Extreme Ozone Loss Following Nuclear War Results in Enhanced Surface Ultraviolet Radiation

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20 Key Points:

- Global nuclear war injecting 150 Tg of stratospheric smoke causes a peak global ozone
 loss of 75% with depletion lasting 15 years.
- Ozone loss leads to a tropical UV Index above 35 after 3 years and lasting 4 years, and a 20% global average increase in UV-B, a hazard to life.
- Including smoke decreases photolytic reactions rates, increasing ozone loss by 15% with decreasing NO_x and increasing HO_x catalytic cycles losses.
- 27

28 Abstract

- 29 For the first time, we use a modern climate model with interactive chemistry including the
- 30 effects of aerosols on photolysis rates to simulate the consequences of regional and global scale
- 31 nuclear wars (injecting 5 and 150 Tg of soot respectively) for the ozone layer and surface
- 32 ultraviolet (UV) light. For a global nuclear war, heating in the stratosphere, reduced photolysis,
- 33 and an increase in catalytic loss from the HO_x cycle cause a 15 year-long reduction in the ozone
- column, with a peak loss of 75% globally and 65% in the tropics. This is larger than predictions
- 35 from the 1980s, which assumed large injections of nitrogen oxides (NO_x) , but did not include the
- 36 effects of smoke. NO_x from the fireball and the fires provide a small (5%) increase to the global
- 37 average ozone loss for the first few years. Initially, soot would shield the surface from UV-B, but
- 38 UV Index values would become extreme: greater than 35 in the tropics for 4 years, and greater 39 than 45 during the summer in the southern polar regions for 3 years. For a regional war, global
- 40 column ozone would be reduced by 25% with recovery taking 12 years. This is similar to
- 41 previous simulations, but with a faster recovery time due to a shorter lifetime for soot in our
- 42 simulations. In-line photolysis provides process specific action spectra enabling future
- 43 integration with biogeochemistry models and allows output that quantifies the potential health
- 44 impacts from changes in surface UV for this and other larger aerosol injections.

45 Plain Language Summary

- 46 Nuclear war would result in many immediate fatalities from the blast, heat, and radiation, but
- 47 smoke from fires started by these weapons could also cause climate change lasting up to 15 years
- 48 threatening food production. For the first time with a modern climate model, we have simulated
- 49 the effects on ozone chemistry and surface ultraviolet light caused by absorption of sunlight by
- 50 smoke from a global nuclear war. This could lead to a loss of most of our protective ozone layer
- 51 taking a decade to recover and resulting in several years of extremely high ultraviolet light at the
- 52 surface further endangering human health and food supplies.

53 1 Introduction

It has been understood since the early days of nuclear weapons testing that nuclear detonations
can initiate large-scale fires in urban and rural settings (Glasstone & Dolan, 1977; OTA, 1979;
Lewis, 1979). The first estimates of the massive amount of smoke that could be generated in a

- 57 global-scale nuclear war were made by Crutzen & Birks (1982), and the potential for this amount
- of smoke injected into the stratosphere to cause global climatic change was shown by the TTAPS
- 59 study (Turco et al., 1983), where the outcome was likened to a "nuclear winter." Subsequent
- simulations by scientists in both the United States and the Soviet Union confirmed these results
- 61 (e.g., Aleksandrov & Stenchikov, 1983; Covey et al., 1984). More recently, these results have
- 62 been reproduced with modern Earth System models by Robock et al. (2007b) and Coupe et al.
- (2019) showing climatic effects lasting for over a decade. Prolonged heating and self-lofting by
 the soot lengthens its lifetime compared to the climatic effects of sulfate from a large volcanic
- 65 eruption like that of Mount Pinatubo in 1991, which lasted for about 2 years (Robock, 2002).
- 66 Originally, it was assumed that a global scale nuclear war between the United States and
- 67 the Soviet Union would involve weapons with a total yield of 5,000-10,000 Mt with individual
- 68 weapons having yields up to 20 Mt (Pittock et al., 1986), where a yield of 1 Mt is the energy
- 69 equivalent of a million tons of TNT. Because of a series of arms control treaties, the inventories
- 70 of the US and Russia have declined since. Under the New START Treaty, the US and Russia
- now have a combined total of approximately 6,000 strategic weapons with a total yield of about

1,500 Mt and a maximum weapon yield of about 800 kt (Kristensen & Korda, 2020a; Kristensen

73 & Korda, 2020b).

74 In the 1970s, it was realized that the fireball from a nuclear weapon explosion would heat 75 the atmosphere producing nitrogen oxides (NO_x) (Bonner, 1971) and that NO_x in the stratosphere 76 could catalytically destroy ozone (O₃) (Crutzen, 1970). The amount of NO_x produced and the 77 altitude to which it is injected depends on the amount of heating in the fireball, which in turn 78 depends on the yield of the weapon (Foley & Rudderman, 1973). Weapons over about 800 kt 79 would have fireballs energetic enough to directly inject NO_x into the stratosphere (Chang & 80 Wuebbles, 1984). Photochemical simulations including NO_x injections from these large 81 inventories with high-yield weapons showed Northern Hemisphere (NH) O₃ losses varying from 82 about 5 to 60% (e.g. Crutzen & Birks, 1982; Turco et al., 1983, Chang & Wuebbles, 1984, 83 Pittock et al. 1986). In the 1980s, it was realized that smoke from the fires could help loft fireball NO_x into the stratosphere and that heating of the stratosphere by the smoke could affect chemical 84 85 reaction rates (Turco et al., 1983), but these effects were not included in those early simulations. 86 Kao et al. (1990) demonstrated that heating of the stratosphere added to the NO_x injection would 87 cause a much larger O_3 loss than the NO_x injection alone, but they were only able to run their simulation for 20 days. Recently Mills et al. (2008, 2014) and others (Stenke et al, 2013; 88 89 Wagman et al. 2020) showed large O_3 loss from a regional nuclear war without including any 90 direct injection of NO_x . The O_3 loss was driven entirely by the heating of the stratosphere by the 91 smoke and the temperature sensitivity of O₃ chemical reactions. 92 O₃ in the stratosphere is a strong absorber of ultraviolet (UV) radiation, which would 93 otherwise be harmful to life at the surface. While sunlight in the UV-C range (200-280 nm) is 94 almost completely absorbed, sunlight in the UV-B (280-300 nm) and UV-A (320-400 nm) ranges 95 does reach the surface. In humans, high levels of UV-B can cause sunburn, photoaging, skin 96 cancer, and cataracts (MacKie, 2000). Photoaging is the premature aging of the skin from 97 exposure to sunlight and can cause pigmentation spots, spider veins, loss of skin tone, and 98 wrinkles. It is also associated with higher skin cancer risks. The World Health Organization uses 99 the UV Index (WHO, 2002) as a metric to inform people about surface UV levels. The scale for 100 UV Index is: low (1-2), moderate (3-5), high (6-7), very high (8-10), and extreme (11+). At the extreme level, unprotected skin can burn within minutes; however, sensitivity does depend on 101 102 the individual. Some places like the Andes typically have UV Index levels that are considered 103 extreme, but the population is mostly dark-skinned and thus not as sensitive to UV. Because of

104 this a threshold of 16 for extreme has been recommended for this region (Para et al., 2019). At 105 highest risk are people with skin phenotype (SPT) I and II, light skinned people who do not tan 106 easily, residing in areas of high UV as is common in Australia (Molina et al., 2000). Melanomas 107 are the most lethal form of skin cancer representing 4% of cases but half of all deaths, and 60-108 70% of melanoma cases are thought to be triggered by UV exposure (Sample & He, 2018). 109 Following a regional nuclear war, Mills et al. (2014) found that UV Index increases by 3-6 in the 110 summer at mid-latitudes resulting in UV Index values of 12-21, which are above the existing 111 scales.

Recent simulations of the climate effects of a global nuclear war between the US and Russia either did not include interactive chemistry (Robock et al., 2007b) or did not report the O₃ loss or surface UV (Coupe et al., 2019). Coupe et al. (2019) did not report their O₃ loss or surface UV because the actinic fluxes used for the photochemistry did not include the attenuation by the smoke. The study in the present paper interactively calculates the actinic flux in the presence of smoke and thus fixes this problem. 118 In this study, we repeat previous simulations of a regional nuclear war between India and 119 Pakistan producing 5 Tg of soot (Robock et al., 2007a; Mills et al. 2008, 2014; Stenke et al., 2013; Toon et al., 2019, Wagman et al., 2020) and a global nuclear war between the US and 120 121 Russia producing 150 Tg of soot (Robock et al., 2007b; Coupe et al., 2019). Our simulations are 122 focused on the changes to O₃ and surface UV using an Earth System model with interactive 123 chemistry and a sophisticated treatment of soot particles that includes the effects of aerosols on 124 photolysis. We include a series of sensitivity tests with varying amounts of NO_x injection, both with and without including aerosols in the calculation of actinic flux, to better characterize the 125 126 importance of smoke, NO_x and these processes on determining the effects of nuclear war on 127 ozone.

