

Effects of ozone air pollution on crop pollinators and pollination

Article

Accepted Version

Creative Commons: Attribution-Noncommercial-No Derivative Works 4.0

Rollin, O., Aguirre-Gutiérrez, J., Yasrebi-de Kom, I. A. R., Garratt, M. P. D., de Groot, G. A., Kleijn, D., Potts, S. G. ORCID: <https://orcid.org/0000-0002-2045-980X>, Scheper, J. and Carvalheiro, L. G. (2022) Effects of ozone air pollution on crop pollinators and pollination. *Global Environmental Change*, 75. 102529. ISSN 09593780 doi: <https://doi.org/10.1016/j.gloenvcha.2022.102529> Available at <https://centaur.reading.ac.uk/105324/>

It is advisable to refer to the publisher's version if you intend to cite from the work. See [Guidance on citing](#).

To link to this article DOI: <http://dx.doi.org/10.1016/j.gloenvcha.2022.102529>

Publisher: Elsevier

All outputs in CentAUR are protected by Intellectual Property Rights law, including copyright law. Copyright and IPR is retained by the creators or other copyright holders. Terms and conditions for use of this material are defined in the [End User Agreement](#).

www.reading.ac.uk/centaur

CentAUR

Central Archive at the University of Reading

Reading's research outputs online

1 **Title:** Effects of ozone air pollution on crop pollinators and pollination

2

3 **Authors:**

4 Orianne Rollin ^{1,2*}, Jesús Aguirre-Gutiérrez ^{3,4}, Izak A.R. Yasrebi-de Kom ^{4,5}, Michael P.D.
5 Garratt ⁶, Arjen de Groot ⁷, David Kleijn ⁸, Simon G. Potts ⁶, Jeroen Scheper ⁸, Luísa G.
6 Carvalheiro ^{1,9}

7

8 ¹ *Centre for Ecology, Evolution and Environmental Changes (cE3c), Faculdade de Ciências da Universidade de*
9 *Lisboa, Campo Grande, 17 49-016 Lisboa, Portugal*

10 ² *Beekeeping Research and Information Centre (CARI), Place Croix du Sud 1, B-1348 Louvain-la-Neuve, Belgium*

11 ³ *Environmental Change Institute, School of Geography and the Environment, University of Oxford, Oxford, UK*

12 ⁴ *Biodiversity Dynamics, Naturalis Biodiversity Center Leiden, The Netherlands*

13 ⁵ *Department of Medical Informatics, Amsterdam UMC location University of Amsterdam, Amsterdam, The*
14 *Netherlands*

15 ⁶ *Centre for Agri-Environmental Research, School of Agriculture, Policy and Development, University of Reading,*
16 *Reading, Berkshire, RG66AR, UK*

17 ⁷ *Wageningen Environmental Research, Wageningen University & Research, Droevendaalsesteeg 3a, 6708 PB*
18 *Wageningen, The Netherlands*

19 ⁸ *Plant Ecology and Nature Conservation Group, Wageningen University, Droevendaalsesteeg 3a, Wageningen*
20 *6708PB, The Netherlands*

21 ⁹ *Departamento de Ecologia, Universidade Federal de Goiás, 74001-970 Goiânia, Brasil*

22

23 * Corresponding author: Dr Orianne Rollin; orianne.rollin@gmail.com;

24 ORCID: <https://orcid.org/0000-0002-1455-9302>

25

26

27

28

29 **Abstract:**

30 Human driven environmental changes increase the concentrations of polluting reactive
31 compounds in the troposphere, such as ozone and nitrogen oxides. These changes lead to
32 biodiversity losses and alter plant physiology and plant-pollinator interactions, essential for
33 pollination services, with potential consequences for agricultural production. Here we used 133
34 unique sampling events from NW Europe to investigate how air pollution (ozone and nitrogen
35 oxides) and other sources of nitrogen is related to pollinator visitation rate and their contribution
36 to agricultural production, also considering possible interactive effects with landscape quality
37 and pesticide input. We showed that ozone modulates the effect of pesticide exposure and
38 temperature on crop pollinators, increasing the probability of negative impacts on crop
39 pollination. Indeed, when ozone levels are highest, the strength of the effect of pesticide on
40 pollinators is more than double then when ozone levels are intermediate. This indicates that air
41 pollution should be considered in management plans and policies aiming to safeguard
42 biodiversity and promote more sustainable food production practices.

43

44

45 **Key words:**

46 air pollution, bees, hoverflies, landscape change, nitrogen, ozone, pesticide risk exposure,
47 pollination

48

49

50

51

52

53

54

55 **1. Introduction**

56

57 Human activity is changing environmental conditions worldwide (Rockström et al., 2009),
58 affecting global biogeochemical flows (e.g. nitrogen, ozone; Fowler *et al.* 2013; Mills *et al.*
59 2013; Lefohn *et al.* 2018; Smil 2000) and, consequently, air, water and soil quality. In addition
60 to habitat loss and climate change (with increased greenhouse-gas contributors), environmental
61 pollution, including nitrogen deposition, is considered a major driver of biodiversity loss
62 (Mazor et al., 2018; Sala et al., 2000) and can negatively impact ecosystem functioning and
63 associated ecosystem services such as crop pollination (González-Varo et al., 2013).

64 Nitrogen deposition (estimated to be 413 Tg N yr⁻¹ in 2010) has more than doubled over the
65 last century (Fowler et al., 2013) due to emissions of ammonia (NH₃, from pecuary and
66 agriculture) and nitrogen oxides (NO_x produced in the combustion of fossil fuels). Such
67 increases have affected plant communities (Carvalho et al., 2020; Tilman et al., 2002), with
68 associated bottom-up impacts on higher trophic levels including pollinators (Carvalho et al.,
69 2020; David et al., 2019; Johnson et al., 2020; Pöyry et al., 2017; Ramos et al., 2018; Wang
70 and Tang, 2019). While scarcity of nitrogen can constrain the positive effect of pollinators on
71 crop production (e.g. sunflower; Tamburini et al., 2016; oilseed rape; Garratt *et al.* 2018),
72 excess nitrogen can also be detrimental and alter the plant's ability to compensate for the
73 absence of pollinators (Marini et al., 2015; Tamburini et al., 2017; Ramos et al., 2018). Such
74 responses are likely mediated by changes in floral resources quality and quantity, which in turn
75 can be moderated by changes in climatic conditions (Flores-Moreno et al., 2016).

76 Another important air pollutant is tropospheric ozone, a major greenhouse-gas which is also
77 phytotoxic (Mills et al., 2013; Lefohn et al., 2018; Ilić and Maksimović, 2021). Ozone levels
78 have increased since the beginning of the industrial period (estimated to up of 35%; Mills et al.,

79 2013; Guerreiro et al., 2014; IPCC, 2014). While there are other sources of ozone (e.g. volatile
80 organic compounds, carbon monoxide and methane), oxidized nitrogen (NO_x) is one of the two
81 major ozone precursors (Mills et al., 2013; Lefohn et al., 2018). Increased concentrations of
82 ozone can reduce photosynthesis and plant growth (Tjoelker and Luxmoore, 1991; Black et al.,
83 2007) and negatively affect the timing of flowering and number of flowers (Feder and Sullivan,
84 1969; Hayes et al., 2012; Leisner and Ainsworth, 2012) (Figure 1). Increased ozone
85 concentration in the air (e.g. 80-120 ppb, frequently found near urban areas; Paoletti et al.,
86 2014) can also change the concentration and emission distance of floral volatile organic
87 compounds (Farré-Armengol et al., 2016; Fuentes et al., 2016; Heiden et al., 1999; Jürgens and
88 Bischoff, 2017; McFrederick et al., 2008) and, consequently, affect pollinator olfaction and
89 foraging behaviour (Farré-Armengol et al., 2016; Fuentes et al., 2016; Vanderplanck et al.,
90 2021) (see Fig. 1). These effects on plant pollinator interactions may partly explain the reported
91 negative effects of ozone on seed and fruit production detected in previous studies (Farré-
92 Armengol et al., 2016; Fuhrer et al., 2016; Mills et al., 2013). Yet, few studies have explored
93 the effects of air pollution (e.g., nitrogen oxides and ozone) on pollinator foraging patterns and
94 efficiency, and if the strength and direction of such effects depends on other important
95 environment drivers, such as pesticide use (Walker and Wu, 2017) or land use (Mazor et al.,
96 2018; Sala et al., 2000).

97 Taking into account potential interactive effects with landscape quality for pollinators (i.e.,
98 natural and semi-natural vegetation composition) and pesticide exposure, we investigated how
99 air pollution by ozone and different sources of nitrogen compromise pollinator visitation rates
100 and their contribution to crop production (apple, blueberry, fava bean, oilseed rape). Given the
101 negative effects on flower abundance and odours described above, and the fact that previous
102 studies detected greater benefit from pollination under lower N availability (Marini et al., 2015;
103 Ramos et al., 2018), we expect that increased ozone and nitrogen will lead to declines in crop

104 pollinator visitation rates and pollination service delivery. However, availability of non-crop
105 habitats is an important determinant of pollinator abundance, richness and pollination services
106 (Dainese et al., 2019; Kennedy et al., 2013). We also expect that the effect of ozone and nitrogen
107 on pollinators and pollination will be weaker in structurally more simple landscapes (less semi-
108 natural habitat and greater risk of exposure to pesticides), where the only potential pollinators
109 would be species with greater resilience to land use intensification (Williams et al., 2010;
110 Bartomeus et al., 2013; Kremen and M’Gonigle, 2015; Kleijn et al., 2015).

111 The results of this study contribute to our understanding of interactive effects among
112 atmospheric pollution, land-use and eutrophication on crop pollinators and pollination to help
113 inform the development of new practices and policies to safeguard pollinators and crop
114 pollination.

115

116 **2. Material and Methods**

117

118 ***2.1. Pollinator and crop production data***

119 A total of 133 unique sampling events with information on pollinator visitation rate and
120 pollinator contribution to crop production were obtained from databases of previous studies,
121 sampled in various crops in the UK (Garratt et al., 2014b, 2014a, 2014c, 2016) and the
122 Netherlands (De Groot et al. *unpublished data*). A unique sampling event is defined by their
123 sampling year, crop species and spatial location (see dataset details in Table 1, Fig. 2).
124 Pollinator data was collected using transects surveys over a defined distance and time, recording
125 visitors to crop flowers as *Apis* or non-*Apis* species (including bees and hoverflies). At each
126 site, pollinator contributions to crop production were measured using pollinator exclusion
127 treatments and compared with open controls to establish a proportional contribution of insect

128 pollinators to production (for further methodological details see Garratt et al., 2014a, 2014c,
129 2016, and De Groot unpublished data in supplementary material).
130 Whenever studies provided more than one crop production metric, we selected the most
131 pertinent variable to define crop production: seed set for oilseed rape, pod set for fava bean (see
132 Garratt et al., 2014b) and fruit set for blueberry (see Kendall et al., 2020). For apple, studies
133 conducted in the Netherlands gave information on fruit quality, i.e., fruit weight. For studies in
134 apple orchards in the UK, data available concerned final fruit set at harvest. For each
135 experimental branch, the number of apples which had developed on experimental
136 inflorescences was recorded (see Garratt et al., 2014a, 2014b, 2016).
137 As data from different studies applied different methodologies to extract information on
138 pollinators and pollination, we calculated z-scores within each study for crop pollination (i.e.,
139 contribution of pollinators to crop production) and pollinator abundances (*Apis* and non-*Apis*
140 pollinators separately). This measure allows for the standardisation of scores with respect to the
141 other scores into the same group (crop/year) (Garibaldi et al., 2011, 2015).

142

143 **2.2. Ozone and NO_x data**

144 Information on atmospheric nitrogen (NO_x) and ozone (O₃) were obtained from the
145 Tropospheric Monitoring Instrument (TROPOMI), hosted by the European Space Agency's
146 (ESA) Sentinel-5P satellite under the Copernicus programme
147 (<https://sentinel.esa.int/web/sentinel/missions/sentinel-5p>). The Sentinel-5 Precursor mission is
148 the first Copernicus mission dedicated to monitoring our atmosphere and provides information
149 and services on air quality, climate, ozone (O₃) and Nitrogen dioxide (NO₂) between the surface
150 and the top of the troposphere and the ozone layer. The spatial resolution of the Sentinel-5P is
151 7×3.5 km. Data of O₃ and NO₂ were extracted using the NASA Panoply 4.11.1 software
152 (NASA, 2020) (Fig. 2).

