Impacts of the production and consumption of biofuels on stratospheric ozone

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[1] Biofuels are becoming increasingly popular sources of renewable energy as economic pressures and environmental consequences encourage the use of alternatives to fossil fuels. However, growing crops destined for use as biofuels incurs large N_2O emissions associated with the use of nitrogen-based fertilizers. Besides being a greenhouse gas, N_2O is also the primary source of stratospheric NO_x ($NO + NO_2$) which leads to stratospheric ozone depletion. In this paper, the potential effects on the ozone layer of a large-scale shift away from fossil fuel use to biofuels consumption over the 21st century are examined. Under such a scenario, global-mean column ozone decreases by 2.6 DU between 2010 and 2100 in contrast to a 0.7 DU decrease under a control simulation (the IPCC SRES B1 scenario for greenhouse gases) and a 9.1 DU increase under the more commonly used SRES A1B scenario. Two factors cause the decrease in ozone in the biofuels simulation: 1) large N_2O emissions lead to faster rates of the ozone-depleting NO_x cycles and; 2) reduced CO_2 emissions (due to less fossil fuel burning) lead to relatively less stratospheric cooling over the 21st century, which decreases ozone abundances. Reducing CO_2 emissions while neglecting to reduce N_2O emissions could therefore be damaging to the ozone layer. **Citation:** Revell, L. E., G. E. Bodeker, P. E. Huck, and B. E. Williamson (2012), Impacts of the production and consumption of biofuels on stratospheric ozone, *Geophys. Res. Lett.*, *39*, L10804, doi:10.1029/2012GL051546.
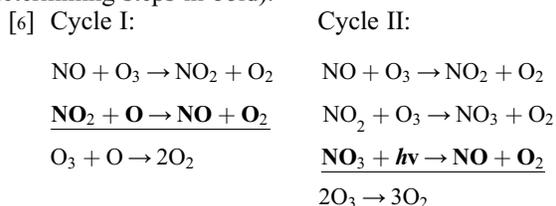
1. Introduction

[2] As fossil fuel reserves diminish and awareness of the detrimental effects of fossil fuel-burning on global climate continues to grow, biofuels are becoming an increasingly attractive supply of fuel, especially in Europe, the USA and Brazil [*Bessou et al.*, 2011]. In 2010, biofuels accounted for 2.7% of road-transportation fuel use globally (http://vitalsigns. worldwatch.org/vs-trend/biofuels-regain-momentum). Biofuels are often considered to be carbon neutral because the CO_2 released on burning had previously been removed from the atmosphere via photosynthesis. However, nitrogen-based fertilizers used in growing the crops from which biofuels are produced lead to N_2O emissions via soil nitrification and denitrification [*Smeets et al.*, 2009]. N_2O is a greenhouse gas (GHG) with a 100-year global warming potential of $\sim$298, and a lifetime of $\sim$114 years [*Forster et al.*, 2007].

[3] *Crutzen et al.* [2008] suggested that the global warming reduction achieved through CO_2 reductions by using first-generation biofuels (those produced from vegetable oil, starch or sugar) instead of fossil fuels, could be countered by the subsequent increase in N_2O emissions resulting from increased nitrogen-based fertilizer use.

[4] Using linked economic and terrestrial biogeochemistry models, *Melillo et al.* [2009] found that carbon emissions resulting from land-use change under a global biofuels programme would be significant. However, they also found that an increase in N_2O emissions due to increases in nitrogen-based fertilizer use would be a more important contributor to climate warming. Additionally, under two different scenarios in which biofuels production was projected to increase, they estimated this would account for $\sim$60% of total annual N_2O emissions by 2100.

[5] As well as being a GHG, N_2O leads to stratospheric ozone destruction through its reaction with $O(^1D)$ to produce NO_x ($NO + NO_2$) [*Crutzen*, 1970]. NO_x depletes ozone by participating catalytically in the following cycles (rate-determining steps in bold):

[6] Cycle I: Cycle II:

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad NO + O_3 \rightarrow NO_2 + O_2$$
$$\mathbf{NO_2 + O \rightarrow NO + O_2} \qquad NO_2 + O_3 \rightarrow NO_3 + O_2$$
$$O_3 + O \rightarrow 2O_2 \qquad \mathbf{NO_3 + h\nu \rightarrow NO + O_2}$$
$$\qquad\qquad\qquad 2O_3 \rightarrow 3O_2$$

