

1 **The impact of biofuel poplar cultivation on ground-level ozone and premature human mortality depends**
2 **on cultivar selection and planting location**

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10 **Abstract**

11 Isoprene and other volatile organic compounds emitted from vegetation play a key role in governing the
12 formation of ground-level ozone. Emission rates of such compounds depend critically on the plant species. The
13 cultivation of biofuel feedstocks will contribute to future land use change, altering the distribution of plant
14 species and hence the magnitude and distribution of emissions. Here we use relationships between biomass yield
15 and isoprene emissions derived from experimental data for 29 commercially available poplar hybrids to assess
16 the impact that the large-scale cultivation of poplar for use as a biofuel feedstock will have on air quality,
17 specifically ground-level ozone concentrations, in Europe. We show that the increases in ground-level ozone
18 across Europe will increase the number of premature deaths attributable to ozone pollution each year by up to
19 6%. Substantial crop losses (up to ~9 Mt y⁻¹ of wheat and maize) are also projected. We further demonstrate that
20 these impacts are strongly dependent on the location of the poplar plantations, due to the prevailing
21 meteorology, the population density and the dominant crop type of the region. Our findings indicate the need for
22 a concerted and centralized decision-making process that considers all aspects of future land use change in
23 Europe, and not just the effect on greenhouse gas emissions.

24 **Introduction**

25 Volatile organic compounds (VOCs) are produced and released to the atmosphere from both anthropogenic and
26 natural sources. Biogenic VOCs (bVOCs) account for over 90% of the non-methane hydrocarbons emitted
27 annually (1). Of these, the reactive compound isoprene (2-methyl-1,3-butadiene) is the most significant in terms
28 of both magnitude of emissions (estimated at 500 TgC y⁻¹ (1)) and subsequent impact on atmospheric
29 composition (2). The photochemical oxidation of isoprene in the presence of the nitrogen oxides (NO_x: NO and
30 NO₂) governs the production rate of ground-level ozone (3), and leads to the formation of low volatility reaction
31 products that can condense into the aerosol phase (4). Both ozone and aerosol are predominantly secondary
32 pollutants with well-documented effects on climate and air quality. Their contribution to radiative forcing since
33 Pre-Industrial times has been quantified as +0.40 (+0.20 to +0.60) W m⁻² and -0.03 (-0.27 to +0.20) W m⁻²
34 respectively (5). The World Health Organization attributes over 3.7 million deaths worldwide to their combined
35 health effects annually, of which around 0.25 million occur in Europe (6). While it is believed that exposure to

36 particulate matter (PM) is responsible for the majority of these premature deaths, ozone pollution has been
37 identified as one of the biggest causes for concern in Europe (7). Owing to the high level of uncertainty involved
38 in modelling the formation of biogenic secondary organic aerosol (SOA) and in attributing health impacts of PM
39 to specific sources given the lack of knowledge of the size distribution and toxicity of aerosols of different
40 origins, this study focuses on changes in ground-level ozone in response to projected land use change associated
41 with biofuel feedstock cultivation. Our estimates of air quality impacts associated with biofuel cultivation
42 therefore represent a lower bound.

43 The synthesis and emission rates of bVOCs are strongly dependent on plant species as well as environmental
44 factors such as light and temperature, and hence are regulated by species distribution (8). Land use and land
45 cover change (LULCC) therefore has the potential to substantially alter emissions of bVOCs by changing the
46 occurrence and distribution of plant species at the regional scale (9,10). Hurtt et al. identify the large-scale
47 cultivation of biofuel feedstock crops together with afforestation initiatives in the mid-latitudes as key drivers of
48 LULCC in the near future in the moderate Intergovernmental Panel on Climate Change Representative
49 Concentration Pathways (RCPs) scenarios (11).

50 Increasing areas of land are already being converted to the production of bioenergy crops (11) in order to meet
51 the growing demand for energy supplies perceived as “carbon-neutral”. In particular, the European Union has
52 set a target of 10% replacement of transportation fuels with biofuels and a 10% replacement of its combined
53 heat and power plant feedstock by 2020 (12). One of the most important short rotation coppice crops currently
54 used for this purpose is hybrid poplar (produced by crossing various *Populus* species) (13), and although the
55 European Union has not mandated that feedstocks are locally grown, environmental and energy security
56 considerations mean that cultivation of poplar is projected to increase.

57 The replacement of land currently given over to grasses and conventional food crops, few of which emit
58 detectable amounts of isoprene (8), with a high isoprene-emitting species such as poplar, will cause the amount
59 of isoprene entering the atmosphere to increase. In the presence of the moderately high concentrations of NO_x
60 found in Europe, emissions of isoprene lead to higher concentrations of ozone and under these conditions
61 isoprene emissions may be one of the most important determinants of ground level ozone concentrations³.
62 Different cultivars of poplar have differing isoprene emission rates (8,13,14) but also produce different biomass
63 yields (13,14).

64 We have previously shown that the large-scale conversion of agricultural and grass lands in Europe to biofuel
65 crops such as poplar increases ground level ozone concentrations sufficiently to have significant impacts on
66 human mortality (more than 1000 additional deaths annually) and crop yields (a 4% reduction) (15). Here, we
67 determine the feasibility of mitigating these impacts through policy intervention, based on either the careful
68 selection of poplar cultivar or well-informed choice of geographic location for future large-scale poplar
69 plantations.

70 We use experimental data on the relationship between biomass yields and isoprene emission rates from 29
71 different commercially available cultivars of poplar (14) in a model of atmospheric transport and chemistry
72 (15,16) to calculate the effects of the large-scale cultivation of these cultivars on ground level ozone
73 concentrations in Europe. We calculate the impacts of this additional ozone on human mortality and crop yields
74 across Europe, for each of a range of cultivar type (low-, mid-, or high-yielding) and defined planting region
75 within the continent.

