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Solar UV radiation and microbial life in the atmosphere

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Abstract

Many microorganisms are alive while suspended in the atmosphere, and some seem to be metabolically active during their time there. One of the most important factors threatening their life and activity is solar ultraviolet (UV) radiation. Quantitative understanding of the spatial and temporal survival patterns in the atmosphere, and of the ultimate deposition of microbes to the surface, is limited by a number factors some of which are discussed here. These include consideration of appropriate spectral sensitivity functions for biological damage (e.g. inactivation), and the estimation of UV radiation impingent on a microorganism suspended in the atmosphere.

We show that for several bacteria (*E. coli, S. typhimurium*, and *P. acnes*) the inactivation rates correlate well with irradiances weighted by the DNA damage spectrum in the UV-B spectral range, but when these organisms show significant UV-A (or visible) sensitivities, the correlations become clearly non-linear. The existence of these correlations enables the use of a single spectrum (here DNA damage) as a proxy for sensitivity spectra of other biological effects, but with some caution when the correlations are strongly non-linear.

The radiative quantity relevant to the UV exposure of a suspended particle is the fluence rate at an altitude above ground, while down-welling irradiance at ground-level is the quantity most commonly measured or estimated in satellite-derived climatologies. Using a radiative transfer model that computes both quantities, we developed a simple parameterization to exploit the much larger irradiance data bases to estimate fluence rates, and present the first fluence-rate based climatology of DNA-damaging UV radiation in the atmosphere.

The estimation of fluence rates in the presence of clouds remains a particularly challenging problem. Here we note that both reductions and enhancements in the UV radiation field are possible, depending mainly on cloud optical geometry and prevailing solar zenith angles. These complex effects need to be included in model simulations of the atmospheric life cycle of the organisms.

1. Introduction

It is well-known that the atmosphere is important for life on our planet, and also that life affects the composition of the atmosphere. But we do not usually consider free air, high above the ground and ocean, as a habitat for continuous life. Recently it has been established that there are birds which spend almost all their life, except when breeding, on the wing[1-3]. They eat, drink, sleep and sometimes mate while flying. But in this paper we shall focus on microbial life in the atmosphere, and its relation with a potentially hostile ultraviolet (UV) environment.

The purpose of the present paper is to review some of the published literature, noting several critical issues that underlie quantitative understanding. We build on existing knowledge to produce a climatology of UV radiation that is relevant to the survival of airborne organisms. We shall start with the kinds, distribution and spread of microorganisms, then continue with the extent to which they can survive in the atmosphere, with particular focus on the effect of UV radiation. Recognized as a major environmental cause of skin cancer in humans and animals[4], UV radiation has been shown in laboratory studies to effectively kill or inactivate many microorganisms. We consider the availability of UV radiation in the atmosphere from the perspective of a suspended biological particle, contrasting clear and cloudy sky conditions. Using the DNA damage spectrum as a proxy for multiple damage mechanisms, a climatology of DNA-damaging UV radiation is obtained with a radiative transfer model and satellite-observed ozone and clouds, that clearly shows the large seasonal and geographical variability in UV exposures experienced by airborne microorganisms.

2 Airborne microorganisms

2.1 Types of airborne microorganisms

Many different types of microorganisms are present in the atmosphere (see Table 1). Most research in this area has been carried out on fungi and bacteria, particularly those which cause disease in man, animals and crops. Pollen has also been extensively researched, the main reason being that pollen from many plants (as are some fungal spores) are allergenic. Another reason for studying pollen carried by wind is that pollen is used to study past plant distribution, and for a correct interpretation of distribution of ancient pollen it is necessary to have an idea of how far it might have travelled before being deposited. A third reason to study how pollen travels carried by air is the risk of unwanted cross-pollination of crops, especially genetically modified crops[10]. Some research has been carried out also on other airborne organisms, such as algae[15,16] and amoebae[17]. So-called dust seeds are so small that we should perhaps include them among microorganisms, and in many cases they are dispersed mainly by wind. The seed of *Bartonia sp.* (Gentianaceae) is 170 μm long[18], that of the orchid *Dichaea panamensis* 160 μm by 70 μm[5].

2.2 Spread of airborne microorganisms

Suspension near sources can occur by many mechanisms. Some organisms are carried aloft in dust storms (e.g., refs. 19-21), but also aquatic microorganisms will escape to the atmosphere in strong wind or by breakage of bubbles at the water surface[22]. Many microorganisms collected from a cloud on a mountain were derived from terrestrial vegetation[23], and Lighthart[13] citing earlier sources also stresses the great contribution from vegetation. Presumably the natural shaking of leaves by wind injects some organisms into the air. Other organisms, such as ferns[24,25], mosses[26,27] and fungi[28-30] (see also review by Sakes et al.[31]) actively eject their spores into the air. Another example is described by Dressaire et al.[32]: cooling of air by evaporation from mushrooms causes air to come down from above and form a horizontal flow outward from the fungus. When the flow encounters a barrier it rises upward again, carrying spores with it.

The terminal velocity (Table 1) is an important limitation to how far suspended particles can be transported vertically as well as horizontally by winds, and multiday transport is unlikely if this velocity is much larger than about 1 cm s^{-1} (≈1 km day^{-1}). The smaller and less dense organisms with lower terminal velocities have a greater chance of long-range dispersal. At the same time small organisms are less able to protect themselves against ultraviolet radiation. It should be noted that although bacteria are among the smallest of the organisms listed in Table 1, they usually appear in air in clusters or attached to other particles. Thus Lighthart[13] reported that bacteria-carrying particles are dominated by sizes ≥ 7 μm as compared to typical single-bacterial sizes of 0.65–1.1 μm. Clauss[33] found that atmospheric particles containing culturable bacteria often had sizes of around 10 μm, while those containing culturable fungi often had sizes around 3 μm. Thus effective particle sizes for bacteria are comparable to those of eukaryotes. By comparison, typical cloud droplets have radii of about 10 μm[34].

Transport to higher altitudes occurs primarily by turbulence and convection in the boundary layer. Near the surface upward air motions are caused by incoming solar radiation heating the ground, the same currents that allow eagles to circle upwards without moving their wings. Large fires also cause upward air currents, and anthropogenic biomass burning is known to propel microorganisms upwards[35]. Convective activity over large forest fires, or the Asian summer monsoon anticyclone[36] might also lift microorganisms all the way to the stratosphere. Some microorganisms are also found in the stratosphere[37-40], where gravito-photophoresis[41] may play a role.