[7] N_2O is projected to be the dominant ozone-depleting gas emitted through the 21st century [*Ravishankara et al.*, 2009]. However, stratospheric cooling resulting from increases in other GHGs, primarily CO_2, decreases the effectiveness of N_2O as an ozone-depleting substance by: 1) increasing the sink for reservoir nitrogen species [*Rosenfield and Douglass*, 1998], and 2) decreasing the O/O_3 ratio [*Jonsson et al.*, 2004], thus slowing the $NO_2 + O$ rate-determining step of Cycle I by decreasing the abundance of atomic oxygen. Furthermore, as sea-surface temperatures (SSTs) increase over the 21st century, the Brewer-Dobson circulation (BDC) is projected to strengthen [*Fomichev et al.*, 2007], thus reducing the yield of NO_x from N_2O [*Cook and Roscoe*, 2009].

[8] Three chemistry-climate model (CCM) simulations – 'A1B', 'B1' and 'biofuels' (Table 1) have been performed to examine the potential effects of increased production and consumption of biofuels on stratospheric ozone. The A1B and B1 simulations were based on the IPCC SRES A1B and B1 GHG emissions scenarios, respectively [*Nakicenovic*

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Table 1. Summary of the Three CCM Simulations

	A1B	B1	Biofuels
CO_2 in 2100 (GtC)[a]	13.1	5.2	3.7
N_2O in 2100 (MtN$_2$O-N)[a]	7.0	5.7	14.3
2090s temperature (K)[b]	215.2	218.6	218.9
ΔO_3 (DU)[c]	9.1	−0.7	−2.6

[a]CO_2 and N_2O emissions.

[b]Global-mean temperatures at 30 hPa simulated by NIWA-SOCOL, and averaged over the 2090s decade.

[c]Change in global-mean total column ozone over the 21st century simulated by NIWA-SOCOL (2090s decade minus 2010s decade), expressed in Dobson Units.

and Swart, 2000]. The SRES A1B scenario portrays an intermediate increase in concentrations of anthropogenic GHGs, while SRES B1 prescribes relatively low GHG emissions. The A1B and B1 simulations display similar N_2O surface concentrations but different CO_2 surface concentrations and SSTs, which influence stratospheric temperatures and the BDC. This facilitated an investigation of how the BDC and CO_2-induced stratospheric cooling influence ozone and ozone-depleting NO_x chemistry. The major difference between the B1 and biofuels simulations was the N_2O surface concentrations, permitting a study of the effect of increased N_2O emissions on ozone as a result of increased nitrogen-based fertilizer use.

2. Computational Methods

[9] CCM simulations were performed using the NIWA-SOCOL (National Institute of Water and Atmospheric Research – SOlar Climate Ozone Links) model. NIWA-SOCOL is based on version 2.0 of the SOCOL model [*Schraner et al.*, 2008; *SPARC CCMVal*, 2010], and includes 41 chemical species, 140 gas-phase reactions, 46 photolysis reactions and 16 heterogeneous reactions. The reaction rate constants used were those recommended by *Sander et al.* [2006, 2009]. NIWA-SOCOL performs chemical and radiative calculations every two hours, and dynamical process calculations every 15 minutes. The adjusted A1 scenario for halocarbon concentrations that accounts for the accelerated phase-out of HCFCs was used [*Daniel et al.*, 2007], while the GHG concentrations prescribed for the three simulations are described in more detail below.

[10] NIWA-SOCOL tracks the contribution to ozone loss from 15 catalytic cycles, similar to the diagnostic approach used by *Lee et al.* [2002]. Odd oxygen ($O + O(^1D) + O_3$) loss rates (in molecules cm^{-3} s^{-1}) were calculated within the model using the rate-limiting steps of the corresponding reaction cycles. Results were recorded and accumulated into daily means within each model grid cell.

[11] A simple climate model, MAGICC6 (Model for the Assessment of Greenhouse-gas Induced Climate Change version 6) [*Meinshausen et al.*, 2011], was used to convert the emissions scenarios to the surface concentration scenarios required as input to the CCM simulations. Prescribing concentrations rather than emissions boundary conditions breaks the feedback of ozone changes on changes in tropospheric lifetimes. However for our study, omitting this feedback does not affect our conclusions. MAGICC6 can be calibrated to emulate any of the 19 CMIP3 (Coupled Model Intercomparison Project phase 3) Atmosphere-ocean General

Circulation Model (AOGCMs) and the 10 C[4]MIP (Coupled Carbon Cycle Climate Model Intercomparison Project) carbon cycle models. For our calculations, MAGICC6 was calibrated to the ECHAM5/MPIOM AOGCM core model parameters, and LLNL model carbon cycle parameters.