76 **Methods and materials**

77 **LULCC scenarios**

78 Eller et al. (14) showed a statistically significant relationship between biomass yield and isoprene emission rate
79 for 29 commercially available poplar hybrids. We categorize these poplar hybrid clones into three groups based
80 on biomass yield; the median yield of each of the low-, medium-, and high-yielding groups is taken as the 10th,
81 50th and 90th percentiles of the yields of the full set respectively. We determine the median isoprene emission
82 rate for each group (see Table 1 and SI). Meeting the 2020 EU targets for biofuel usage will require the
83 production of 260 Mt (dry weight) of biomass per year (12,17). We calculate the land area required to meet this
84 biomass yield target, using the average yield for each of the three groups. We also use emissions and yield data
85 for a fourth poplar clone, genetically manipulated so that it does not emit isoprene. Such a genetically modified
86 organism (GMO) has already been engineered and has been shown to have a biomass yield close to the median
87 of conventional cultivars, with negligible isoprene emission (13).

88 The observed range of biomass yields for the cultivar groups is 4.3-11.5 t(dry weight) ha⁻¹ y⁻¹, resulting in land
89 requirements between 23 and 61 Mha for these types. Fischer et al. (2010) (18) demonstrated that up to 72Mha
90 of land in Europe currently used for food crop or livestock production could be converted to biofuel feedstock
91 cultivation without jeopardising food security. We distribute this land area required for the additional cultivation
92 of poplar across Europe according to previously identified land availability (15,18), under three broad LULCC
93 scenarios, shown in Table 1.

94 In the first planting scenario, a “fixed area” approach, we assume that 33 Mha of land is converted to poplar
95 cultivation across the EU. This is the land area required to reach the biofuel production target using the medium-
96 yield group of poplar cultivars. The total biomass yield produced from this 33 Mha then depends on the cultivar
97 type used.

98 In the second LULCC scenario, taking a “fixed yield” approach, we assume that sufficient land is turned over
99 for each cultivar group to ensure that the EU’s biomass requirement is harvested from the poplar plantations
100 annually. In this experiment, the area required differs, depending on the assumed yield of the cultivar used. We
101 also estimate upper and lower bounds for the air quality impacts of the different poplar types for the fixed yield
102 cultivation scenarios.

103 In a final “regional” approach we assume that a medium-yield cultivar is grown, but that the required 33 Mha of
104 land used are confined to one of four distinct regions within Europe: NW Europe, NWEu; the Mediterranean

105 region, Med; Eastern Europe, EEU; Ukraine, Ukr. The differing environmental conditions in these regions lead
 106 to differences in the ozone production resulting from the increase in isoprene emissions, and differences in
 107 population density and crop production then also determine the air quality impacts of the modelled land use
 108 change. Differences in environmental conditions other than temperature and light within the regions of
 109 cultivation (e.g. differences in soil moisture availability) may result in different total biomass yields, but these
 110 second-order effects are not accounted for here.

111
 112 Table 1 shows isoprene emission rates and total emissions for Europe under each of our biofuel cultivation
 113 scenarios. Figures in parentheses for the “fixed yield” scenarios indicate the upper and lower bounds used in the
 114 sensitivity tests performed to constrain the uncertainties in our estimates (see text above and SI for further
 115 details).

116

		Isoprene emission rate ($\mu\text{g m}^{-2} \text{h}^{-1}$)	Biomass yield (Mt odw)	Land area required (Mha)	Total isoprene emissions (Tg y^{-1})
Base case		35	-	-	11.4
Fixed area (33.3 Mha)	Hi	82.8	340	33.3	15.4
	Mid	55.3	260	33.3	14.1
	Lo	28.9	190	33.3	12.8
	GMO	2.0	240	33.3	11.4
Fixed yield (260 Mt)	Hi	82.8 (80.3, 85.2)	260	22.6 (22.0, 23.3)	14.1
	Mid	55.3 (52.9, 57.8)	260	33.3 (32.0, 34.8)	14.1
	Lo	28.9 (27.5, 31.3)	260	61.2 (56.8, 66.3)	13.9
	GMO	2.0	260	35.6	11.4
Regional (33.3 Mha)	Ukr	55.3	260	33.3	14.4
	EEU	55.3	260	33.3	14.1
	NW	55.3	260	33.3	14.0
	Med	55.3	260	33.3	15.1

117

118 Under each of the three approaches taken here we account for the impacts of land use change on isoprene
 119 emission rates, surface roughness, leaf area indices and deposition processes, factors which have been
 120 previously shown to substantially affect ozone concentrations (19,20).

121 **Experimental data**

122 Leaf-level isoprene emission rates and total first growth year biomass increases for 29 commercially available
 123 hybrid poplar clones (14) were used to determine emission factors (basal emission rates at standard conditions

124 (*I*) and estimated total biomass yields (per hectare) for a four year growing cycle (*13,14,21*). The following
125 Reduced Major Axis regression relationship (*22*) between the two was determined:

126
$$y=0.13449\varepsilon+0.35812$$

127 where *y* is the yield (t ha⁻¹) and ε is the isoprene emission factor (mg m⁻² h⁻¹). “Average” emission factors
128 (shown in Table 1) and yields for three groups: low- (taken as the 10th percentile), mid- (median) and high- (90th
129 percentile) yielding, were quantified. The Standard Error of the Mean were also derived and used for a series of
130 sensitivity simulations to provide an upper and lower bound estimate of the air quality impacts of the fixed yield
131 cultivation scenarios. See SI for further details of the data analysis performed. The isoprene emission factor for
132 the GMO poplar clone was derived by assuming a 5% isoprene “leakage” rate based on emissions from a
133 conventional poplar cultivar (*13*).

