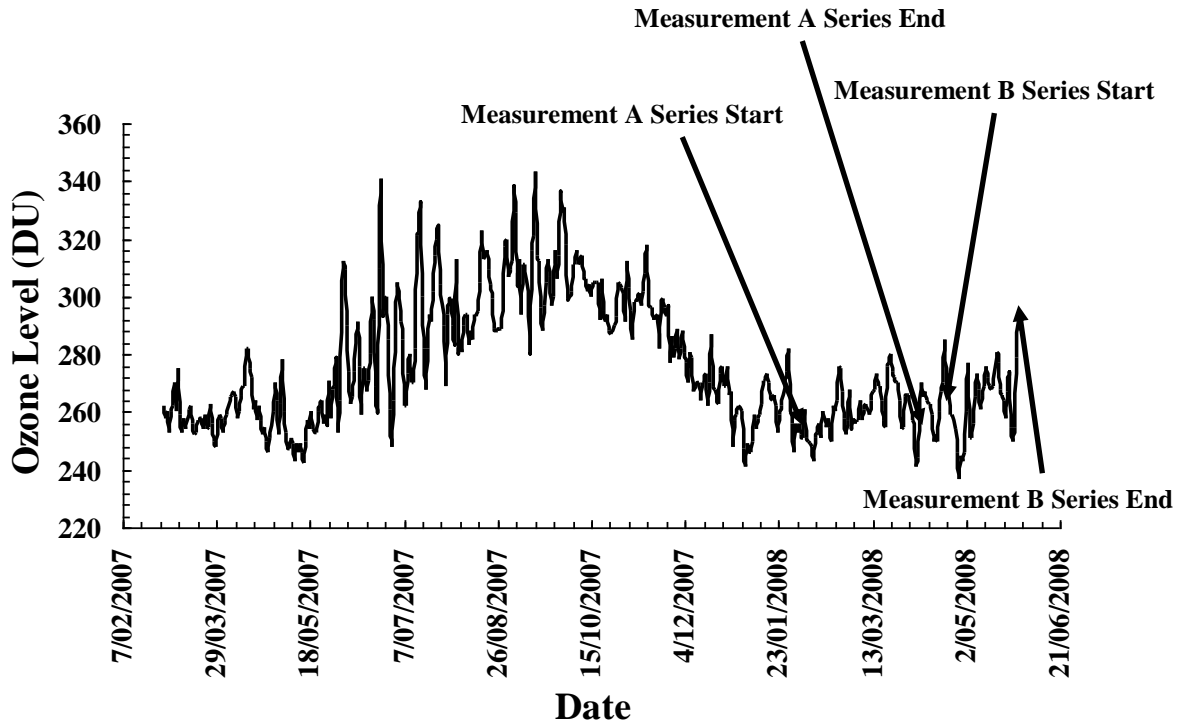


Figure 2. Ozone over the Toowoomba region from March 2007 to May 2008.

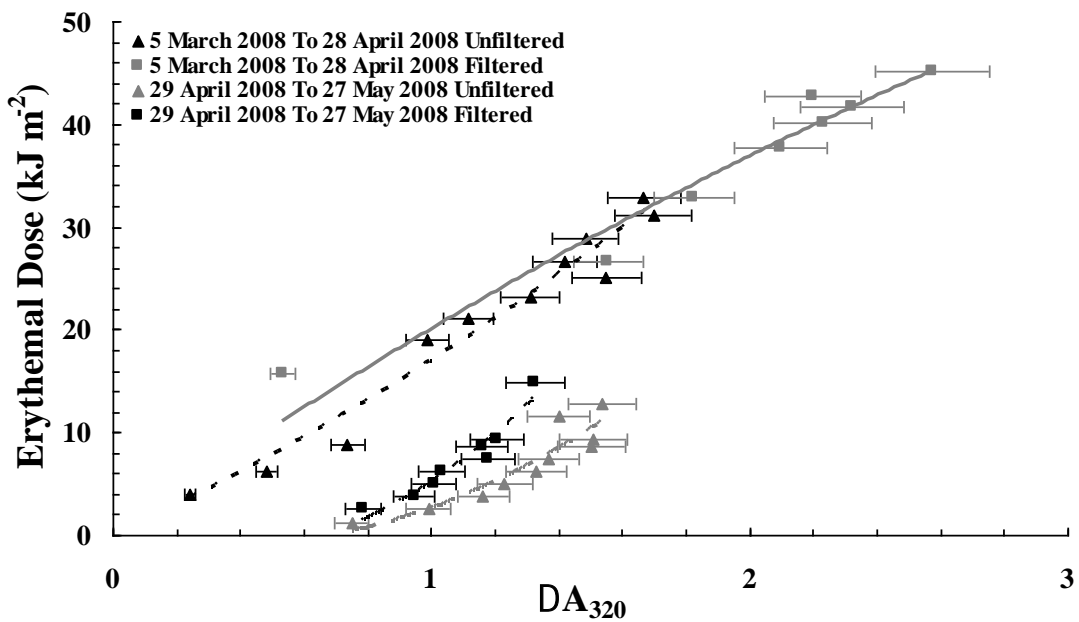


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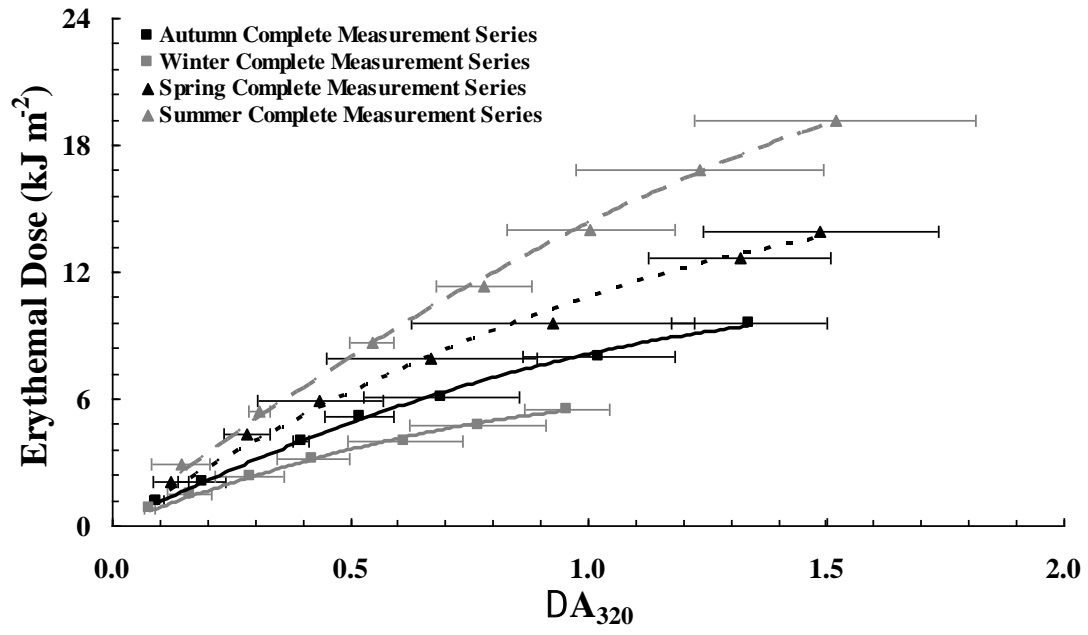


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