- 128 Both of these cases were originally defined based upon older weapons inventories. The 129 weapons inventories held by India and Pakistan have likely increased in both quantity and yield, making the 5 Tg estimate on the conservative side of what might actually occur (Toon et al., 130 131 2019). The US and Russia have decreased the number and size of their weapons, so the 150 Tg 132 estimate is likely an overestimate for a counterforce war with targets in just those two countries, 133 but Toon et al. (2008) estimated 180 Tg or more was possible if these countries included targets 134 in Europe and Asia. The exact amount of soot injected depends on details of the weapons used 135 and the specific targets, so it is highly uncertain and an area of active research. Using these 136 previously defined cases allows us to explore the range of possible values and to more easily 137 compare with prior results.
- We compare the resulting O₃ losses to those found in prior studies, perform sensitivity tests for including aerosols in photolysis and various NO emissions, and discuss the implications of the O₃ and UV changes on the biota. For the first time, these simulations have an in-line calculation for the actinic flux that includes aerosols, which provides improved photolysis rates, and high spectral resolution fluxes allowing the output of spectral action functions, weighted integrals over specific wavelength ranges, for biologically important processes. Section 2 describes the model setup, Section 3 presents the results and discusses UV impacts, and Section
- 145 4 discusses the implications of these results.

146 **2 Methods**

147 2.1 WACCM4 Model

148 We use the Community Earth System Model (CESM) (Hurrell et al., 2013), a chemistry climate

- 149 model with interactive atmosphere, land, ocean, and sea ice. The ocean model simulates the
- evolution of physical and biogeochemical parameters at 1° horizontal resolution with 60 vertical
- 151 layers of varying depth by coupling the Parallel Ocean Program (POP) version 2 ocean physical
- model (Danabasoglu et al., 2012) with the Biogeochemical Elemental Cycling ocean
- 153 biogeochemical model (Lindsay et al., 2014). The land model is the Community Land Model
- 154 (CLM) version 4 with a carbon-nitrogen cycle and simulates the evolution of the land physical
- state, characteristics of the land surface, exchanges of energy and material with the atmosphere,
- and run-off into the ocean. It has a horizontal resolution of $1.9^{\circ}x2.5^{\circ}$ with 15 vertical layers for
- 157 the land and 10 vertical layers for lakes. (Oleson et al., 2010; Bonan et al., 2011).
- 158The atmospheric model is the Whole Atmosphere Community Climate Model version 4
- 159 (WACCM4) (Marsh et al., 2013) which has a model top around 140 km, a horizontal resolution
- 160 of 1.9°x2.5°, 66 vertical levels, and includes interactive chemistry. The chemistry mechanism
- 161 used in this study includes a detailed representation of the middle atmosphere, with a
- 162 sophisticated suite of gas-phase and heterogeneous chemistry reactions including the odd-oxygen

163 (O_x) , odd-nitrogen (NO_x) , odd-hydrogen (HO_x) , odd-chlorine (ClO_x) , and odd-bromine (BrO_x)

- reaction families. WACCM4 has been used successfully in numerous studies of stratospheric O₃
- 165 chemistry (e.g., Solomon et al., 2015; Solomon et al., 2016; Polvani et al., 2017; Randel et al.,
- 166 2017; Stone et al., 2019; Froidevaux et al., 2019). Vertical resolution in WACCM4 is about 1.25
- 167 km in the troposphere and stratosphere and 2.5 km in the mesosphere. To stabilize the model
- against the large soot perturbations, the changes described in Bardeen et al. (2017), including the
- addition of the Rapid Radiative Transfer Model for GCMs (RRTMG) (Iacono et al., 2000), are
- 170 used for all these simulations.
- 171 All simulations are free running and external forcings, other than soot and NO_x , are
- 172 configured for repeated year 2000 conditions. A control simulation was run for 19 years and each
 173 war simulation starts at year 4 of the control and lasts for 15 years allowing most simulations to
- 174 return to steady state.
- 175 2.2 War Scenarios
- 176 In this study, two types of wars are simulated: a regional nuclear war between India and
- 177 Pakistan, and a global nuclear war between the United States and Russia. The regional war
- 178 follows the 5 Tg of soot scenario described in Toon et al. (2019) using 44 15 kt-weapons with
- 179 urban targets in a war lasting 3 days and is similar to that used by Mills et al. (2008, 2014).
- 180 Smoke is emitted instantaneously when a target is attacked. The global war follows Coupe et al.
- 181 (2019) and is similar to that used by Robock et al. (2007b). It assumes that 150 Tg of soot is
- 182 emitted uniformly over the continental US and Russia. The war lasts 7 days and the smoke
- 183 emissions ramp down linearly during that time. As was done in previous studies, these wars are
- assumed to start on May 15th of the first year. Coupe et al. (2019) and Robock et al. (2007b) did
- 185 not include weapon counts or yields needed for determining NO_x emissions, so we have
- 186 performed sensitivity tests with weapons inventories from: Toon et al. (2008), Kristensen &
- 187 Korda (2020a, 2020b), and NRC (1985).

188 2.3 Soot Treatment and Emissions

- 189 Soot aerosol is treated using the Community Aerosol and Radiation Model for Atmospheres
- 190 (CARMA) (Toon et al., 1988; Bardeen et al., 2006), a sectional microphysics model that allows
- aerosols and the processes that affect them, including coagulation and sedimentation, to be
- treated in a size-resolved manner. The CARMA treatment for soot follows Bardeen et al. (2017)
- and has been used in previous studies of smoke from nuclear war (Coupe et al., 2019; Toon et
 al., 2019). Rather than treating the soot as one large spherical particle of pure black carbon (BC),
- as is often done in aerosol models, we treat the soot as fractal particles, clusters of smaller
- spheres of a uniform size (monomers) whose spatial arrangement is described by its fractal
- 197 properties. We assume a monomer radius of 30 nm, a fractal dimension of 2.2 and a packing
- 198 coefficient of 1, which creates a particle whose overall shape is more like a sheet than a sphere.
- 199 This affects the fall velocity, coagulation rate, and optical properties of the soot. The particles are
- 200 emitted in a log-normal size distribution with a mean of the radius of a sphere with equivalent
- 201 mass of $0.11 \,\mu\text{m}$ and a width of 1.6. All soot is emitted in the vertical as a constant mixing ratio
- 202 between 150 and 300 hPa.
- 203 2.4 NOx Emissions
- NO_x is a shorthand for the combination of nitric oxide (NO) and nitrogen dioxide (NO₂). It can
- 205 be created by the heat from the fireball of the nuclear explosion as well as being emitted from the
- surface fires cause by the explosion. NO_x in the model is emitted only as NO. There is a rapid
- 207 cycling between NO and NO₂ driven by NO₂ photolysis during the day and rapid reformation of

208 NO₂ at night, so the choice of the NO_x species to use for emissions is not important (Brasseur & 209 Solomon, 2005).