134 **Atmospheric chemistry modelling**

135 We used the Frontier Research System for Global Change/University of California Irvine (FRSGC/UCI) global
136 chemistry transport model (CTM) to simulate isoprene emissions and atmospheric chemistry (*16*). The CTM
137 calculates biogenic emissions on-line using the Parameterized Canopy Environment Emission Activity
138 algorithms of the Model of Emissions of Gases and Aerosols from Nature (MEGAN) model v2.04 (*9*), here with
139 isoprene emission factors at standard conditions taken from the experimental data outlined above. For the poplar
140 scenarios, the baseline vegetation distribution (*9*) was altered to include a broadleaf tree biofuel crop in place of
141 current crops or grasses. Dry deposition velocities were altered to reflect the changes in land cover (*15*) (see SI
142 for further details). Other biogenic VOCs were not included in the simulations as they have a substantially
143 smaller effect on tropospheric ozone (*2*). Anthropogenic emissions were taken from the International Institute
144 for Applied Systems Analysis inventory for the year 2003 (*23*). Emissions associated with the production of
145 ligno-cellulosic ethanol from woody biomass and the final combustion of the biofuel have not been considered.
146 The CTM was driven by meteorological data from the European Centre for Medium-Range Weather Forecasts
147 at T42L37 resolution (2.8° by 2.8°) for 2001, with sub-gridscale structure captured using the second-order
148 moment scheme resulting in an effective diagnostic resolution of 0.9° by 0.9° (*16*). The capability of the CTM to
149 capture observed ozone concentrations in Europe has been assessed previously (*15,24*) against measurements
150 taken from EMEP (European Monitoring and Evaluation Program) monitoring stations. The small high bias of
151 the CTM output during the summer months was corrected using monthly scaling factors as outlined in our
152 earlier study (*15*). Given the non-linear response of ground-level ozone concentrations to increased isoprene
153 emissions, the use of a large-scale model is likely to introduce a high bias in projections of the number of
154 premature deaths while under-estimating crop production losses. However, these errors, associated with spatial
155 averaging of ozone concentrations across disparate chemical regimes (urban vs. rural), have been shown to be
156 small (*15*), as have the effects of changes in anthropogenic NO_x emissions since 2003 (*15*).

157 **Impacts analysis**

158 **Human mortality**

159 We applied the following dose-response relationship to each gridcell and summed the results over the domain
160 for a year:

$$161 \quad \Delta\text{Mort} = y_0(1-\exp(-\beta\Delta x))\text{Pop}$$

162 where ΔMort is the number of additional daily mortalities resulting from the LULCC scenario, y_0 is the baseline
163 mortality rate in the population, β is the concentration-response factor, Δx is the change in 8-hour ozone above a
164 threshold value of 35 ppbv, and Pop is the grid cell population (25).

165 Although there is considerable uncertainty in the quantification of human health impacts arising from increased
166 exposure to ground-level ozone, the above relationship has been developed from meta-analyses of
167 epidemiological studies (26). The use of a threshold concentration, while not physiologically realistic, is in
168 accordance with WHO guidelines (6) and increases the robustness of disentangling the effects of ozone from
169 confounders such as temperature (26). The values of both the concentration-response factor β (set at a 0.67%
170 increase in mortalities for every 10 ppbv increase in ozone (26)), and the baseline mortality y_0 (10 per 1000
171 deaths (27)) are Europe-specific values.

172 The chronic (morbidity) effects of increasing ground-level ozone concentrations are not well established (6) and
173 we restrict our analysis to the impacts on mortality.

174 **Crop production losses**

175 We estimate crop production losses for wheat and maize, two of the most commercially important food crops in
176 Europe, based on relative yield reductions in response to increasing ground-level ozone concentrations based on
177 the following expressions:

$$178 \quad \text{For wheat} \quad \text{RY} = -0.0161 \cdot \text{AOT40} + 0.99$$

$$179 \quad \text{For maize} \quad \text{RY} = -0.0036 \cdot \text{AOT40} + 1.02$$

$$180 \quad \text{CPL} = (1 - \text{RY}) \cdot \text{CP}$$

181 where RY is the yield reduction relative to the theoretical yield without ozone damage, CPL is the crop
182 production loss, CP is the actual crop production for 2000 and AOT40 is the accumulated exposure to ozone
183 concentrations above a threshold of 40 ppbv (28). AOT40 is accumulated during daylight hours (08:00 to 20:00)
184 for the three-month growing season, May to July, for Europe (29). These parameterizations are based on
185 extensive field studies and use Europe-specific values for the intercepts and gradients (28,29). While the
186 response of vegetation to increasing atmospheric concentrations of ozone is highly uncertain and expected to
187 depend on the actual flux of ozone through plant stomata (30), the use of the AOT40 metric represents current
188 policy best practice (31).

189 We do not consider the impact of ozone damage on the biomass yield of the poplar cultivated for biofuel
190 production in our scenarios. While some studies have previously suggested that carbon assimilation and hence
191 productivity are reduced in poplar clones exposed to high levels of atmospheric ozone (32), we assume that such
192 a reduction in yield would necessarily lead to the expansion of the poplar plantations in order to meet the target

193 yield of 260 Mt y⁻¹. We further assume that this would have a negligible effect on the magnitude or spatial
194 distribution of the increased isoprene emissions.

195 **Economic losses**

196 The economic losses associated with the projected number of premature deaths for each scenario was based on
197 OECD analysis of Value of a Statistical life for Europe for 2005 (33). Crop prices for the most recent 3-year
198 period (2009-2011) were taken from Eurostat (34) and averaged to estimate the cost of the simulated yield
199 reductions. Costs were converted to 2010 USD values using average exchange rates (35) and estimates of
200 deflation (36) from the US Government.

201 **Food security**

202 The calories lost for each 1 Mt loss of wheat harvest were estimated from FAO statistics for the nutritional
203 content of wheat flour and assume 5% post-harvesting losses and a 73% flour recovery during the milling
204 process (37).

205 **Results and discussion**

206 **Effects on isoprene** When large areas of very low-isoprene emitting grasses and traditional agricultural crops
207 are replaced by high-isoprene emitting poplar, total European annual isoprene emissions increase, as shown in
208 Table 1. Planting 33 Mha of commercially available poplar cultivar as biofuel crops (i.e. the land area required
209 to meet EU biomass targets for the medium-yielding group of cultivars – the “fixed area” scenario) results in
210 increases of isoprene emissions across the model domain of between 12 and 36% relative to the base case in
211 which no additional poplar is cultivated for biofuel use. It should be noted that, by contrast, planting poplar that
212 has been genetically-modified not to emit isoprene, instead of commercial cultivars, does not affect annual
213 isoprene emissions, as the assumed isoprene “leakage rate” (5%) (13) from such cultivars is roughly equal to the
214 emissions from the replaced vegetation (crops and pasture) (9).

