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Influence of Tropospheric Ozone Control on Exposure to Ultraviolet Radiation at the Surface

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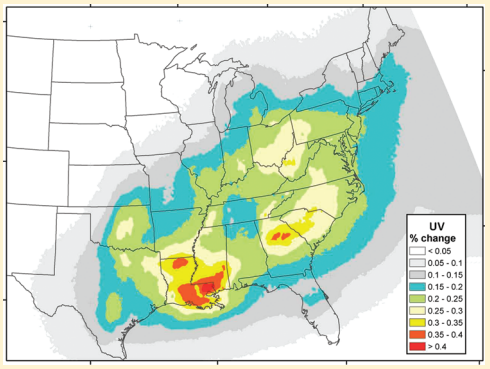
Supporting Information

ABSTRACT: Improving air quality by reducing ambient ozone (O_3) will likely lower O_3 concentrations throughout the troposphere and increase the transmission of solar ultraviolet (UV) radiation to the surface. The changes in surface UV radiation between two control scenarios (nominally 84 and 70 ppb O_3 for summer 2020) in the Eastern two-thirds of the contiguous U.S. are estimated, using tropospheric O_3 profiles calculated with a chemistry-transport model (Community Multi-Scale Air Quality, CMAQ) as inputs to a detailed model of the transfer of solar radiation through the atmosphere (tropospheric ultraviolet-visible, TUV) for clear skies, weighed for the wavelengths known to induce sunburn and skin cancer. Because the incremental emission controls differ according to region, strong spatial variability in O_3 reductions and in corresponding UV radiation increments is seen. The geographically averaged UV increase is 0.11 ± 0.03%, whereas the population-weighted increase is larger, 0.19 ± 0.06%, because O_3 reductions are greater in more densely populated regions. These relative increments in exposure are non-negligible given the already high incidence of UV-related health effects, but are lower by an order of magnitude or more than previous estimates.



INTRODUCTION

Atmospheric ozone (O_3) absorbs solar ultraviolet (UV) radiation at wavelengths that are particularly active biologically (UV-B, 280−315 nm, and to a lesser extent UV-A, 315−400 nm). While the majority of O_3 (~90%) is located in the stratosphere, O_3 in the troposphere is even more effective, on a per molecule basis, in absorbing UV radiation because Rayleigh scattering by the denser air in the lower atmosphere can increase photon path-lengths and the probability of encountering O_3 molecules.[1] Since preindustrial times the amount of O_3 in the troposphere has increased substantially over natural background levels, and model calculations suggest that this has lowered surface UV radiation by 3−8% over the U.S. and Europe.[2,3] However, these changes are small relative to those from variations in stratospheric O_3, clouds, or aerosols, and observational studies have not shown a clear signature of tropospheric O_3 variability on surface UV radiation.[4,5]

Tropospheric O_3 is also a main constituent of urban and regional smog,[6,7] with well-known effects on human (esp. respiratory) and ecosystem health.[8,9] The question arises then to what extent lowering tropospheric O_3 concentrations could increase surface UV radiation. The only previously published analysis linking tropospheric O_3 control strategies to increased surface UV radiation reached a preliminary conclusion that the incremental costs of skin cancer and cataract may be of the same order of magnitude as the corresponding benefits of improved respiratory health.[10] If tropospheric O_3 concentrations were reduced by 10 parts per billion (ppb) in the United States, the average erythemal UV radiation levels would increase by 1.3−2.5%, leading to an additional 2000−11 000 nonmelanoma skin cancer and 13 000−28 000 cases of cataract per year. Although the assumptions inherent in that analysis have been discussed extensively,[8,11] to date no improved estimates have been presented.

Here, state of the art models of atmospheric chemistry, transport, and radiation are combined to provide a more realistic calculation of how surface UV radiation responds to changes in tropospheric O_3 under different control scenarios. The modeling framework features (i) geographic and temporal distributions of the changes in tropospheric O_3; (ii) an accurate UV radiation model including coupling between Rayleigh scattering and O_3 absorption; and (iii) high spatial resolution (county level) overlap between population and the predicted UV radiation changes. Compared with the previous study[10] in which a fixed 10 ppb reduction in surface O_3 used, our analysis accounts for the detailed temporal, horizontal, and vertical variability of the O_3 changes, and yields UV radiation changes that are about an order of magnitude smaller.