210 2.4.1 Fireball NO_x

211 The amount of NO created in the fireball is proportional to the yield of the weapon. These

simulations assume a rate of 1×10^{32} molecules of NO injected per Mt of yield (NAS, 1985). The 212

- 213 emissions are spread uniformly between the top and bottom of the fireball. The fireball height is
- 214 a function of yield determined using equations adapted from Chang et al. (1979) that fit results 215 from Foley & Rudderman (1973):
- 216

217

$$Z_{top} = 21.64Y^{0.2} + (Z_{trop} - Z_{ref})$$
(1)

(2)

 $Z_{bot} = 13.41Y^{0.2} + (Z_{trop} - Z_{ref})$ (2) where Z_{trop} is the tropopause height at the target (km), Y is the yield of the weapon (Mt), Z_{ref} is a 218 219 reference height of the tropopause (17 km), and Ztop (km) and Zbot (km) are the heights of the top and bottom of the fireball. These equations have been adapted to account for the local tropopause 220 221 height as the data from Foley & Rudderman (1973) were based upon atmospheric tests that were 222 at low to mid-latitudes while a nuclear war between the US and Russia would occur mostly at 223 mid- to high latitudes where the tropopause is lower. Fireball NO is emitted at the time of the

224 weapon detonation. For the regional case the weapon use is spread out over 3 days (Toon et al.,

- 225 2019), but for the global war case all the NO is released at the start of the war. Since we do not
- 226 here have a detailed war plan for the global war case indicating when the weapons would be
- 227 used, we have assumed that all the weapons are used and thus all the fireball NO is released on
- 228 the first day.
- 229 2.4.2 Fire NO_x
- 230 NO can be emitted from fires as part of the combustion process. Wildfire data from Andreae
- 231 (2019) are used to estimate that ~ 2 g of NO is emitted per kg of fuel burned. Using the
- 232 assumption of 20 g of black carbon (BC) per kg of fuel (Toon et al., 2007) that was made to
- 233 determine the soot emissions for the war scenarios, gives us a ratio of 0.1 g NO per g BC. NO is
- 234 assumed to be emitted at the same location and with the same timing as the soot emissions.
- 235 2.5 In-line Photolysis
- 236 In WACCM4, photolysis was previously calculated by first using a lookup table (LUT) to
- 237 calculate the actinic flux in 100 wavelength bands between 121 and 750 nm and then by applying
- 238 the cross-section and quantum yield for each photolysis reaction adjusted for the local
- 239 temperature and pressure (Kinnison et al., 2007). Thus the cross-sections and quantum yields
- 240 were affected by heating caused by solar absorption from aerosols, but photolysis rates did not
- 241 include the direct effects on actinic flux from aerosol scattering and absorption. This approach
- 242 was used by Mills et al. (2008, 2014), Toon et al. (2019), and Coupe et al. (2019). For this study
- 243 we added the Tropospheric Ultraviolet and Visible (TUV) model (Madronich & Flocke, 1997)
- 244 version 4.2 to provide an in-line calculation of the actinic flux.
- 245 The older calculation of the photolysis coefficients is divided into two regions: (1) 120 246
- nm to 200 nm (33 wavelength intervals); and (2) 200 nm to 750 nm (67 wavelength intervals). 247 The total photolytic rate constant for each absorbing species was derived during model execution
- 248 by integrating the product of the wavelength dependent top of the atmosphere flux; the
- 249 atmospheric transmission function (or normalized actinic flux), the molecular absorption cross-
- 250 section, and the quantum yield. The top of the atmosphere flux over these wavelength intervals
- 251 can be specified from observations and varied over the 11-year solar sunspot cycle. The
- 252 wavelength-dependent transmission function was derived as a function of the model abundance

of O₃ and molecular oxygen. For wavelengths beyond 200 nm a flux LUT approach was used,
based on the 4-stream off-line version of TUV. The transmission function was interpolated from
the LUT as a function of altitude, column O₃, surface albedo, and zenith angle. The LUT

assumed a mid-latitude O_3 profile and scaled the result based upon the total column O_3 . This older approach did not include the effects of aerosols or sulfur dioxide (SO₂).

258 The temperature and pressure dependences of the molecular cross sections and quantum 259 vields for each photolytic process were also represented by another LUT in these wavelength regions. At wavelengths less than 200 nm, the wavelength-dependent cross section and quantum 260 yields for each species are specified and the transmission function is calculated explicitly for 261 262 each wavelength interval. There are two exceptions to this approach. In the case of the 263 photolysis rates for NO and molecular oxygen (O_2) , detailed photolysis parameterizations are 264 included in-line. In the Schumann-Runge band region, the parameterization of NO photolysis in the δ -bands is based on Minschwaner & Siskind (1993). This parameterization includes the 265 effect of self-absorption and subsequent attenuation of atmospheric transmission by the model-266 267 derived NO concentration. For O₂ photolysis, the Schumann-Runge band and Lyman-alpha parameterizations are based on Koppers & Murtagh (1996) and Chabrillat & Kockarts (1997, 268 269 1998), respectively.

For the current study we added the TUV model to provide an in-line calculation of the actinic flux. The in-line version differs from the LUT approach discussed above in that a 2stream version of TUV was used for computational efficiency. For consistency with prior work, the same band structure, cross-sections, and quantum yields are used. Using in-line TUV allows

- 274 the actual O_3 , SO_2 , and aerosol profiles in the column to be included in the calculation. For the
- aerosols, the optical properties calculated for the coarser band structure used by the radiative
 transfer code have been interpolated onto the wavelength grid used by TUV. TUV is also able to
- output spectral integrals of useful radiative fluxes and biologically important action spectra such
- as UV-B and photosynthetically available radiation (PAR, 400-700 nm) that can aid in
- understanding the effects on the biota of the changes to O_3 and to the aerosols.
- 280 3 Results and Discussion
- 281 We first describe changes to O₃ and surface UV from a regional nuclear war compared to results
- from Mills et al. (2008, 2014) and then evaluate O_3 depletion following a global nuclear war
- compared to results from the 1980s (Pittlock et. al, 1986; Kao et al., 1990). All the WACCM4
- simulations made for this study are summarized in Table 1. The notation has the name of the
- potential conflict (e.g., IP is India-Pakistan) and the base cases include TUV with aerosols
- affecting photolysis and NO emission from the fireball and the fires. A minus sign (–) indicates
- that a process is not included (e.g., IP–AOD means the India-Pakistan case, but not accounting
- for the effects of aerosols, using AOD to mean aerosol optical depth, in the chemistry
- 289 calculations) and a plus sign (+) indicates that alternate fireball NO emissions were used.
- 290 Climate effects other than those related to O₃ and surface UV have already been described in
- 291 Coupe et al. (2019) for the global war case and Toon et al. (2019) for several regional war cases.
- Our climate results are similar to these and thus will not be discussed here; however, we do show
- the evolution of the global (Figs. 1, S1) and polar (Figs. S2, S3) average vertical profile changes
- for soot, temperature, H_2O , HO_x , NO_x , and O_3 from our regional and global war cases.
- 295 3.1 Regional Nuclear War
- 296 Mills et al. (2008, 2014) assumed that a nuclear conflict involving India and Pakistan generated 5
- 297 Tg of soot with the soot spread uniformly over the region. Mills et al. (2008) used WACCM3
- and Mills et al. (2014) used WACCM4 with a fixed size of 100 nm for the soot to simulate the

299 resulting climate effects. Toon et al. (2019) recreated a similar case; however, they used 300 emissions from a specific set of targets and weapon yields, a fractal representation for the soot, and the soot was allowed to coagulate. This study repeats Toon et al. (2019) but includes in-line 301 302 photolysis from TUV as well as NO_x emissions from the fireballs and the fires. Figure 2 shows 303 the evolution of the change in the monthly global average O₃ column for several cases compared 304 to their control cases. All these cases show a peak O_3 loss of ~25% 2-3 years after the war, but 305 the fractal soot cases (IP, IP–NO, IP–AOD, and Toon et al., (2019)) all show a faster recovery of 306 12 years than Mills et al. (2008, 2014), because the soot particles grow larger (> 500 nm) and 307 thus have a shorter lifetime in the stratosphere. Mills et al. (2008) showed faster O₃ recovery than 308 that in Mills et al. (2014) because Mills et al. (2008) used specified sea surface temperatures and 309 had a horizontal resolution of 4° latitude by 5° longitude, which led to a faster stratospheric 310 circulation and shorter soot lifetime than Mills et al. (2014), which has an interactive ocean and a 311 horizontal resolution of 1.9° latitude by 2.5° longitude (Mills et. al, 2014). Including NO 312 emissions (IP versus IP-NO) does slightly increase O₃ loss, mostly by slowing the recovery, but

the effects are minor because the amount of NO injected is small and the temperature effects are so strong. Including aerosols in the photolysis calculations (IP versus IP–AOD) slightly decreases the amount of O₃ loss, but the effect is not significant.