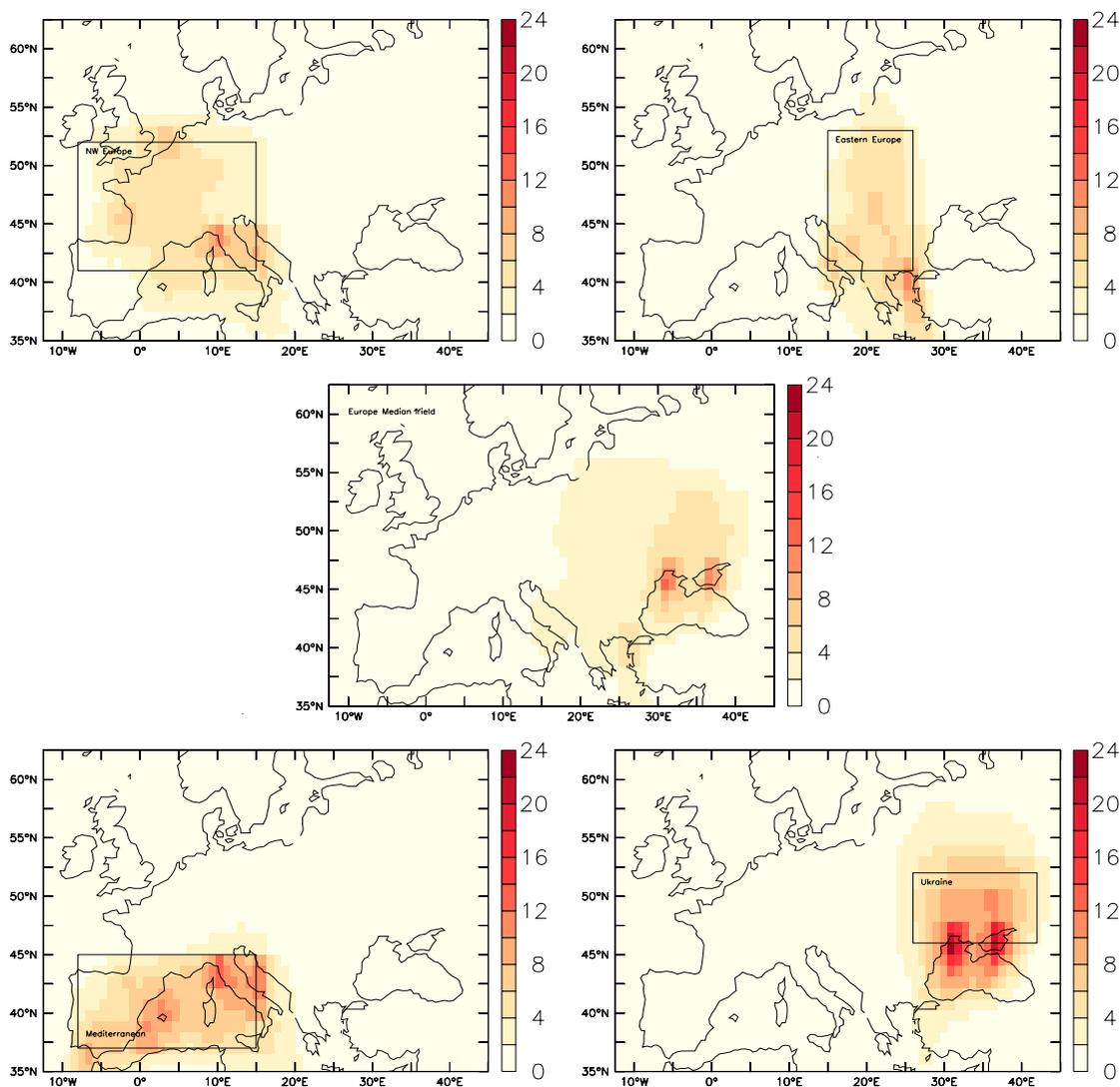
215 The effect of planting only sufficient areas of poplar to yield the woody biomass required to meet the EU yield
216 target of 260 Mt y⁻¹ (the “fixed yield” scenario) is much less variable, with increases of between 22 and 24% in
217 total isoprene emissions, due to the compensating effects of planting density and isoprene emission rate for the
218 cultivar groups – using high-yielding cultivars requires less land to be converted to plantations.

219 If all of the 33 Mha of land replanted with poplar is concentrated in specific geographical regions instead of
220 being distributed across the continent as a whole (the “regional” scenarios), total annual isoprene emissions are
221 increased by between 23 and 33% compared with baseline emissions (see Table 1). The differences between the
222 emission increases in different regions are due to regional differences in temperature and light intensity, the key
223 environmental drivers of isoprene emissions (6). Under these planting scenarios, the maximum increase in
224 emissions occurs when the plantations are located in the hot, sunny Mediterranean region, while biofuel
225 cultivation in cooler, cloudier north-west Europe results in the smallest increase.

226 The spatial distribution of the absolute changes in isoprene emissions is strongly dependent on the distribution
227 of cultivation. As isoprene is relatively reactive with respect to atmospheric oxidizing species (atmospheric
228 lifetime of around 90 minutes (38)), increases in its atmospheric concentration are confined to the vicinity of the
229 location of the emissions.

230 **Effects on ozone** Boundary-layer concentrations of NO_x are moderately high across Europe, while emissions of
231 VOCs are generally relatively low (39). Hence the boundary-layer atmospheric chemistry of the region is
232 sensitive to increased emissions of volatile organic compounds, with the cycle of radical reactions resulting in
233 enhanced production of ozone in the lower troposphere as emissions rise (3,39). While increases in annual mean
234 concentrations of ground-level ozone are modest when considered over the entire domain in all scenarios
235 (reaching around 2% for the commercially available poplar cultivars), increases in monthly mean concentrations
236 for July (when isoprene emissions peak in Europe) can be over 2 ppbv when averaged across the domain as a
237 whole and as high as 18 ppbv for some source locations.

