Variations and trends of biologically effective doses of solar ultraviolet radiation in Asia, Europe and South America from 1999 to 2007†

Nobuo Munakata,*[a] Stelios Kazadzis,[b,e] David Bolseé,[c] Nelson Schuch,[d] Tapani Koskela,[e] Alex Karpetchko,[f] Charoula Meleti,[b] Claudio Casiccia,[g] Marcelo Barcellos da Rosa,[f,h] Toshiaki Saida,[i] Chikako Nishigori,[j] Katsumi Ogata,[k] Kazuhiro Imafuku,[l] Chung-Ming Liu,[m] Sri Lestari,[n] Mpu Kanoko,[o] Santoso Cornain,[o] Ketut Mulyadi[p] and Kotaro Hieda[a]

Received 6th April 2009, Accepted 27th May 2009
First published as an Advance Article on the web 23rd June 2009
DOI: 10.1039/b906975e

Biological monitoring of solar UV radiation using spore dosimeters has been undertaken since the year 1999 at more than 20 sites in Asia, Europe and South America. The monthly-cumulative data to the end of the year 2004 have been presented before. In this paper, successive data to the end of the year 2007 are compiled and the trends and correlation analyses with yearly and monthly average amounts of columnar ozone are presented. Mean yearly doses at 10 northern and 6 southern hemisphere sites exhibited exponential latitudinal gradients with similar slopes indicating a doubling of the dose with the decline of about 14 degrees. Among 12 sites where continual data for more than 6 years were available, increasing trends in yearly UV doses were observed at 11 sites. At one European (Brussels), two tropical Asian (Padang and Denpasar), and two South American (São Martinho and Punta Arenas) sites, decreasing trends of ozone amounts were noted, whereas at the remaining 6 sites (five sites in Japan and Thessaloniki), increasing trends of the UV doses were observed without notable changes, or with an increase at one site (Kiyotake), of the average ozone amounts. At one site (Taipei), the UV doses and the ozone amounts stayed constant. In the monsoon areas, climatic variations and changes, particularly in the extent of cloudiness and frequency of rainfall in summer months, might have been largely responsible for the trends of the UV doses. However, even at these sites, the decreases in the ozone amounts in summer months were frequently observed and might have contributed to the increasing trends of the UV doses. Since each region and locality is unique in climatic and atmospheric conditions, it is not easy to generalize the global trends. However, at many sites involved in this monitoring project, the increases in the biological UV doses during this period seemed to be linked to the decreases in the ozone amounts.

Introduction

Destruction of the stratospheric ozone layer by human releases of chlorofluorocarbon compounds manifested itself in the spring-time ozone hole in Antarctic region and gave dire warnings of the fragility of atmospheric environment.[1,2] Despite international restrictions imposed on the release of ozone-depleting chemicals, it still seems uncertain when the trend of ozone loss might be halted.[3,4] Furthermore, the climate change linked to the release of greenhouse gases could also affect the atmospheric conditions and

[a]*Biophysics Laboratory and Research Information Center for Extremophiles, Faculty of Science, Rikkyo University, Tokyo, Japan*
[b]*Physics Department, Aristotle University of Thessaloniki, Thessaloniki, Greece*
[c]*Belgian Institute of Space Aeronomy, Brussels, Belgium*
[d]*Southern Regional Space Research Centre (CRSPE/INPE-MCT), Santa Maria, Brazil*
[e]*Finnish Meteorological Institute, Research and Development, Helsinki, Finland*
[f]*Finnish Meteorological Institute, Arctic Research Centre, Sodankylä, Finland*
[g]*Ozone and UV Radiation Laboratory, University of Magallanes, Punta Arenas, Chile*
[h]*Laboratory of Environmental Chemistry, University Vale do Rio Doce-UNIVALE, Gavernador, Valadares, Brazil*
[i]*Department of Dermatology, Shinshu University School of Medicine, Matsumoto, Japan*
[j]*Department of Dermatology, Kobe University, Kobe, Japan*
[k]*Department of Dermatology, Miyazaki University, Kiyotake, Miyazaki, Japan*
[l]*Division of Dermatology, Department of Organ Oriented Medicine, University of the Ryukyus, Nishihara, Okinawa, Japan*
[m]*Department of Atmospheric Sciences, National Taiwan University, Taipei, China*
[n]*Dermatology and Venereology Department, Andalas University, Padang, Indonesia*
[o]*Department of Anatomic Pathology, University of Indonesia, Jakarta, Indonesia*
[p]*Department of Anatomic Pathology, Udayana University Denpasar, Indonesia*

† Electronic supplementary information (ESI) available: Summary of solar UV doses determined with spore dosimetry from 2005 to 2007. See DOI: 10.1039/b906975e

result in changes and variations of UV doses.[5] Also, UV increases are possible due to the improvement in air quality by effective pollution control.[5] The uncertainties in the changes of ozone amounts, aerosol, and climate demand dedicated and coordinated efforts to monitor solar UV doses prevalent in the environment of terrestrial and marine organisms.

Physical measurements of solar UV radiation have been mainly carried out by the use of spectrophotometers.[6] This requires a spectroscopic instrument with fine resolution to the short-wavelength end of solar UV spectrum (290 nm or shorter), and human and material resources for instalment and maintenance including calibrations and comparisons of the instruments. Therefore the measurements could only be carried out at well-equipped institutions mostly in the mid-latitudinal sites in the northern hemisphere. Another problem inherent to physical measurements is how to guarantee the correlation with biological effects. The biological effectiveness, such as the induction of DNA damage, and solar spectral irradiance behave in the opposite way in the UV region: as the wavelength is shortened, the effectiveness increases while the irradiance decreases. Therefore, to estimate effective doses from irradiance spectra, it is necessary to use some simplified and hypothetical models such as an erythemal spectrum assuming strictly independent activity of each wavelength.[7] Since it is not easy to experimentally validate these model spectra and the absence of wavelength interactions, we are not sure which model to use for a particular concern and purpose.