316 Mills et al. (2008, 2014) did not include NO_x emissions and found that O₃ loss was 317 primarily driven by heating of the stratosphere and temperature dependencies in ozone 318 decomposition and NO_x catalytic losses along with additional N_2O transport and longer NO_x 319 lifetimes. Figure 3 shows the global average total column odd-oxygen chemical reaction rates for 320 the IP (Fig. 3b), IP-AOD (Fig. 3a), and IP-NO (Fig. 3c) cases as solid lines. The individual 321 chemical reactions that are important to odd-oxygen chemistry and their contribution to these 322 subtotals (Brasseur & Solomon, 2005) are shown in Table 2. Mills et al. (2008) did not output all 323 of these individual rates, but their results should be similar to the IP-AOD case. Both cases show 324 an increase in O production, which is primarily because of a temperature sensitivity in the 325 photolysis cross section for the $O_2 + hv \rightarrow 2O$ reaction. The IP case has lower overall reaction 326 rates, because photolysis is reduced compared to the IP-AOD case by the reduced actinic flux caused by solar absorption from the soot. However, the total production and loss rates are greater 327 328 than the control, indicating that the temperature effects are greater than the effects caused by the 329 reduced actinic flux. There is a small increase in NOx loss in the IP case compared to the IP-NO 330 case, but the effects of the NO injection are negligible.

331 Looking at these rates relative to the total production of odd-oxygen (Figure 4) shows 332 that HO_x, ClO_x, and BrO_x catalytic cycles decrease relative to the control while the NO_x catalytic 333 cycle and O_3 decomposition (O_x) increase. The magnitude of these changes is less when the 334 aerosols are included in the photolysis calculation. Though HO_x remains the dominant odd-335 oxygen loss mechanism, as reported by Mills et al. (2008), increases in NO_x and O_x are the drivers of increased O₃ loss following a nuclear conflict. The absolute contribution from HO_x 336 337 stays fairly constant leading to a decreased relative contribution as odd-oxygen production 338 increases. We also see both an absolute and relative decrease in the ClO_x and BrO_x catalytic 339 cycles that was not seen by Mills et al. (2008). The O_x, ClO_x, and BrO_x changes last about 2 340 years while NO_x changes last about 6 years. HO_x changes switch from relative decrease for 6 vears to a slight relative increase for another 4 years. 341

Figure 5 shows the vertical profile of the global average chemical rates and O₃ profile for the first year after the war for the IP–AOD and IP cases and the corresponding change in temperature is shown in Figure S4. The O₃ profiles are similar for the IP–AOD and IP cases with reductions in the stratosphere and upper mesosphere. The IP–AOD case shows about a 20%

346 increase in O production relative to the control in the stratosphere, but the IP case shows little

347 change in O production relative to the control case when aerosols affect the actinic flux.

348 Similarly, there is a large increase relative to the control in odd-oxygen loss from NO_x and O_x in

349 the IP–AOD case in the stratosphere that is smaller in the IP case. In both cases, HO_x driven loss

is increased in the stratosphere relative to the control, but reduced in the lower mesosphere and is nearly identical to the control in the troposphere and upper mesosphere contributing to the small

351 incarly identical to the control in the troposphere and upper mesosphere in HO_x loss rates seen in the total column (Figure 3).

Figure 6a shows the evolution of the change in zonal average column O₃ for the IP case, while absolute amounts are shown in Figure S5b. Similar to Mills et al. (2014), we see a greater percentage loss, up to 60%, at higher latitudes and losses on the order of 10% in the equatorial tropics. As seen in the global average (Fig. 2), the O₃ recovers faster in our simulation than in Mills et al. (2014) because the soot particles get larger and have a shorter lifetime.

358 These O₃ changes cause an increase in the surface UV Index (Fig. 7b). The values shown 359 are the monthly average of the daily maximum clear-sky values calculated by TUV. The daily 360 maximum value should be close to the noon local time value that is commonly reported for UV 361 Index; however, observed values at exactly noon local time or averaged over 30 minutes around 362 noon local time may be lower than the maximum daily value by several points (Para et al., 2019). 363 Compared to the control run (Figs. 7a, S6a), there is an increased surface UV immediately 364 following the war lasting for about 6 years. Since surface UV is a function of the Sun, aerosols, clouds, and O₃, the areas of the largest O₃ depletion do not necessarily show the largest increases 365 in surface UV. Values of 20 or larger are found between 30°S to 20°N the control case, but 366 367 extend to 40°S to 40°N in the IP case. The largest increases in the IP case occur in the southern 368 polar region around 70°S, which is far removed from where the smoke was injected. The

369 consequences of these changes in surface UV on the biota will be discussed in section 3.3.

370 3.2 Global Nuclear War

Coupe et al. (2019) used WACCM4 to simulate the effects of a nuclear war between the United

372 States and Russia using a scenario similar to Robock et al. (2007b), but with the same soot

representation used in Toon et al. (2019). This study repeats Coupe et al. (2019) but includes in-

374 line photolysis from TUV and NO_x emissions from the fireballs and the fires. Figure 8 shows the

evolution of the change in column O_3 for the NH (Fig. 8a) and globally (Fig. 8b). Connell &

Wuebbles (shown in Pittock et al., 1986) used a photochemical model to assess O₃ changes from 377 3 different weapons inventories from AMIBIO (1982), Knox (1983), and NRC (1985) but only

included effects from the fireball NO_x with no heating caused by the soot. The largest reduction

they found was about 45% using the Knox (1983) inventory and the losses lasted about 10 years.

380 Kao et al. (1990) included the effects of the soot and the fireball NO_x, but only in a short

simulation that lasted 20 days. They used the NRC (1985) inventory, but achieved much larger
loss of 15% in 20 days than Connell & Wuebbles did with the same inventory.

383 Coupe et al. (2019) showed that the stratospheric heating in the global war case is

384 stronger than for a regional war and we show this heating results in O₃ losses of up to 80% in the

NH with losses lasting for 15 years. The difference in O_3 loss between our extreme cases for NO_x emissions (UR–NO and UR+NO3) is up to 15% in the NH and 10% globally. Removing the

387 aerosols from the actinic flux calculation (UR–AOD) results in less O₃ loss by over 20% in the

388 NH and 15% globally. The cause for this lower loss of O₃ can be seen in the global average

chemical reaction rates (Fig. 9). The heating alone causes an almost 3 times increase in reaction

rates (Fig. 9a); however, when the aerosols are included in the actinic flux (Fig. 9b), the reaction

- 391 rates drop to less than a quarter of the unperturbed rates and more than compensate for the
- heating induced increase. Thus unlike the IP case, the reduction in actinic flux has a bigger
- 393 impact on O₃ than the temperature changes in the UR case as the total production and loss are
- much lower than seen in the control. Including the NO injection (Fig. 9c) causes a small increase
- in the NO_x loss for the first few years. The UR–AOD (Fig. 10a) and UR (Fig. 10b) cases show
- 396 very different relative reaction rates. UR–AOD suggests that the HO_x cycle decreases initially 397 and that the NO_x cycle and O_x decomposition drive the O₃ loss, similar to what was seen in the
- 398 IP case. However, in the UR case, the HO_x cycle actually increases for 15 years, while the NO_x
- 399 cycle increases for 6 years and then decreases for 9. O_x decomposition decreases slightly and
- 400 ClO_x and BrO_x cycles decrease for 10 years. O₃ production in the troposphere takes on a more 401 important role as O production is greatly diminished.
- The global average vertical profile for the first year also shows significant differences between the UR–AOD (Fig. 11a) and UR (Fig. 11b) cases. In the UR case, all reaction rates in the stratosphere are significantly reduced compared to UR–AOD. Both simulations show a greatly reduced stratospheric O₃ layer, with UR having a larger peak in the mesosphere than UR– AOD, where O production by photolysis is largely unaffected.
- Figure 6b shows the evolution of the zonal average change in the O₃ column for the UR
 case, while absolute amounts are in Figure S5c. Losses at mid to high latitude exceed 75% and
 over 65% in the tropics during the first few years. Recovery in the tropics is faster than at high
- 410 latitude, but still takes 10 years to recover to within 5% of the control case. Initially, the soot
- 411 compensates for the O₃ loss and UV is actually reduced at the surface (Figs. 7c, S6b) compared
- 412 to the control (Fig. 7a). However, the UV Index begins to increase relative to the control after 3
- 413 years as the soot clears reaching a peak 8 to 9 years after the war. UV Index values of 35 are seen
- 414 in the tropics from years 5 to 8, and are greater than 45 during the summer in the southern polar
- 415 regions from years 5 to 8. Maps of UV Index for selected months averaged over years 8 to 10
- show a generally zonal structure but with regions of extreme UV including higher altitudes and
- 417 deserts (Fig. S8).