The smaller a microorganism is, the longer it is likely to stay in the air, but many of the types mentioned can spread over vast distances, given sufficiently strong winds[20,42]. Pollen can blow across the Alps[43], from Germany and northern France to Spain across the Pyrénées[44], from Texas or Oklahoma to Canada[45], and probably from the Baltic states to northern Sweden[46]. Two moss species were found on the new volcanic island Surtsey as early as 1967 when eruptions ceased, and several new bryophyte species were discovered every year until 1973 when regular sampling ended[47]. Presumably moss spores had arrived by wind from the nearest larger island some 6 km away. Smaller particles such as

viruses, bacteria and fungal spores may spread globally in air. For instance, fungal spores have spread from Australia to New Zealand[48] and hundreds of km over USA[49].

2.3 Ultraviolet radiation and survival of microorganisms in air

Environmental factors that may shorten life-span of microorganisms in air are ultraviolet (and to a lesser extent visible) solar radiation, desiccation and repeated freeze-thaw cycles[50-52]. In a simulation of various factors in the stratosphere UV radiation was the most severe one for spores of *Bacillus subtilis*[53].

Singer and Ames[54] observed that bacteria normally well-exposed to sunlight tend to have a higher content of guanine plus cytosine in their DNA than bacteria living more in the dark, and explained how this may contribute to increased UV resistance. Members of the genus *Bacillus* have UV protecting spore coats and DNA repair machinery in the endospores which counteract damage from solar UV radiation[55]. It was found that many of the living bacteria in dust at high altitude were either actinomycetes with high guanine plus cytosine content in their DNA, or spore-forming bacilli[56]. The survival of several different bacteria to specific doses of UV-A (315–400 nm) and UV-B (280–315 nm) in the laboratory are shown in Table 2.

We can draw the following conclusions from Table 2: (1) On an energy basis UV-B (in the absence of UV-A) is much more potent than UV-A. (2) There are large variations in sensitivity among bacteria. (3) There is no correlation between UV-A and UV-B sensitivity among the various bacteria. (4) For some bacteria, such as *Acinetobacter*, *Brevibacterium* or *Micrococcus*, solar UV-A, because of its higher fluence rate, is probably more important than UV-B. A review of the literature on photoinactivation of bacteria by monochromatic visible light of various wavelengths is provided by Hessling et al.[58]

Unfortunately, experimental studies of the impact of solar radiation on pollen[59,60], bacteria and other microorganisms have been conducted mostly on hydrated cells (usually on agar gel). This leads to an underestimation of sensitivity in dry air, in which repair of damage is inhibited. In the real atmosphere, conditions intermediate between dry and wet can be encountered,

e.g. at high relative humidity for microbes attached to hygroscopic aerosol particles. Furthermore, because repair is partly light-dependent (photo-repair) and driven mainly by UV-A radiation, experiments with only UV-B radiation will generally overestimate effects of solar UV-B.

2.4 Action Spectra

The spectral sensitivity of organisms is commonly represented by action spectra, for which a particular endpoint, e.g. photo-inactivation, is measured as a function of wavelength. A few examples of spectral sensitivity of bacteria are given in Fig. 1. Many other sensitivity spectra have also been published (Table 3). For several reasons they should be regarded as rather approximate: (1) There are often large deviations from exponential decay of viability with fluence, e.g. sometimes with slower initial or final decay, so the inactivation constant depends on fluence (i.e. is not really a constant); (2) in general the organisms have been in an aqueous medium, in which the radiation sensitivity is likely to differ from that in air; (3) in air the sensitivity also depends on relative humidity, temperature and the metabolic state of the organism. Good spectral resolution in these action spectra is particularly important in the UV-B band, where absorption by stratospheric ozone modulates transmission by several order of magnitudes over a mere few tens of nanometers.

The action spectra point to DNA as a dominant chromophore for photoinactivation (Fig. 1), and this absorption is mirrored in the inactivation spectra of *Escherichia coli* and *Salmonella typhimurium.* For *E. coli,* we show both the spectrum for aerobic conditions measured by Webb and Brown[65], and with a UV-A tail observed in lab-grown *E. coli* by Silverman and Nelson[69]. Other chromophores such as porphyrins may impress their signatures (e.g., in *Propionibacterium acnes*). Thus, the action spectra shown in Fig. 1 cover a range of qualitatively different spectral behaviors, and illustrate several issues related to their use.

The absolute sensitivity to different wavelengths depends not only on the biological action spectrum, but also on the spectrum of the available solar radiation. Such spectra can be measured directly (see Sect. 3.1), or calculated with radiative transfer models that simulate the propagation of solar radiation

198 through the atmosphere. Here, we use the Tropospheric Ultraviolet-Visible
199 (TUV) model[83], described in the Supplementary Materials, to provide UV spectra
200 applicable to a wide range of conditions.
201 Figure 2 shows the contributions of different wavelengths to total inactivation
202 rates, for high sun conditions at sea level, using the action spectra of Fig. 1. The
203 steep *E. coli* (aerobic) and *S. typhimurium* are dominated by UV-B radiation, with
204 only a few percent contribution from longer wavelengths. The UV-A tail observed
205 from lab-grown *E. coli* causes UV-A contributions that are comparable to those of
206 UV-B wavelengths, while for *P. acnes* the contributions are fairly evenly
207 distributed over the observed range 320–440 nm, but with unclear possible
208 contributions from outside this range.
209 The different relative importance of different wavelengths, evident in Fig. 2,
210 has notable consequences. One of these is that the inactivation rates of different
211 organisms may or may not be simply proportional to one another. This is shown
212 in Fig. 3, where (relative to DNA damage) good linearity is seen for *S.*
213 *typhimurium* and *E. coli* (aerobic), but correlations are much poorer (see
214 flattening curves) for the two spectra with large UV-A contributions. The
215 linearity (or lack of) depends on the causes of the variations: If – as is the case in
216 Fig. 3 – the variations in DNA damage are largely due to clear-sky variations in
217 the solar zenith angle (particularly in the slant crossing of stratospheric O_3
218 layers), the UV-A variations will be disproportionately smaller than UV-B
219 variations; while variations in cloudiness (not shown) would affect UV-A and UV-
220 B similarly and thus retain simple proportionality.
221 Notwithstanding this and other limitations, plots such as Fig. 3 provide a quick
222 way to estimate inactivation rates for a number of different effects if one
223 standard metric, e.g. DNA-damaging radiation, is known. For example, a typical
224 mid-latitude summer day with DNA damage radiation of 100 J m^{-2} day^{-1}, implies
225 for *S. typhimurium* a survival reduction by 400 e-folds day^{-1}, or equivalently an
226 1/e reduction (to 37% of initial value) in ≈3 minutes. In these cases, the
227 maximum transport distances are clearly limited.
228
229 **3. UV Exposure of DNA in the Atmosphere**
230

We consider next the DNA-weighted radiation specifically normalized at 254 nm, as a key indicator for the most UV-B sensitive processes, such as *S. typimurium* and *E. coli* (aerob.) inactivation described above. The normalization at 254 nm is somewhat arbitrary, but possibly useful given that many inactivation studies have used the 254 nm line of low-pressure Hg lamps[84]. Thus, tropospheric irradiances calculated with this normalized DNA spectrum are equivalent to the energy (e.g., J s^{-1} m^{-2}) of 254 nm photons that would have the same effect (e.g. inactivation). However, this assumes that the action spectrum is accurate over several orders of magnitude (from 254 to tropospheric UV-B wavelengths), so this conversion of laboratory data at 254 to tropospheric wavelengths can be problematic[85].