3. Emissions Scenarios

[12] For the B1 simulation, surface concentrations of CO_2, N_2O and CH_4, commensurate with the SRES B1 emissions scenario, were calculated using MAGICC6 tuned as described above. SSTs and sea-ice concentrations (SICs) were prescribed using output from the UKMO-HadCM3 AOGCM, based on the SRES B1 emissions scenario [*Nakicenovic and Swart*, 2000]. The SST and SIC datasets were obtained from the World Climate Research Programme's CMIP3 multi-model dataset.

[13] Since the biofuels simulation was constructed to follow the same radiative forcing pathway as the B1 simulation (see below), it used the same SSTs, SICs and CH_4 surface concentrations as the B1 simulation. The biofuels simulation was based on findings from *Melillo et al.* [2009], which suggest that biofuels production and the associated use of nitrogen-based fertilizers could account for ~60% of total annual N_2O emissions by 2100. To capture this impact, an emissions scenario for N_2O, with emissions of 14.3 MtN$_2$O-N in 2100 compared with 5.7 MtN$_2$O-N in the B1 scenario, was constructed. MAGICC6 calculated the corresponding surface N_2O concentrations, which were 443 ppb in 2100 compared with 368 ppb in the B1 scenario (Figure 1a).

[14] CO_2 emissions in the biofuels scenario were decreased relative to those prescribed in the B1 scenario such that the global-mean radiative forcing in the biofuels simulation was the same as in the B1 simulation. By keeping the radiative forcing the same in both simulations, we can ensure that any differences seen are attributable to changes in chemistry and are not potentially compromised by changes in dynamics. The reduction in CO_2 concentrations also served to simulate the expected reduction in CO_2 emissions resulting from biofuels use rather than fossil fuels use. This resulted in CO_2 emissions of 3.7 GtC in 2100 in the biofuels scenario, compared with 5.2 GtC in the B1 scenario (summarized in Table 1, along with N_2O emissions). The equivalent CO_2 surface concentrations in 2100, calculated using MAGICC, were 532 ppm in the biofuels scenario, compared with 551 ppm in the B1 scenario (Figure 1b).

[15] The A1B simulation used GHG concentrations, SSTs and SICs consistent with the SRES A1B scenario. SSTs and SICs were taken from ECHAM5/MPIOM AOGCM output. SSTs for the A1B and B1 simulations are shown in Figure 1d. CH_4 concentrations for the three scenarios are displayed in Figure 1c.

4. Results and Discussion

[16] The projected changes in global-mean total column ozone over the 21st century for the three simulations are summarized in Table 1. In our A1B simulation, global-mean total column ozone is projected to increase by 9.1 DU through the 21st century, due to a slowing of the halogen ozone-loss cycles (as concentrations of stratospheric
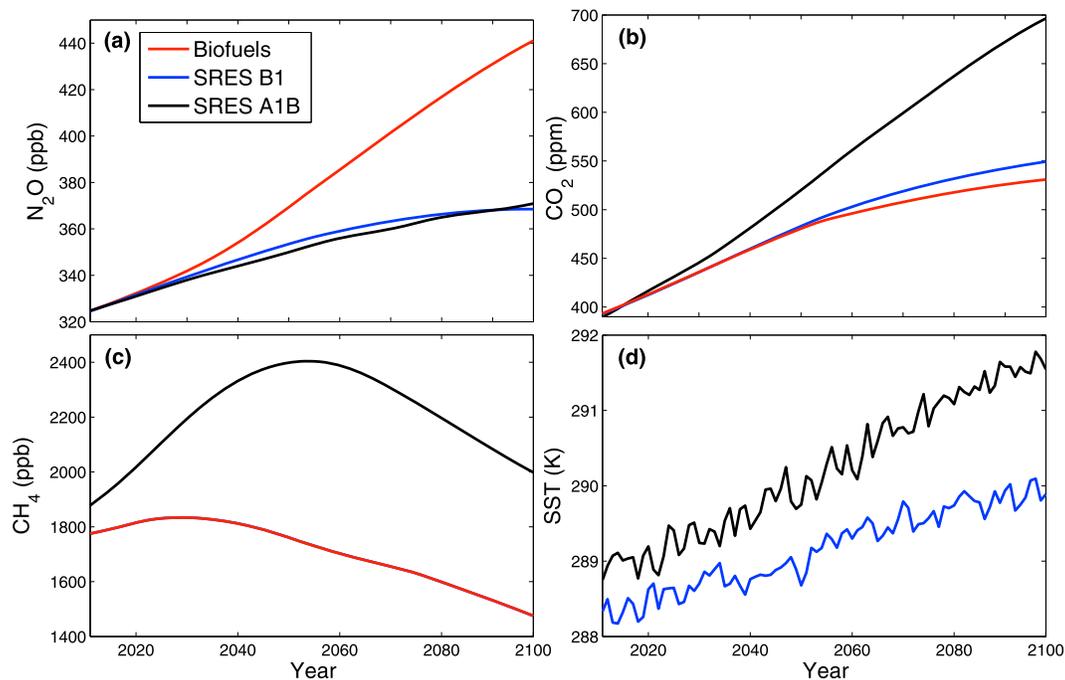
Figure 1. (a) N_2O, (b) CO_2, (c) CH_4 surface concentrations for the A1B, B1 and biofuels emissions scenarios. CH_4 concentrations are identical for the B1 and biofuels scenario such that the red and blue traces overlay each other. (d) Global-mean SSTs for the A1B and B1 simulations (the biofuels simulation used B1 SSTs).