153 To generate mean NO₂ and O₃ values over our specific sites, we extract daily values from
154 TROPOMI layers between May 2019 (first of the TROPOMI-Sentinal5P products was released
155 at the end of April 2019) and September 2019. This specific period covers the period of activity
156 of most pollinators in the study region (Balfour et al., 2018; Peeters et al., 2012) and therefore
157 to the period when they may be most exposed to atmospheric pollutants. We did not include
158 data from 2020 in our mean calculation, due to the unusual change in human activity caused by
159 covid-19 health crises. While nitrogen oxides are one of the several precursor of ozone (Mills
160 et al., 2013), O₃ and NO₂ are not correlated ($cor = 0.070$; $p\text{-value} = 0.421$).

161

162 ***2.3. Agricultural nitrogen input data***

163 Estimated average total annual application of manufactured nitrogen (1km resolution,
164 kg/km²/year) was extracted for England from the raster CEH Land Cover® plus Fertilisers
165 (CEH, Wallingford, UK; <https://www.ceh.ac.uk>). CEH dataset used data from Defra British
166 Survey of Fertiliser practice (2010-2015) to derive average annual application of manufactured
167 fertilisers for each crop type and then derived total application at 1km resolution using crop
168 areas from CEH Land Cover® plus: Crops (averaged 2015-2017) (Osório et al., 2019). As
169 changes in land use intensity in the UK were limited, with trends stable overall since 1994
170 (Martay et al., 2018), we assume that values based on these maps are representative for the
171 sampling years (2011 and 2012).

172 For the Netherlands, mean values of nitrogen fertilizer application rate by crop were extracted
173 from the database of the Netherlands Enterprise Agency (RVO) for 2016 (*Gewascodelijst*
174 *Stikstofgebruiksnormen*; <https://english.rvo.nl>). To estimate the mean value of nitrogen applied
175 as fertilizer at 1km resolution (in kg/km²/year), we calculated a weighted average, taking into
176 account the proportion of each crop in the landscape. Crop coverage per site were extracted for

177 each 1km² cell as an average of the BRP (*Basisregistratie gewaspercelen*) shapefiles 2015 and
178 2016 obtained from the RVO (<https://english.rvo.nl/>).

$$179 \quad N_{Fer} = \sum (Proportion\ of\ each\ crop\ category \times mean\ annual\ application\ rate\ for\ the\ category)$$

180 These years (2015-2016) correspond to the median of study years in the Netherlands included
181 in the analyses, that are 2013, 2014, 2017 and 2018.

182

183 **2.4. Pesticide input data**

184 To estimate average level of pesticide applied per crop at each field site (1km buffer), we
185 calculated a pesticide risk index (*RI*), including herbicides, insecticides, molluscicides and
186 fungicides, using the methodology described by Yasrebi-de-Kom et al., (2019) as:

$$187 \quad RI = \sum HQ = \left(\frac{Application\ rate\ (g.\ ha^{-1})}{Toxicity\ (LD_{50}\ in\ \mu g\ per\ bee)} \right) > 50$$

188 with *HQ* the hazard quotient (HQ) of each active molecule and the median lethal dose per bee
189 (*LD*₅₀). The median lethal dose is one way to measure the short-term poisoning potential (acute
190 toxicity) of a substance. The *LD*₅₀ is the amount of a substance, given all at once, which causes
191 the death of 50% of a group of test animals. The hazard quotient ratio gives an approximation
192 of how close the likely exposure of bees is to a toxicologically significant level. The pesticide
193 risk index (*RI*) was defined as the number of high risk active ingredients (*HQ*>50; see EPPO,
194 2010) that were applied. If *HQ*<50, the active ingredient was categorized as low risk to bees.

195 The *LD*₅₀ of 390 active ingredients used in the UK and the Netherlands were extracted from the
196 “*Pesticides Properties DataBase*” (PPDB) from the University of Hertfordshire, UK
197 (<https://sitem.herts.ac.uk/aeru/ppdb/en/index.htm>; Lewis et al., 2016; Lewis and Tzilivakis,
198 2019) (see list of active ingredients in Appendix A in Supporting Information). As proposed by
199 EPPO (2010), the risk assessment was carried out selecting the lowest of the oral and contact
200 *LD*₅₀ values available across the different bee species (honey bees, bumble bees and other wild
201 bees), to take the most conservative approach for the entire bee community (see Table S2).

202 However, LD_{50} values were mainly available for honey bees, sometimes for bumble bees, and
203 much less frequently for other pollinators (Lewis et al., 2016; Lewis and Tzilivakis, 2019;
204 Yasrebi-de Kom et al., 2019).
205 For the UK, the average annual application rate at 1km resolution (in $\text{kg}/\text{km}^2/\text{year}$) was obtained
206 for 130 pesticide active ingredients from the “*CEH Land Cover® plus: Pesticides 2012-2016*”
207 (Jarvis et al., 2019) of the Centre for Ecology and Hydrology (CEH, Wallingford, UK;
208 <https://www.ceh.ac.uk>), across a four-year period (from 2012, 2013, 2014 and 2016). For the
209 Netherlands, we created the average annual allowed application rate at 1km^2 (in $\text{kg}/\text{km}^2/\text{year}$)
210 across a two-year period (2015 and 2016) for 179 pesticide active ingredients, combining
211 allowed application rates produced by Yasrebi-de-Kom et al. (2019) and the BRP shapefiles for
212 crops in 2015 and 2016 obtained from the RVO (<https://english.rvo.nl>).

213

214 **2.5. Land cover composition data**

215 The availability of crop pollinators strongly depends on landscape composition (Kennedy et al.,
216 2013; Dainese et al., 2019). Previous studies in Western Europe focused on the same crops (e.g.
217 Dainese et al., 2018; Gathmann and Tscharntke, 2002; Holzschuh et al., 2006; Shaw et al.,
218 2020; Steffan-Dewenter et al., 2002) have shown that a 1km buffer is an appropriate scale at
219 which to characterize landscape composition in agricultural contexts for the specific pollinator
220 assemblages studied here. We therefore calculated the proportion of forest and (semi-) natural
221 habitats combined in a 1km radius buffer zone for each sampling site. For the UK, data were
222 extracted from the Land Cover Map for 2015 (LCM2015; 25m resolution raster) (CEH Data
223 Licence Agreement – 1338). For the Netherlands, we merged data from the BRP shapefiles
224 2015 and 2016 (<https://www.pdok.nl/introductie/-/article/basisregistratie-gewaspercelen-brp->)
225 and the BBG (*Bestand Bodemgebruik*) shapefile 2015 ([https://www.pdok.nl/introductie/-](https://www.pdok.nl/introductie/-/article/cbs-bestand-bodemgebruik)
226 [/article/cbs-bestand-bodemgebruik](https://www.pdok.nl/introductie/-/article/cbs-bestand-bodemgebruik)) obtained from the RVO (<https://english.rvo.nl>) and

227 Statistics Netherlands (CBS, <https://www.cbs.nl/en-gb>) respectively for an optimal coverage
228 (especially for unimproved grasslands). These habitats included the proportion of forest areas
229 and (semi-) natural areas (including grasslands), but excluded agricultural improved grasslands
230 and pastures due to their generally intensive management strategy and low habitat quality for
231 pollinators (Ekroos et al., 2020).

232

233 ***2.6. Statistical analyses***

234 We used linear mixed models to analyse effects of ozone, nitrogen enrichment (i.e., including
235 both the mean values of N fertilizer application on the agricultural fields and the NO_x
236 concentration in the air from satellite data), the risk of pesticide exposure and the proportion of
237 (semi-) natural habitats and their two-way interactions on the abundance of pollinators and their
238 contribution to crop production (see correlation matrix in Appendix B).

239 The relation between temperature and ozone is not yet well established. While some studies
240 show that temperature can be a good predictor of ozone concentration (e.g. Stathopoulou et al.,
241 2008) others show no clear relationship between the temperature and ozone (e.g., Mahmood et
242 al., 2020). In our study tropospheric ozone and temperature are not correlated ($cor = -0.086$; p -
243 $value = 0.336$). Therefore, as temperature can be an important parameter altering pollinator
244 assemblages (Bartomeus et al., 2011; Duchenne et al., 2020) and visitation rates (Abou-Shaara
245 et al., 2017; Clarke and Robert, 2018; Usha et al., 2020), we included the mean annual
246 temperature (using for each sampling event data extracted from the Global Climate Monitor
247 System; Camarillo-Naranjo et al., 2018) in each pollinator model, also taking into account
248 potential interactive effects with other variables.

249 The local abundance of honey bees is primarily determined by beekeeper behaviour rather than
250 local effects of habitats (Büchler et al., 2014; Wood et al., 2020). As managed species they are
251 influenced differently by environmental pressures compared to wild pollinators, and we

252 therefore analysed *Apis mellifera* separately from non-*Apis* pollinators (i.e., other bees and
253 hoverflies).

254 To account for variation associated with the crop system on pollinators and pollination, crop
255 identity was included as a random effect in all models. Moreover, to remove potential
256 confounding effects with study region or country, all explanatory variables included in each
257 model were centered within study-year combinations (Van de Pol and Wright, 2009).

258

259 As previous studies have also shown that densities of non-*Apis* pollinators can in some
260 circumstances be negatively affected by honey bee densities (e.g., Lindström et al., 2016;
261 Geslin et al., 2017; Mallinger et al., 2017), honey bee abundance was included as an explanatory
262 variable in non-*Apis* pollinator models. For the analysis of the contribution of pollinators to
263 crop production, in addition to sources of eutrophication, ozone pollution, pesticide risk and
264 proportion of (semi-) natural habitats, we included abundance of honey bees (*Apis mellifera*)
265 and non-*Apis* pollinators as covariates.

266 First, to test for spatial autocorrelation, we compared models with different spatial correlation
267 structures (exponential, Gaussian, Linear, rational quadratics and spherical spatial
268 autocorrelation) with those including no spatial correlation structure, and defined the best
269 random structure of the model based on their AICc scores (Akaike Information Criterion for
270 small samples). Then, we applied model selection to the fixed terms of the model ($\Delta\text{AICc} < 2$
271 with the best model; Anderson et al., 2001). To not overfit the global model in relation to our
272 sample size, the number of parameters in each tested model was restricted to 5 (including
273 potential interaction effects). Selection of the best candidate models are presented in
274 Supplementary material (see Appendix C, D and E in Supporting Information).

275 We used Variance Inflation Factors (VIF) to detect potential multicollinearity between
276 variables in our selected statistical models, and no correlation was detected. Indeed, all VIF

277 values were less than 1.5. In the selected models of crop pollinator abundance (*Apis* and non-
278 *Apis*), VIF values ranged between 1.176 and 1.467, and in for the selected model of pollinator
279 contribution to crop production, VIF values ranged between 1.001 and 1.114.

280 All analyses were computed using the *ape* (Paradis et al., 2019), *nlme* (Pinheiro et al., 2020)
281 and *MuMIn* (Bartoń, 2011) packages in R software, version 3.4.2 (R Development Core Team,
282 2018). All spatial extraction or landscape index calculation from shapefile and raster maps were
283 made using QGIS software version 3.10 A Coruña (QGIS Development Team, 2020).

284

285 **3. Results**

286

287 The observed abundances of pollinators at each sampling transect varied from 0 to 367 for
288 honey bees (*Apis*) and from 0 to 154 non-*Apis* pollinators (i.e., wild bees and hoverflies).

289 Mean ozone value per study site varied from 0.140 to 0.144 mol.m⁻² in the Netherlands and
290 from 0.142 to 0.147 mol.m⁻² in UK. These values represent ca. 7% of the full range of ozone
291 values throughout the world (0.079 and 0.222 mol.m⁻², including areas of high ozone pollution
292 as well as areas of low tropospheric ozone concentration). Previous studies that also used
293 remote sensing data Sentinel-5P (e.g., Naqvi et al., 2022; Roman-Gonzalez et al., 2020), show
294 a similar ranges of tropospheric ozone (and NO₂) concentration for areas of comparable size
295 (i.e., country scale: Peru, India). The mean tropospheric NO₂ per study site ranged from 27.8 to
296 76.5 μmol.m⁻² (from 0 to 2.14 mmol.m⁻² worldwide) and the gradient of fertilizer N input varied
297 from 2.28 to 21.09 t.km⁻² (2.28 to 12.32 in UK and 3.89 to 21.09 in the Netherlands). The risk
298 index of pesticide exposure varied between 2 to 8 in the Netherlands and between 2 to 10 in
299 UK, (i.e., between 2 and 10 high risk active molecules were applied in the 1km buffer around
300 study sites). The proportion of natural and semi-natural habitats in the 1km² surrounding buffer

301 varied from 0.1 to 37% in UK (with a mean=8.9 and median=4.4) and from 0 to 47% in the
302 Netherlands (with mean=7.0 and median=1.8).