3.1 Irradiance Incident on a Horizontal Surface

The most commonly measured quantity of UV radiation is the spectral irradiance incident on a horizontal plane (usually the detector), at the surface of the Earth. Measurements from high elevation mountain stations are available but must be distinguished from the fewer observations available from balloon or aircraft above ground.

A climatology of observations of ground-based spectral UV-B and UV-A irradiance has been established previously, using high-quality spectral data from the Network for the Detection of Atmospheric Composition Change (NDACC)[86-88]. Relationships between monthly mean UV-B and DNA-weighted UV at a subset of those sites were used to estimate DNA-weighted irradiances (see Supplementary Material). These were then re-normalized to unity at 254 nm for comparability with the data shown in Fig. 3. The weighted irradiances thus derived are shown in Fig. 4. The data show strong latitudinal variation. Seasonal changes are relatively small in the tropics, but become more pronounced at mid to high latitudes. At the highest latitudes, irradiances are zero during the polar night. As noted previously for other UV weightings[89] southern mid-latitude doses in summer tend to be significantly higher than at comparable Northern Hemisphere locations. Highest values, ca. 220 J m^{-2} day^{-1}, are achieved at Mauna Loa, Hawaii (3.4 km elevation), while in Barrow, Alaska summer values are notably lower

than in Antarctica. The difference between dry and wet climates is also seen, e.g., with Alice Springs and Darwin, the latter experiencing frequent cloud and rainfall during December – March. These large seasonal and latitudinal variations in DNA-damaging UV could potentially have profound effects on survival rates of airborne species.

3.2 Scalar Irradiance, or Actinic Flux, or Fluence Rate

Irradiance, the radiation impinging on a horizontal surface, is not the radiative quantity most relevant to airborne particles. A horizontal plate (a typical irradiance detector) will heat more/less when tilted to/away from the incident solar beam, while an airborne particle is indifferent to the direction from which the light originates, at least to the extent that it is spherical or randomly oriented. This total radiation, independent of direction, is known by various names including actinic flux, scalar irradiance, or fluence rate[85,90], with the latter used here.

The relationship between fluence rate and irradiance depends on the angular distribution of the radiation field, and can be complex. Nader and White[91] measured fluence rates in urban Los Angeles by placing sensors on the six faces of a cube (with appropriate geometric corrections), and compared thisto irradiance measured by a single upward facing sensor. The ratio of fluence rate to irradiance varied from about 1.2 at high sun angle, to about 2.2 at low sun angle. Similar values have been measured using spherically shaped detectors[92], or by systematic sampling of radiation incident from different sky directions with irradiance sensors[93]. A major controlling factor is the direct/diffuse ratio (the solar beam compared to the sky radiation) which in turn depends on atmospheric conditions (clouds, ground reflectivity), wavelength, solar zenith angle, and altitude. Models[94-97] generally reproduced the observed values[92,98-100] fairly well if the surrounding atmosphere is well known (though that is often not true). Measurements at high spectral resolution have allowed more accurate estimates of molecular photo-dissociation coefficients important in the formation of smog[101-103].

The ratio of fluence rate at any altitude to the irradiance at the surface is shown in Fig. 5, for daily DNA-weighted radiation and cloud-free conditions. The

fluence rate is systematically larger than the irradiance, by a factor of 2 or more near the surface and increasing with altitude. This is due to the importance of photons reaching the biological target from the sides, whereas such photons would be barely detected by a horizontal plate (irradiance detector). It should be noted that this illustration assumed a minimal surface albedo (5%). With larger surface albedo, e.g. 90% as possible over snow, the fluence rate enhancements would be even larger.

A fit of the data presented in Fig. 5 (described in the Supplementary Materials) led to the simple parameterization for the ratio, *R*, of fluence rate at any altitude z, to the irradiance at ground level.

$$R \approx 2.6 + 0.8\,z - (0.8 + 0.5\,z)\cos\Theta_N \qquad \text{Eq. (1)}$$

Θ_N = solar zenith angle at noon

z = altitude, km

This simple formula provides an estimate of the enhancement in DNA-damaging radiation as a function of altitude and location/season (through the noon solar zenith angle). It should be recalled that this parameterization is based on DNA-damaging radiation, 24-hour average, clear sky conditions, and should be tested for validity for extended uses. Note that the enhancement factor *R* increases with altitude, due to the contribution of increased reflections from the atmosphere below.

3.3 A Global Climatology

The development of a simple parameterization (Eq. 1) relating irradiance and fluence rate means that available climatologies for surface irradiance, whether from observations (as discussed in Section 3.1), or from modeling, can be used to estimate corresponding climatologies of fluence rates. This is illustrated in Fig. 6, in which we extended the climatology of surface UV irradiance given by Lee-Taylor et al.[104] by normalizing the DNA values to 254 nm and converting to fluence using Eq. 1. The model calculations in Fig. 6 show similar geographic and temporal patterns as the irradiance measurements shown in Fig. 4, with fluences being typically larger by a factor of two or more (However, note that for the spatial resolution used in Fig. 6, the altitude of the Mauna Loa Observatory (MLO) is not fully resolved, so the maximum fluence there appears less than

twice the measured maximum irradiance). For cloud-free skies, the ground-based climatology can be extended to higher altitudes using Eq. 1; in cloudy conditions Eq. 1 is a reasonable approximation only near the ground, but not within and above cloud (see Sect. 4.2, and Sect. S3 of the Suppl. Materials)

The DNA fluences and hence survival times depend strongly on latitude and season. Survival times with respect to UV damage are very much longer in winter than in summer, particularly at higher latitudes, and are shortest in the tropics where seasonal variations are also small.