418 3.3 Effects of Surface UV Changes

- Soot injected into the atmosphere absorbs sunlight heating the stratosphere and reducing the 419 420 solar radiation at the surface causing lower surface temperatures and reduced precipitation. The 421 heating in the stratosphere causes increased O₃ destruction that allows more UV radiation to the 422 surface. Effects from these climatic changes for regional wars have been discussed in Robock et 423 al. (2007a), Stenke et al. (2013), Mills et al. (2014), and Toon et al. (2019). Jägermeyr et al. 424 (2020) found large agricultural changes and Scherrer et al. (2020) predicted significant effects on 425 fisheries in response to climatic changes from a regional nuclear war. Scherrer et al. (2020) also 426 found larger effects for the global war case. However, neither study included the effects of O_3 or 427 UV changes, which would likely lead to additional losses especially related to global grain 428 productivity. The effects of a global nuclear war were studied in the 1980s (e.g., Crutzen & Birks 429 (1982), Turco et al. (1983), Harwell et al. (1986)), but these studies did not include the effects of 430 stratospheric heating on O₃ loss. Robock et al. (2007) and Coupe et al. (2019) revisited these 431 global war studies, but did not include estimates of changes to stratospheric O₃ or surface UV. 432 Figure 12 shows the evolution of global average aerosol optical depth (AOD), surface
- 432 reperture, precipitation, O₃, PAR, UV-A, and UV-B for the IP and UR cases. In both cases,
- PAR and UV-A is reduced by the presence of the aerosol, but UV-B responds differently in the
- 434 FAR and UV-A is reduced by the presence of the aerosol, but UV-B responds differently in the 435 two cases. The major absorber for UV-B is O₃, so in the IP case (Fig. 12a), which added a
- 436 smaller amount of soot but still has significantly lowered O₃, UV-B increases by up to 10% in

437 proportion to the O₃ destruction. For the UR case (Fig. 12b), so much soot is injected that even 438 though the O₃ loss is greater than in the IP case, there is still a dramatic reduction in UV-B, UV-439 A, and PAR. However, in the UR case, the soot is removed faster than the O₃ recovery, so after 7 440 years there is a net 20% increase in UV-B. UV-B remains elevated for 8 more years, while PAR 441 recovers after 10 years. Photosynthetic organisms in terrestrial and marine environments are 442 sensitive to the ratios of UV-B, UV-A, and PAR (Krizek, 2004). High levels of UV-B can 443 contribute to inhibition of photolysis affecting photosystem II (Hakala-Yatkin et al., 2010, Ragni 444 et al., 2008, Kataria et al., 2014) reducing leaf expansion (e.g. Searles et al., 2001) and plant growth rate (Allen et al. 1998; Ballaré et al. 2001; Ballaré et al. 2011). However, some UV-B 445 446 damage may be offset by supplemental PAR and UV-A radiation involved in the repair process 447 (Krizek, 2004) with plants adapting to higher UV-B by producing compounds such as plant 448 pigmentation which might improve the quality of certain crops (Bassman, 2004; Bornman et al., 449 2015). Unfortunately, in our nuclear war scenarios we see increased UV-B along with decreased 450 UV-A and PAR, suggesting photosystem II damage and a simultaneous slowing of repair 451 processes. In the IP case, PAR, UV-A, and UV-B return to within a few percent of normal levels at the same time; however, in the UR case UV-A and PAR return to within a few percent of 452 453 normal levels 5 years before UV-B which may offset some of the potential damage.

454 UV can have both beneficial and harmful effects on humans. UV-B is needed for vitamin 455 D synthesis and UV-A helps prevent seasonal affective disorder, a type of depression associated 456 with the change of seasons (MacKie, 2000). Vitamin D deficiency can lead to rickets, a disease that can cause weak or soft bones in children and a similar disease called osteomalacia in the 457 458 elderly. Because of this, vitamin D is often added as a food supplement, for example, to milk. 459 Vitamin D deficiency has also been linked to chronic diseases like cardiovascular disease and 460 cancer, and it is especially prevalent in minority groups, likely due to a combination of genetic 461 (pigmentation) and socioeconomic factors (Forrest and Struhldreher, 2011). On the other hand, there are several serious harmful effects of increased UV-B including sunburn, photoaging, skin 462 463 cancer, and cataracts (MacKie, 2000). People with either a genetic or drug-induced sensitivity to 464 UV or who are genetically unable to repair DNA damage may be particularly affected. UV-C 465 (200-280 nm) is readily absorbed by O₃ and thus does not reach the surface and is generally not considered in terms of effects on human health (MacKie, 2000). 466

467 To better understand the role of UV radiation on biota, various action spectra have been identified that integrate a portion of the spectrum that is important for particular biological 468 469 processes. Figure 13 shows examples of changes in the action spectra calculated by TUV for 470 plant growth (Flint, 2003), phytoplankton inhibition (Boucher, 1994), vitamin-D synthesis (Bouillon et al., 2006), cataract formation (Oriowo, 2001), solar induced erythema (Anders, 471 472 1995), and DNA damage (Setlow, 1974) along with the UV-B spectrum for the IP and UR cases. 473 The changes in the action spectra generally follow the shape of UV-B, but the magnitudes may 474 be different. For the IP case (Fig. 13a) these are generally positive for 10 years following the 475 war. Plant growth, which refers to the height of light-grown oat seedlings, is the only one that experiences a negative change which persists for 2 years. The largest increase of up to 40% is for 476 477 DNA damage, a proxy for forming human skin cancers. The next largest is erythema (sunburn) 478 which increases at up to 28% closely followed by vitamin-D synthesis. Cataract damage, the 479 formation of cloudy spots on pig eye lenses, closely matches the changes in UV-B, with up to a 480 19% increase. Finally, UV inhibition of phytoplankton carbon absorption, increases by up to 481 11%. The UR case (Fig. 13b) shows the same general relationship between the magnitudes of the 482 different action spectra. However, each action spectra first experiences negative changes with a

90% reduction and then becomes positive after a period of time. Vitamin-D synthesis is negative
for the first 4 years, indicating a brief period of extreme risk for rickets. The DNA damage
metric becomes positive within a year and reaches a peak increase of 140%. The others show
positive values after 4 to 7 years, with peak increases between 10 and 60%.

487 UV that could cause DNA damage increases rapidly in the UR case because of the shapes 488 of the O₃ and soot absorption spectra. Figure 14a shows the O₃ absorption cross-section (Gorshelev et al., 2014) and the soot mass absorption coefficient for 0.16 µm particles in the 489 490 range of 230 to 370 nm, both normalized by the maximum value within that range. O₃ has a very 491 strong peak around 255 nm, but does absorb across this entire wavelength range. Soot absorption 492 decreases by 40% from 230 to 370 nm. Figure 14 shows the evolution of changes in UV-B and 493 action spectra related to human skin cancer: erythema, DNA damage, and skin cancer (de Gruijl 494 & Van der Leun, 1994; CIE 2006) for the IP and UR cases. In the control (not shown) O₃ 495 absorbs essentially all of the UV-C and a large fraction of the UV-B light. In the IP case (Fig. 496 14b), the moderate loss of O₃ causes an increase in surface UV. In the UR case (Fig. 14c), even 497 though there is a much larger loss of O₃, there is a decrease in surface UV-B because of the large 498 UV absorption by the soot. As the soot is removed, the surface UV increases gradually over 8 499 years. UV-B is larger compared to the control starting 4 to 7 years after the war, depending on 500 the exact spectral weights of the action function. However, lost O₃ absorption in the UV-C range, 501 that could cause DNA damage and where O_3 is most absorbing, is not compensated for by the 502 increased but relatively weaker soot absorption. This leads to the rapid and large increase of 503 DNA damaging UV in the UR case; however, this increase is from very low levels, so UV-C is 504 still probably not a health threat for either nuclear war case. UV-C changes may need to be 505 accounted for when O_3 loss is larger, for example following fires generated by an asteroid impact 506 at the K-Pg boundary (Bardeen et al., 2017).