303

304 We observed effects of pesticide risk exposure and ozone on crop pollinator abundance as well
305 as interacting effects between these two factors, but such effects differed between *Apis* and non-
306 *Apis*. We found that abundance of honey bees (*Apis mellifera*) in crops was negatively related
307 to concentration of ozone (*t-value* = -1.12) but positively related to the risk of pesticide
308 exposure (*t-value* = 1.27; see Appendix C). However, the positive relationship observed
309 between the abundance of honey bee and the risk of pesticide exposure became less strong with
310 increasing ozone levels (Fig. 3A; see Appendix C). In other words, the negative effect of ozone
311 on *Apis* pollinators is more accentuated when this risk of exposure increases (see Appendix F).
312 As for non-*Apis*, the negative effect of pesticides on abundance (*t-value* = -1.77) was more
313 accentuated at higher ozone exposition (Figure 3B; see Appendix D). As expected, temperature
314 influenced bee abundance. It had a positive effect on honey bees' abundance (*t-value* = 1.11;
315 see appendix C) and a negative effect on the abundance of non-*Apis* pollinators (*t-value* = -
316 1.33; see Appendix D).

317 As hypothesized, we found a negative correlation between ozone and the contribution of
318 pollinators to crop production (i.e., crop pollination assessed by the difference of production
319 between open and close treatments; *t-value* = -1.50, see Appendix E and F), but also an
320 interacting effect between ozone and the risk of pesticide exposure on crop production. While
321 at low concentration of ozone the risk index (RI) of pesticide exposure was positively related
322 to the contribution of pollinators to crop production, the relationship became negative when
323 ozone levels were high (Figure 3C; see Appendix E).

324

325 Contrary to our expectations, we found no evidence of a relationship of any Nitrogen sources
326 studied here (i.e., atmospheric nitrogen dioxide deposition and mean application rate of nitrogen
327 fertilisers at 1km resolution) on the abundance of *Apis* and non-*Apis* pollinators nor on the
328 contribution of pollinators to crop production (see Appendix C, D and E). We also did not
329 observe evidence of a correlation between the proportion of semi-natural habitats and the
330 abundance of crop pollinators or on their contribution to crop production.

331

332 **4. Discussion**

333

334 Despite the recognised influence that ongoing human driven changes on nitrogen and ozone
335 availability have on plant communities (Fowler et al., 2013; Mills et al., 2013; Guerreiro et al.,
336 2014), little is known about how such changes impact pollinators and the services they provide
337 to crop pollination, or how this interacts or is moderated by other drivers of pollinator decline.
338 Recent studies showed that ozone pollution can impact directly and indirectly on many
339 fundamental ecological processes with consequences on biodiversity and sustainability of
340 ecosystem services, such as pollination (Duque et al., 2020; Emberson, 2020; Fuhrer et al.,
341 2016; Tai et al., 2014). Here, we highlighted that ozone is part of a complex interacting system,
342 mediating the strength of the effects pesticide exposure has on crop pollinators and the
343 contribution of these pollinators to crop production. Below, we discuss in detail the potential
344 mechanisms behind the patterns detected and the implications of our findings for conservation
345 and management of crop pollination.

346

347 ***4.1. Interacting effect of ozone with pesticide exposure***

348 As expected (Hayes et al., 2012; Leisner and Ainsworth, 2012; Mills et al., 2013), ozone levels
349 were negatively correlated to crop pollination. Recent studies have estimated that global

350 agricultural losses due to high ozone levels totalled 79–121 million metric tons in 2000 with
351 global economic losses ranging from \$11 to \$26 billion (Van Dingenen et al., 2009; Avnery et
352 al., 2011a) and predicted increases of between \$17 and \$35 billion annually by 2030 (Van
353 Dingenen et al., 2009; Avnery et al., 2011a). Such effects may be partly related to a reduction
354 in pollen germination (Leisner and Ainsworth, 2012; Taia et al., 2013; Gillespie et al., 2015).
355 Our results suggest that changes in pollination by insects (due to changes in flower visitation
356 patterns) may also play an important role.

357 The fact that increasing ozone levels modified the response of crop pollination to pesticide
358 exposure (which turns from positive to negative) may be related to pest control. Farmers widely
359 use pesticides to minimize infestations by pests and protect crops from potential reduction of
360 crop production, both in quality and quantity (Damalas, 2009), and hence positive effects of
361 pesticide use on production are expected if pests are more limiting than pollinators to
362 production.

363 It is however possible, that in more degraded environments, i.e., with a higher level of ozone
364 pollution, the cost/benefit ratio of pesticides on crop production changes. In less intensive
365 landscapes with a higher pollinator pool, the negative impact of pesticides on pollinators and
366 resulting crop pollination is compensated for by improvement in pest regulation by pesticides.
367 However, in highly intensive landscapes, due to scarcity of pollinators limiting pollination and
368 crop production, the negative effects of pesticides on crop pollinators (which are more
369 accentuated under high ozone levels, Fig 3) may outweigh the positive effects on pest reduction
370 on crop production.

371 The negative relationship between ozone pollution and flower visitor abundance could be due
372 to changes in plant-pollinator communication and flower attractiveness affecting crop
373 pollinator foraging behaviour. Previous studies have shown that ozone induces changes in
374 availability of floral resources by modifying flowering time and number of flowers, with some

375 plant species being particularly sensitive (Hayes et al., 2012; Leisner and Ainsworth, 2012;
376 Mills et al., 2013). Ozone also alters pollinator decision-making, modifying and reducing the
377 volatile floral scents (Farré-Armengol et al., 2016; Fuentes et al., 2016; Saunier and Blande,
378 2019; Vanderplanck et al., 2021) and damaging pollinators olfactory organs (Dötterl et al.,
379 2016; Vanderplanck et al., 2021).

380 The fact that the negative effect of pesticide exposure on non-*Apis* pollinators (Mancini et al.,
381 2019; Walker and Wu, 2017; Woodcock et al., 2017) was more accentuated under high ozone
382 concentration (Table S1) could be due to pollinator communities being less diverse and/or
383 abundant in regions with high ozone, but also to changes in pollinator assemblages. In more
384 degraded areas (high pesticide exposure, high ozone concentration), crop pollinator
385 communities are dominated by a handful of dominant widespread species that are more resilient
386 to intensive land use (Kleijn et al., 2015), which often have a more generalist diet and may be
387 more mobile (Biesmeijer et al., 2006; Goulson et al., 2008; Connop et al., 2010). Consequently,
388 in such regions, i.e., under high level of pesticide exposure, the negative effect of ozone on non-
389 *Apis* crop pollinators might be less detectable.

390 Although the negative impact of pesticides on honey bees is well known (e.g. Mancini et al.,
391 2019; Walker and Wu, 2017; Woodcock et al., 2017; Park et al., 2015; Tosi et al., 2017), we
392 found that pesticide exposure was positively related to honey bee density in crops. This result
393 is probably due to beekeeping management strategies that are likely more frequent in intensive
394 agricultural areas where the demand for colony supply to ensure efficient pollination is high
395 (Garibaldi et al., 2017; Rollin and Garibaldi, 2019), masking (and even compensating) the
396 negative effects of pesticides. However, the positive relationship between abundance of honey
397 bees in crops and pesticide exposure was lower when ozone concentration increased. This can
398 reflect the negative effect of pesticides on honey bees, decreasing the pollination efficiency and

399 survival of honey bees (Prado et al., 2019), despite the local increase of individuals due to the
400 import of colonies by beekeepers in intensive farming systems.

401

402 *4.2. Effect of the temperature on pollinators*

403 The negative effect of the increase in mean annual temperature on the abundance of wild
404 pollinators are in agreement with recent studies showing that climate change characterised by
405 events of extreme heat, shifts European pollinator phenology with consequences on pollinators
406 assemblages (Bartomeus et al., 2011; Duchenne et al., 2020; Vasiliev and Greenwood, 2021).
407 The decade 2011-2020 (during which our studies were carried out) has been the hottest on
408 record since the preindustrial period, with several summer heat waves being recorded in
409 Northern and Western Europe (see WMO; <https://public.wmo.int/en>). Therefore, regions with
410 slightly milder temperatures might be preferable for native pollinators. The fact that the
411 negative effect of annual temperature on the abundance of non-*Apis* pollinators was reduced or
412 became null at higher ozone concentration, could be due to the nature of pollinator species
413 assemblages in regions with higher ozone levels. Further studies are needed to better understand
414 if species persisting in regions of higher ozone levels have traits that affect their response to
415 temperature. Nevertheless, this result is consistent with other studies that have found that the
416 impact of temperature on pollination and crop production was less accentuated under high
417 ground-level ozone concentrations (Mahmood et al., 2020). Yet, some species with a broad
418 temperature range are likely to cope well with higher temperatures, since higher temperatures
419 reduces the energy cost necessary to maintain optimum body temperature improving bee
420 foraging activity (Abou-Shaara et al., 2017; Corbet et al., 1993; Kwon and Saeed, 2003). This
421 could explain the positive effect of temperature on honey bee abundance observed here
422 (foraging temperature ranging from 21.0 to 33.5 °C, Usha et al., 2020). Moreover, as it is a
423 managed species, abundance of honey bees is primarily determined by beekeeper behaviour

424 and do not have to adapt in the same way to environmental changes (Büchler et al., 2014; Wood
425 et al., 2020).

426

427 *4.3. Effect of nitrogen enrichment*

428 Contrary to our expectations, we did not observe effects of nitrogen enrichment sources on crop
429 pollinators and pollination. It is possible that the proxies used in our study do not adequately
430 represent the real nitrogen exposure levels in our study fields. Indeed, while pollinators can be
431 affected by local (i.e., within field) changes in nutrient availability (David et al., 2019), our
432 proxies for nitrogen levels are taken at much broader scales. The amount of nitrogen that is in
433 reality deposited in a specific location of the biosphere may not be well represented by the NO₂
434 levels measured in the troposphere at much larger spatial resolution of the available data from
435 the Sentinel-5 satellite (i.e., 7×3.5 km). Similarly, the estimated mean application rate of
436 fertilizers at each study region (which is based on average application levels for each crop at
437 country level, and do not consider personal decisions of landowners) may not be of a sufficient
438 resolution to detect changes in fertilization rate and its effects at the local scale. For example,
439 the presence of (semi-)natural habitats in the landscape will inevitably decrease the estimated
440 average fertilizer application rate at 1km² resolution, while a high proportion of highly enriched
441 crops, such as cereals, maize or fertilised grasslands (e.g., ray grass) will tend to increase the
442 estimated average application rate. Future work involving farmer interviews asking for the
443 actual amount of fertilizer applied to better characterize nutrient availability would be
444 important. Moreover, although we had a clear gradient of N fertilizer input across sites, all study
445 sites were located in landscapes with a critical positive surplus of nitrogen inputs (that goes up
446 to 20 t.km⁻² for the year 2010) (European Environment Agency, 2020). Consequently, it is
447 possible that throughout the study region pollinator communities are dominated by nitrophilous
448 species (Carvalho et al., 2020) well adapted to high nitrogen conditions and the negative

449 effects of nitrogen on pollinators and their contribution to crop production are no longer
450 detectable in our specific study sites.

451 Finally, it is possible that functional composition of pollinators has changed along the nitrogen
452 availability gradient but with no net change in pollinator abundance, or their contribution to
453 crop production. Indeed, N enrichment can have contrasting effects on pollinator species.
454 Pollinators with more diversified diets might be less affected by landscape eutrophication
455 potentially due to their ability to forage on a greater richness of flowers in a diverse set of
456 habitats (Pöyry et al., 2017; Carvalheiro et al., 2020). N deposition that changes soil nutrient
457 availability is an important driver of plant species composition change and result in the decline
458 of oligotrophic plant species, such as nitrogen fixing Fabaceae species (Roth et al., 2019, 2013).
459 Fabaceae are the main food resource of most bumble bee species and many other solitary bees
460 (Connop et al., 2010; Goulson et al., 2008; Kleijn and Raemakers, 2008). Thus, species
461 specialised on Fabaceae (and other N sensitive plants), can have more difficulty in finding
462 adequate resources and thus be more susceptible to the effects of N enrichment than other
463 pollinator species (Stevens et al., 2018). But if, for the crops studied here, species that prefer
464 nitrophilous environments (see Carvalheiro et al., 2020) are equally efficient for crop
465 pollination than species which are negatively affected, pollinator community compositional
466 changes would not affect the net crop pollination outcome.