238 **Figure 1. Increases in July monthly mean ground-level ozone concentrations across all of Europe for each of the regional planting**
239 **scenarios.** The centre panel shows the same for the median yielding fixed-land planting scenario for comparison. The boxes drawn on
240 each panel show the extent of the area in which the biofuel poplar plantations were located in each scenario.



241

242 Under the regional planting scenarios, where substantial increases in isoprene emissions and concentrations are
243 confined to small areas, the effects on ground-level ozone are more pronounced although localized to the region
244 of cultivation. Although domain-wide changes are of similar magnitude to those simulated under the fixed area
245 and fixed yield scenarios, increases of up to 9 ppbv occur in the July monthly mean ozone concentration in
246 Ukraine, as shown in Figure 1, where high background levels of NO_x are exacerbated by ideal photochemical
247 conditions for ozone production. When cultivation is limited to the Mediterranean ground level annual and July
248 monthly mean concentrations reach 44 and 51 ppbv respectively, compared with 40 and 45.5 ppbv in the base
249 case with no LUC. The smallest increases (of 2.5 and just under 4 ppbv, up from 36 and 41 ppbv) are seen in the
250 cooler, cloudier north-west of Europe.

251 Because background levels of ground-level ozone across Europe are rising (39), even the small increases
252 resulting from the realistic planting scenarios developed in this study are sufficient to raise ozone mixing ratios
253 above 40 ppbv in many locations. This is the concentration of ground-level ozone above which adverse effects
254 on both human health and crop yields are thought to be observable (26,29).

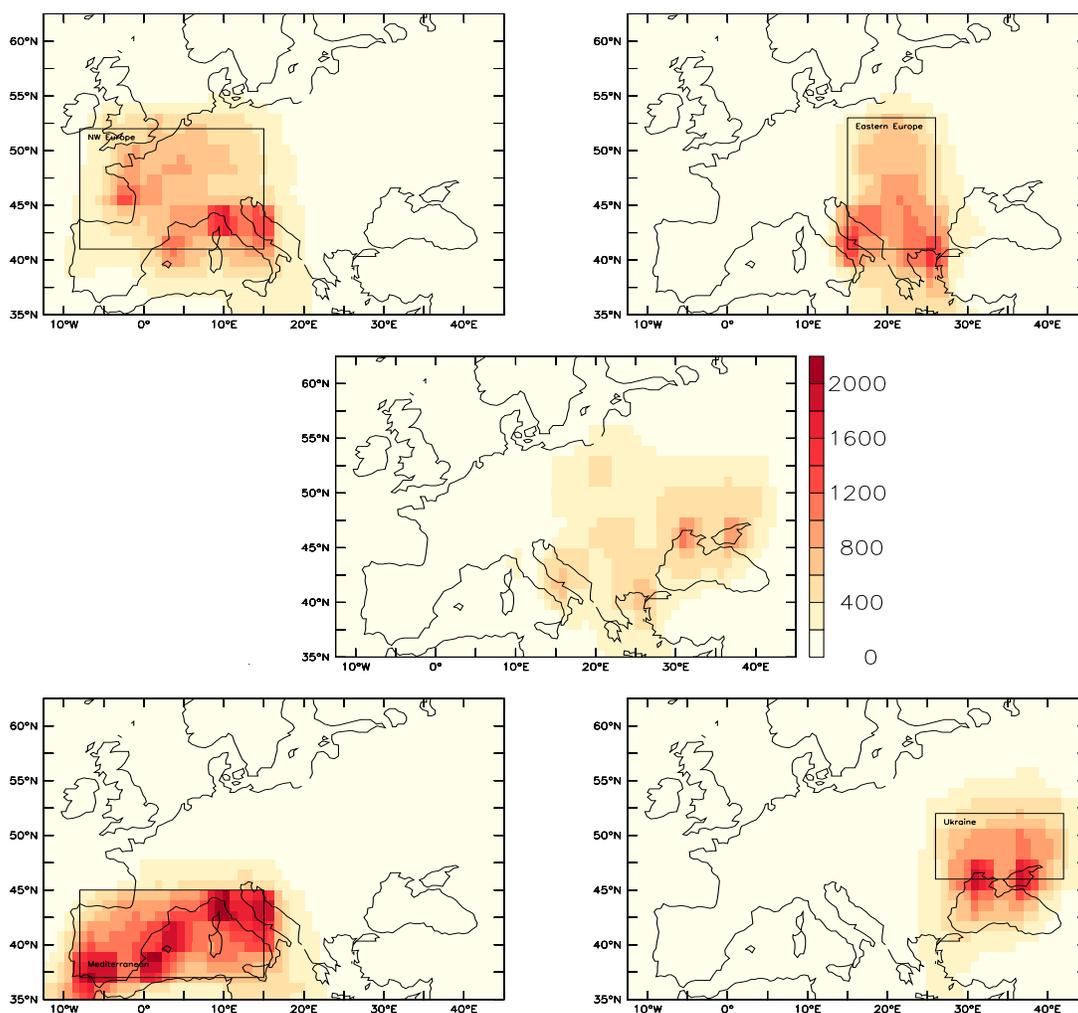
255 To put the increases simulated in this study into context, recent modeling studies show that projections of
256 ground-level ozone concentrations in Europe are strongly dependent on changes in both climate and precursor
257 emissions. Most agree that meteorological changes will enhance ozone production over most of the region,
258 although decreases may be observed in the Mediterranean. In particular, changes in climate associated with
259 RCP8.5 are projected to increase summertime domain-averaged mean ozone concentrations by around 1.5 ppb
260 per decade (40). However, taken in combination with assumed future decreases in NO_x emissions in the region,
261 some regions may experience decreases in ozone concentrations as the chemistry becomes NO_x rather than VOC
262 limited (40).