Some meters that directly record biologically weighted doses might be less cumbersome for operation, but they still require controlled and coordinated efforts to ensure the uniformity and consistency of the meter response. Earlier efforts with Robertson–Berger type meters in many cities in USA have shown that the increase in the air pollution around the measuring sites is probably a more crucial factor determining the dose changes than ozone amounts.[8,9] Recently, several projects have been carried out to monitor UVB doses in Europe[10] and in Japan[11] with dedicated instruments. Unfortunately, these attempts are difficult to integrate and compare from the aspect of biologically effective doses, because the interpretation of the dose measured with these meters is based on different presumptions and models. We considered it was important to develop dosimeters that directly measured biologically effective UV doses in order to obtain more focused data pertaining to the effects on human health, productivity and the biosphere.[12,13]

The spore dosimeter has been developed for this purpose and exhibits several characteristics that make it suitable for global comparisons and long-term monitoring.[14–17] The samples are compact (about 5 cm square with 5 mm thickness) and require only an un-shaded horizontal plane on which to fix them. Thence, the monitoring can be performed without any extra equipment or electricity. Necessary operations at the exposure site are exchanging several samples at the end of each month and occasional dusting off the surface. The assay of inactivation depends on the strictly exponential survival that reflects the amount of DNA photoproducts mostly identified as 5-thyminyl-5,6-dihydrothymidine.[18,19] Therefore, the resultant dose directly provides the effectiveness of solar radiation to induce DNA damage. On the other hand, the need for filtering and protecting material introduces complications that should have been overcome through the experience in the exposure under various environmental conditions.

We started measurements of monthly-cumulative doses using spore dosimeters at several sites in the beginning of the year 1999. The result to the end of the year 2004 has been published.[17] In this paper, we present the data of the period 2005 to 2007, together with the analyses of variations and trends of the doses of solar-UV radiation in relation to the amounts of columnar ozone.

Experimental

Spore dosimetry

Preparations and assays of spore dosimeter samples, and the characterization of the blue polyethylene sheet used to protect the samples from rain and to reduce the dose have been described before.[17] The action spectra of the spore dosimeter.[16] and CIE erythemal dose (ED)[20] are shown in Fig. 1 together with a typical irradiance and effectiveness spectrum[7,16] Spore inactivation dose (SID) is derived from the absolute amount of the natural logarithm of the fractional survival multiplied by the factor of dose reduction of the covering sheet. The changes in UV transmittance of the covering sheet[17] determined before and after the exposure were less than 10%.

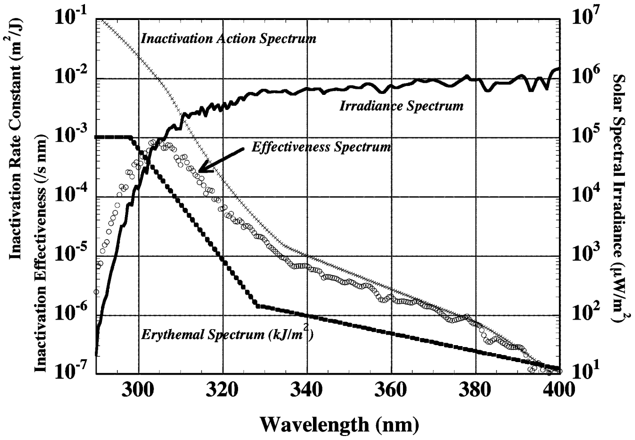

Fig. 1 Spectral characteristics of the spore dosimeter: the inactivation action spectrum (×) of the spore dosimeter and the effectiveness spectrum (○) derived by multiplication of an irradiance spectrum obtained at Naha on 1996/7/20 at 13:00 pm (line) and the action for each 0.5 nm interval.[16] A reference CIE erythemal spectrum[20] (dotted line) is shown for comparison.

Exposure sites

In addition to the sites described before,[17] three sites joined during this period: one South American site at University Vale do Rio Doce in Governador Valadares, Minas Gerais, Brazil, abridged as Valadares (18°51′S, 41°56′W; altitude, 171 m) and two northern European sites at Finnish Meteorological Institute at Jokioinen, Helsinki, Finland, abridged as Jokioinen (60°49′N, 23°30′E; altitude, 108 m) and at Arctic Research Centre at Sodankylä, Rovaniemi, Finland, abridged as Sodankylä (67°22′N, 26°39′E; altitude, 179 m).

Ozone column

The values of monthly average columnar ozone in Dobson units (DU) were obtained from the database of Ozone Monitoring Instrument (OMI).[21,22] The data up to the year 2004 were retrieved from the database of Total Ozone Mapping Spectrometer (TOMS).[23] Since the two data sets for the year 2005 were closely matched, we did not apply any correction for the transition. Also, the corrections for the TOMS data proposed from the comparisons with ground based measurements were not applied.[24]

Results

Project outlines

Measurements of monthly doses of solar UV radiation were performed with spore dosimeters and the doses were expressed as the values of spore inactivation dose (SID). The project started at the beginning of the year 1999 and continued to the end of the year 2007. The results up to the end of 2004 have been presented.[17] In the three year period from 2005 to 2007, monthly doses have been obtained at 10 sites without breaks except for accidental losses. At two sites (Nishihara and Denpasar), the measurements were terminated during the period due to movement of the investigators, and at one site (Jakarta) the data between July 2005 and January 2007 were unavailable due to disturbances of the site. Three new sites (Valadares, Jokioinen and Sodankylä) were added in the course and provided records for more than one year. In total, 444 points of monthly data were accumulated during this period in addition to the 896 points obtained up to the end of 2004. The numbers of missing months in the measurement period were 48 (9.8%) due to spoilage of samples, most prominently by combination of heavy rain and heat, and by the pecking of birds. A summary table is presented in the ESI.†