4 Life in Clouds

A particularly interesting and complex topic is the effect of clouds on microbial survival, all the more so because much data about bioaerosols comes from collected cloud and rain water. Clouds offer refuge from dehydration, which was previously mentioned as one of the environmental factors most damaging to microbial life. But they also have complex temperature and radiative effects that require consideration.

4.1 Microbial Activity in Clouds

Generally, temperatures decrease as we ascend through the troposphere, and life processes usually proceed more slowly at colder temperatures. But we should not overestimate the ability of low temperature to stop life processes. Mykytczuk et al.[105] found that the bacterium *Planococcus halocryophilus* is able to grow and divide at minus 15°C, (although the optimum temperature is around plus 25°C) and is still metabolically active at -25°C; and *Psychromonas ingrahamii* is still able to grow at -12°C [106]. *P. ingrahamii* normally lives in and on sea ice for which temperatures range from -1.8°C and -30°C, and the sea ice surface accounts for a large part of the primary production in the polar oceans.

While it has been known for a long time that microorganisms are present in air, only recently has evidence started to show that some microorganisms carry out life processes while aloft[107,108]. Many of the organisms are carried aloft in dry conditions (dust storms, e.g., refs. 19-21) and although spore germination has been observed at a water activity of only 0.64 [109] and bacterial cell division

below a water activity of 0.69 [110], biological activity takes place primarily in low clouds with life-friendly temperatures. Delort et al.[111] have given an overview of the microbial population in clouds. Most bacteria reaching the stratosphere are rapidly killed by ultraviolet radiation[112]. Repeated freeze-thaw cycles are particularly detrimental to microorganisms.

Cloud droplets and dry aerosols can be sampled using balloons[113] or aircraft, or from the ground on mountains[114]. The easiest way to get information about organisms in tropospheric clouds is to investigate rainwater collected under stringent conditions, although direct collection of cloud droplets is preferable. Also investigation of hailstones[115,116] provides a way to sample cloud organisms.

Hu et al.[117] found that a large fraction of the bacteria in rainwater were viable. However, this does not mean that they necessarily were biologically active while in the atmosphere prior to precipitation. Klein et al.[118] tried to obtain information on biological activity by measuring ribosomal DNA and RNA molecules. Since metabolically active cells have more ribosomes (e.g., ref. 119) they (often) have a higher rRNA to rDNA ratio. Krumins et al.[120], using rRNA abundance as a proxy, came to the conclusion that a bacterium, *Sphingomonas aerolata*, originally isolated from air, can be metabolically active while suspended in air. However, the positive correlation between rRNA/rDNA and growth rate does not hold for all bacteria[121], and thus some of their conclusions may need further support. Since RNA is more UV-resistant than DNA[122], UV radiation may be a confounding factor here. By studying the incorporation of added ^{3}H -thymidine into bacterial cells in cloud-water, Sattler et al.[123] estimated a cell number doubling time varying from 3.6 to 19. 5 days. Protein synthesis was estimated by incorporation of ^{14}C-leucine. From this they deduced an average carbon assimilation rate of 12 ng L^{-1} day^{-1}, with a maximum of 28 ng L^{-1} day^{-1}. They conclude that the global bacterial production of organic carbon in clouds may be in the range of 1-10 Tg carbon per year, a very small amount compared to the carbon cycle as a whole. Vaïtilingom et al[114] stress that for realistic simulation of biological activity in cloud water, ultraviolet radiation must be provided, and studied how its presence accelerated the destruction of hydroxyl radicals.

More certain evidence for microbial activity in the atmosphere could be obtained if one could define and measure chemical reactions taking place in the atmosphere that can be carried out only by living organisms. Direct monitoring of reactions in clouds is difficult, but Amato et al.[124], as well as Matulová et al.[125] showed that bacteria collected from cloud water are able to transform various organic substances present in the atmosphere and cloud water. Adenosine triphosphate was generated at 17°C by organisms in collected cloud water. *Pseudomonas* species, such as *P. syringae*, are considered to be among the more active bacteria in clouds, since they can develop at low temperature. A review of organic compounds present in fogs and clouds is provided by Herckes et al.[126].

Other photo-biological processes are also possible: Does photosynthesis take place in organisms suspended in the atmosphere? Klein[127] found that members of *Rhodospirillales* were abundant in both the total and potentially active communities in cloudwater. This order comprises non-sulfur purple bacteria, and one species (*Acidisphaera rubrifaciens*), otherwise known from its presence in hot springs, is also present in cloud-water. It contains bacteriochlorophyll, and grows on a number of carbon compounds present in cloud water. Growth is stimulated by illumination, but whether the bacterium carries out photosynthesis is not quite clear[128]. Cyanobacteria have been found in cloud-water[129] and rainwater[130]. Potentially they should be able to carry out photosynthesis in the atmosphere in the absence of organic carbon or a reductant other than water. Whether they do that has not been established.

The residence time in the atmosphere is probably limiting the extent to which organisms are able to reproduce while aloft. Burrows et al.[131] estimated the residence time for 1 μm particles to range from less than a week if they finally act as condensation nuclei to around 180 days if they do not form condensation nuclei. Based on this, the generation times of organisms in cloud water measured by Sattler et al.[123], temperatures in the atmosphere and other factors, Klein[127] in her thesis drew the conclusion that at least some organisms could go through more than 50 generations while in the atmosphere. This is probably an overestimate, since it is based on the size of "naked" individual bacteria, while bacteria are usually clumped together or attached to other particles, and in any

case they would need to be associated with water containing nutrients to grow and divide.

We may conclude that although bacteria can be metabolically active in clouds, their growth and production of organic compounds there is probably negligible in comparison to their activity in other environments, but may have a non-negligible impact on cloud water composition[111,132].

4.2 Clouds and DNA-damaging UV Radiation

Clouds present some particular challenges also because they dramatically modify the sky radiation field at UV wavelengths as well as visible wavelengths. The effects of clouds on fluence rates are as complex as the great variety of clouds themselves.

Considering an isolated, simple single-layer, horizontally extended cloud, three regimes can be identified: (1) above cloud, the fluence rate is generally enhanced by the reflection from the cloud below; (2) below cloud, the fluence rate is typically reduced due to attenuation of sunlight by overhead cloud; and (3) within the cloud, where strong vertical gradients are experienced. This is illustrated in Fig. 7 for DNA-damaging fluences, computed at two latitudes (the polar circle and Equator) for a range of cloud optical depths. Although the absolute values differ by more than an order of magnitude between the two locations, the relative effect of cloud is mostly similar, except near cloud top.