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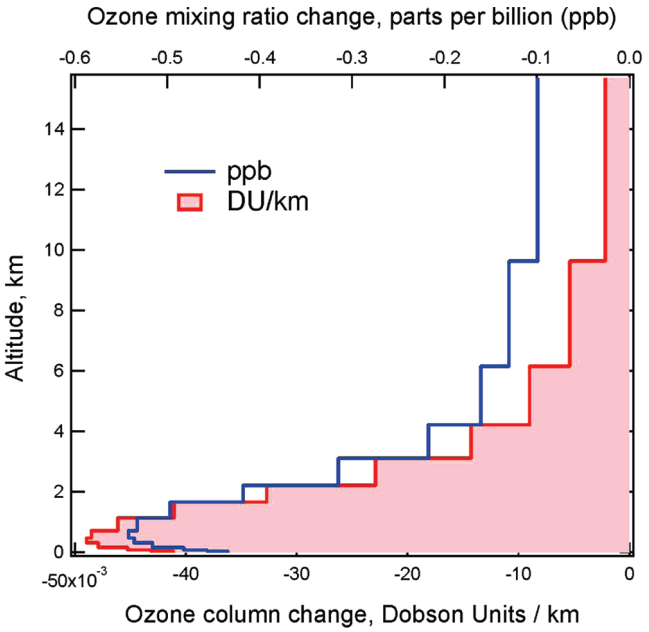

Figure 1. Domain-averaged O_3 reductions from the 084 to 070 scenarios, expressed as mixing ratio (blue, top scale in units of parts per billion, ppb), and as O_3 column density (red, bottom scale in Dobson Units (DU) km^{-1} where 1 DU = 2.69×10^{16} molecules cm^{-2}). The step structure reflects the CMAQ model vertical layering. The shaded area gives the total vertically integrated O_3 column change (0.18 DU).

MATERIALS AND METHODS

The spatial and temporal distribution of tropospheric O_3 was obtained from simulations with the Community Multi-Scale Air Quality (CMAQ) model as carried out for, and described in, the Ozone Regulatory Impact Analysis.[12] The CMAQ model is a three-dimensional chemistry transport model (CTM) that includes emissions, chemical transformations, horizontal and vertical transport, and removal by deposition or outflow from the domain.[13] For this application, the domain consisted of the contiguous U.S. eastward of about 140°W, subdivided into a 12 × 12 km horizontal grid and 14 altitude layers extending from the ground to about 16 km. Meteorological fields were generated offline with a mesoscale model for historical meteorological conditions,[12] and chemical boundary conditions were provided by a global CTM. Ozone concentrations, expressed as molar mixing ratio relative to air in parts per billion (ppb), were computed for each grid point (latitude, longitude, altitude) at hourly intervals for 24 h of each day from 1 June to 31 August 2020.

Two scenarios were selected to represent a hypothetical control strategy toward attainment of historical standards (the 084 scenario), and with additional known controls designed to bring areas predicted to exceed 70 ppb in 2020 into attainment (the 070 scenario). Both scenarios are based on 8 h averages. The controls include a multitude of strategies depending on economic sector and geographic region, and include current state and federal programs plus controls to attain the current O_3 standard and $PM_{2.5}$ standards (see http://www.epa.gov/ttnecas1/ria.html for a complete list of controls). More detailed descriptions of the processes and parametrizations implemented in the CMAQ model are given in ref 12. Importantly, the use of the same model simulations here ensures that the evaluation of UV increments is made in the same framework as that of the benefits (e.g., respiratory) of tropospheric O_3 reductions.

Figure 1 shows how the domain-averaged vertical O_3 profiles change in going from the 084 to the 070 scenario. The largest changes in the O_3 mixing ratio as well as in the contribution to the total O_3 vertical column amount occur below 3 km altitude, but contributions from higher layers are significant (e.g., with about 7% of the column change in the top layer) and must be included. The changes at the surface are somewhat smaller than the peak values because of the nighttime deposition of O_3. The uncertainty in CMAQ predicted surface O_3 mixing ratios has been estimated as 10−20%, based on comparisons with observations,[14,15] from which we estimate the in-quadrature uncertainty for the O_3 difference between two scenarios as about ±20%.