Dependence of yearly SID to latitudinal gradient

The monthly values of SID obtained between 2005 and 2007 were summed for each calendar year. When the values for some months were missing, they were filled by the mean values of the nearest two years. Yearly total SID values covered the minimum 3490 (Sodankylä, 2007) to the maximum 96 500 (Padang, 2006), the ratio being about 28. This ratio was similar to that reported previously, 30 between 3200 (Oulu, 2001) and 96 047 (Denpasar, 2001).[17] Yearly total values at 20 sites since 1999 are shown in Fig. 2 together with the yearly average amounts of columnar ozone at 12 sites. The changes and variations in the UV doses and the ozone amounts are analyzed in the sections below.

The means of yearly total values from the year 2005 to 2007 at each site are plotted against the latitude in Fig. 3 and the regressions are made for each hemisphere. The data for each hemisphere could be fitted to exponential regressions. In the regressions, extrapolation values for the equator (0°) are 111 000 and 104 000, and the exponents for the latitudinal gradient are 0.050 and 0.048, for the southern and northern hemisphere, respectively. Both values were slightly larger in the southern hemisphere, but the significance is not clear, since this seems to depend largely on the particular choices of sites. As noted previously,[17] some points deviating from the regressions could be explained by local climate and atmospheric conditions. The value at Jakarta is lower than the regression, suggesting a reduction by air pollution, whereas the higher value at Valadares might be due to prevalence of clear weather. The value at Thessaloniki is higher than the regression made mainly from eastern Asian sites, suggesting the former site has more cloudless days due to the difference between Mediterranean *versus* monsoon climate.

Dependence of monthly SID to the effective ozone column

The diurnal and seasonal solar altitude geometrically defines radiation flux incident to the horizontal plane. Moreover, the major absorption of solar UV radiation is by atmospheric ozone,

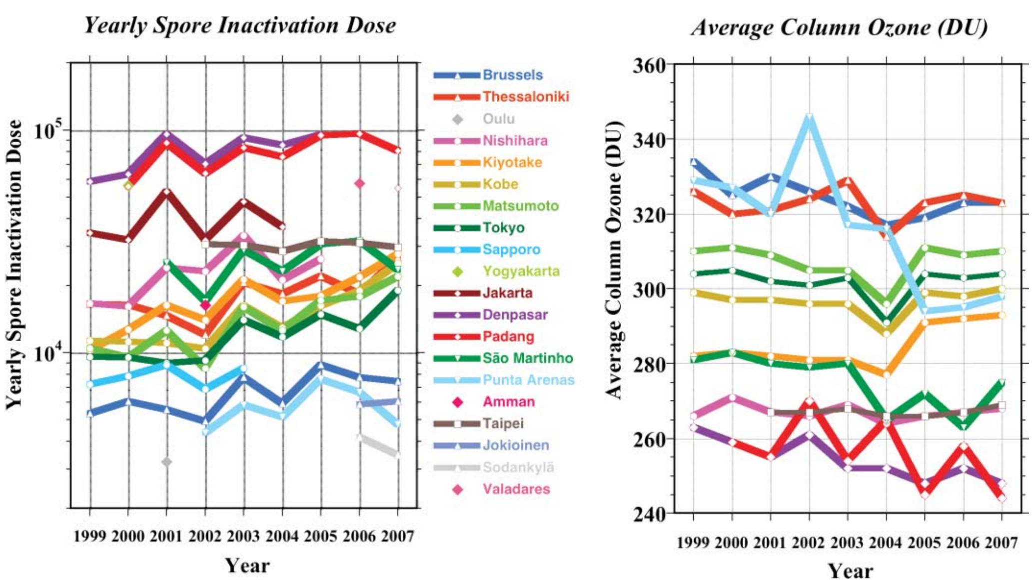

Fig. 2 Yearly total doses (SID) at 20 sites (left panel) and average column ozone amounts (DU) at 12 sites (right panel). In the left panel, the solitary markers without lines are from the sites where data from only one year are available.

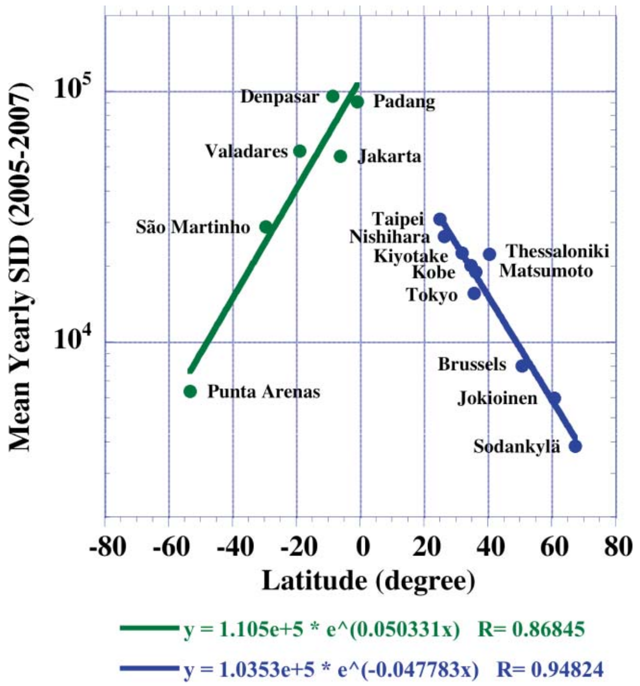

Fig. 3 Mean yearly doses against latitudes for the data obtained from 2005 to 2007. Negative and positive values of latitude are for the southern and northern hemisphere, respectively.