chlorine and bromine decline) and GHG-induced stratospheric cooling (which slows the temperature-dependent ozone loss cycles [*Rosenfield et al.*, 2002]). This result is consistent with other CCM projections in the SPARC (Stratospheric Processes and their Role in Climate) CCM Validation (CCMVal-2) assessment [*SPARC CCMVal*, 2010], which also used the SRES A1B scenario.

[17] In the B1 simulation, column ozone decreases by 0.7 DU over the 21st century since stratospheric cooling (which induces ozone increases) is relatively less than in the A1B simulation where larger stratospheric cooling leads to a 9.1 DU increase in ozone. A larger decrease in column ozone of 2.6 DU is calculated in the biofuels simulation, because greater N_2O emissions lead to enhanced rates of the ozone-depleting NO_x cycles.

[18] Ozone differences between the B1 and A1B simulations at the end of the 21st century are displayed in Figure 2a as a percentage of ozone in the A1B simulation, and in Figure 2b as the difference in total column ozone (B1 minus A1B). This facilitates an assessment of the effect of GHG forcing (for example, driving changes in stratospheric temperature and the BDC) on ozone. The ozone concentrations in the B1 simulation in the upper and lower Antarctic stratosphere and the tropical lower stratosphere are greater than those in the A1B simulation, consistent with the findings of *Eyring et al.* [2010]. Elsewhere, the B1 simulation ozone concentrations are lower by as much as 20%.

[19] The lower ozone concentrations in the B1 simulation are likely due to: 1) the weaker stratospheric cooling, with the result that the temperature-dependent gas-phase ozone loss cycles slow less than in the A1B simulation; 2) lower CH_4 concentrations than in the A1B scenario [*Portmann and Solomon*, 2007; *Fleming et al.*, 2011] and; 3) reduced strengthening of the BDC, as a result of cooler SSTs

[*Fomichev et al.*, 2007; *Bekki et al.*, 2011], which means that the ozone flux into the southern mid-latitude lower stratosphere is weaker than in the A1B simulation.

[20] Increased tropical lower stratospheric ozone in the B1 simulation is consistent with a difference in the BDC between the B1 and A1B simulations. The slower ascent rate in the tropical lower stratosphere in the B1 simulation allows more time for ozone to form in the rising air parcels than in more quickly ascending air in the A1B simulation [*Avallone and Prather*, 1996]. Elevated lower-stratospheric Antarctic ozone in the B1 simulation is caused by a relatively warmer lower stratosphere; over the 21st century, Antarctic winter temperatures at 50 hPa average 197 K in the B1 simulation compared to 189 K in the A1B simulation. As a result, polar stratospheric cloud formation, and the associated heterogeneous chlorine and bromine ozone-depleting chemistry, is suppressed in the B1 simulation compared to the A1B simulation. In the 2090s, there is ~5% more upper-stratospheric Antarctic ozone in the B1 simulation than in the A1B simulation because there is less CH_4, and hence the ozone-depleting HO_x cycles (which are dominant in the upper stratosphere) are slower.