507 While CESM is capable of simulating the ocean physical, biogeochemical, and ecological 508 response to nuclear war (e.g., Lovenduski et al., 2020; Scherrer et al. 2020; Coupe et al., 2021), 509 the version of CESM used in this study does not include the effect of UV inhibition on 510 phytoplankton growth, which may affect each taxa differently (Jeffrey et al., 1999; Xu et al., 511 2016; De Tommasi et al., 2018). Diatoms, which are the dominant phytoplankton functional type 512 outside the oligotrophic subtropical gyres, have a silica shell that may provide UV protection, while in the gyres small phytoplankton dominate, which with the exception of coccolithophores 513 514 have no known UV protection. Future work will address this issue by including a representation 515 of UV inhibition in lower trophic levels. Changes in phytoplankton will in turn influence their 516 consumers, i.e. fish and other higher trophic-level organisms (Stock et al., 2017). In this way, the 517 war-driven changes in UV might have bottom-up effects on fish and fisheries that could be 518 estimated in future work. Experiments indicate that many shallow-living marine organisms and 519 freshwater fish are directly harmed by increasing levels of UV-A and UV-B (Llabrés et al., 2013; 520 Alves & Agustí, 2020). However, the direct effects of UV on the fish in the wild are poorly 521 known, leaving the impact of UV changes on fish and fisheries highly uncertain.

522 UV effects on plant growth are complex and currently not represented in most crop and 523 land surface models (Wargent and Jordan, 2013), but recent model development is trying to 524 address and quantify related processes. Surface O₃ also changes in these simulations (Fig. S7) 525 and can have severe impacts on plant growth and crop yields by decreasing plant photosynthesis 526 rates and stomatal conductance (Lombardozzi et al., 2012; Lombardozzi et al., 2013). Surface O₃ 527 is strongly affected by the Stratosphere-Troposphere-Exchange (STE) and tropospheric 528 photosphere to prove the stratosphere to provide the stratosphere to prove to prove to pr

528 photochemistry (Xia et al., 2017). While these simulations do show a significant change in

- 529 surface O₃ from STE, they do not include detailed tropospheric chemistry and thus cannot fully
- assess the changes to surface O₃. Impacts of surface O₃ on plants and crop yields are not
- 531 currently simulated by Earth System models or most process-based crop models (Schauberger et
- al. 2019); however, there are an increasing number of pilot studies including an O₃ damage
- function in land models (Lombardozzi et al., 2015) and crop models (Emberson et al. 2018).
- 534 CESM does not have a built-in capability to track effects upon human health, but the
- 535 availability of these action spectra could enable additional capabilities in the future. Humans if 536 properly informed and equipped have the ability to adapt to these changes by staving indoors,
- 536 properly informed and equipped have the ability to adapt to these changes by staying indoors, 537 only going out at night, or by covering up with clothes and sunscreens. Plants and animals are
- also sensitive to these changes. While ranchers may be able to make adaptations for livestock,
- wild populations of plants and animals will be exposed to the direct effects of these UV changes.

540 4 Conclusions

541 In this study, we conducted simulations of regional and global scale nuclear war impacts on

- 542 ozone using a modified version of WACCM4 that for the first time includes in-line calculations
- of actinic flux using TUV. This allows for the inclusion of aerosols in the photolysis calculation
- and for the diagnostic output of action spectra that can help quantify ecosystem and health
- 645 effects. We also included the emissions of NO from the fireball and the surface fires. For
- regional wars, we find that while including aerosols in the photolysis calculation does change
- 547 reactions rates, the effects of this and NO emissions does not make a significant change in the O_3
- 548 loss relative to earlier calculations that did not include them. This results in a peak global
- 549 average loss of \sim 25% with much larger losses of up to 55% at high latitudes. We find that the 550 HO_x catalytic cycle remains the largest loss mechanism, but is largely unchanged as found by
- 550 HO_x catalytic cycle remains the largest loss mechanism, but is largely unchanged as found by 551 Mills et al. (2008). Also, as reported by Mills et al. (2008), the increase in the NO_x cycle and O_x
- decomposition drive the increased O_3 loss. We see a reduction in loss from the ClO_x and BrO_x
- 553 cycles that was not reported previously.

554 For global nuclear war, we provide the first estimates of O₃ and surface UV from modern 555 climate models and find much greater O₃ loss than was predicted in the 1980s, with a peak global 556 average loss of 75%. While the weapon sizes and therefore NO emissions have decreased since 557 the 1980s, the inclusion of smoke and the heating of the stratosphere causes significant O₃ loss as 558 demonstrated by Kao et al. (1990) and shown for regional wars by Mills et al. (2008). For the 559 global war, the inclusion of aerosols in the photolysis calculations show large changes in 560 chemical reaction rates and 15% larger peak O₃ loss. We also show that reduced O production 561 and the HO_x cycle dominate the increased O₃ loss with a contribution from the NO_x cycle for the 562 first few years.

563 The warming of the higher polar latitudes (Figs. S2, S3, S4) in both the IP and UR cases greatly reduces the importance of the standard chlorine and bromine heterogeneous chemistry for 564 565 O₃ depletion. For the UR case, the zonal winds are significantly altered compared to the control and the overall stratospheric Brewer Dobson circulation changes affect the transport of odd-566 567 oxygen loss precursors into the polar lower stratosphere. Poleward and downward transport of NO_x is increased by more than 150% in the IP case and more than 400% in the UR case relative 568 569 to the control. Increased stratospheric H₂O from a warmer tropopause increases H₂O in the SH 570 polar region to values greater than 8 ppmv and 150 ppmv relative to the control in the IP and UR 571 cases respectively. Consequently HO_x is also increased by over 100% and 500% in these two 572 cases (Fig. S2). Air depleted of O_3 is also transported from the upper/middle stratosphere into the

573 lower polar stratosphere. Depletion of total column ozone (TCO) is much greater for the six

574 years after the IP event than in the control (Figs. 6, S5). This depletion is not limited to just the

575 September and October periods, but throughout the SH polar season due to increases in NO_x and

576 HO_x. The IP event would cause a short-term hiatus in the recovery of TCO (e.g., Solomon et al.,

577 2016) of approximately 10 years. TCO depletion from the UR case in the SH polar latitudes is

578 extreme, with a minimum TCO of less than 75 DU extending from the pole to 50°S for the first 579 six years following the war. This depletion is not only seen in the SH polar region, but also the

579 six years following the war. This depletion is no 580 NH polar region.

581 Surface UV is affected by both the O₃ loss, which increases surface UV, and the smoke 582 injection, which decreases surface UV. For the regional war, the smoke injection is small enough relative to the O₃ destruction that there is an increase in surface UV for the entire period 583 584 following the war that is proportional to the amount of the O_3 loss. For the global war, while the 585 O₃ loss is extremely large, the smoke injection is so big that it leads to reduced UV at the surface for the first few years following the war. The rate of O₃ recovery lags the smoke removal and 586 587 thus there are several years of very high surface UV, with a UV Index over 35 following a global 588 nuclear war. Thus, following a regional war the climatic changes and the surface UV changes 589 occur at the same time, shortening the period of large change and providing a uniform threat that 590 decreases with time. However, for the global war the climatic changes come first followed by the 591 elevated surface UV. This extends the period of changed conditions and causes the nature of the 592 change to switch from reduced light, surface temperature and precipitation to one of increased 593 surface UV. This means that adaptions that may have worked in first few years following the 594 global war may not work as the surface UV increases. Changes in spectral integrals show that 595 greatest increase is for DNA damage, which increases by up to 140% in the global war case and 596 there is a risk of insufficient vitamin-D during the first few years following a global war. 597 Assessments of impacts on agriculture and fisheries like those done by Jägermeyr et al. (2020) 598 and Scherrer et al. (2020) for a regional nuclear war need to include the effects of UV and 599 surface O₃ changes, and need to be done for the larger and differently sequenced effects that are 600 generated by a global nuclear war.

601 A new version of the WACCM model has been released (Gettelman et al., 2019) with 602 improved atmospheric physics including interactive modal aerosols, stratospheric aerosols, a new boundary layer and shallow convection scheme, a new cloud scheme that includes the aerosol 603 604 indirect effect, and improved biogeochemistry in the land and the ocean models. Initial 605 experiments with this model have shown it to be unstable when presented with the large 606 perturbation considered here, so further work is needed before using it for nuclear war 607 simulations. We also plan to explore the addition of organic and sulfate coatings on the soot. Yu 608 et al. (2019, 2020) have shown that wildfire smoke lofted into the stratosphere is coated with 609 large amounts of organics. They also speculated on whether it might become coated in sulfate 610 and contribute surface area to heterogenous chemistry. Because of the different fuels, urban fires 611 (as in a nuclear war scenario) would likely have less of an organic coating than wildfire smoke, but such a coating would provide a "lensing" effect that increases the particle absorption (Bond 612 et al., 2006) and the larger particles would likely have a shorter lifetime in the stratosphere. This 613 614 could affect both the intensity and the duration of climatic changes following a nuclear war. 615 The ability to calculate and output spectral action functions allows for a better assessment

of the threats posed to the biota by a nuclear war. It also opens up the possibility for these data to be coupled with surface models allowing the effects of the detailed changes in surface light spectra to be included in parameterizations of the effects on the biota including those for

619 phytoplankton and vegetation. There is a need for parameterizations of biological effects to be

- 620 developed for the large UV increases that are predicted here and for climatic conditions that are
- 621 cooling rather than for the warming conditions that have generally been evaluated for studies of
- 622 the ozone hole and anthropogenic climate change. Action spectra also provide data that can be
- 623 used for assessment of human health impacts. These model improvements would be beneficial
- 624 for simulation of other types of climate change driven by large aerosol injections including
- 625 volcanic eruptions, mass fires, dust storms, and solar geoengineering.