467

468 ***5. Conclusions and implications for conservation of crop pollinators and pollination***

469 Increased air pollution can affect plant and animal physiology in many ways (Emberson, 2020;
470 Mills et al., 2013; Van Dingenen et al., 2009). In Europe, a significant problem today is the
471 increased concentrations of tropospheric ozone due to its harmful effects on human health and
472 ecosystems (Ilić and Maksimović, 2021). Air pollution does not constitute a single problem,
473 but is one of many threats and opportunities to plants and animals (Dudley and Stolton, 2021).

474 Plants are more sensitive to ozone than animals, but air pollution, by modifying the physiology
475 and biochemistry of plants, has a decisive influence on the interactions of plants and insects
476 (Ilić and Maksimović, 2021). Therefore, changes in plant communities can propagate
477 throughout the food webs to affect other organisms (Dudley and Stolton, 2021; Ilić and
478 Maksimović, 2021; Lovett et al., 2009).

479 While we were not able to detect effects of oxide nitrates, our results highlight potential
480 negative effects of ozone on crop pollinators and changes in the contribution of pollinators to
481 crop production, as well as affecting the sensitivity of pollinators to pesticide exposure. Indeed,
482 different air pollutants (such as ozone and NO_x) can act at different spatio-temporal scales and
483 interact with other natural and anthropogenic factors that also alter ecosystem functioning
484 (Dudley and Stolton, 2021).

485 Even if more detailed studies are required and further evidence from other regions and crops is
486 needed, our findings suggest management plans involving changes in pesticide use, should take
487 into account the ongoing increase in air pollution, and specifically of the predicted increased
488 concentration of tropospheric ozone in the near future (Archibald et al., 2020; Avnery et al.,
489 2011b; Van Dingenen et al., 2009). Our results also highlight those negative impacts of ozone
490 pollution on pollinators and pollination exist, and should be considered when developing
491 transport, manufacturing and renewable energy policies in favor of the protection of air quality
492 and the conservation of biodiversity and associated ecosystem services.

493

494 **Acknowledgements:**

495 LGC, OR were funded by Fundação para Ciência e Tecnologia (FCT) and European Union via
496 the programa operacional regional de Lisboa 2014/2020 (project EUCLIPO-028360). LGC was
497 also funded by the Brazilian National Council for Scientific and Technological Development
498 (CNPq. Universal 421668/2018-0; PQ 305157/2018-3). J.A.G. was funded by the Natural

499 Environment Research Council (NERC; NE/ T011084/1) and the Netherlands Organization for
500 Scientific Research (NWO) under the Rubicon programme (019.162LW.010). SGP and MG
501 were supported by the Safeguard, Safeguarding European Wild Pollinators, project (EC H2020:
502 101003476). DK and JS received funding from the EU's Horizon 2020 research and innovation
503 programme under grant agreement No 862480 [The Showcase project]. This research was
504 carried out as part of the Insect Pollinators Initiative funded jointly by a grant from BBSRC,
505 Defra, NERC, the Scottish Government and the Wellcome Trust, under the Living With
506 Environmental Change Partnership.

507

508 **Statement of authorship:**

509 OR and LGC conceived the study idea. MPDG, AG, DK, SP and JS performed field
510 experimentations. OR managed all the data received. OR, JAG and IYK extracted spatial
511 dataset. OR run all data analyses in collaboration with LGC. OR and LGC wrote the first draft
512 and all authors provided input and approved the final manuscript.

513

514

515 **References**

516

- 517 Abou-Shaara, H.F., Owayss, A.A., Ibrahim, Y.Y., Basuny, N.K., 2017. A review of impacts
518 of temperature and relative humidity on various activities of honey bees. *Insect. Soc.* 64,
519 455–463. <https://doi.org/10.1007/s00040-017-0573-8>
- 520 Anderson, D.R., Link, W.A., Johnson, D.H., Burnham, K.P., 2001. Suggestions for presenting
521 the results of data analyses. *The Journal of Wildlife Management* 65, 373–378.
- 522 Archibald, A.T., Neu, J.L., Elshorbany, Y.F., Cooper, O.R., Young, P.J., Akiyoshi, H., Cox,
523 R.A., Coyle, M., Derwent, R.G., Deushi, M., Finco, A., Frost, G.J., Galbally, I.E.,

524 Gerosa, G., Granier, C., Griffiths, P.T., Hossaini, R., Hu, L., Jöckel, P., Josse, B., Lin,
525 M.Y., Mertens, M., Morgenstern, O., Naja, M., Naik, V., Oltmans, S., Plummer, D.A.,
526 Revell, L.E., Saiz-Lopez, A., Saxena, P., Shin, Y.M., Shahid, I., Shallcross, D., Tilmes,
527 S., Trickl, T., Wallington, T.J., Wang, T., Worden, H.M., Zeng, G., 2020. Tropospheric
528 Ozone Assessment Report: A critical review of changes in the tropospheric ozone
529 burden and budget from 1850 to 2100. *Elementa: Science of the Anthropocene* 8.
530 <https://doi.org/10.1525/elementa.2020.034>

531 Avnery, S., Mauzerall, D.L., Liu, J., Horowitz, L.W., 2011a. Global crop yield reductions due
532 to surface ozone exposure: 1. Year 2000 crop production losses and economic damage.
533 *Atmospheric Environment* 45, 2284–2296.
534 <https://doi.org/10.1016/j.atmosenv.2010.11.045>

535 Avnery, S., Mauzerall, D.L., Liu, J., Horowitz, L.W., 2011b. Global crop yield reductions due
536 to surface ozone exposure: 2. Year 2030 potential crop production losses and economic
537 damage under two scenarios of O₃ pollution. *Atmospheric Environment* 45, 2297–2309.
538 <https://doi.org/10.1016/j.atmosenv.2011.01.002>

539 Balfour, N.J., Ollerton, J., Castellanos, M.C., Ratnieks, F.L.W., 2018. British phenological
540 records indicate high diversity and extinction rates among late-summer-flying
541 pollinators. *Biological Conservation* 222, 278–283.
542 <https://doi.org/10.1016/j.biocon.2018.04.028>

543 Bartomeus, I., Ascher, J.S., Gibbs, J., Danforth, B.N., Wagner, D.L., Hedtke, S.M., Winfree,
544 R., 2013. Historical changes in northeastern US bee pollinators related to shared
545 ecological traits. *PNAS* 110, 4656–4660. <https://doi.org/10.1073/pnas.1218503110>

546 Bartomeus, I., Ascher, J.S., Wagner, D., Danforth, B.N., Colla, S., Kornbluth, S., Winfree, R.,
547 2011. Climate-associated phenological advances in bee pollinators and bee-pollinated
548 plants. *PNAS* 108, 20645–20649. <https://doi.org/10.1073/pnas.1115559108>

549 Bartoń, K., 2011. MuMIn: Multi-model inference.

550 Biesmeijer, J.C., Roberts, S.P.M., Reemer, M., Ohlemuller, R., Edwards, M., Peeters, T.,
551 Schaffers, A.P., Potts, S.G., Kleukers, R., Thomas, C.D., Settele, J., Kunin, W.E., 2006.
552 Parallel declines in pollinators and insect-pollinated plants in Britain and the
553 Netherlands. *Science* 313, 351–354. <https://doi.org/10.1126/science.1127863>

554 Black, V.J., Stewart, C.A., Roberts, J.A., Black, C.R., 2007. Ozone affects gas exchange,
555 growth and reproductive development in *Brassica campestris* (Wisconsin Fast Plants).
556 *New Phytologist* 176, 150–163. <https://doi.org/10.1111/j.1469-8137.2007.02163.x>

557 Büchler, R., Costa, C., Hatjina, F., Andonov, S., Meixner, M.D., Conte, Y.L., Uzunov, A.,
558 Berg, S., Bienkowska, M., Bouga, M., Drazic, M., Dyrba, W., Kryger, P., Panasiuk, B.,
559 Pechhacker, H., Petrov, P., Kezić, N., Korpela, S., Wilde, J., 2014. The influence of
560 genetic origin and its interaction with environmental effects on the survival of *Apis*
561 *mellifera* L. colonies in Europe. *Journal of Apicultural Research* 53, 205–214.
562 <https://doi.org/10.3896/IBRA.1.53.2.03>

563 Carvalho, L.G., Biesmeijer, J.C., Franzén, M., Aguirre-Gutiérrez, J., Garibaldi, L.A., Helm,
564 A., Michez, D., Pöyry, J., Reemer, M., Schweiger, O., Leon van den, B.,
565 WallisDeVries, M.F., Kunin, W.E., 2020. Soil eutrophication shaped the composition of
566 pollinator assemblages during the past century. *Ecography* 43, 209–221.
567 <https://doi.org/10.1111/ecog.04656>

568 Clarke, D., Robert, D., 2018. Predictive modelling of honey bee foraging activity using local
569 weather conditions. *Apidologie* 49, 386–396. [https://doi.org/10.1007/s13592-018-0565-](https://doi.org/10.1007/s13592-018-0565-3)
570 3

571 Connop, S., Hill, T., Steer, J., Shaw, P., 2010. The role of dietary breadth in national
572 bumblebee (*Bombus*) declines: Simple correlation? *Biological Conservation* 143, 2739–
573 2746. <https://doi.org/10.1016/j.biocon.2010.07.021>

574 Corbet, S.A., Fussell, M., Ake, R., Fraser, A., Gunson, C., Savage, A., Smith, K., 1993.
575 Temperature and the pollinating activity of social bees. *Ecological Entomology* 18, 17–
576 30. <https://doi.org/10.1111/j.1365-2311.1993.tb01075.x>

577 Dainese, M., Martin, E.A., Aizen, M.A., Albrecht, M., Bartomeus, I., Bommarco, R.,
578 Carvalheiro, L.G., Chaplin-Kramer, R., Gagic, V., Garibaldi, L.A., Ghazoul, J., Grab,
579 H., Jonsson, M., Karp, D.S., Kennedy, C.M., Kleijn, D., Kremen, C., Landis, D.A.,
580 Letourneau, D.K., Marini, L., Poveda, K., Rader, R., Smith, H.G., Tschardtke, T.,
581 Andersson, G.K.S., Badenhausser, I., Baensch, S., Bezerra, A.D.M., Bianchi, F.J.J.A.,
582 Boreux, V., Bretagnolle, V., Caballero-Lopez, B., Cavigliasso, P., Četković, A.,
583 Chacoff, N.P., Classen, A., Cusser, S., Silva, F.D. da S. e, Groot, G.A. de, Dudenhöffer,
584 J.H., Ekroos, J., Fijen, T., Franck, P., Freitas, B.M., Garratt, M.P.D., Gratton, C.,
585 Hipólito, J., Holzschuh, A., Hunt, L., Iverson, A.L., Jha, S., Keasar, T., Kim, T.N.,
586 Kishinevsky, M., Klatt, B.K., Klein, A.-M., Krewenka, K.M., Krishnan, S., Larsen,
587 A.E., Lavigne, C., Liere, H., Maas, B., Mallinger, R.E., Pachon, E.M., Martínez-
588 Salinas, A., Meehan, T.D., Mitchell, M.G.E., Molina, G.A.R., Nesper, M., Nilsson, L.,
589 O'Rourke, M.E., Peters, M.K., Plečáš, M., Potts, S.G., Ramos, D. de L., Rosenheim,
590 J.A., Rundlöf, M., Rusch, A., Sáez, A., Scheper, J., Schleuning, M., Schmack, J.M.,
591 Sciligo, A.R., Seymour, C., Stanley, D.A., Stewart, R., Stout, J.C., Sutter, L., Takada,
592 M.B., Taki, H., Tamburini, G., Tschumi, M., Viana, B.F., Westphal, C., Willcox, B.K.,
593 Wratten, S.D., Yoshioka, A., Zaragoza-Trello, C., Zhang, W., Zou, Y., Steffan-
594 Dewenter, I., 2019. A global synthesis reveals biodiversity-mediated benefits for crop
595 production. *Science Advances* 5, eaax0121. <https://doi.org/10.1126/sciadv.aax0121>

596 Dainese, M., Riedinger, V., Holzschuh, A., Kleijn, D., Scheper, J., Steffan-Dewenter, I.,
597 2018. Managing trap-nesting bees as crop pollinators: Spatiotemporal effects of floral
598 resources and antagonists. *Journal of Applied Ecology* 55, 195–204.

599 <https://doi.org/10.1111/1365-2664.12930>

600 Damalas, C.A., 2009. Understanding benefits and risks of pesticide use. *Scientific Research*
601 *and Essays* 4, 945–949.