The O_3 changes shown in Figure 1 are averaged over all locations and times for illustrative purposes, but cannot be translated directly into changes in surface UV radiation by simple scaling. The altitude of the O_3 perturbations is critical because of the coupling between Rayleigh scattering and absorption.[1] The diurnal dependence is also important, e.g., the domain-averaged change in the surface O_3 concentration (for 15 July) is −0.7 ppb for high sun (solar zenith angle, sza, < 45°), −0.6 ppb for low sun (45° < sza <90°), and −0.5 ppb at night (sza >90°). To capture accurately the effects of spatial and temporal variability of tropospheric O_3 and its changes, the UV radiation is calculated using the full O_3 vertical profile every hour (from 1 June to 31 August) at each of the 66 920 geographic locations.

The surface spectral UV irradiance was calculated with the tropospheric ultraviolet-visible (TUV, version 4.5) model,[16] which accounts for the scattering and absorption of solar UV by atmospheric gases and particles. This spectral irradiance was multiplied by a sensitivity function (action spectrum) for the induction of nonmelanoma skin cancer in mice corrected for human skin transmission,[17] then integrated over UV wavelengths to obtain the biologically effective irradiance. Additional details of the radiation modeling are provided as Supporting Information. The overall uncertainty associated with the atmospheric configuration and radiative transfer calculation is estimated as ±20%. Combined with the uncertainty in predicting O_3 concentrations, the estimated uncertainty of the UV increments between scenarios is ±30%.

RESULTS AND DISCUSSION

The relative change in skin cancer-weighted surface UV radiation between O_3 scenarios 084 and 070 is shown in Figure 2, for the summer months (June, July, and August) of 2020. The largest changes are seen to occur near the Mississippi River delta, the southeast United States, and the Ohio Valley, while negligible changes are seen in the modeled region westward of the central United States. Figure 3 shows the number of locations incurring specific increases in monthly UV radiation. Of the 66 920 locations, about 20% have negligible UV increments (within 0.01% of zero), and few show increments larger than 0.4% (or 0.3% in June). The median UV increment was about 0.07% in June and 0.09% in July and August.

The temporal variability of the domain-averaged UV is shown in Figure 4. An overall increasing trend is seen from early June through early August, with values more than doubling (from ∼0.06% to ∼0.15%) before declining again, due largely to the meteorological conditions (stagnation, high temperatures) that are conducive to O_3 photochemical production, which then also