626 **Data**

- 627 The CESM model is freely available from NCAR, but requires registration at
- 628 www.cesm.ucar.edu/models/cesm1.2. All the data and scripts needed for the plots presented in
- this paper and the supplement are publicly available from osf.io/Bardeen 2021 OzoneUV
- 630 (doi:10.17605/OSF.IO/KVY86). The full model output is very large and is stored on the
- 631 PetaLibrary at the University of Colorado, which is not available to the public. However,
- 632 additional data from these runs can be provided upon request.

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- authors helped edit the manuscript. Thanks to Sasha Madronich for his help with TUV and to
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642 Tables

- 643 **Table 1.** List of the WACCM4/CARMA simulations performed in this study and our prior work.
- 644 IP indicates a regional conflict between India and Pakistan, and UR indicates a global conflict
- between the US and Russia. Soot shows the total soot emission, NO the total NO emission
- 646 (fireball and fire), TUV indicates that in-line TUV was used for photolysis, and AOD indicates
- 647 that the aerosol optical depth was included in the TUV photolysis calculation. Yields indicate the 648 total yield, the count and size (Mt) of the individual weapons that are assumed for the NO_x
- total yield, the count and size (Mt) of the individual weapons that are assumed for the NO_x production, and the source of the weapon inventory. For Toon et al. (2008), the weapon yield
- was assumed to be 300 kt rather than 100 kt. For NRC (1985), 1500 Mt of ground bursts were
- not included in the total. The different NO emission cases (NO, NO2, and NO3) are sensitivity
- tests that differ only in the amount and altitude extent of the fireball NO emission.
- 653

Table 2. List of the main chemical reactions used in WACCM that are important to odd-oxygen

655 (O and O₃) concentrations. These reactions are grouped by production and loss and by catalytic

656 cycles for analysis purposes. The weight indicates the net odd-oxygen production or loss from a

reaction. These weights are used to sum the individual reaction rates to calculate total production

and loss rates.

660 Figures

- **Figure 1**. Evolution of the change in the global average vertical profile for soot (a, g),
- temperature (b,h), $H_2O(c, i)$, $HO_x(d, j)$, $NO_x(e, k)$, and $O_3(f, l)$ for the IP (top) and UR
- 663 (bottom) cases from the control. Soot, temperature, and H₂O are shown as differences, while
- HO_x , NO_x , and O_3 are shown as percentage differences. Scales are the same for the IP and UR
- 665 cases. HO_x is the sum of H, OH, and HO_2 , and NO_x is the sum of N, NO, and NO_2 .
- **Figure 2**. Evolution of the change in global average column O₃ for the 5 Tg soot injection
- 667 regional nuclear war cases: Mills et al. (2008), Mills et al. (2014), Toon et al. (2019), IP, IP–NO,
- and IP–AOD. See Table 1 for the abbreviations and descriptions of the India/Pakistan (IP) cases
- 669 from this study.
- 670 Figure 3. Evolution of column integrated absolute chemical reaction rates (color) and the O₃
- 671 column (black) for the IP-AOD (left), IP (center), and IP-NO (right) cases. The total reaction
- rates are on the top (a-c), and the percentage difference from the control case are on the bottom
- 673 (d-f). The IP–AOD case does not include aerosols in the photolysis and the IP-NO case does not
- 674 include any NO injection. The solid lines are the named case and the dotted lines are the control
- 675 case. See Table 2 for the reactions included in each grouping.
- **Figure 4**. Similar to Figure 2, but for column integrated chemical reaction rates relative to the evolving total production rate of the respective case. Thus, by definition the total production for each of these cases, both the perturbation and the control, is 1.0.
- **Figure 5**. Global average vertical profile of absolute reaction rates (colors) and O₃ (black) for the first year following the war for the IP–AOD and IP cases. In both cases, the solid lines are the named case and the dotted lines are the control. Note the O₃ scale is increasing to the left.
- Figure 6. Evolution of the zonal average percent change in the O₃ column from the control for
 the IP (left) and UR (right) cases.
- 684 **Figure 7**. Evolution of zonal and monthly averages of the daily maximum clear-sky surface UV
- Index for the control (left), IP (middle), and UR (right) cases. The maximum values are 26, 33,
 and 50 respectively. The gray line shows the UV Index of the 20 contour to highlight the
- 687 expansion of areas of extreme UV in the IP and UR cases.
- **Figure 8**. Evolution of the change in global average column O_3 in the Northern Hemisphere from Pittock et al. (1986) using scenarios from AMBIO (1982), Knox (1983), and NRC (1985) along
- 690 with WACCM4 simulations from Coupe et al. (2019), UR, UR-NO, UR-AOD, UR+NO2, and
- 691 UR+NO3 (left) and global averages (right) for just the WACCM4 simulations. Kao et al. (1990)
- 692 only provided information about the end of their 20-day simulation. See Table 1 for the
- 693 abbreviations and descriptions of the United States/Russia (UR) cases from this study.
- **Figure 9**. Similar to Figure 3, but for the UR–AOD (left), UR (center), and UR-NO (right) cases.
- 695 **Figure 10**. Similar to Figure 4, but for the UR–AOD (left) and UR (right) cases.

- 696 Figure 11. Similar to Figure 5, but for the UR–AOD (left) and UR (right) cases. Note the O₃
- 697 scale is increasing toward the left in panels a and b.
- 698 Figure 12. Evolution of the change in the global average for surface values of aerosol optical
- depth, temperature, O₃ column, and precipitation compared with the spectral integrals of PAR
- 700 and UV-B for the IP (left) and UR (right) cases. Temperature (black) is on the right scale and the
- percentage change of the other fields (color) are on the left scale. AOD is the total aerosol optical
- depth including both the soot and the background aerosol and has been reduced by a factor of 3
- in the IP case and 30 in the UR case to fit on the plot. The maximum global average AOD values
- are 0.13 for the control, 0.20 for the IP case, and 2.92 for the UR case.
- **Figure 13**. Evolution of the change in the global average surface values for UV-B compared
- with the action functions for plant growth, inhibition of phytoplankton, cataract formation,
- vitamin-D synthesis, erythema, and DNA damage for the IP (left) and UR (right) cases. Note thedifferent vertical scales for the two panels.
- **Figure 14**. UV spectra for O₃ absorption cross-section (black) and soot mass absorption
- 710 coefficient (gray) along with spectral weighting functions for UV-B, erythema, DNA damage,
- and two metrics for non-melanoma skin cancer (colors) over the range 230-370 nm (left).
- 712 Evolution of the change in the global average spectral integrals of UV-B compared with the
- action functions for erythema, DNA damage and non-melanoma skin cancer for the IP (center)
- and UR (right) cases. Note the different vertical scales for these two panels.
- 715

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1010Figure 1. Evolution of the change in the global average vertical profile for soot (a, g),1011temperature (b,h), H_2O (c, i), HO_x (d, j), NO_x (e, k), and O_3 (f, l) for the IP (top) and UR1012(bottom) cases from the control. Soot, temperature, and H_2O are shown as differences,1013while HO_x , NO_x , and O_3 are shown as percentage differences. Scales are the same for the1014IP and UR cases. HO_x is the sum of H, OH, and HO_2 , and NO_x is the sum of N, NO, and1015 NO_2 .



Figure 2. Evolution of the change in global average column O₃ for the 5 Tg soot injection
regional nuclear war cases: Mills et al. (2008), Mills et al. (2014), Toon et al. (2019), IP,
IP–NO, and IP–AOD. See Table 1 for the abbreviations and descriptions of the
India/Pakistan (IP) cases from this study.