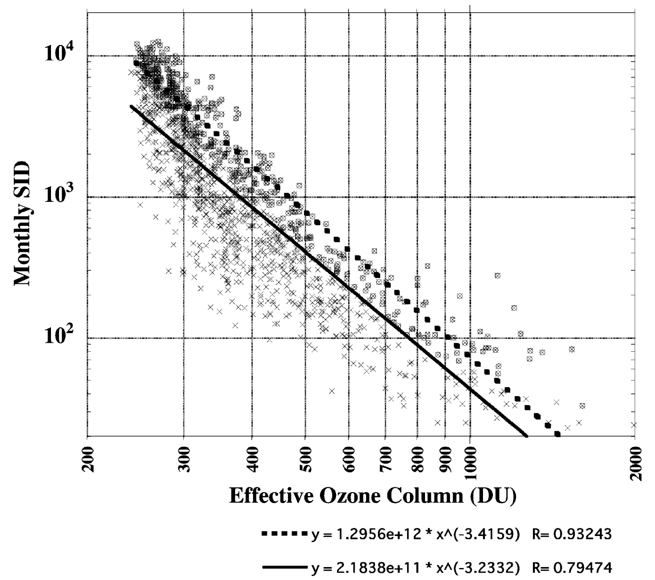

Fig. 4 Dependence of monthly UV doses (SID) on effective ozone column (DU). The monthly average amount of columnar ozone is divided by the cosine of the minimum solar zenith angle on the 15th day of each month at each site and shown as the amount of the effective ozone column. Dotted and filled lines are from the regressions for upper third and total data, respectively.

the vertical amount of which is given as the columnar ozone amount. These two primary factors determining the terrestrial UV dose could be combined to the ozone amount corrected for the path length ("the effective ozone column"). The maximum altitude, the minimum solar zenith angle at solar-noon time on the 15th day of each month, was used for derivation of the value. Thus, for each month at each site, the monthly average value of ozone was divided by the cosine of the zenith angle in degrees, and against this value, the observed value of SID is plotted in Fig. 4. The values of the effective ozone column in DU are distributed from minimum 241 to maximum 4109. Looking at the figure, monthly SID values scatter in formidable amounts at each value of the effective ozone column. This is expected since the effective ozone column only takes into account solar altitude and ozone amounts and neglects other components in the atmosphere that play a large role in modulating the UV dose.

To see general correlations, two types of regressions were performed. One is for all ("total" data) and the other is for the higher SID values ("upper" data) at each value of the effective ozone column selecting about one third of the data (471 data shown as ⊗ among 1336 points in Fig. 4). For both, the best fit was obtained with the power functions shown in the figure. These empirical relationships provide "expected" monthly SID values under "average" conditions or the prevalence of clear weather at any location, wherever the value of solar altitude and the amount of ozone are known. When the abscissa values are taken as solely dependent on ozone amounts, 1% decrease corresponds to the increase of SID values by 3.3 or 3.5% for the "total" and "upper" data, respectively. This value is larger than the ozone amplification factors derived from clear-sky irradiance and biological weighting factors, including the calculated values (about 1.9) for spore dosimeters. It is not possible to explain the discrepancy between empirical data and spectral analyses but some pertinent points are mentioned in relation to the difference between SID and erythemal doses in the Discussion.

Analysis of trends in yearly doses

To examine the trends of UV doses, the data at 12 sites were chosen where more than 6 years continual data between 1999 and 2007 were available. Yearly total values of SID and the average values of ozone amount at each site are subjected to linear regressions. Resulting values of yearly changes are shown in Fig. 5. In the analyses, the correlation coefficients are used to evaluate the reliability: those regressions with $R < 0.4$ are unmarked, $0.4 < R < 0.7$ are marked with a star, and $R > 0.7$ have two stars. Out of twelve sites, nine sites exhibit increasing trends for UV dose ($0.4 < R$). They include 2 European, 5 Japanese, and 2 Indonesian sites. Two South American sites exhibit increasing trends with

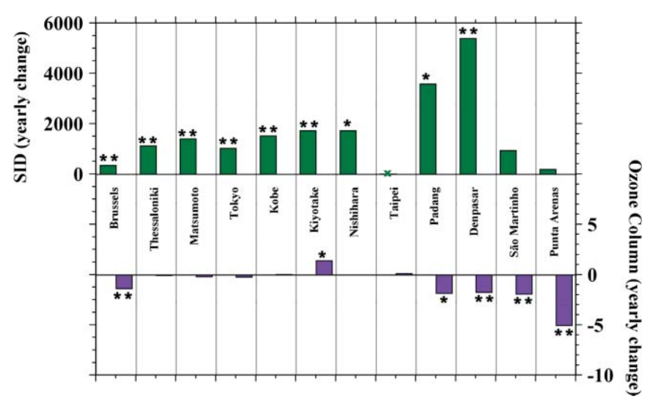

Fig. 5 Analysis of yearly trends in total UV doses (upper columns) and average ozone amounts (lower columns) at 12 sites.

low correlation coefficients and one site (Taipei) does not show significant change in the dose during this period.

With regard to ozone amounts, five sites exhibit decreases and one site (Kiyotake) exhibits an increase, while the other six sites do not show significant changes. Notably, the southern-most site (Punta Arenas) exhibits a striking decline (about 5 DU per year), despite small changes in the extent of the ozone hole during this period.[25] Thus, during the monitoring period, at all four sites in the southern hemisphere and at one northern European site, decreases in average yearly amounts of columnar ozone have been observed. Since ozone amount is considered to be stable in the tropics,[25] the two Indonesian sites involved in this study might be exceptional.