[21] Figure 2c displays the same quantity as Figure 2a, but calculated for the difference between the biofuels and B1 simulations. This facilitates an assessment of the effects of larger N_2O emissions on ozone. Figure 2c shows that under the biofuels scenario, ozone concentrations are (relatively) suppressed throughout the middle stratosphere and enhanced in the lower stratosphere. However, because the middle stratosphere dominates the ozone column, total column amounts are smaller in the biofuels simulation and up to 11 DU smaller at northern high latitudes (Figure 2d).

[22] In the biofuels simulation, ozone is greater by up to 4% in the troposphere compared to that in the B1 simulation.
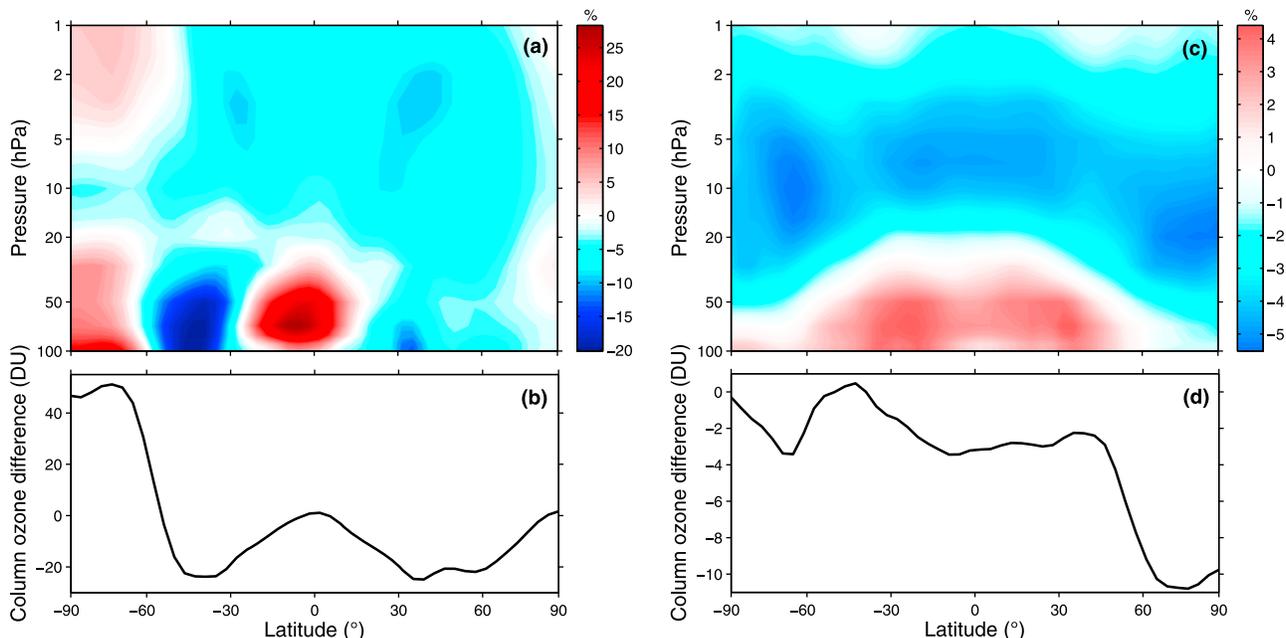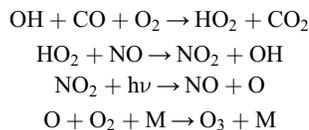
Figure 2. (a) B1 ozone minus A1B ozone in the 2090s decade, calculated as a percentage of ozone in the A1B simulation. (b) 2090s decade B1 total column ozone minus A1B total column ozone. (c) Biofuels ozone minus B1 ozone in the 2090s decade, calculated as a percentage of ozone in the B1 simulation. (d) 2090s decade biofuels total column ozone minus B1 total column ozone.

N_2O leads to an increase in tropospheric ozone production, via the following reactions involving NO_x:

$$OH + CO + O_2 \rightarrow HO_2 + CO_2$$
$$HO_2 + NO \rightarrow NO_2 + OH$$
$$NO_2 + h\nu \rightarrow NO + O$$
$$O + O_2 + M \rightarrow O_3 + M$$

Ozone production by this mechanism is generally insignificant in the stratosphere, where the concentration of CO is too small [*Lanzendorf et al.*, 2001].