1023Figure 3. Evolution of column integrated absolute chemical reaction rates (color) and the1025O3 column (black) for the IP–AOD (left), IP (center), and IP-NO (right) cases. The total1026reaction rates are on the top (a-c), and the percentage difference from the control case are1027on the bottom (d-f). The IP–AOD case does not include aerosols in the photolysis and the1028IP-NO case does not include any NO injection. The solid lines are the named case and the1029dotted lines are the control case. See Table 2 for the reactions included in each grouping.



1034Time (years)Time (years)1035Figure 4. Similar to Figure 2, but for column integrated chemical reaction rates relative1036to the evolving total production rate of the respective case. Thus, by definition the total1037production for each of these cases, both the perturbation and the control, is 1.0.



Figure 5. Global average vertical profile of absolute reaction rates (colors) and O₃ 1041 (black) for the first year following the war for the IP-AOD and IP cases. In both cases, 1042 the solid lines are the named case and the dotted lines are the control. Note the O₃ scale is 1043 increasing to the left.

104610471047Figure 6. Evolution of the zonal average percent change in the O_3 column from the10481049

1051 surface UV Index for the control (left), IP (middle), and UR (right) cases. The maximum values are 26, 33, and 50 respectively. The gray line shows the UV Index of the 20 contour to highlight the expansion of areas of extreme UV in the IP and UR cases.

1056Time (years)Time (years)1057Figure 8. Evolution of the change in global average column O3 in the Northern1058Hemisphere from Pittock et al. (1986) using scenarios from AMBIO (1982), Knox1059(1983), and NRC (1985) along with WACCM4 simulations from Coupe et al. (2019),1060UR, UR-NO, UR-AOD, UR+NO2, and UR+NO3 (left) and global averages (right) for1061just the WACCM4 simulations. Kao et al. (1990) only provided information about the1062end of their 20-day simulation. See Table 1 for the abbreviations and descriptions of the1063United States/Russia (UR) cases from this study.1064United States/Russia (UR) cases from this study.

1077 1078 Figure 12. Evolution of the change in the global average for surface values of aerosol 1079 optical depth, temperature, O₃ column, and precipitation compared with the spectral 1080 integrals of PAR and UV-B for the IP (left) and UR (right) cases. Temperature (black) is 1081 on the right scale and the percentage change of the other fields (color) are on the left 1082 scale. AOD is the total aerosol optical depth including both the soot and the background 1083 aerosol and has been reduced by a factor of 3 in the IP case and 30 in the UR case to fit 1084 on the plot. The maximum global average AOD values are 0.13 for the control, 0.20 for 1085 the IP case, and 2.92 for the UR case. 1086

1087Time (years)1088Figure 13. Evolution of the change in the global average surface values for UV-B1089compared with the action functions for plant growth, inhibition of phytoplankton, cataract1090formation, vitamin-D synthesis, erythema, and DNA damage for the IP (left) and UR1091(right) cases. Note the different vertical scales for the two panels.

Figure 14. UV spectra for O₃ absorption cross-section (black) and soot mass absorption coefficient (gray) along with spectral weighting functions for UV-B, erythema, DNA damage, and two metrics for non-melanoma skin cancer (colors) over the range 230-370 nm (left). Evolution of the change in the global average spectral integrals of UV-B compared with the action functions for erythema, DNA damage and non-melanoma skin cancer for the IP (center) and UR (right) cases. Note the different vertical scales for these two panels.

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Figure S1. Similar to Figure 1, but with a different scale for the IP case to allow the structure of the IP case to be seen more clearly compared with the UR case. Most UR scales are 5 times the IP scale, except for H_2O which is 50 times and O_3 which is 2 times.

Figure S4. Vertical profiles of temperature differences between the IP (blue) and UR (red) cases with the control case for the southern polar (left), global (center), and northern polar (right) averages for the same time period as used in Figures 5 and 11.

Figure S5. Evolution of the zonal monthly column O₃ for the control (left), IP (middle), and UR (right) cases.

1128Image: right red with red wi

1134 (left), and UR (right) cases.

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Case	Soot	NO	T	A	Yields (Mt)
Control	0 Tg	0 Tg	X	Х	None
Toon et al. (2019)	5 Tg	0 Tg			Total: 0.66 Mt
					(44x0.015)
IP	5 Tg	0.5 Tg	Х	Х	Same as above
IP-AOD	5 Tg	0.5 Tg	X		Same as above
IP–NO	5 Tg	0 Tg	Х	Х	Same as above
Coupe et al. (2019)	150 Tg	0 Tg			None
UR	150 Tg	22.5 Tg	X	Х	Total: 1494 Mt
					(R: 48x0.05, 1232x0.1, 580x0.25, 574x0.8;
					US: 1486x0.09, 528x0.15, 200x0.3, 600x0.335, 322x0.36, 384x0.455)
					Kristensen & Korda (2020a; 2020b)
UR–AOD	150 Tg	22.5 Tg	Х		Same as above
UR–NO	150 Tg	0 Tg	X	Х	None
UR+NO2	150 Tg	21.6 Tg	X	Х	Total: 1320 Mt
					(4400x0.3)
					Toon et al. (2008) (0.1 Mt \rightarrow 0.3 Mt)
UR+NO3	150 Tg	39.9 Tg	X	Х	Total: 5000 Mt
					(220x1.5, 2200x1.0, 330x0.75, 1600x0.5, 2400x0.25, 3200x0.1, 12500x0.04)
					NRC (1985) (no ground bursts)

1143 Table 1. List of the WACCM4/CARMA simulations performed in this study and our prior work. IP indicates a regional conflict between India and Pakistan, and UR indicates a global 1144 1145 conflict between the US and Russia. Soot shows the total soot emission, NO the total NO emission (fireball and fire), TUV indicates that in-line TUV was used for photolysis, and 1146 1147 AOD indicates that the aerosol optical depth was included in the TUV photolysis calculation. Yields indicate the total yield, the count and size (Mt) of the individual 1148 weapons that are assumed for the NO_x production, and the source of the weapon 1149 1150 inventory. For Toon et al. (2008), the weapon yield was assumed to be 300 kt rather than 1151 100 kt. For NRC (1985), 1500 Mt of ground bursts were not included in the total. The

- different NO emission cases (NO, NO2, and NO3) are sensitivity tests that differ only in the amount and altitude extent of the fireball NO emission. 1153

Туре	Name	Weight	Reactions			
Production						
	0	2	$O_2 + h \rightarrow O + O(^1D)$			
		2	$O_2 + h \rightarrow 2 O$			
	O ₃	1	$NO + HO_2 \rightarrow NO_2 + OH$			
	1		$CH_3O_2 + NO \rightarrow CH_2O + NO_2 + HO_2$			
Loss						
	O _x	2	$O + O_3 \rightarrow 2 O_2$			
		1	$O(^{1}D) + H_{2}O \rightarrow 2 OH$			
	NO _x 2		$NO_2 + O \rightarrow NO + O_2$			
		2	$NO_3 + h \rightarrow NO + O_2$			
	HO _x	1	$HO_2 + O \rightarrow OH + O_2$			
		1	$HO_2 + O_3 \rightarrow OH + 2 O_2$			
		1	$OH + O \rightarrow H + O_2$			
		1	$OH + O_3 \rightarrow HO_2 + O_2$			
			$H + O_3 \rightarrow OH + O_2$			
	ClO _x BrO _x	2	$ClO + O \rightarrow Cl + O_2$			
	2 2 2 2 2 2 2 2 2 2 2 2 2 1	2	$Cl_2O_2 + h \rightarrow 2 Cl$			
		2	$ClO + ClO \rightarrow 2 Cl + O_2$			
		2	$ClO + ClO \rightarrow Cl_2 + O2$			
		$BrO + ClO \rightarrow Br + Cl + O_2$				
		$BrO + ClO \rightarrow BrCl + O_2$				
			$BrO + BrO \rightarrow 2 Br + O_2$			
			$BrO + O \rightarrow Br + O_2$			
			$ClO + HO_2 \rightarrow O_2 + HOCl$			
			$BrO + HO_2 \rightarrow HOBr + O_2$			

1155	Table 2. List of the main chemical reactions used in WACCM that are important to odd-oxygen
1156	(O and O ₃) concentrations. These reactions are grouped by production and loss and by
1157	catalytic cycles for analysis purposes. The weight indicates the net odd-oxygen
1158	production or loss from a reaction. These weights are used to sum the individual reaction
1159	rates to calculate total production and loss rates.