Those five sites with decreases in ozone amount exhibited increases in UV doses at various degrees, though two trends are not necessarily correlated in quantitative terms: for example, despite a large decrease in ozone amount, the absolute amount of UV change is small at Punta Arenas, while at tropical Denpasar and Padang the striking increases in UV doses occurred with small decreases of the ozone amounts. It should be pointed out that the amounts of total UV dose differ greatly among the sites: the ratios of mean yearly doses at Punta Arenas and the two tropical Indonesian sites are more than 14, thus the amount of dose change looks smaller at the former site. Also the fact that the average ozone amounts are higher at the former site may be attributable for relatively small changes in the UV dose.

On the other hand, no obvious correlation exists at six mid-latitude sites in the northern hemisphere including one European (Thessaloniki) and 5 Japanese sites where large increases in the UV doses occurred without decreases in ozone amounts, one site (Kiyotake) exhibiting the opposite trends (increases in both UV and ozone amounts). In Japan, since the rainy season overlaps with the period of high solar altitude, the yearly variations of the length and magnitude of the rainy season could affect the UV doses greatly. Thus, the large year-to-year variations and the increases in the UV doses observed in Japan might be attributable to the changing pattern of summer weather. It is especially notable that the year 2007 was very dry and exceptionally sunny during May to September at most sites in central Japan resulting in the highest UV doses observed.

Analysis of trends in monthly doses

To pursue further the analyses of variability and trends of the UV dose and ozone amounts, the trends for each separate month are investigated at the 12 sites. In Fig. 6, the yearly changes of UV doses and ozone amounts for each month are illustrated as double columns. As in the previous graph for the yearly total amount, the regressions for each month are marked with stars in three categories regarding the values of the regression coefficient.

At Brussels, small increases in the UV dose were observed throughout spring and summer, whereas the ozone decreases were seen from April to October except August. Thus, the UV increase from April to July was coincidental to the ozone decrease in these months. On the other hand, the ozone decrease in autumn to winter did not seem to have affected the UV dose. When the doses were low, the effect of ozone decrease did not manifest clearly.

At Thessaloniki, the large increases in UV doses were observed from May to July, while the amount of ozone showed no significant change. The only month in which ozone change was clear was March, when a small increase in the UV dose was seen in opposition to the expectation. It seems that the large increase in UV doses occurred without significant changes in ozone amounts at this site.

At three sites in Honshu Island in Japan, Matsumoto, Tokyo and Kobe, the patterns of changes in UV doses and ozone amounts were mostly similar with minor variations. Large increases in UV doses were observed from May to September, while the ozone amounts decreased from July to October and increased in March. The changes in ozone during winter to early spring seemed rather sporadic and were not reflected in the UV dose. As mentioned above, the variations in summer weather could affect the UV doses especially in June corresponding to the middle of rainy season. At these three sites, the large changes in the UV doses in June were observed without significant changes in the ozone amounts. On the other hand, the UV increases in July and August were coincidental to ozone decreases.

At Kiyotake in Kyushu Island in Japan, large increases in UV doses were observed from May to September, while large increases in ozone amount were observed from November to April. This was the only site in this work where the yearly average ozone amounts showed an increasing trend, conflicting with the trend of the UV dose. However, this seemed at least partly explainable by the observation that the large ozone increases were mostly confined to the low UV dose months. In July and August, there were decreases in ozone amounts similar to the three Honshu sites.

At Nishihara in Okinawa Island, the largest increase in UV dose was observed in July concurrent to the decrease in the ozone amount. The seasonal pattern of the ozone change was similar to the other sites in Japan, showing decreases from late spring to late autumn, while increases in winter to spring. At Taipei in Taiwan Island, neither UV nor ozone amounts exhibited significant changes in yearly total. A UV increase was observed from February to July with decreases in August and from November to January. On the other hand, the seasonal changes in the ozone amounts were similar to Nishihara; decreases from April to July and increases from August to February except January. Though the large decrease in UV dose in August was concurrent to the small ozone increase, it seemed unlikely that this was the major cause of this change.

At two tropical Indonesian sites, the yearly averaged ozone amounts decreased steadily as seen in Fig. 2, with a biannual wave at the near-equatorial Padang. When the trend was analyzed monthly, the ozone changes were fairly uniform throughout year with a peak change in January at Denpasar, and two peaks in January and June at Padang. On the other hand, the increasing trends in UV were scattered without simple patterns. Large peaks of increases from February to May were observed at Denpasar, while scattered peaks at February, June, July, November and December were seen at Padang.

At two South American sites, São Martinho and Punta Arenas, the yearly average amounts of ozone exhibited decreases during the period. When analyzed monthly, the decreases occurred from September to April, coincident to the higher dose months. The increases in UV doses were seen clearly in late spring to mid-summer (November to January) at both sites, though the amounts were small at Punta Arenas, in contrast to the large decreases in the ozone amounts.