[23] In the middle and upper stratosphere, ozone in the biofuels scenario is up to 5% less than that in the B1 simulation, since N_2O concentrations are higher, leading to a faster rate of the ozone-depleting NO_x cycles. These cycles
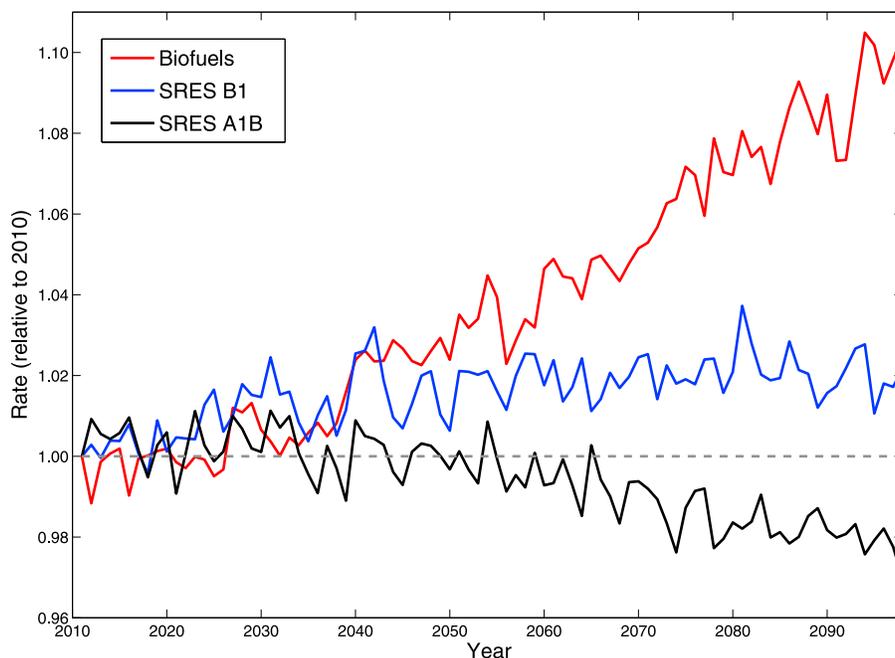

Figure 3. Global-mean rate of the NO_x ozone loss cycles, averaged over 1–100 hPa and normalized to 1.0 in 2010. The differences between the biofuels and B1 simulations are largely a consequence of different N_2O emissions, while those between the B1 and A1B simulations are largely due to temperature differences.

do not occur in the troposphere where O and O_3 concentrations are minimal.

[24] Figure 3 shows the combined rate of the two NO_x ozone-loss cycles (Cycles I and II), averaged over 1–100 hPa, relative to their rate in 2010 for each of the three simulations. Despite increasing N_2O emissions in the A1B simulation, NO_x-induced ozone depletion slows over the 21st century (as described by L. E. Revell et al., The effectiveness of N_2O in depleting stratospheric ozone, submitted to *Geophysical Research Letters*, 2011). The B1 simulation has N_2O emissions almost identical to those in the A1B simulation, yet because the stratosphere cools relatively less, the rate at which the NO_x cycles deplete ozone increases by ~2% between 2010 and 2100. Finally, in the biofuels simulation, the rate of ozone-depleting NO_x chemistry increases by ~10% over the 21st century owing to the large increase in N_2O concentrations.

5. Conclusions

[25] We have presented the effects of three different GHG emissions scenarios on stratospheric ozone. The SRES A1B scenario is the scenario on which the majority of the SPARC CCMVal-2 simulations were based. In our A1B simulation, global-mean total column ozone increased by 9.1 DU over the 21st century due to decreasing halogen concentrations and GHG-induced stratospheric cooling. Such an increase was not observed in the simulation based on the SRES B1 scenario (0.7 DU decrease), which has lower CO_2 and CH_4 emissions. Increasing N_2O emissions increase the rate of NO_x-catalyzed ozone loss, and this effect becomes more pronounced when the stratosphere cools relatively less, as it does in the biofuels simulation (2.6 DU decrease). Therefore, the reduction in CO_2 emissions achieved by switching to biofuels is not large enough to compensate for the deleterious effects of associated increases in N_2O emissions on stratospheric ozone. Increased biofuels production and consumption could therefore be damaging to the ozone layer.

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