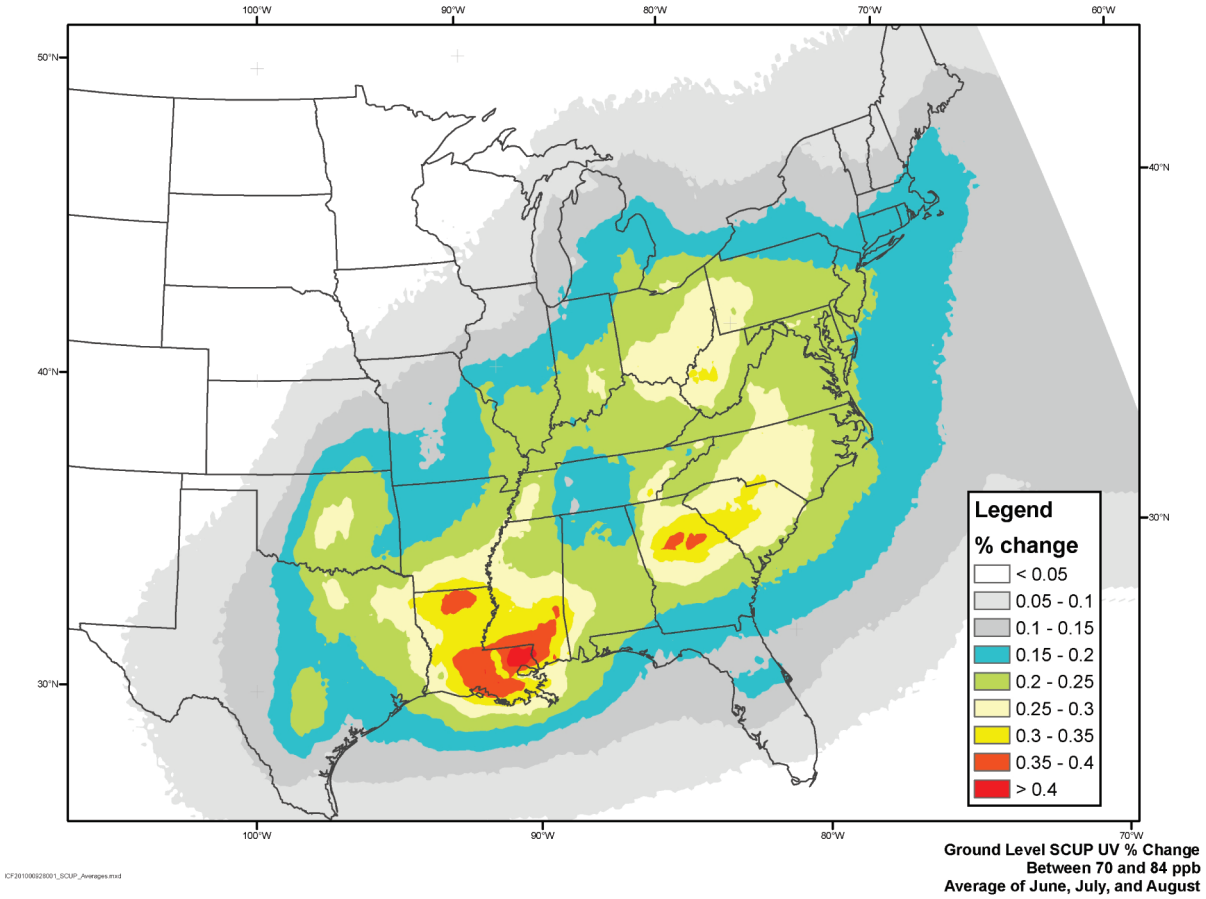

Figure 2. Increase (%) in ground level skin cancer-weighted UV irradiance, from scenario 084 to 070, for summer.

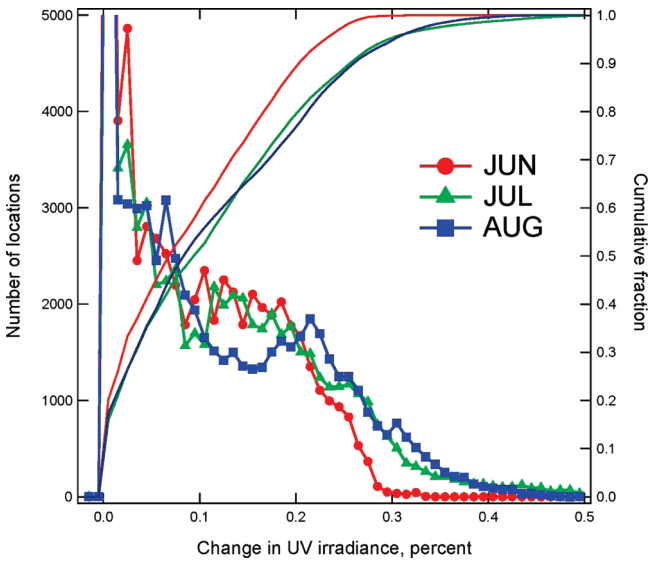

Figure 3. Frequency distribution of increases in skin cancer-weighted monthly UV radiation, in steps of 0.01%, from scenario 084 to 070. Lines with markers show number of locations (66 920 total, left scale) while smooth lines show cumulative fraction (right scale).

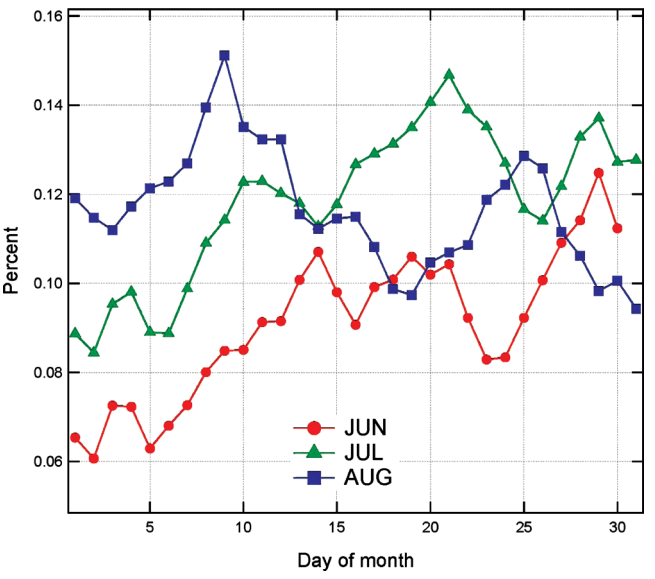

Figure 4. Domain-averaged increases (%) in skin cancer-weighted daily UV doses, from scenario 084 to 070.