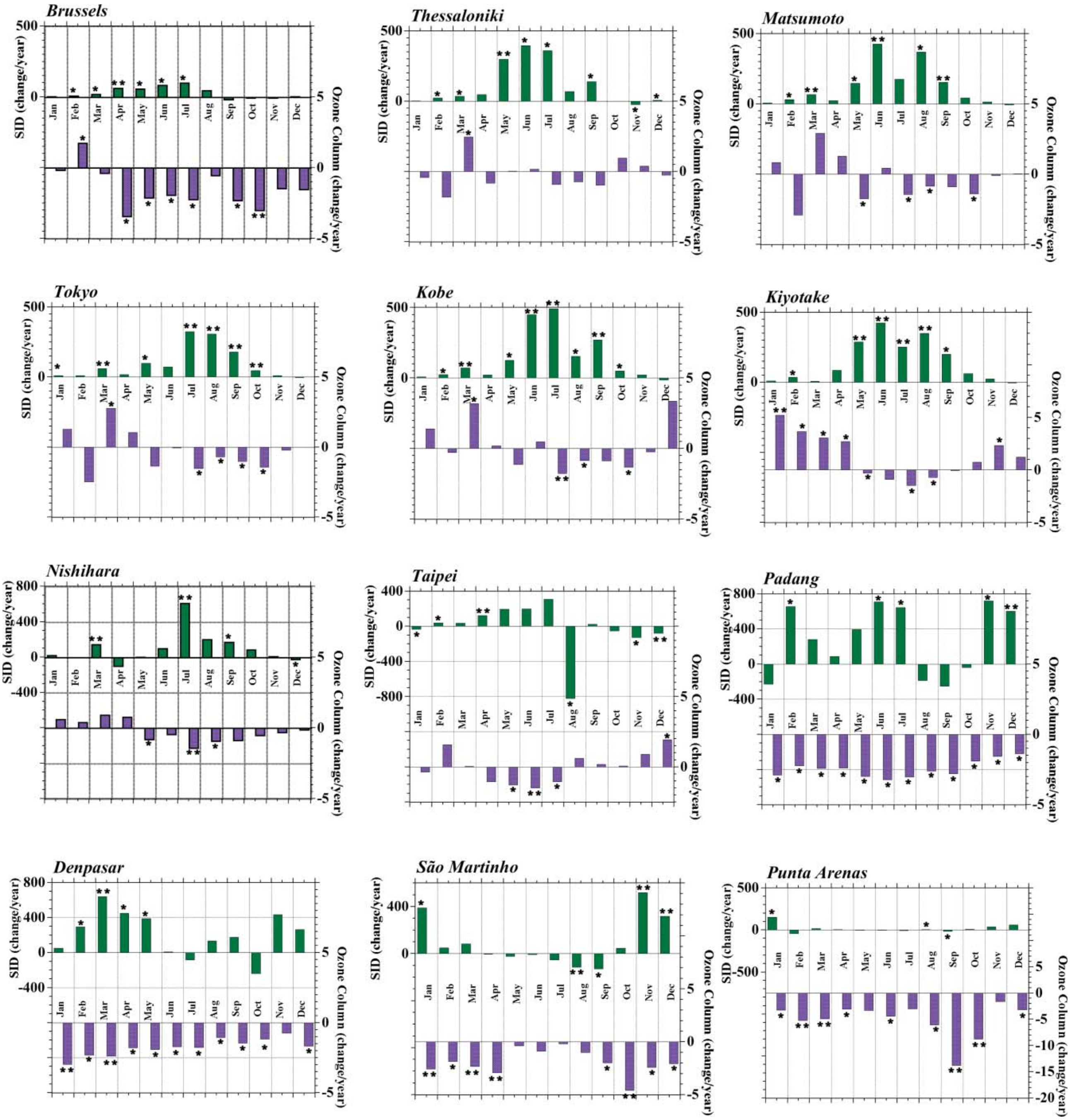

Fig. 6 Analysis of trends in monthly UV doses and average ozone amounts at 12 sites.

Correlations of SID with ED values

At four sites (Brussels, Thessaloniki, São Martinho, and Valadares), concurrent measurement of spectral irradiance using spectrophotometers has been in operation, and the values of monthly erythemal dose (ED) were available from the calculation based on the CIE erythemal spectrum. Correlations of monthly values of SID and ED are analyzed using the data and shown in Fig. 7. Although the values scatter considerably, it seems that the correlations between them are consistent among the four sites. This indicates the instrumental biases and variations for the ED measurements are small, and the causes for the scatters between these two values are not site-specific. The exponent values of the power regressions of SID to ED values were between 1.04 at Jokioinen and 1.57 at Thessaloniki, while the value for all data was 1.27. It seems that these exponent values become larger as the dose increases, suggesting that the sensitivity difference between SID and ED becomes larger at higher doses.

Discussion

Among the various environmental stresses of life, solar UV radiation is one of the most ubiquitous, wide-ranging and variable.

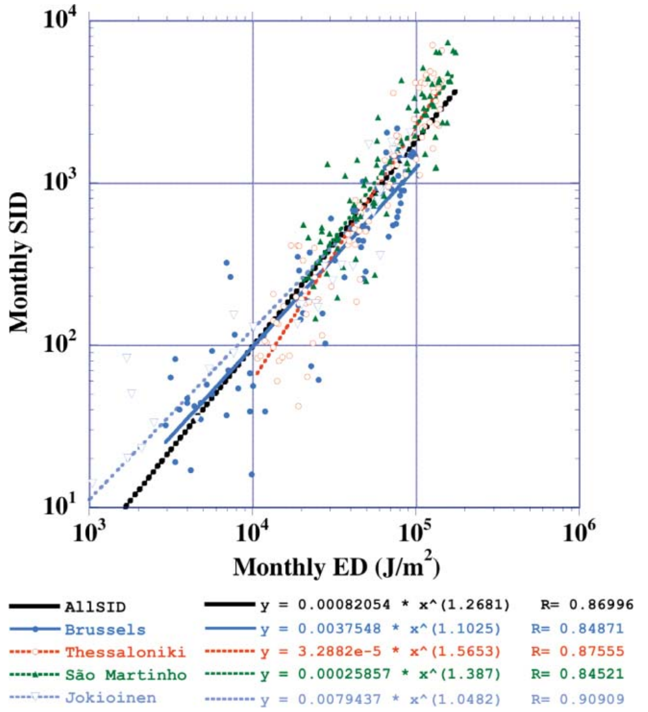

Fig. 7 Correlation analyses between monthly SID and ED values at 4 sites. Lines are drawn by power regressions for all sites (filled) and for each site (dotted).