263 The increases in ground-level ozone concentrations affect daily maximum 8-hour ozone, the metric used to
264 assess potential health and ecosystem impacts. Figure 2 shows the increase in the accumulated exposure to 8-
265 hour ozone above a threshold of 35 ppbv for each of the regional cultivation scenarios. As in Figure 1, the
266 changes are mostly limited to the region of LUC, although some downwind transport is observable. By contrast,
267 however, the changes are highest in the Mediterranean, where background levels of ozone are already high.
268 Although the absolute changes are higher in the Ukraine, these are not always sufficient to raise ozone above 35
269 ppbv as background concentrations are lower. While increases in 8-hour ozone in NW Europe are lower still, the
270 magnitude of the changes in accumulated exposure is similar, particularly in areas where ozone levels are
271 already high.

272 In addition, the EU sets a limit of 60ppbv in 8-hour ozone, as recommended by WHO (5). Days on which this is
273 exceeded (known as “exceedance days” are reportable, with a limit on the number of exceedance days at any
274 monitoring location set at 3 per year. Table 2 shows the increase in total number of reportable days across
275 Europe for each cultivation scenario. Panels (c) and (f) in Figure 2 show the spatial distribution of these changes
276 for the Ukraine and NW Europe cultivation scenarios respectively. While the total number of exceedance days

277 reflect the changes in mean concentrations for the fixed land and fixed yield scenarios in which cultivation
278 occurs throughout Europe, when confined to a small region there are considerable differences, as the current
279 background level of ozone varies markedly between the regions. In line with the increases in 8-hour ozone
280 outlined above, the biggest increase in number of exceedance days occurs in the Mediterranean. By contrast,
281 however, the relatively low concentrations in the Ukraine under current land cover mean that in spite of the
282 large increases in ground-level ozone in this region, 8-hour ozone concentrations still exceed 60 ppbv less often.
283 However, many locations would exceed the 3 day per year reporting threshold.

284 **Figure 2. Increases in annual accumulated ozone exposure across all of Europe for each of the regional planting scenarios. As**
285 **Figure 1, but showing the increases in the annual accumulated exposure to daily maximum 8-hour ozone concentrations over a threshold**
286 **of 35 ppv (in ppm days).**



287
288 The atmospheric lifetime of ozone in the lower troposphere is sufficiently long (of the order of a few days) to
289 allow transport from source locations over 100s of km. Transport from rural areas (i.e. the areas of cultivation)
290 to urban areas is significant in terms of human health impacts as the additional ozone generated as a result of

291 biofuel cultivation penetrates into areas with high population densities. Transport over these distances allows
 292 sufficient time for the air mass to become well-mixed and ozone concentrations to become relatively uniform.

293 **Impacts of changes in ground-level ozone concentrations**

294 Table 2 shows the changes in ground-level ozone concentrations and resulting impacts for Europe under each
 295 biofuel planting scenario. Figures in parentheses for the “fixed yield” scenarios indicate the upper and lower
 296 bounds used in the sensitivity tests performed to constrain the uncertainties in our estimates (see text above and
 297 SI for further details).

		Changes in ground-level ozone concentrations (ppbv)		Changes in number of exceedance days	Impacts (annual)		Economic losses (annual) (2010 US\$ billion)	
		Annual mean	Monthly mean	Additional days/year	Additional mortality/year	Crop losses (Mt/year)	Additional mortality/year	Crop losses (Mt/year)
Base case		(35.2)	(38.8)	(24680)	(22,000)	(14.3)	-	-
Fixed area (33.3 Mha)	Hi	0.59	2.04	13921	1040	7.87	5.4	1.5
	Mid	0.40	1.42	9146	710	5.42	3.7	1.1
	Lo	0.21	0.77	4495	380	2.90	2.0	0.6
	GMO	0.007	0.026	163	15	0.14	0.08	0.03
Fixed yield (260 Mt)	Hi	0.41	1.44	9324	720 (700, 820)	5.49 (5.31, 5.66)	3.7	1.1
	Mid	0.40	1.42	9146	710 (700, 825)	5.42 (5.39, 6.33)	3.7	1.1
	Lo	0.39	1.36	8754	680 (650, 850)	5.20 (4.99, 6.53)	3.5	1.0
	GMO	0.007	0.027	173	15	0.15	0.10	0.03
Regional (33.3 Mha)	Ukr	0.33	1.45	7985	490	3.96	2.6	0.8
	EEu	0.39	1.06	10075	725	5.42	3.8	1.1
	NW	0.54	1.43	10011	1210	8.65	6.3	1.7
	Med	0.64	1.50	11283	990	4.25	5.2	0.8

298

299 *Impacts on human mortality*

300 Ozone is a powerful oxidant known to cause cellular damage with consequential effects on both chronic and
 301 acute cardio-respiratory diseases (39,6). Such diseases result in increased ill-health, hospital admissions,
 302 morbidity and mortality. Epidemiological studies have shown clear and statistically significant links between
 303 high-ozone events and health impacts (25,26). Meta-analyses of such studies have led to the quantification of
 304 increased mortality arising from measured increases in ground-level ozone concentrations (6,26). Here we use a
 305 numerical relationship developed specifically for Europe (26), further details of which are given in the Methods
 306 section.