Blue skies become white (or grey) when clouds appear, so a shift in the spectral distribution is expected. The optical properties of cloud particles are relatively independent of wavelength[133], but the interactions with Rayleigh scattering cause a wavelength dependence to appear: Back-scattering between the top of the cloud and the overlying air molecules gives to some photons multiple opportunities to re-enter the cloud, traverse it, and reach the ground[134-135]. This effect becomes greater at the shorter wavelengths where Rayleigh scattering is stronger. In general, clouds show greater contrast relative to clear skies at visible wavelengths, while at UV wavelengths Rayleigh scattering already contributes substantial haziness[136].

In practice, clouds can exhibit considerable horizontal and vertical structure, and are still difficult to predict or even describe quantitatively. Measurements of

fluence rates (actinic fluxes) in the presence of clouds have been made from balloons and aircraft, but are few and have been limited to UV-A and visible wavelengths[137,138]. The measurements generally confirm radiative transfer calculations of the vertical profiles (such as those in Fig. 7), but also re-affirm the large uncertainties that arise from incomplete knowledge of clouds, from their microscopic drop or ice particle size distributions that determine optical properties, to larger scale often-complex three-dimensional morphology possibly including multiple layers, etc.

Clouds can also complicate the relationship between fluence rate at any altitude, and ground-level irradiances, Eq. 1, which was derived from modeled cloud-free skies. Model calculations with clouds show (e.g. Fig. S4) that above and inside clouds the ratio R can achieve much larger values due to reflections from the cloud, but below it, i.e. between cloud base and ground, it is remarkably similar to the clear-sky value, ca. 2 (since in both cases scattered radiation from all directions is important).

Satellites can provide cloud information, such as location and optical depth, needed to estimate fluence rates at various altitudes. Ryu et al.[139] compared UV-A fluence rates measured from aircraft during cloudy conditions, with model calculations that used observations of reflected radiance from the Geostationary Operational Environmental Satellite to infer cloud optical properties, and from these the fluence rates at aircraft locations. The assimilation of satellite-derived clouds led to good agreement with fluence rate observations, and an improvement over using clouds predicted by a weather forecasting model.

An additional optical consideration is that the fluence rates inside cloud droplets are themselves enhanced by diffraction[140] which can be understood as a lensing effect in the geometric (large particle) limit. For typical spherical cloud droplets this enhancement factor is 1.6, i.e., the in-drop average fluence rate is 60% larger than in the interstitial space.

5. Conclusions

Solar radiation is just one of several detrimental factors facing microorganisms aloft. Survival times against UV damage calculated here can therefore be

regarded as maximum estimates, provided the organisms appear as single cell particles. For the bacteria considered here, e-folding inactivation (37% survival) occurs in a few minutes when DNA damaging fluences are of order 100 J m^{-2} day^{-1}. Survival times calculated with the e-folding criterion can be extremely short, but because of the great number of individuals in some microbial populations, survival times of populations are longer.

Bacteria are often clumped together with one another or with other material, which can result in increased resistance to UV radiation. Sensitivity varies greatly among organisms, even for the small subset of species for which data exist. Under solar radiation, the UV-B component usually dominates killing, and the effect of visible light is negligible. Bacterial spores are much more resistant than vegetative cells of the same species, but there exist also bacterial species that are very radiation-tolerant in the vegetative stage. To some extent microorganisms may multiply while suspended in the atmosphere.

In any particular situation, the UV exposure for airborne spores can in principle be computed using a radiative transfer code in conjunction with atmospheric dynamical modelling by considering the fluence rate received along trajectories of atmospheric winds that carry the organism in question (both in the vertical and horizontal directions). However, allowance for the effects of clouds, which are ubiquitous in most areas, will always lead to large uncertainties in exposure, and therefore in estimated survival times. Measurements of the action spectrum for UV damage for the species in question are also required.

Sophisticated numerical models have been developed in recent years to simulate the physical and chemical state of the atmosphere, i.e. to better understand and predict issues important to human society including weather, climate change, and air pollution. But to our knowledge, analogous models for the biological state of the atmosphere have not yet been developed or remain rather crude (compared to the physico-chemical models). Such models could be quite useful for a wide range of problems, for example, in studying specific episodes leading to acute health impacts (e.g. from transport of allergens), or in helping to understand the geographic distribution of species over evolutionary time scales. They may also provide better estimates for biological influences on

526 atmospheric physics, e.g. the nucleation of ice clouds by biological particles[111].
527 The need for an interdisciplinary approach in building such a model is self-
528 evident. In this paper we discussed three issues at the intersection of
529 microbiology and the atmospheric sciences: (i) the selection and applicability of
530 representative biological sensitivity spectra, (ii) the importance of including
531 radiation arriving from all directions of the atmosphere (not just the direct solar
532 beam), and (iii) the spatial and temporal variability of atmospheric radiation,
533 including its dependence on season, latitude, longitude (or time of day), altitude,
534 and atmospheric constituents such as ozone and clouds. We have shown that
535 these problems are complex but tractable, and note increasing opportunities for
536 including photo-biological processes in interdisciplinary atmospheric models.
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Table 1. Various organisms found in air, together with their terminal sedimentation velocities[a] and typical sizes.

Species	V_{term}, cm s^{-1}	size, µm	Reference
Orchid seeds			
Encyclia diurna	11	410x120	Zotz et al. 2016 [5]
Epidendrum difforme	10	520x60	Zotz et al. 2016 [5]
Stenorrhynchos speciosum	9	1500x150	Zotz et al. 2016 [5]
Brassavola nodosa	16	540x80	Murren & Ellison 1998 [6]
Pollen[b]			
Abies pectinata	39		Szczepanek et al. 2017 [7]
Picea abies	8.7		Szczepanek et al. 2017 [7]
Pinus sylvestris	2.5-4.0		Szczepanek et al. 2017 [7]
Juniperus, various species	0.73-1.29	19-24	Bunderson & Levetin 2015 [8]
Zea mays	17-31		Chamecki et al. 2011 [9]
Zea mays	23-28	80-105	Marceau et al, 2012 [10]
Moss spores	0.49- 8.52	12-53	Zanatta et al. 2016 [11]
Fungal spores	0.002-0.5	1.5-7.2	Hussein et al. 2013 [12]
Bacteria	very low	0.65-1.1	Lighthart 1997 [13]
Virus	very low	0.004-0.1	various sources

[a] *The velocities given in the table are for sea level. At higher elevation the terminal velocities are larger, because air pressure decreases more rapidly with elevation than does gravity.*

[b] *For more data on conifer pollen, see ref. 14.*

Table 2. Survival of bacteria in aqueous suspension after exposure to UV radiation (from Santos et al.[57]). The dominant wavelength for UV-A is 365 nm, for UV-B 302 nm.