Its primary effects are harmful due to the induction of damage to cellular genetic material. However, it also prevents vitamin D deficiency in mammals in the case of insufficient intake from diet. From these aspects, it is remarkable that humankind has managed to inhabit the lowest and highest extremes, with at least a 30-fold difference in yearly total doses as shown in this work. This extreme variability in the dose is primarily caused by solar altitude that has a latitudinal dependency and the patterns of seasonal and diurnal changes. In concert with the previous work,[17] the exponential latitudinal gradient of the UV dose exhibits slopes of 0.050 and 0.048 for the southern and northern hemisphere, respectively, that translate to a doubling of the dose for a decline of about 14° in latitude. A decadal result of the ELDONET project,[10] mostly focused on European sites, shows a latitudinal gradient for yearly UV-B (280–315 nm) doses for the northern hemisphere with an exponential slope of 0.045 (calculated from Table 4 in ref. 10), and seems to be congruent with our estimate.

Since major absorption of solar UV radiation occurs by ozone distributed mainly in the stratosphere, the dose received on the earth depends on the amounts of columnar ozone. The ozone is produced and destroyed by photochemical processes in the stratosphere and the patterns of vertical and horizontal distribution are affected largely by the atmospheric dynamics that produce uniquely changing patterns at each region and site. Moreover, other materials in the atmosphere, especially various forms of water, could affect greatly the radiation flux, and modify the irradiance on the earth. For example, clouds generally reduce the amounts of UV flux by absorption, reflection and scattering, but under some conditions they might increase the dose by diffused radiation and reflection from the ground. The patterns and intensity of clouds and rainfall are diverse and unique to each region and locality. These complex interactions of multiple factors in the atmosphere made it very difficult to disclose the patterns and the causes of variations in UV doses. At Thessaloniki, a long-term record of spectral UV irradiance is available, and it was reported that monthly erythemal irradiance increased about 6% per decade between 1994 and 2004. This is attributed to the reduction of clouds and aerosols.[26,27]

Although the correlations between the SID values directly observed by the spore dosimeters and the ED values from the spectroscopic data are found, the proportionality seems to hold only under low-dose conditions. The monthly SID exhibits exponents greater than one in the power regressions of SID to ED in the high-dose region. It means that either the ED values underestimate or the SID values exaggerate the biological effectiveness under high irradiance. One possible cause for this divergence is the spectral difference between the weighting function for ED and the inactivation for spore dosimeter as shown in Fig. 1, but preliminary calculations at two sites showed the same tendency for the calculated values of SID with those of the ED. Previous findings revealed that the discrepancy between observed and calculated SID rates in field comparison experiments becomes larger under high dose rates.[15,16] A possible cause for this difference is that the spores on the membrane filter might behave as partially oval objects receiving radiation from various directions including the reflection from the membrane surface. Another possibility is that the biological dosimeter might not be totally free from the interactions among various wavelength components or from the influence of environmental factors such as heat and humidity, though to disclose such interactions in the laboratory has so far been unsuccessful. On the other hand, spectral photometers currently in use might be missing the fine details, especially in the short wavelength end (shorter than 290 nm), where the biological effectiveness increases steeply. In any case, to find out the causes for the non-proportionality between spore dosimeter and spectral photometer needs be pursued in future projects.

Despite these problems and uncertainties, the results have shown clearly that the changes and variations of ozone amounts could profoundly affect biologically effective doses. The thesis that solar UV dose received on the earth is dependent on the amounts of ozone has been established by the model calculations and measurements under severe ozone loss such as the ozone hole over Antarctica. In this work, we analyzed the trends in the UV doses and the ozone amounts at 12 sites where more than 6 years continuous data were available and showed that the trend of the increases in yearly UV doses was concurrent to the decreasing trend of ozone at 5 sites (Brussels, two Indonesian and two South American sites). At five sites in Japan exhibiting UV increases, the decreasing trends in yearly-average ozone amounts were not clearly observed, but the decreasing trends in the ozone amounts in summer months were seen. The generality and causes for such seasonal trends in the ozone amounts are not clear at this point. At one site in Japan (Kiyosato), despite the ozone increase, the UV dose increased. At one European site (Thessaloniki), despite the UV increase, no change in the ozone amount was observed. At one Asian site (Taipei), neither the UV dose nor the ozone amount changed. In conclusion, the decreasing trends in the ozone amount at several sites correlated with the increase in the UV doses, but the regional and local changes and variations in weather and air transparency also greatly affected the UV doses, and often compromised the effect of the ozone changes.

References

1 M. J. Molina and F. S. Rowland, Stratospheric sink for chlorofluoromethanes: chlorine atom-catalysed destruction of ozone, *Nature*, 1974, **249**, 810–812.

2 F. S. Rowland, Chlorofluorocarbons and the depletion of stratospheric ozone, *Am. Sci.*, 1989, **77**, 36–45.

3 A. F. Bais, D. Lubin, A. Arola, G. Bernarhd, M. Blumthaler, N. Chubarova, C. Erlick, H. P. Gies, N. Krotkov, K. Lantz, B. Mayer, R. L. McKenzie, R. Piacentini, G. Seckmeyer, J. R. Slusser, C. Zerefos, V. Fioletov, J. Groebner, E. Kyro and H. Slaper, Surface Ultraviolet Radiation: Past, Present and Future, *Report No. 50: Global Ozone Research and Monitoring Project - Scientific Assessment of Ozone Depletion*, World Meteorological Organization, 2007, Chapter 7.