602 David, T.I., Storkey, J., Stevens, C.J., 2019. Understanding how changing soil nitrogen affects
603 plant–pollinator interactions. *Arthropod-Plant Interactions* 13, 671–684.
604 <https://doi.org/10.1007/s11829-019-09714-y>

605 Dötterl, S., Vater, M., Rupp, T., Held, A., 2016. Ozone Differentially Affects Perception of
606 Plant Volatiles in Western Honey Bees. *J Chem Ecol* 42, 486–489.
607 <https://doi.org/10.1007/s10886-016-0717-8>

608 Duchenne, F., Thébault, E., Michez, D., Elias, M., Drake, M., Persson, M., Rousseau-Piot,
609 J.S., Pollet, M., Vanormelingen, P., Fontaine, C., 2020. Phenological shifts alter the
610 seasonal structure of pollinator assemblages in Europe. *Nat Ecol Evol* 4, 115–121.
611 <https://doi.org/10.1038/s41559-019-1062-4>

612 Dudley, N., Stolton, S., 2021. Air pollution and biodiversity: a review.

613 Duque, L., Poelman, E.H., Steffan-Dewenter, I., 2020. Effects of ozone stress on flowering
614 phenology, plant-pollinator interactions and plant reproductive success. *Environmental*
615 *Pollution* 115953. <https://doi.org/10.1016/j.envpol.2020.115953>

616 Ekroos, J., Kleijn, D., Batáry, P., Albrecht, M., Báldi, A., Blüthgen, N., Knop, E., Kovács-
617 Hostyánszki, A., Smith, H.G., 2020. High land-use intensity in grasslands constrains
618 wild bee species richness in Europe. *Biological Conservation* 241, 108255.
619 <https://doi.org/10.1016/j.biocon.2019.108255>

620 Emberson, L., 2020. Effects of ozone on agriculture, forests and grasslands. *Philosophical*
621 *Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences*
622 378, 20190327. <https://doi.org/10.1098/rsta.2019.0327>

623 EPPO, 2010. Environmental risk assessment scheme for plant protection products. EPPO

624 Bulletin 40, 323–331. <https://doi.org/10.1111/j.1365-2338.2010.02419.x>

625 European Environment Agency (EEA), 2020. Nitrogen surplus and exceedances of critical
626 nitrogen inputs to agricultural land in view of adverse impacts on water quality [WWW
627 Document]. European Environment Agency. URL [https://www.eea.europa.eu/data-and-](https://www.eea.europa.eu/data-and-maps/figures/nitrogen-surplus-and-exceedances-of)
628 [maps/figures/nitrogen-surplus-and-exceedances-of](https://www.eea.europa.eu/data-and-maps/figures/nitrogen-surplus-and-exceedances-of) (accessed 6.16.20).

629 Farré-Armengol, G., Peñuelas, J., Li, T., Yli-Pirilä, P., Filella, I., Llusia, J., Blande, J.D.,
630 2016. Ozone degrades floral scent and reduces pollinator attraction to flowers. *New*
631 *Phytologist* 209, 152–160. <https://doi.org/10.1111/nph.13620>

632 Feder, W.A., Sullivan, F., 1969. Ozone: Depression of Frond Multiplication and Floral
633 Production in Duckweed. *Science* 165, 1373–1374.
634 <https://doi.org/10.1126/science.165.3900.1373>

635 Flores-Moreno, H., Reich, P.B., Lind, E.M., Sullivan, L.L., Seabloom, E.W., Yahdjian, L.,
636 MacDougall, A.S., Reichmann, L.G., Alberti, J., Báez, S., Bakker, J.D., Cadotte, M.W.,
637 Caldeira, M.C., Chaneton, E.J., D’Antonio, C.M., Fay, P.A., Firn, J., Hagenah, N.,
638 Harpole, W.S., Iribarne, O., Kirkman, K.P., Knops, J.M.H., La Pierre, K.J., Laungani,
639 R., Leakey, A.D.B., McCulley, R.L., Moore, J.L., Pascual, J., Borer, E.T., 2016.
640 Climate modifies response of non-native and native species richness to nutrient
641 enrichment. *Philosophical Transactions of the Royal Society B: Biological Sciences*
642 371, 20150273. <https://doi.org/10.1098/rstb.2015.0273>

643 Fowler, D., Coyle, M., Skiba, U., Sutton, M.A., Cape, J.N., Reis, S., Sheppard, L.J., Jenkins,
644 A., Grizzetti, B., Galloway, J.N., Vitousek, P., Leach, A., Bouwman, A.F., Butterbach-
645 Bahl, K., Dentener, F., Stevenson, D., Amann, M., Voss, M., 2013. The global nitrogen
646 cycle in the twenty-first century. *Philosophical Transactions of the Royal Society B:*
647 *Biological Sciences* 368, 20130164. <https://doi.org/10.1098/rstb.2013.0164>

648 Fuentes, J.D., Chamecki, M., Roulston, T., Chen, B., Pratt, K.R., 2016. Air pollutants degrade

649 floral scents and increase insect foraging times. *Atmospheric Environment* 141, 361–
650 374. <https://doi.org/10.1016/j.atmosenv.2016.07.002>

651 Fuhrer, J., Martin, M.V., Mills, G., Heald, C.L., Harmens, H., Hayes, F., Sharps, K., Bender,
652 J., Ashmore, M.R., 2016. Current and future ozone risks to global terrestrial biodiversity
653 and ecosystem processes. *Ecology and Evolution* 6, 8785–8799.
654 <https://doi.org/10.1002/ece3.2568>

655 Garibaldi, L.A., Bartomeus, I., Bommarco, R., Klein, A.M., Cunningham, S.A., Aizen, M.A.,
656 Boreux, V., Garratt, M.P.D., Carvalheiro, L.G., Kremen, C., Morales, C.L., Schüepp,
657 C., Chacoff, N.P., Freitas, B.M., Gagic, V., Holzschuh, A., Klatt, B.K., Krewenka,
658 K.M., Krishnan, S., Mayfield, M.M., Motzke, I., Otieno, M., Petersen, J., Potts, S.G.,
659 Ricketts, T.H., Rundlöf, M., Sciligo, A., Sinu, P.A., Steffan-Dewenter, I., Taki, H.,
660 Tschardtke, T., Vergara, C.H., Viana, B.F., Woyciechowski, M., 2015. Trait matching
661 of flower visitors and crops predicts fruit set better than trait diversity. *J Appl Ecol* 52,
662 1436–1444. <https://doi.org/10.1111/1365-2664.12530>

663 Garibaldi, L.A., Requier, F., Rollin, O., Andersson, G.K.S., 2017. Towards an integrated
664 species and habitat management of crop pollination. *Current Opinion in Insect Science*.

665 Garibaldi, L.A., Steffan-Dewenter, I., Kremen, C., Morales, J.M., Bommarco, R.,
666 Cunningham, S.A., Carvalheiro, L.G., Chacoff, N.P., Dudenhöffer, J.H., Greenleaf,
667 S.S., Holzschuh, A., Isaacs, R., Krewenka, K., Mandelik, Y., Mayfield, M.M.,
668 Morandin, L.A., Potts, S.G., Ricketts, T.H., Szentgyörgyi, H., Viana, B.F., Westphal,
669 C., Winfree, R., Klein, A.M., 2011. Stability of pollination services decreases with
670 isolation from natural areas despite honey bee visits. *Ecology Letters* 14, 1062–1072.
671 <https://doi.org/10.1111/j.1461-0248.2011.01669.x>

672 Garratt, M.P.D., Bishop, J., Degani, E., Potts, S.G., Shaw, R.F., Shi, A., Roy, S., 2018. Insect
673 pollination as an agronomic input: Strategies for oilseed rape production. *Journal of*

674 Applied Ecology 55, 2834–2842. <https://doi.org/10.1111/1365-2664.13153>

675 Garratt, M.P.D., Breeze, T.D., Boreux, V., Fountain, M.T., McKerchar, M., Webber, S.M.,
676 Coston, D.J., Jenner, N., Dean, R., Westbury, D.B., Biesmeijer, J.C., Potts, S.G., 2016.
677 Apple Pollination: Demand Depends on Variety and Supply Depends on Pollinator
678 Identity. PLOS ONE 11, e0153889. <https://doi.org/10.1371/journal.pone.0153889>

679 Garratt, M.P.D., Breeze, T.D., Jenner, N., Polce, C., Biesmeijer, J.C., Potts, S.G., 2014a.
680 Avoiding a bad apple: Insect pollination enhances fruit quality and economic value.
681 Agriculture, Ecosystems & Environment 184, 34–40.
682 <https://doi.org/10.1016/j.agee.2013.10.032>

683 Garratt, M.P.D., Coston, D.J., Truslove, C.L., Lappage, M.G., Polce, C., Dean, R.,
684 Biesmeijer, J.C., Potts, S.G., 2014b. The identity of crop pollinators helps target
685 conservation for improved ecosystem services. Biological Conservation 169, 128–135.
686 <https://doi.org/10.1016/j.biocon.2013.11.001>

687 Garratt, M.P.D., Truslove, C.L., Coston, D.J., Evans, R.L., Moss, E.D., Dodson, C., Jenner,
688 N., Biesmeijer, J.C., Potts, S.G., 2014c. Pollination deficits in UK apple orchards.
689 Journal of Pollination Ecology 12, 9–14.

690 Gathmann, A., Tschardtke, T., 2002. Foraging ranges of solitary bees. J Anim Ecology 71,
691 757–764. <https://doi.org/10.1046/j.1365-2656.2002.00641.x>

692 Geslin, B., Gauzens, B., Baude, M., Dajoz, I., Fontaine, C., Henry, M., Ropars, L., Rollin, O.,
693 Thébault, E., Vereecken, N.J., 2017. Massively Introduced Managed Species and Their
694 Consequences for Plant–Pollinator Interactions 56.
695 <http://dx.doi.org/10.1016/bs.aacr.2016.10.007>

696 Gillespie, C., Stabler, D., Tallentire, E., Goumenaki, E., Barnes, J., 2015. Exposure to
697 environmentally-relevant levels of ozone negatively influence pollen and fruit
698 development. Environmental Pollution 206, 494–501.

699 <https://doi.org/10.1016/j.envpol.2015.08.003>

700 González-Varo, J.P., Biesmeijer, J.C., Bommarco, R., Potts, S.G., Schweiger, O., Smith,
701 H.G., Steffan-Dewenter, I., Szentgyörgyi, H., Woyciechowski, M., Vilà, M., 2013.
702 Combined effects of global change pressures on animal-mediated pollination. *Trends in*
703 *Ecology & Evolution* 28, 524–530. <https://doi.org/10.1016/j.tree.2013.05.008>

704 Goulson, D., Lye, G.C., Darvill, B., 2008. Decline and conservation of bumble bees. *Annual*
705 *Review of Entomology* 53, 191–208.
706 <https://doi.org/10.1146/annurev.ento.53.103106.093454>

707 Guerreiro, C.B.B., Foltescu, V., de Leeuw, F., 2014. Air quality status and trends in Europe.
708 *Atmospheric Environment* 98, 376–384.
709 <https://doi.org/10.1016/j.atmosenv.2014.09.017>

710 Hayes, F., Williamson, J., Mills, G., 2012. Ozone pollution affects flower numbers and timing
711 in a simulated BAP priority calcareous grassland community. *Environmental Pollution*
712 163, 40–47. <https://doi.org/10.1016/j.envpol.2011.12.032>

713 Heiden, A.C., Hoffmann, T., Kahl, J., Kley, D., Klockow, D., Langebartels, C., Mehlhorn, H.,
714 Sandermann, H., Schraudner, M., Schuh, G., Wildt, J., 1999. Emission of Volatile
715 Organic Compounds from Ozone-Exposed Plants. *Ecological Applications* 9, 1160–
716 1167. [https://doi.org/10.1890/1051-0761\(1999\)009\[1160:EOVOCF\]2.0.CO;2](https://doi.org/10.1890/1051-0761(1999)009[1160:EOVOCF]2.0.CO;2)

717 Holzschuh, A., Steffan-Dewenter, I., Kleijn, D., Tschardtke, T., 2006. Diversity of flower-
718 visiting bees in cereal fields: effects of farming system, landscape composition and
719 regional context. *Journal of Applied Ecology* 44, 41–49. <https://doi.org/10.1111/j.1365-2664.2006.01259.x>

720

721 Ilić, P., Maksimović, T., 2021. Air Pollution and Biodiversity.

722 IPCC, 2014. Climate Change 2013: The Physical Science Basis: Working Group I
723 Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate

724 Change. Cambridge University Press.

725 Jarvis, S.G., Henrys, P.A., Redheard, J.W., Osório, B.M., Pywell, R.F., 2019. CEH Land
726 Cover plus: Pesticides 2012-2016 (England and Wales). NERC Environmental
727 Information Data Centre. [https://doi.org/10.5285/a72f8ce8-561f-4f3a-8866-](https://doi.org/10.5285/a72f8ce8-561f-4f3a-8866-5da620c0c9fe)
728 [5da620c0c9fe](https://doi.org/10.5285/a72f8ce8-561f-4f3a-8866-5da620c0c9fe)

729 Johnson, B., Standish, R., Hobbs, R., 2020. Non-native plants and nitrogen addition have little
730 effect on pollination and seed set in 3-year-old restored woodland. *Austral Ecology* 45,
731 1156–1168. <https://doi.org/10.1111/aec.12949>

732 Jürgens, A., Bischoff, M., 2017. Changing odour landscapes: the effect of anthropogenic
733 volatile pollutants on plant–pollinator olfactory communication. *Functional Ecology* 31,
734 56–64. <https://doi.org/10.1111/1365-2435.12774>

735 Kendall, L.K., Gagic, V., Evans, L.J., Cutting, B.T., Scalzo, J., Hanusch, Y., Jones, J.,
736 Rocchetti, M., Sonter, C., Keir, M., Rader, R., 2020. Self-compatible blueberry cultivars
737 require fewer floral visits to maximize fruit production than a partially self-incompatible
738 cultivar. *Journal of Applied Ecology* 57, 2454–2462. [https://doi.org/10.1111/1365-](https://doi.org/10.1111/1365-2664.13751)
739 [2664.13751](https://doi.org/10.1111/1365-2664.13751)

740 Kennedy, C.M., Lonsdorf, E., Neel, M.C., Williams, N.M., Ricketts, T.H., Winfree, R.,
741 Bommarco, R., Brittain, C., Burley, A.L., Cariveau, D., Carvalheiro, L.G., Chacoff,
742 N.P., Cunningham, S.A., Danforth, B.N., Dudenhöffer, J.-H., Elle, E., Gaines, H.R.,
743 Garibaldi, L.A., Gratton, C., Holzschuh, A., Isaacs, R., Javorek, S.K., Jha, S., Klein,
744 A.M., Krewenka, K., Mandelik, Y., Mayfield, M.M., Morandin, L., Neame, L.A.,
745 Otieno, M., Park, M., Potts, S.G., Rundlöf, M., Saez, A., Steffan-Dewenter, I., Taki, H.,
746 Viana, B.F., Westphal, C., Wilson, J.K., Greenleaf, S.S., Kremen, C., 2013. A global
747 quantitative synthesis of local and landscape effects on wild bee pollinators in
748 agroecosystems. *Ecol Lett* 16, 584–599. <https://doi.org/10.1111/ele.12082>

749 Kleijn, D., Raemakers, I., 2008. A Retrospective Analysis of Pollen Host Plant Use by Stable
750 and Declining Bumble Bee Species. *Ecology* 89, 1811–1823.
751 <https://doi.org/10.1890/07-1275.1>

752 Kleijn, D., Winfree, R., Bartomeus, I., Carvalheiro, L.G., Henry, M., Isaacs, R., Klein, A.-M.,
753 Kremen, C., M’Gonigle, L.K., Rader, R., Ricketts, T.H., Williams, N.M., Lee
754 Adamson, N., Ascher, J.S., Báldi, A., Batáry, P., Benjamin, F., Biesmeijer, J.C., Blitzer,
755 E.J., Bommarco, R., Brand, M.R., Bretagnolle, V., Button, L., Cariveau, D.P., Chifflet,
756 R., Colville, J.F., Danforth, B.N., Elle, E., Garratt, M.P.D., Herzog, F., Holzschuh, A.,
757 Howlett, B.G., Jauker, F., Jha, S., Knop, E., Krewenka, K.M., Le Féon, V., Mandelik,
758 Y., May, E.A., Park, M.G., Pisanty, G., Reemer, M., Riedinger, V., Rollin, O., Rundlöf,
759 M., Sardiñas, H.S., Scheper, J., Sciligo, A.R., Smith, H.G., Steffan-Dewenter, I., Thorp,
760 R., Tschamntke, T., Verhulst, J., Viana, B.F., Vaissière, B.E., Veldtman, R., Westphal,
761 C., Potts, S.G., 2015. Delivery of crop pollination services is an insufficient argument
762 for wild pollinator conservation. *Nat Commun* 6. <https://doi.org/10.1038/ncomms8414>

763 Kremen, C., M’Gonigle, L.K., 2015. EDITOR’S CHOICE: Small-scale restoration in
764 intensive agricultural landscapes supports more specialized and less mobile pollinator
765 species. *J Appl Ecol* 52, 602–610. <https://doi.org/10.1111/1365-2664.12418>

766 Kwon, Y.J., Saeed, S., 2003. Effect of temperature on the foraging activity of *Bombus*
767 *terrestris* L. (Hymenoptera: Apidae) on greenhouse hot pepper (*Capsicum annuum* L.).
768 *Applied Entomology and Zoology* 38, 275–280.

769 Lefohn, A.S., Malley, C.S., Smith, L., Wells, B., Hazucha, M., Simon, H., Naik, V., Mills, G.,
770 Schultz, M.G., Paoletti, E., De Marco, A., Xu, X., Zhang, L., Wang, T., Neufeld, H.S.,
771 Musselman, R.C., Tarasick, D., Brauer, M., Feng, Z., Tang, H., Kobayashi, K., Sicard,
772 P., Solberg, S., Gerosa, G., 2018. Tropospheric ozone assessment report: Global ozone
773 metrics for climate change, human health, and crop/ecosystem research. *Elementa*

774 (Wash D C) 1, 1. <https://doi.org/10.1525/elementa.279>

775 Leisner, C.P., Ainsworth, E.A., 2012. Quantifying the effects of ozone on plant reproductive
776 growth and development. *Global Change Biology* 18, 606–616.
777 <https://doi.org/10.1111/j.1365-2486.2011.02535.x>

778 Lewis, K.A., Tzilivakis, J., 2019. Wild Bee Toxicity Data for Pesticide Risk Assessments.
779 Data 4, 98. <https://doi.org/10.3390/data4030098>

780 Lewis, K.A., Tzilivakis, J., Warner, D.J., Green, A., 2016. An international database for
781 pesticide risk assessments and management. *Human and Ecological Risk Assessment: An International Journal* 22, 1050–1064.
782 <https://doi.org/10.1080/10807039.2015.1133242>

783

784 Lindström, S.A.M., Herbertsson, L., Rundlöf, M., Bommarco, R., Smith, H.G., 2016.
785 Experimental evidence that honeybees depress wild insect densities in a flowering crop.
786 *Proceedings of the Royal Society B: Biological Sciences* 283, 20161641.
787 <https://doi.org/10.1098/rspb.2016.1641>

788 Lovett, G., Tear, T., Evers, D., Findlay, S., Cosby Jr, B., Dunscomb, J., Driscoll, C.,
789 Weathers, K., 2009. Effects of Air Pollution on Ecosystems and Biological Diversity in
790 the Eastern United States. *Annals of the New York Academy of Sciences* 1162, 99–135.
791 <https://doi.org/10.1111/j.1749-6632.2009.04153.x>

792 Mahmood, F., Khokhar, M.F., Mahmood, Z., 2020. Examining the relationship of
793 tropospheric ozone and climate change on crop productivity using the multivariate panel
794 data techniques. *Journal of Environmental Management* 272, 111024.
795 <https://doi.org/10.1016/j.jenvman.2020.111024>

796 Mallinger, R.E., Gaines-Day, H.R., Gratton, C., 2017. Do managed bees have negative effects
797 on wild bees?: A systematic review of the literature. *PLOS ONE* 12, e0189268.
798 <https://doi.org/10.1371/journal.pone.0189268>

799 Mancini, F., Woodcock, B.A., Isaac, N.J.B., 2019. Agrochemicals in the wild: Identifying
800 links between pesticide use and declines of nontarget organisms. *Current Opinion in*
801 *Environmental Science & Health, Environmental Pollution: Wildlife* 11, 53–58.
802 <https://doi.org/10.1016/j.coesh.2019.07.003>

803 Marini, L., Tamburini, G., Petrucco-Toffolo, E., Lindström, S.A.M., Zanetti, F., Mosca, G.,
804 Bommarco, R., 2015. Crop management modifies the benefits of insect pollination in
805 oilseed rape. *Agriculture, Ecosystems & Environment* 207, 61–66.
806 <https://doi.org/10.1016/j.agee.2015.03.027>

807 Martay, B., Pearce-Higgins, J.W., Harris, S.J., Gillings, S., 2018. Monitoring landscape-scale
808 environmental changes with citizen scientists: Twenty years of land use change in Great
809 Britain. *Journal for Nature Conservation* 44, 33–42.
810 <https://doi.org/10.1016/j.jnc.2018.03.001>

811 Mazor, T., Doropoulos, C., Schwarzmüller, F., Gladish, D.W., Kumaran, N., Merkel, K., Di
812 Marco, M., Gagic, V., 2018. Global mismatch of policy and research on drivers of
813 biodiversity loss. *Nature Ecology & Evolution* 2, 1071–1074.
814 <https://doi.org/10.1038/s41559-018-0563-x>

815 McFrederick, Q.S., Kathilankal, J.C., Fuentes, J.D., 2008. Air pollution modifies floral scent
816 trails. *Atmospheric Environment* 42, 2336–2348.
817 <https://doi.org/10.1016/j.atmosenv.2007.12.033>

818 Mills, G., Wagg, S., Harmens, H., 2013. Ozone Pollution: Impacts on ecosystem services and
819 biodiversity. Centre for Ecology and Hydrology, Gwynedd, UK.

820 Naqvi, H.R., Mutreja, G., Shakeel, A., Siddiqui, M.A., 2022. Air Quality and Its Relationship
821 with COVID-19 Mortality in Hotspot Places of India: A Post-lockdown Analysis.
822 <https://doi.org/10.21203/rs.3.rs-56228/v1>

823 NASA, 2020. Panoply v. 4.11.1. NASA Goddard Institute for Space Studies, USA.

824 Osório, B.M., Redheard, J.W., Javis, S.G., May, L., Pywell, R.F., 2019. CEH Land Cover
825 plus: Fertilisers 2010-2015 (England). NERC Environmental Information Data Centre.
826 <https://doi.org/10.5285/15f415db-e87b-4ab5-a2fb-37a78e7bf051>

827 Paoletti, E., De Marco, A., Beddows, D.C.S., Harrison, R.M., Manning, W.J., 2014. Ozone
828 levels in European and USA cities are increasing more than at rural sites, while peak
829 values are decreasing. *Environmental Pollution* 192, 295–299.
830 <https://doi.org/10.1016/j.envpol.2014.04.040>

831 Paradis, E., Blomberg, S., Bolker [aut, B., cph, Brown, J., Claude, J., Cuong, H.S., Desper,
832 R., Didier, G., Durand, B., Dutheil, J., Ewing, R.J., Gascuel, O., Guillerme, T., Heibl,
833 C., Ives, A., Jones, B., Krah, F., Lawson, D., Lefort, V., Legendre, P., Lemon, J.,
834 Marcon, E., McCloskey, R., Nylander, J., Opgen-Rhein, R., Popescu, A.-A., Royer-
835 Carezzi, M., Schliep, K., Strimmer, K., Vienne, D. de, 2019. ape: Analyses of
836 Phylogenetics and Evolution.

837 Park, M.G., Blitzer, E.J., Gibbs, J., Losey, J.E., Danforth, B.N., 2015. Negative effects of
838 pesticides on wild bee communities can be buffered by landscape context. *Proceedings*
839 *of the Royal Society B: Biological Sciences* 282, 20150299.
840 <https://doi.org/10.1098/rspb.2015.0299>

841 Peeters, T., Nieuwenhuijsen, H., Smit, J., van der Meer, F., Raemakers, I., Heitmans, W., van
842 Achterberg, C., Kwak, M., Loonstra, A.J., De Rond, J., Roos, M., Reemer, M., 2012.
843 *De Nederlandse bijen (Hymenoptera: Apidae S. L.)*.

844 Pinheiro, J., Bates, D., DebRoy, S., Sarkar, D., Heisterkamp, S., Bert, V.W., 2020. nlme:
845 Linear and Nonlinear Mixed Effects Models.