Bacterial strain	Surviving fraction	
	UV-A 300 kJ m^{-2}	UV-B 90 kJ m^{-2}
Acinetobacter (EU545154.1)	0.14	0.045
Bacillus thuringiensis (JN084031.1)	0.10	0.0041
Brevibacterium (JF905605.1)	0.0050	0.0068
Micrococcus (HM352362.1)	0.44	0.18
Paracoccus (AB681547.1)	0.59	0.0040
Pseudomonas (JF749828.1)	0.34	0.11
Psychrobacter piscidermidis (EU127295.1)	0.43	0.012
Sphingomonas (AM900788.1)	0.48	0.015
Staphylococcus saprophyticus (HQ407261.1)	0.15	0.078

Table 3. Literature describing photoinactivation spectra for various organisms. For Escherichia coli many different strains and effect of aerobic and anaerobic conditions have been investigated. In some cases both exponentially growing bacteria and those in stationary phase have been used. There are also many spectra for virus, which are not listed in the table.

Organism	Wavelength range	Reference
Bacteria	nm	
"B. coli"	254–302	Gates 1930 [61]
Escherichia coli	254–434	Peak et al. 1983, 1984 [62,63]
Escherichia coli	254–405	Kelland et al. 1983 [64]
Escherichia coli *	254–460	Webb & Brown 1979 [65]
Escherichia coli	254–365	Webb & Tuveson 1982 [66]
Escherichia coli	270–740	Lui et al. 2016 [67]
Escherichia coli	240–460	Mackay et al. 1976 [68]
Escherichia coli *	300-450	Silverman & Nelson 2016 [69]
Enterococcus faecalis	270–660	Lui et al. 2016 [67]
Staphylococcus aureus	254–302	Gates 1930 [61]
Staphylococcus aureus	400–430	Maclean et al. 2008 [70]
Salmonella typhimurium	222–303	Chen et al. 2009 [71]
Salmonella typhimurium *	240–550	Mackay et al. 1976 [68]
Propionibacterium acnes *	320–440	Kjeldstad & Johnsson 1986 [72]
Bacillus subtilis spores	222–303	Chen et al. 2009 [71]
Bacillus subtilis spores	200–293	Cabaj et al. 2002 [73]
Bacillus subtilis spores	217–294	Mamane-Gravetz et al. 2005 [74]
Bacillus subtilis spores	172–254	Wang et al. 2010 [75]
Bacillus subtilis spores	0.1–300	Munakata et al. 1991, 1992 [76,77]
Bacillus subtilis spores	254–365	Tyrrell 1995 [78]
Bacillus pumilis spores	220–290	Beck et al. 2015 [79]
Eukaryotes		
Saccharomyces cerevisiae	254–313	Zölzer & Kiefer 1983 [80]
Cryptosporidium parvum	210–290	Beck et al. 2015 [79]
Cryptosporidium parvum	216–290	Linden et al. 2001 [81]

* *Indicates those used in Figs. 1-3.*

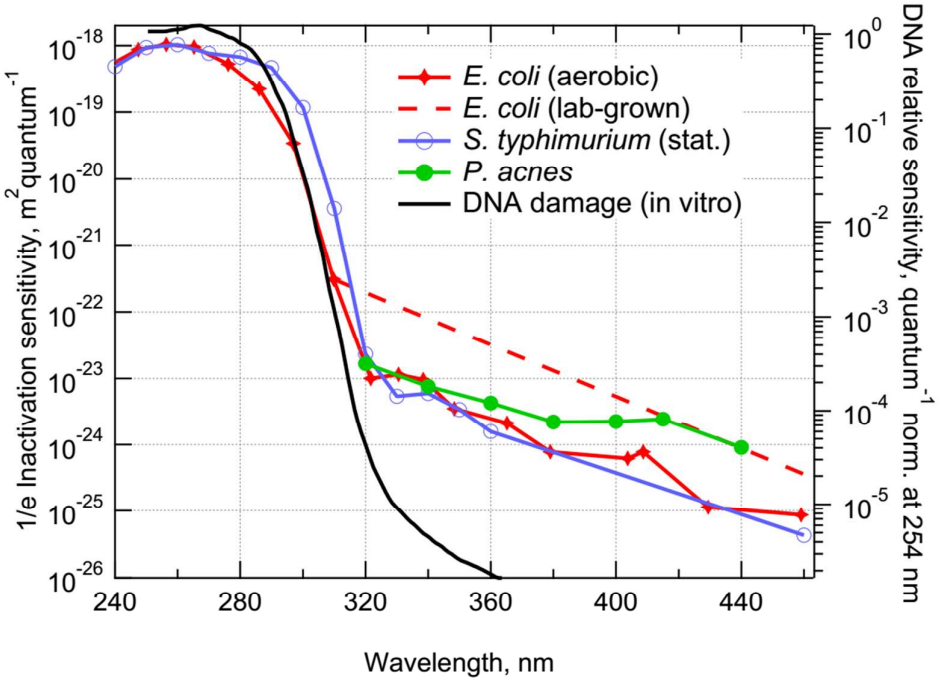

Figure 1. Inactivation spectra for some bacteria compiled from various sources, expressed on a logarithmic scale as (spectral) inactivation cross section, i.e. the inverse of number of photons per m² necessary to reduce the amount of living bacteria by the factor e (left scale). Also shown is the spectrum for in vitro DNA damage normalized at 254 nm (right scale), from Setlow[82]. E. coli (aerobic) from Webb and Brown[65]; UV-A tail for lab-grown E. coli (also aerobic) from Silverman and Nelson[69]; for anaerobic conditions the curves are lower for wavelengths over 320 nm; Propionibacterium acnes from Kjeldstad and Johnsson[72]; salmonella typhimurium (stationary phase) from Mackay et al.[68].