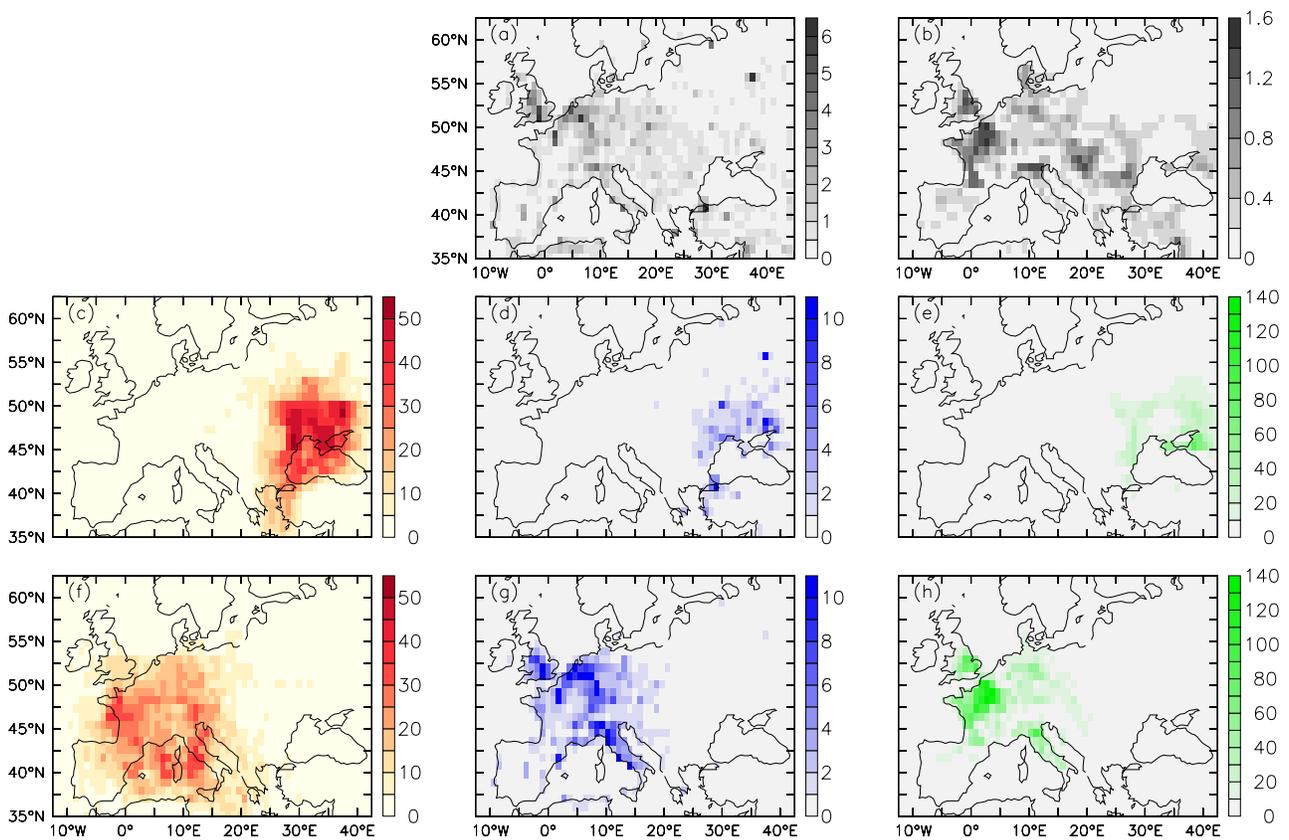
307 When a fixed area of land (33 Mha) is identified and used for biofuel cultivation, the projected increases in
 308 ground-level ozone concentrations are substantial enough under all our scenarios to increase ozone-associated
 309 mortality. If high-yielding commercially available cultivars are planted at locations throughout Europe our
 310 simulations suggest around 1040 additional deaths per year may result, an increase of ~5% in the 22000 annual

311 premature deaths currently attributed to ground-level ozone in Europe (31). Increases in mortality of around 3%
312 are projected for the medium- and low-yielding cultivars.

313 Again, the impact of the biofuel cultivation is strongly dependent on the region in which the poplar plantations
314 are located, as shown in the central column of Figure 3. If the 33 Mha of medium-yielding cultivars is planted
315 only in the populous NW of the continent (a scenario which yields sufficient biomass to meet the EU's 2020
316 targets), the impact on mortality is more substantial (~6%). Under our regional planting scenarios, the minimum
317 increase in the number of premature deaths (~2%) occurs when the plantations are located in the more sparsely
318 populated Ukraine. Although small in number, an increase of this magnitude may be sufficient to offset the
319 number of deaths avoided through implementation of emissions control policies in Europe (7). By contrast, the
320 use of a genetically modified poplar that does not emit isoprene produces no additional ozone and hence causes
321 no additional mortality across the domain.

322

323 Comparison of the different cultivars on a fixed yield (260 Mt) basis with the planting area distributed across



324 Europe shows that the number of additional deaths due to high levels of ground-level ozone is around 3%
325 relative to the base case for all commercial cultivar types. Based on upper and lower bounds for the isoprene
326 emission factors and biomass yields for the three cultivar types, we estimate the total number of premature
327 deaths to lie between 650 and 850 per annum.

328 **Figure 3 Human health and crop impacts of the regional cultivation of poplar for biofuels.** Panel (a) shows the population
329 distribution for Europe for 2006 (taken from the LandScan database (45)) and (b) the yield of wheat and maize (in Mt) for 2000 (46).
330 Panel (c) shows the increases in the number of days on which the daily maximum 8-hour ozone concentration exceeded the EU reporting
331 threshold of 60ppbv (“exceedance days”) for the Ukraine scenario; (d) the number of deaths brought forward annually as a result of the
332 changes in ozone for the Ukraine scenario; (e) the loss of wheat and maize production (in kt) as a result of the changes in ozone during
333 the growing season for the Ukraine scenario. Panels (f) to (h) show the same as (c) to (e) for the NW Europe scenario.
334

335 Economically, these additional deaths represent a cost to Europe of 2 - 6 billion USD (based on the 2010 dollar
336 value). Increases in morbidity and the associated workdays lost and hospital admissions are not accounted for in
337 this analysis. Furthermore, we have not assessed the impacts on human health of changes in the formation of
338 SOA arising from the increased bVOC emissions. SOA formation is critically dependent on precursor
339 emissions, and observations suggest that condensable products of biogenic origin mainly partition to the aerosol
340 phase in areas of high anthropogenic influence (41). The increases in isoprene emissions projected here are
341 expected to result in elevated concentrations of fine particulate matter in urban areas. As no threshold has been
342 observed for health impacts of fine particles any increase in concentration would result in increased mortality
343 (6). Thus, our assessment of the number of premature deaths resulting from the cultivation of poplar for biofuel
344 feedstocks should be seen as a lower bound.

345 *Impacts on crop yields*

346 High concentrations of ground-level ozone result in damage to plant cells, impairing photosynthesis and leading
347 to reduced carbon assimilation and ultimately lower biomass yields (29). Field studies have demonstrated
348 quantifiable reductions in yields from agricultural crops in Europe resulting from exposure to high
349 concentrations of ground-level ozone during the growing season, leading to the development of numerical
350 relationships used by regulators and policy-makers in the EU to estimate crop damage resulting from ozone
351 pollution (28,29). Details are given in the Methods section.