846 Pöyry, J., Carvalheiro, L.G., Heikkinen, R.K., Kühn, I., Kuussaari, M., Schweiger, O.,
847 Valtonen, A., Bodegom, P.M. van, Franzén, M., 2017. The effects of soil eutrophication
848 propagate to higher trophic levels. *Global Ecology and Biogeography* 26, 18–30.

849 <https://doi.org/10.1111/geb.12521>

850 Prado, A., Pioz, M., Vidau, C., Requier, F., Jury, M., Crauser, D., Brunet, J.-L., Le Conte, Y.,
851 Alaux, C., 2019. Exposure to pollen-bound pesticide mixtures induces longer-lived but
852 less efficient honey bees. *Science of The Total Environment* 650, 1250–1260.
853 <https://doi.org/10.1016/j.scitotenv.2018.09.102>

854 QGIS Development Team, 2020. QGIS Geographic Information System. Open Source
855 Geospatial Foundation.

856 R Development Core Team, 2018. R: A language and environment for statistical computing.
857 R Foundation for Statistical Computing, Vienna, Austria. ISBN 3-900051-07-0, URL.

858 Ramos, D. de L., Bustamante, M.M.C., Silva, F.D. da S. e, Carvalheiro, L.G., 2018. Crop
859 fertilization affects pollination service provision – Common bean as a case study. *PLoS*
860 *One* 13. <https://doi.org/10.1371/journal.pone.0204460>

861 Rockström, J., Steffen, W., Noone, K., Persson, Å., Chapin, F.S., Lambin, E.F., Lenton, T.M.,
862 Scheffer, M., Folke, C., Schellnhuber, H.J., Nykvist, B., Wit, C.A. de, Hughes, T.,
863 Leeuw, S. van der, Rodhe, H., Sörlin, S., Snyder, P.K., Costanza, R., Svedin, U.,
864 Falkenmark, M., Karlberg, L., Corell, R.W., Fabry, V.J., Hansen, J., Walker, B.,
865 Liverman, D., Richardson, K., Crutzen, P., Foley, J.A., 2009. A safe operating space for
866 humanity. *Nature* 461, 472–475. <https://doi.org/10.1038/461472a>

867 Rollin, O., Garibaldi, L.A., 2019. Impacts of honeybee density on crop yield: A meta-
868 analysis. *Journal of Applied Ecology* 0. <https://doi.org/10.1111/1365-2664.13355>

869 Roman-Gonzalez, A., Navarro-Raymundo, A.F., Vargas-Cuentas, N.I., 2020. Air Pollution
870 Monitoring in Peru Using Satellite Data During the Quarantine Due to COVID-19.
871 *IEEE Aerospace and Electronic Systems Magazine* 35, 73–79.
872 <https://doi.org/10.1109/MAES.2020.3018895>

873 Roth, T., Kohli, L., Bühler, C., Rihm, B., Meuli, R.G., Meier, R., Amrhein, V., 2019. Species

874 turnover reveals hidden effects of decreasing nitrogen deposition in mountain hay
875 meadows. PeerJ 7, e6347. <https://doi.org/10.7717/peerj.6347>

876 Roth, T., Kohli, L., Rihm, B., Achermann, B., 2013. Nitrogen deposition is negatively related
877 to species richness and species composition of vascular plants and bryophytes in Swiss
878 mountain grassland. *Agriculture, Ecosystems & Environment* 178, 121–126.
879 <https://doi.org/10.1016/j.agee.2013.07.002>

880 Sala, O.E., Chapin, F.S., Armesto, J.J., Berlow, E., Bloomfield, J., Dirzo, R., Huber-Sanwald,
881 E., Huenneke, L.F., Jackson, R.B., Kinzig, A., Leemans, R., Lodge, D.M., Mooney,
882 H.A., Oesterheld, M., Poff, N.L., Sykes, M.T., Walker, B.H., Walker, M., Wall, D.H.,
883 2000. Global biodiversity scenarios for the year 2100. *Science* 287, 1770–1774.
884 <https://doi.org/10.1126/science.287.5459.1770>

885 Saunier, A., Blande, J.D., 2019. The effect of elevated ozone on floral chemistry of
886 Brassicaceae species. *Environmental Pollution* 255, 113257.
887 <https://doi.org/10.1016/j.envpol.2019.113257>

888 Shaw, R.F., Phillips, B.B., Doyle, T., Pell, J.K., Redhead, J.W., Savage, J., Woodcock, B.A.,
889 Bullock, J.M., Osborne, J.L., 2020. Mass-flowering crops have a greater impact than
890 semi-natural habitat on crop pollinators and pollen deposition. *Landscape Ecol* 35, 513–
891 527. <https://doi.org/10.1007/s10980-019-00962-0>

892 Smil, V., 2000. PHOSPHORUS IN THE ENVIRONMENT: Natural Flows and Human
893 Interferences. *Annual Review of Energy and the Environment* 25, 53–88.
894 <https://doi.org/10.1146/annurev.energy.25.1.53>

895 Stathopoulou, E., Mihalakakou, G., Santamouris, M., Bagiorgas, H.S., 2008. On the impact of
896 temperature on tropospheric ozone concentration levels in urban environments. *J Earth*
897 *Syst Sci* 117, 227–236. <https://doi.org/10.1007/s12040-008-0027-9>

898 Steffan-Dewenter, I., Münzenberg, U., Christof Bürger, Thies, C., Tschardtke, T., 2002.

899 Scale-dependent effects of landscape context on three pollinator guilds. *Ecology* 83,
900 1421–1432. <https://doi.org/10.2307/3071954>

901 Stevens, C.J., David, T.I., Storkey, J., 2018. Atmospheric nitrogen deposition in terrestrial
902 ecosystems: Its impact on plant communities and consequences across trophic levels.
903 *Functional Ecology* 32, 1757–1769. <https://doi.org/10.1111/1365-2435.13063>

904 Tai, A.P.K., Martin, M.V., Heald, C.L., 2014. Threat to future global food security from
905 climate change and ozone air pollution. *Nature Climate Change* 4, 817–821.
906 <https://doi.org/10.1038/nclimate2317>

907 Taia, W., Basahi, J., Hassan, I., 2013. Impact of ambient air on physiology, pollen tube
908 growth, pollen germination and yield in pepper (*Capsicum annuum* L.). *Pakistan*
909 *Journal of Botany* 45, 921–926.

910 Tamburini, G., Berti, A., Morari, F., Marini, L., 2016. Degradation of soil fertility can cancel
911 pollination benefits in sunflower. *Oecologia* 180, 581–587.
912 <https://doi.org/10.1007/s00442-015-3493-1>

913 Tamburini, G., Lami, F., Marini, L., 2017. Pollination benefits are maximized at intermediate
914 nutrient levels. *Proceedings of the Royal Society B: Biological Sciences* 284, 20170729.
915 <https://doi.org/10.1098/rspb.2017.0729>

916 Tilman, D., Cassman, K.G., Matson, P.A., Naylor, R., Polasky, S., 2002. Agricultural
917 sustainability and intensive production practices. *Nature* 418, 671–677.
918 <https://doi.org/10.1038/nature01014>

919 Tjoelker, M.G., Luxmoore, R.J., 1991. Soil nitrogen and chronic ozone stress influence
920 physiology, growth and nutrient status of *Pinus taeda* L. and *Liriodendron tulipifera* L.
921 seedlings. *New Phytologist* 119, 69–81. [https://doi.org/10.1111/j.1469-](https://doi.org/10.1111/j.1469-8137.1991.tb01009.x)
922 [8137.1991.tb01009.x](https://doi.org/10.1111/j.1469-8137.1991.tb01009.x)

923 Tosi, S., Burgio, G., Nieh, J.C., 2017. A common neonicotinoid pesticide, thiamethoxam,

924 impairs honey bee flight ability. *Scientific Reports* 7, 1201.
925 <https://doi.org/10.1038/s41598-017-01361-8>

926 Usha, Mishra, V.K., Devi, M.S., 2020. Effect of environmental factors on the foraging
927 activities of major bee pollinators. *J. Entomol. Zool. Stud.* 8, 450–454.

928 Van de Pol, M., Wright, J., 2009. A simple method for distinguishing within- versus between-
929 subject effects using mixed models. *Animal Behaviour* 77, 753–758.
930 <https://doi.org/10.1016/j.anbehav.2008.11.006>

931 Van Dingenen, R., Dentener, F.J., Raes, F., Krol, M.C., Emberson, L., Cofala, J., 2009. The
932 global impact of ozone on agricultural crop yields under current and future air quality
933 legislation. *Atmospheric Environment* 43, 604–618.
934 <https://doi.org/10.1016/j.atmosenv.2008.10.033>

935 Vanderplanck, M., Lapeyre, B., Brondani, M., Opsommer, M., Dufay, M., Hossaert-McKey,
936 M., Proffit, M., 2021. Ozone Pollution Alters Olfaction and Behavior of Pollinators.
937 *Antioxidants* 10, 636. <https://doi.org/10.3390/antiox10050636>

938 Vasiliev, D., Greenwood, S., 2021. The role of climate change in pollinator decline across the
939 Northern Hemisphere is underestimated. *Science of The Total Environment* 775,
940 145788. <https://doi.org/10.1016/j.scitotenv.2021.145788>

941 Walker, L., Wu, S., 2017. Pollinators and Pesticides, in: Steier, G., Patel, K.K. (Eds.),
942 International Farm Animal, Wildlife and Food Safety Law. Springer International
943 Publishing, Cham, pp. 495–513. https://doi.org/10.1007/978-3-319-18002-1_17

944 Wang, C., Tang, Y., 2019. Responses of plant phenology to nitrogen addition: a meta-
945 analysis. *Oikos* 128, 1243–1253. <https://doi.org/10.1111/oik.06099>

946 Williams, N.M., Crone, E.E., Roulston, T.H., Minckley, R.L., Packer, L., Potts, S.G., 2010.
947 Ecological and life-history traits predict bee species responses to environmental
948 disturbances. *Biological Conservation* 143, 2280–2291.

949 <https://doi.org/10.1016/j.biocon.2010.03.024>

950 Wood, T.J., Michez, D., Paxton, R.J., Drossart, M., Neumann, P., Gérard, M., Vanderplanck,
951 M., Barraud, A., Martinet, B., Leclercq, N., Vereecken, N.J., 2020. Managed honey
952 bees as a radar for wild bee decline? *Apidologie*. <https://doi.org/10.1007/s13592-020->
953 [00788-9](https://doi.org/10.1007/s13592-020-00788-9)

954 Woodcock, B.A., Bullock, J.M., Shore, R.F., Heard, M.S., Pereira, M.G., Redhead, J.,
955 Ridding, L., Dean, H., Sleep, D., Henrys, P., Peyton, J., Hulmes, S., Hulmes, L.,
956 Sárospataki, M., Saure, C., Edwards, M., Genersch, E., Knäbe, S., Pywell, R.F., 2017.
957 Country-specific effects of neonicotinoid pesticides on honey bees and wild bees.
958 *Science* 356, 1393–1395. <https://doi.org/10.1126/science.aaa1190>

959 Yasrebi-de-Kom, I.A.R., Biesmeijer, J.C., Aguirre-Gutiérrez, J., 2019. Risk of potential
960 pesticide use to honeybee and bumblebee survival and distribution: A country-wide
961 analysis for The Netherlands. *Diversity and Distributions* 25, 1709–1720.
962 <https://doi.org/10.1111/ddi.12971>

963

964

965

966

967 **Table 1.** Sources of data for crop production and pollinator abundance included in the analyses.