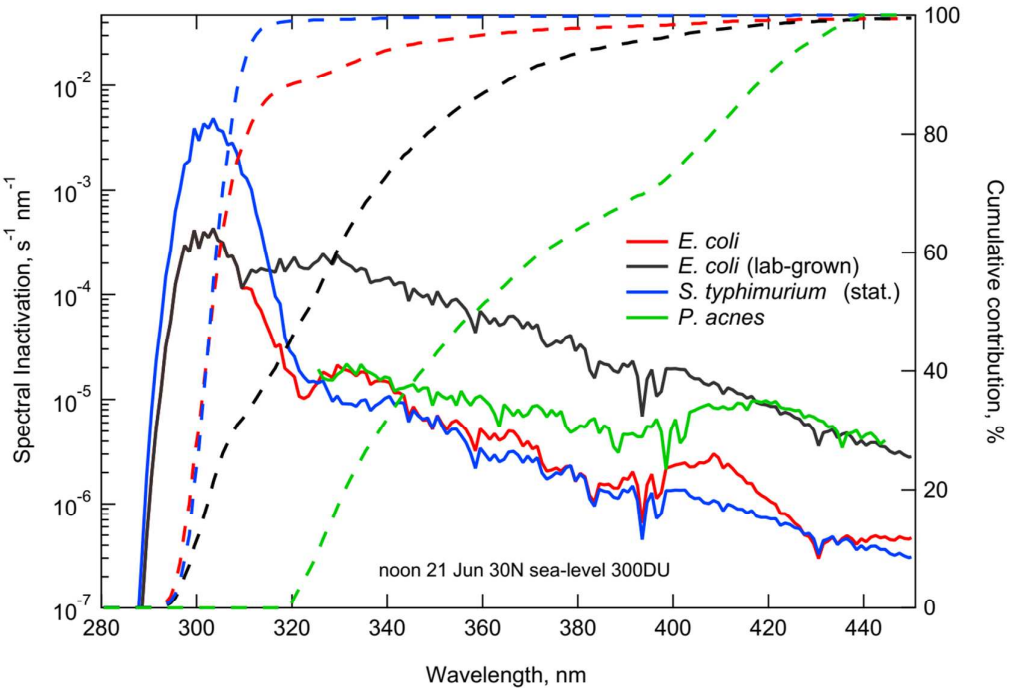

Figure 2. Contribution of different wavelengths to the total inactivation (left scale), for the spectra shown in Fig. 1. The dashed lines (right scale) give the cumulative contribution from short to long wavelengths. Calculations were made with the TUV model (see Supp. Mat.)

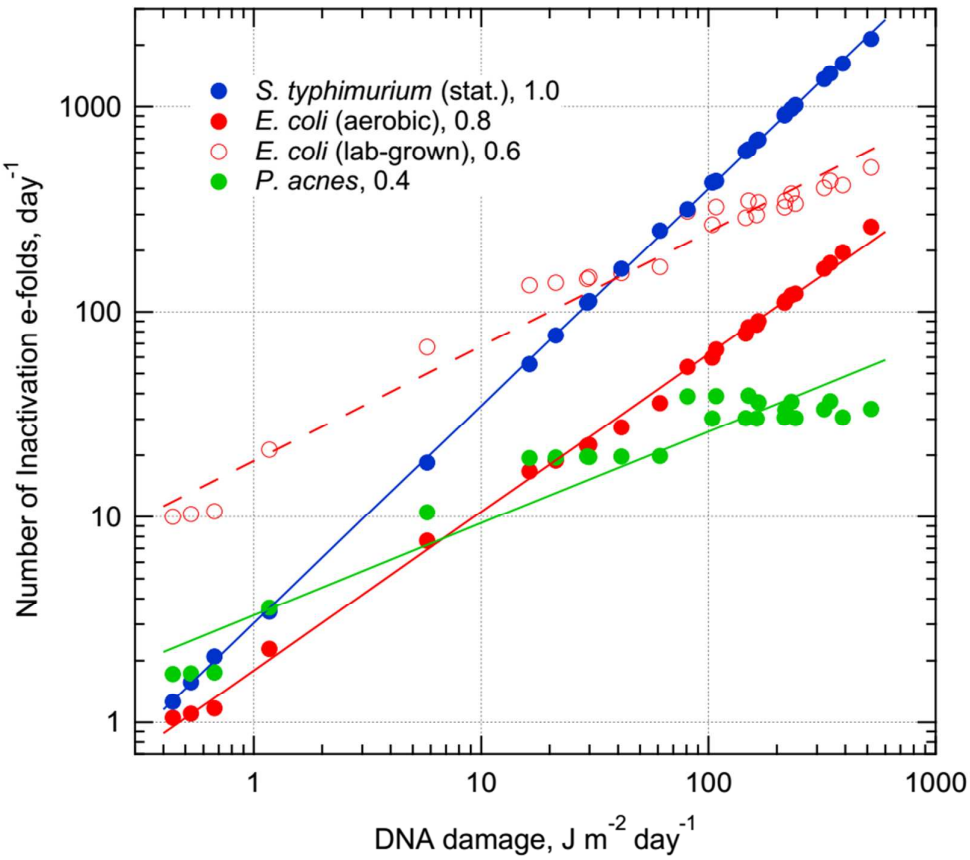

Figure 3: Correlations between DNA damaging fluence with inactivation rates for several bacteria. TUV model for clear skies, range of latitudes and seasons. The correlation exponents (log-log slopes) are given in the legend.