352 Wheat and maize are two of the most important crops in Europe, with annual yield losses due to ozone damage
353 currently estimated to be around 14 Mt y⁻¹ (28). Under the biofuel cultivation scenarios used in this study we
354 estimate that further losses ranging from just under 3 to 9 Mt y⁻¹ of wheat and maize could occur due to the
355 increases in ground-level ozone concentrations arising from enhanced isoprene emissions associated with poplar
356 cultivation. For the fixed yield scenarios, we estimate that crop production losses lie between 5.0 and 6.5 Mt y⁻¹.
357 This represents additional losses of as much as 60% of those currently attributed to elevated ground-level ozone
358 in Europe.

359 Again the impacts are highly dependent on the planting region, as can be seen in the right-hand column of
360 Figure 3. Locating poplar plantations in NW Europe, where the dominant crop is wheat which is highly sensitive

361 to ozone damage, results in higher crop losses ($\sim 8.7 \text{ Mt y}^{-1}$) than locating the same plantations in the Ukraine,
362 where the primary crop of maize is less sensitive ($\sim 4 \text{ Mt y}^{-1}$).

363 The economic costs of these additional reductions in crop yield are between 600 and 1700 million USD (at 2010
364 values). While this may be partially offset by the net value of the biofuel produced, the further reduction in crop
365 yields also jeopardises food security. The loss of 1 Mt y^{-1} of wheat is equivalent to the loss of total calorific
366 intake for ~ 2.9 million people for a year (37).

367 Our model results show that the large scale planting of poplar as a biofuel feedstock in Europe will increase
368 ground-level ozone concentrations across the region. This deterioration in air quality will lead to small but
369 quantifiable impacts on human health and mortality and crop yields, the magnitudes of which will vary with the
370 type of poplar cultivars used and the chosen locations of large plantations.

371 Recent international efforts to mitigate greenhouse gas emissions and climate change, coupled with concerns
372 about the wider environmental impacts of first-generation biofuels, and concerns regarding fuel security, have
373 led the EU to re-affirm its commitment to the increasing use of second-generation biofuel feedstocks to meet its
374 renewable energy policy and reduce its dependence on fossil fuels (42). While the land currently under poplar
375 cultivation in Europe is reported to be low ($<5\text{Mha}$) (43), several further initiatives by the EU are likely to drive
376 a rapid expansion in poplar plantations. Small trial plantations of both poplar and willow have demonstrated that
377 yields are high even on degraded and other marginal land, and that both species have beneficial effects on such
378 poor quality land (43). Furthermore, both can be used as a component of wastewater treatment processes. In
379 addition, the re-classification of the use of so-called short rotation coppice species such as poplar and willow as
380 agricultural practice, thereby including these as crops eligible for subsidies (43), makes their cultivation
381 economically attractive (particularly on poor quality land) and is likely both to drive an expansion in the area of
382 land under cultivation and encourage full and accurate reporting of this land use. While the LULCC scenarios
383 adopted in this study assume a highly aggressive expansion from the current situation, the land used has been
384 identified as available for conversion by previous research (18). It is assumed here that all biofuel plantations are
385 poplar, rather than a mix of poplar and willow in order to demonstrate the effect of the use of different cultivars
386 for which we have experimentally determined yields and isoprene emission rates. It should be noted that the
387 average yield and emission from willow species (9,15,21) is almost the same as that of our medium-yielding
388 poplar cultivar type.

389 The current focus within both policy-making circles and the biomass industry is on maximizing yields at all
390 stages of fuel production. Here we show that the choice of poplar cultivar has wider socio-economic
391 implications than climate change mitigation and profit – cultivars that are high-yielding also produce most
392 isoprene and hence have the greatest impacts on air quality. Our results clearly demonstrate that perturbations in
393 the emissions of VOCs arising from the cultivation of poplar for biofuel in Europe result in adverse effects on
394 air quality that are both cultivar and location dependent.

395 We show that the environmental conditions (light and temperature) associated with the proposed site of poplar
396 cultivation are of greatest importance in determining the effects of that site on local and regional air quality. But,

397 the impacts of the deterioration in air quality depend critically on the population density and agricultural crops
398 in the region. Further, we demonstrate that mitigation of these impacts could be achieved through European-
399 wide strategic planning of plantation siting. For example, a decision could be made to cultivate poplar on a large
400 scale in areas of Europe with low population density and geographically removed from areas of high population
401 and intensive agriculture.

402 Our findings indicate the need for a wide-reaching in-depth assessment of the implications of the cultivation of
403 biofuel feedstocks, and highlight the need for detailed local impact assessments accounting for specific cultivar
404 to be conducted on an individual case-by-case basis. Such assessments should be fully inter-disciplinary in
405 approach and include cost-benefit analyses of all aspects of the replacement of fossil fuels with cultivated
406 biofuels, including environmental effects (climate, air quality and ecosystem services), human behavior,
407 dynamics and public opinion, human health, ecosystem health and biodiversity, economic costs, energy and
408 food security, and feedbacks between changes in atmospheric composition and the Earth system. Assessments
409 such as these should focus on specific local situations, but must also consider the region as a whole, as air
410 pollutants are transported long distances and transport of the feedstock to the final market should also be a
411 consideration.

412 In addition, research is required to constrain the substantial uncertainties involved in such assessments (44).
413 These include uncertainties in the modeled ozone concentrations due to up-scaling of experimentally determined
414 isoprene emission rates and biomass yields, assumptions regarding planting location and density, uncertainties
415 associated with risk analysis using dose-response relationships derived for the population as a whole, and the
416 use of absolute concentrations rather than fluxes to assess damage to vegetation. As we account only for the
417 effects of changes in ozone our work should be seen as a lower bound estimate for the impacts associated with
418 the effect of land use change on air quality in Europe..

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424 **Supporting Information Available**

425 Supporting Information is available as a single 6-page Word document containing 1 figure (Figure S1) and 3 tables
426 (Tables S1-S3). This information is available free of charge via the Internet at <http://pubs.acs.org>.

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