scales the magnitude of O_3 reductions between the 084 and 070 scenarios. The UV radiation increases in Figures 2−4 are not weighted by population, and therefore reflect changes in the physical state of the atmosphere rather than a measure of change in human exposure. The population-weighted increments, shown in Table 1 as averages over the domain, are considerably larger than for geographic averages, as expected if the incremental

Table 1. Domain-Averaged Changes from O_3 Control Scenarios 084 to 070

ΔO_3, DU	ΔO_3, %	geographically averaged change in UV radiation, %	population-weighted change in UV adiation, %
-0.18 ± 0.04	-0.051 ± 0.01	0.11 ± 0.03	0.19 ± 0.06

controls of the 070 scenario are most effective for densely populated areas.

The population-weighted increment in UV exposure associated with the lower tropospheric O_3 scenario estimated here, $0.19 \pm 0.06\%$, is an order of magnitude smaller than the 1.3−2.5% increment estimated by ref 10. The main reason for the discrepancy is that the earlier study applied a 10 ppb reduction in O_3 uniformly over the fraction of the population in nonattainment areas, and throughout the vertical extent of the troposphere, while the CMAQ model predicts much smaller O_3 changes on average. The incremental control strategy between the 084 and 070 scenarios has the greatest impact on reducing surface O_3 in the most polluted areas, but these reductions are not proportionate throughout the vertical extent of the atmosphere or the geographic domain. As seen in Figure 1, the geographically averaged surface O_3 reduction is only ~0.5 ppb and decreases rapidly with altitude. Thus, the change in tropospheric O_3 content was previously overestimated. Detailed temporal and spectral variations neglected in the earlier work are now also included.

Skin cancer is widespread in the United States, with an estimated annual 2 000 000 medical procedures for nonmelanoma skin cancer[18] and 60 000 new cases of melanoma.[19] Therefore even small relative increases in UV exposure may lead to substantial numbers of new cases. However, a direct translation of our result into incremental skin cancer incidence is difficult because the computed UV increments apply to a single year (2020) while the occurrence of skin cancer is related to life-long exposure, with induction times of up to several decades. Significant uncertainties also exist in the dose−response (exposure-incidence) relations, and for melanoma on the relative importance of UV-B and UV-A wavelengths.[20] A detailed analysis of the health impacts of such incremental UV exposures is beyond the scope of this work. Published estimates do exist for the increases in nonmelanoma skin cancer associated with sustained stratospheric O_3 depletion.[21−23] Although the temporal and spatial variabilities of stratospheric and tropospheric O_3 differ greatly, some perspective may be gained by comparing the $0.19 \pm 0.06\%$ UV increment calculated here with the 6−14% increase estimated to have occurred at middle and high latitudes of both hemispheres due to stratospheric O_3 depletion in the 1980s and 1990s,[22] or with the more than doubling by 2060 at northern midlatitudes if the Montreal Protocol agreements to protect stratospheric O_3 had not been implemented.[23]

A fundamental premise of this analysis is that the relative (percent) change in UV exposure resulting from a given change in tropospheric O_3 can be assessed separately from all other changes, for example, in stratospheric O_3, clouds, aerosols, other gaseous pollutants, surface reflectivity, behavioral exposure, migration or other demographic trends. Some interactions are easily considered, for example, if behavioral patterns change so that more people are exposed to UV radiation, the relative increments calculated here apply to this larger population. Other less direct, but potentially important, effects may occur. Emissions reductions aimed at diminishing ambient O_3 may also affect other atmospheric gases and aerosols that alter atmospheric UV transmission directly through absorption and scattering, and indirectly by changing cloud cover and more generally climate. Such indirect effects are poorly known and not included here.

■ ASSOCIATED CONTENT

Supporting Information. Additional information including a figure and table. This material is available free of charge via the Internet at http://pubs.acs.org.

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