968 UK: United Kingdom; NL: Netherlands.

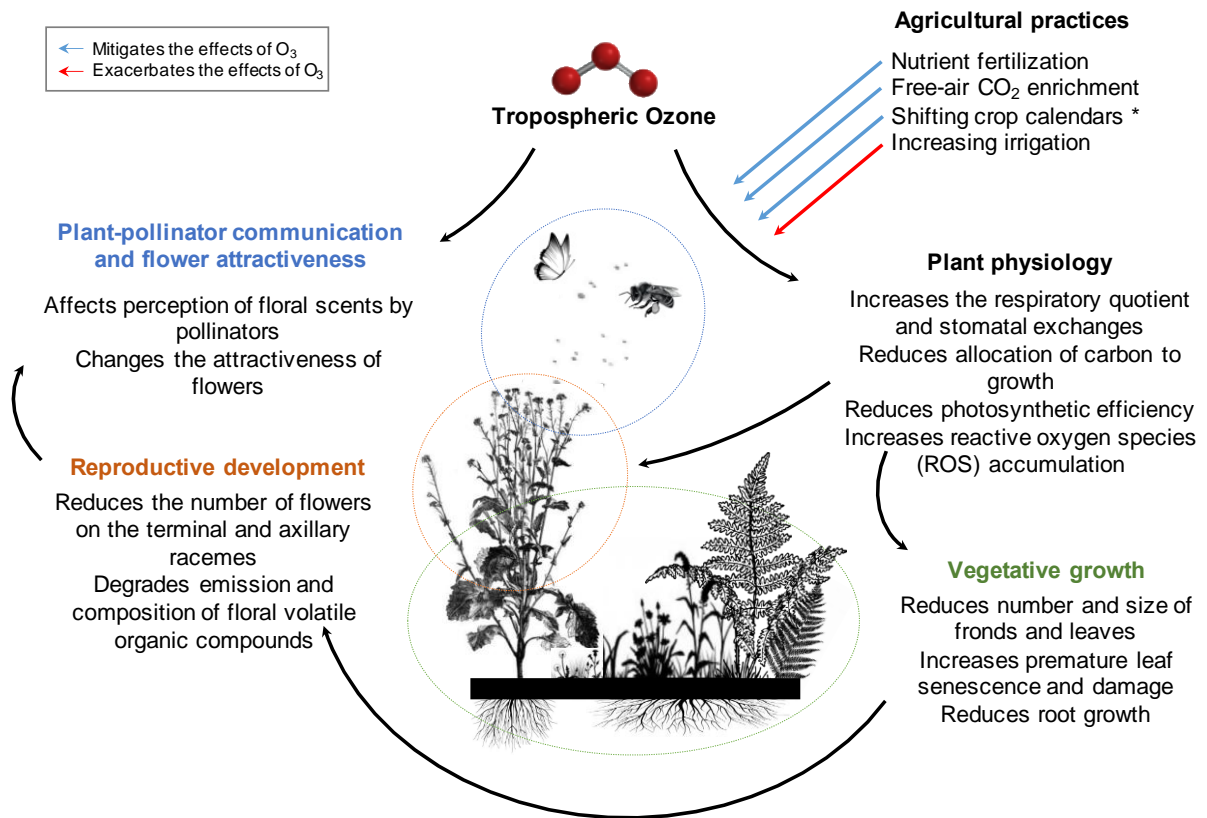
969

Study	Crop	Country	Sampling Year	Number of Sites
Garratt et al 2013. Journal of Pollination Ecology	<i>Malus domestica</i>	UK	2011	8
Garratt et al. 2014. Agriculture Ecosystems & Environment	<i>Malus domestica</i>	UK	2011	6
Garratt et al. 2016. PLOSOne	<i>Malus domestica</i>	UK	2012	5
Garratt et al. 2014. Biological conservation	<i>Vicia faba</i>	UK	2011	8
Garratt et al. 2014. Biological conservation	<i>Brassica napus</i>	UK	2012	8
De Groot et al. unpublished data: Blueberry	<i>Vaccinium corymbosum</i>	NL	2013	15
De Groot et al. unpublished data: Blueberry	<i>Vaccinium corymbosum</i>	NL	2014	15
De Groot et al. unpublished data: Blueberry	<i>Vaccinium corymbosum</i>	NL	2017	10
De Groot et al. unpublished data: Blueberry	<i>Vaccinium corymbosum</i>	NL	2018	10
De Groot et al. unpublished data: Apple	<i>Malus domestica</i>	NL	2013	15
De Groot et al. unpublished data: Apple	<i>Malus domestica</i>	NL	2014	15
De Groot et al. unpublished data: Apple	<i>Malus domestica</i>	NL	2017	8
De Groot et al. unpublished data: Apple	<i>Malus domestica</i>	NL	2018	10
<i>Total</i>				<i>133</i>

970

971

972

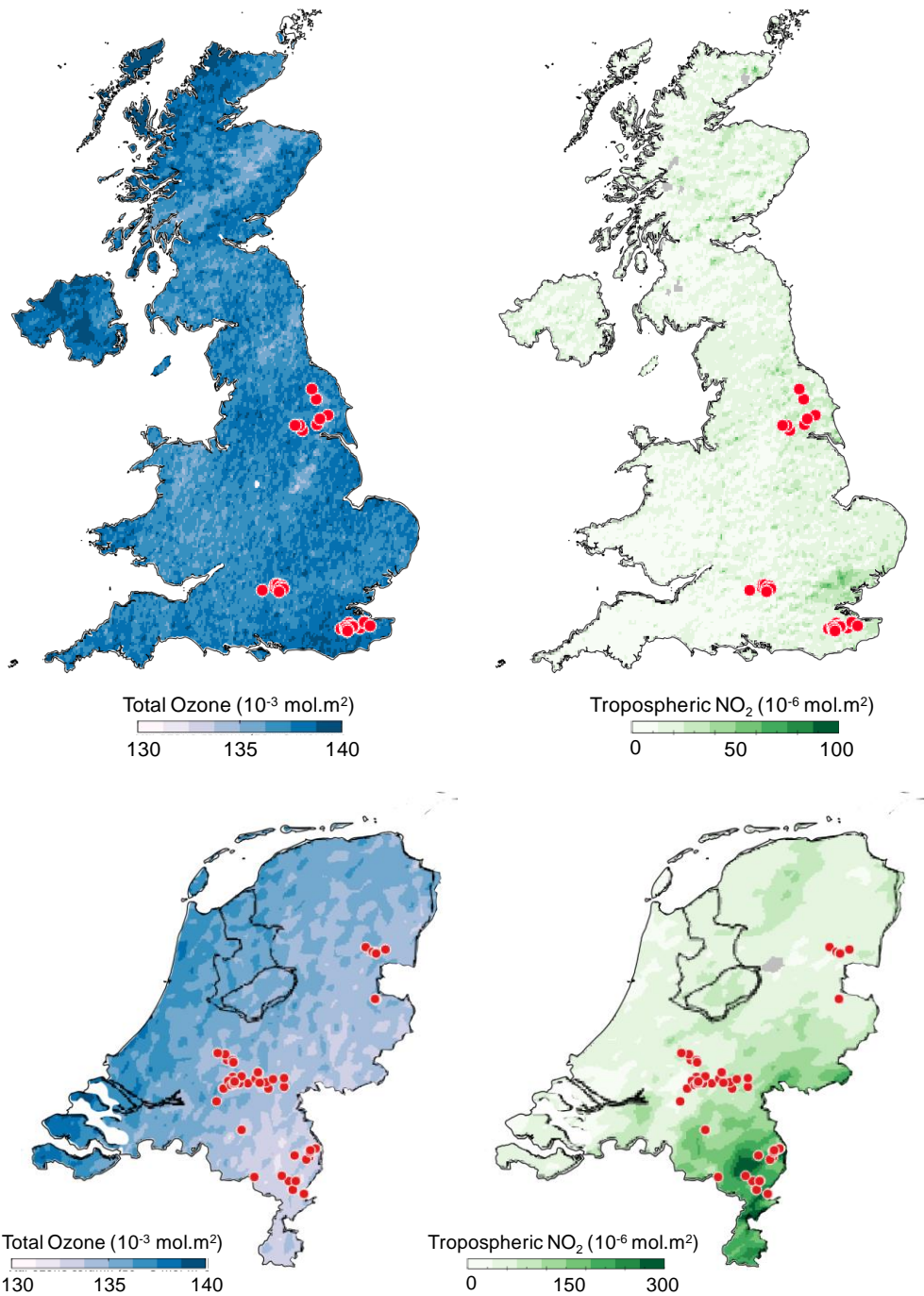


974

975 **Figure 1.** Review of known effects of tropospheric ozone on plants and plant-pollinator
 976 interactions. Blue and red arrows indicate agricultural practices that can respectively, mitigate
 977 or exacerbate effects of ozone on plant physiology (*Shifting crop calendars consists of a
 978 change in the sowing period to dissociate the peak of flowering and production of sensitive
 979 crops from the peak of atmospheric ozone concentration).

980

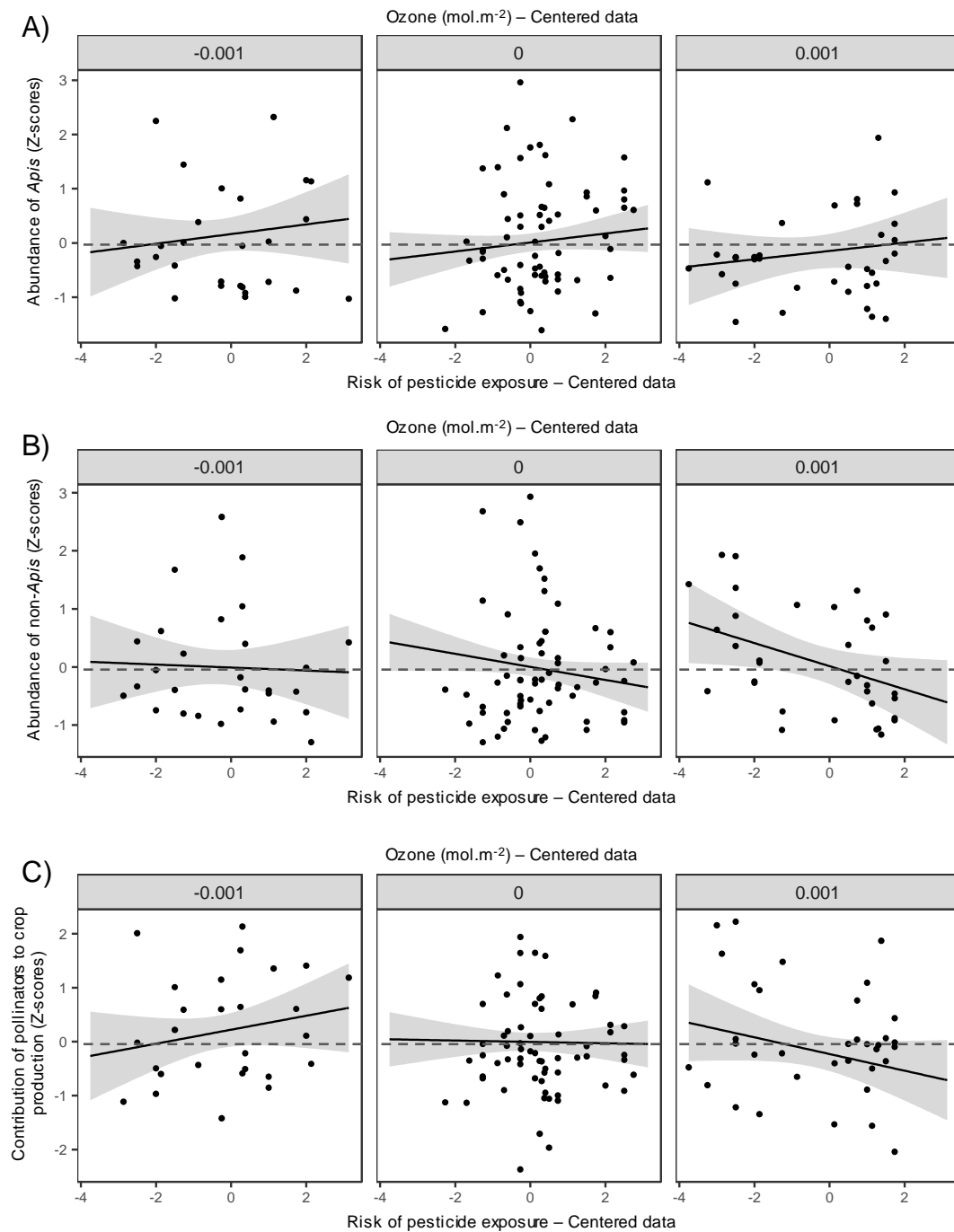
981



983

984 **Figure 2.** Sampling sites (red dots) included in the study and gradient of ozone (O_3) and dioxide
 985 nitrogen (NO_2) in the United Kingdom (UK) and the Netherlands (NL). O_3 and NO_2 gradients
 986 were mapped using the software NASA Panoply v.4.11.1 (e.g. *Sentinel-5 satellite data*
 987 *extraction for August 2019*) (NASA, 2020) and QGIS v.3.6 (QGIS Development Team, 2020).

988



989

990 **Figure 3.** The increase in ozone concentration modifies the relationship between the risk of
 991 pesticide exposure and (A) the abundance of honey bees, (B) the abundance of non-*Apis*
 992 pollinators and (C) the contribution of pollinators on crop production. The dashed lines show a
 993 null difference of the response variable with the mean of the study (combination
 994 crop/year/country).

995