4 R. L. McKenzie, P. J. Aucamp, A. F. Bais, L. O. Björn and M. Ilyas, Changes in biologically-active ultraviolet radiation reaching the Earth's surface, *Photochem. Photobiol. Sci.*, 2007, **6**, 218–231.

5 S. R. Wilson, K. R. Solomon and X. Tang, Changes in tropospheric composition and air quality due to stratospheric ozone depletion and climate change, *Photochem. Photobiol. Sci.*, 2007, **6**, 301–310.

6 *EC Air Pollution Report No. 63: Advances in Solar Ultraviolet Spectroradiometry*, ed. A. R. Webb, European Commission, Luxembourg, 1997.

7 R. B. Setlow, The wavelength in sunlight effective in producing skin cancer: a theoretical analysis, *Proc. Natl. Acad. Sci. U. S. A.*, 1974, **71**, 3363–3366.

8 B. C. Kennedy and W. E. Sharp, A validation study of the Robertson-Berger meter, *Photochem. Photobiol.*, 1992, **56**, 133–141.

9 J. Scotto, G. Cotton, F. Urbach, D. Berger and T. Fears, Biologically effective ultraviolet radiation: surface measurements in the United States 1974 to 1985, *Science*, 1988, **239**, 762–764.

10 D.-P. Häder, M. Lebert, M. Schuster, L. del Ciampo, E. W. Helbling and R. McKenzie, ELDONET - A decade of monitoring solar radiation on five continents, *Photochem. Photobiol.*, 2007, **83**, 1348–1357.

11 S. Takeshita and M. Sasaki, Solar radiation monitoring network of Tokai University, Japan, *Proc. SPIE*, 2003, **5156**, 303–310.

12 G. Horneck, Quantification of the biological effectiveness of environmental UV radiation, *J. Photochem. Photobiol., B*, 1995, **31**, 43–49.

13 *EC Air Pollution Research Report No. 71, Biological UV dosimetry, a tool for assessing the impact of UV radiation on health and ecosystems*, ed. P. Rettberg, G. Horneck, C. Baumstark-Khan and G. Amanatidis, European Commission, Luxembourg, 1999.

14 N. Munakata, Killing and mutagenic action of sunlight upon *Bacillus subtilis* spores: a dosimetric system, *Mutat. Res.*, 1981, **82**, 263–268.

15 N. Munakata, F. Morohoshi, K. Hieda, K. Suzuki, Y. Furusawa, H. Shimura and T. Ito, Experimental correspondence between spore dosimetry and spectral photometry of solar ultraviolet radiation, *Photochem. Photobiol.*, 1996, **63**, 74–78.

16 N. Munakata, S. Kazadzis, A. F. Bais, K. Hieda, G. Rontó, P. Rettberg and G. Horneck, Comparisons of spore dosimetry and spectral photometry of solar-UV radiation at four sites in Japan and Europe, *Photochem. Photobiol.*, 2000, **72**, 739–745.

17 N. Munakata, S. Cornain, M. Kanoko, K. Mulyadi, S. Lestari, W. Wirohadidjojo, D. Bolseé, S. Kazadzis, V. Meyer-Rochow, N. Schuch, C. Casiccia, M. Kaneko, C.-M. Liu, K. Jimbow, T. Saida, C. Nishigori, K. Ogata, K. Inafuku, K. Hieda and M. Ichihashi, Biological monitoring of solar ultraviolet radiation at 17 sites in Asia, Europe and South America from 1999 to 2004, *Photochem. Photobiol.*, 2006, **82**, 689–694.

18 R. Tyrrell, Solar dosimetry with repair deficient bacterial spores: action spectra, photoproduct measurements and a comparison with other biological systems, *Photochem. Photobiol.*, 1978, **27**, 571–579.

19 T. Douki, B. Setlow and P. Setlow, Effects of the binding of a/b-type small, acid-soluble spore proteins on the photochemistry of DNA in spores of *Bacillus subtilis* and *in vitro*, *Photochem. Photobiol.*, 2005, **81**, 163–169.

20 A. F. McKinlay and B. L. Diffey, A reference action spectrum for ultraviolet induced erythema in human skin, *CIE Res. Note*, 1987, **6**, 17–22.

21 Total Ozone Mapping Spectrometer Ozone Processing Team, NASA/GSFC, ftp://toms.gsfc.nasa.gov/pub/omi/data/monthly_averages/ozone.

22 R. McPeters, M. Kroon, G. Labow, E. Brinksma, D. Balis, I. Petropavlovskikh, J. P. Veefking, P. K. Bhartia and P. F. Levelt, Validation of the Aura Ozone Monitoring Instrument total column ozone product, *J. Geophys. Res.*, 2008, **113**, D15S14.

23 Total Ozone Mapping Spectrometer Ozone Processing Team, NASA/GSFC, ftp://toms.gsfc.nasa.gov/pub/eptoms/data/monthly_averages/ozone.

24 G. E. Bodeker, J. C. Scott, K. Kreher and R. L. McKenzie, Global ozone trends in potential vorticity coordinates using TOMS and GOME intercompared against the Dobson network: 1978–1998, *J. Geophys. Res.*, 2001, **106**, 23029–23042.

25 *Environmental effect of ozone depletion and its interaction with climate changes: 2006 assessment.*, UNEP, http://ozone.unep.org/Assessment_Panels/SAP/Scientific_Assessment_2006.

26 C. S. Zerefos, Long-term ozone and UV variations at Thessaloniki, Greece, *Phys. Chem. Earth, Parts A/B/C*, 2002, **27**, 455–460.

27 K. Garane, A. Bais, K. Tourpali, C. Meleti, C. Zerefos and S. Kazadzis, Variability of spectral UV irradiance at Thessaloniki, Greece, from 15 years measurements, *Proc. SPIE*, 2005, **5886**, 601–610.