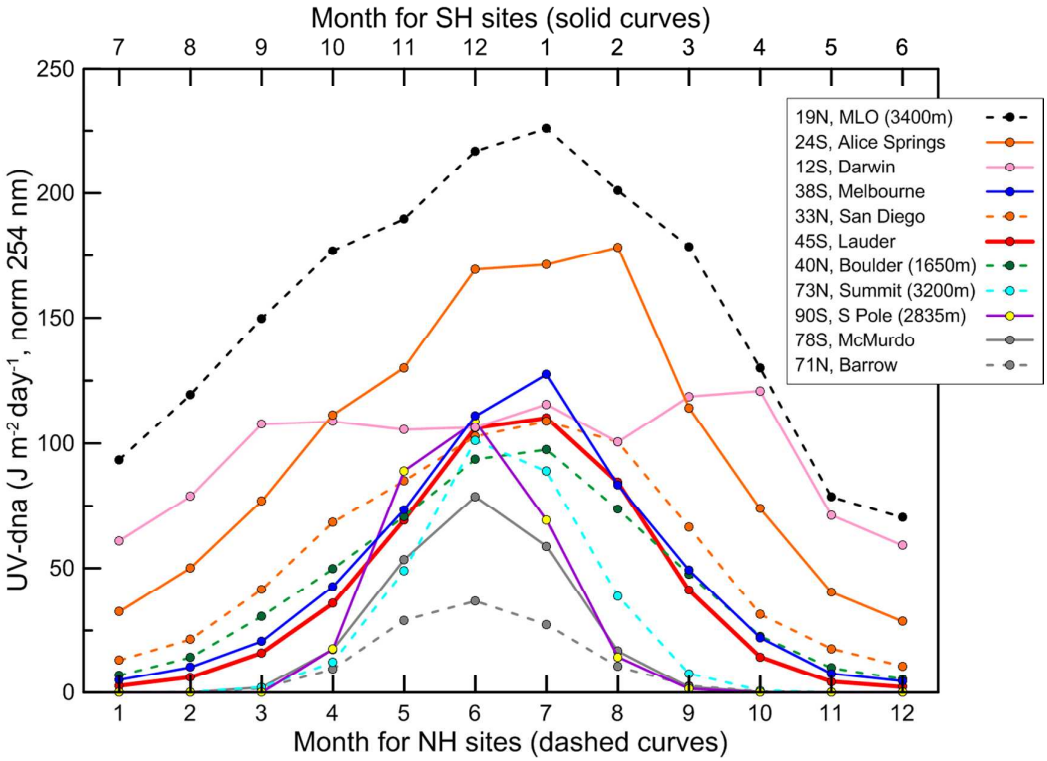

Figure 4. Monthly means of DNA-weighted irradiance based on measurements at NDACC sites (see text for explanation). Solid lines are used to denote Southern Hemisphere sites, while dashed curves are used to denote Northern Hemisphere sites. Note that the x-axis for Southern Hemisphere sites has been shifted by 6 months to allow direct comparability with Northern Hemisphere sites.

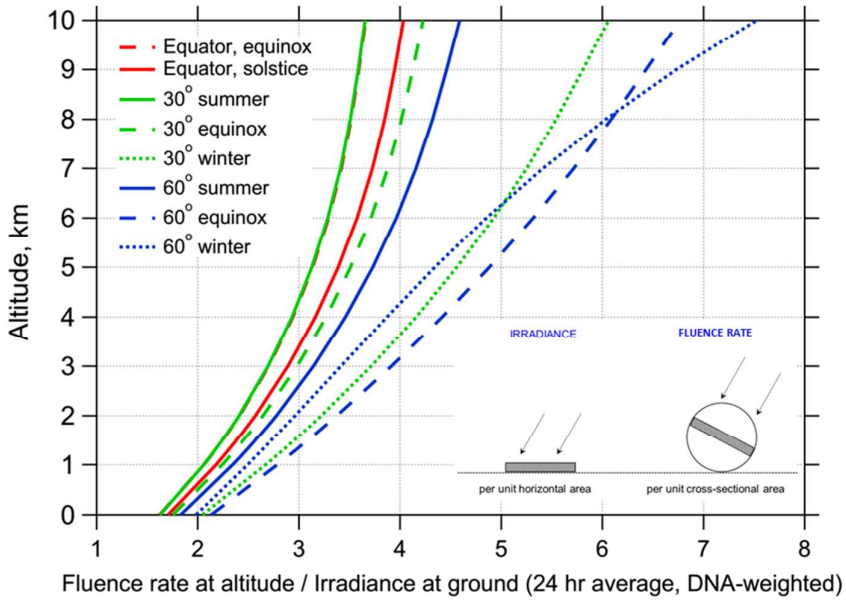

Figure 5: Geometric factor relating the horizontal irradiance measured at Earth's surface, to the fluence rate at any altitude, based on the DNA damage action spectrum, 24 hr averages, calculated with the TUV model for representative locations and dates, cloud-free conditions. Inset in lower right shows difference between irradiance on a horizontal surface and fluence rate incident on a spherical or randomly oriented particle.

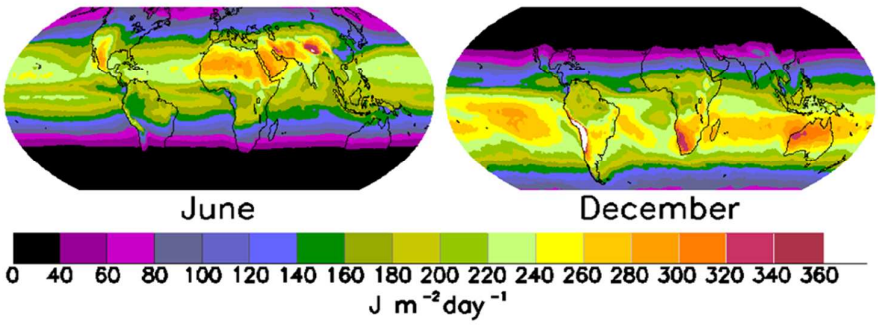

Figure 6. DNA-damaging UV fluence at ground-level. Climatology derived using satellite-observed ozone and clouds 1979-2000 as input to the TUV model[104].

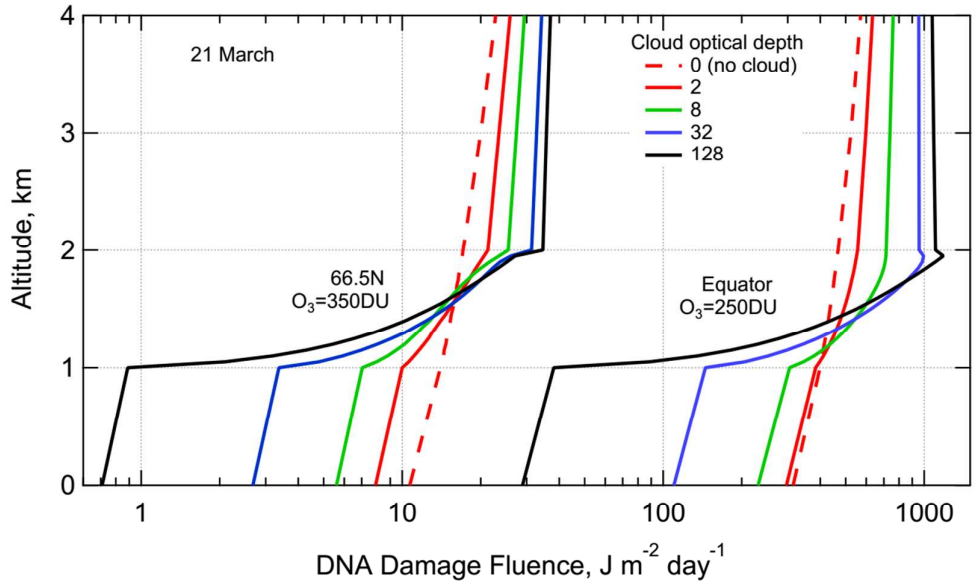

Figure 7. The vertical structure of DNA-damaging fluence in the presence of clouds, at two locations (polar circle and equator) for several cloud optical depths given in legend. Dashed curves are for cloud-free conditions. Calculated with the TUV model.