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

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Abstract:

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

Key words:

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

1. Introduction

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

Nitrogen deposition (estimated to be 413 Tg N yr^{-1} in 2010) has more than doubled over the last century (Fowler et al., 2013) due to emissions of ammonia (NH_3, from pecuary and agriculture) and nitrogen oxides (NOx produced in the combustion of fossil fuels). Such increases have affected plant communities (Carvalheiro et al., 2020; Tilman et al., 2002), with associated bottom-up impacts on higher trophic levels including pollinators (Carvalheiro et al., 2020; David et al., 2019; Johnson et al., 2020; Pöyry et al., 2017; Ramos et al., 2018; Wang and Tang, 2019). While scarcity of nitrogen can constrain the positive effect of pollinators on crop production (e.g. sunflower; Tamburini et al., 2016; oilseed rape; Garratt *et al.* 2018), excess nitrogen can also be detrimental and alter the plant’s ability to compensate for the absence of pollinators (Marini et al., 2015; Tamburini et al., 2017; Ramos et al., 2018). Such responses are likely mediated by changes in floral resources quality and quantity, which in turn can be moderated by changes in climatic conditions (Flores-Moreno et al., 2016).

Another important air pollutant is tropospheric ozone, a major greenhouse-gas which is also phytotoxic (Mills et al., 2013; Lefohn et al., 2018; Ilić and Maksimović, 2021). Ozone levels have increased since the beginning of the industrial period (estimated to up of 35%; Mills et al., 2013; Guerreiro et al., 2014; IPCC, 2014). While there are other sources of ozone (e.g. volatile organic compounds, carbon monoxide and methane), oxidized nitrogen (NO_x) is one of the two major ozone precursors (Mills et al., 2013; Lefohn et al., 2018). Increased concentrations of ozone can reduce photosynthesis and plant growth (Tjoelker and Luxmoore, 1991; Black et al., 2007) and negatively affect the timing of flowering and number of flowers (Feder and Sullivan, 1969; Hayes et al., 2012; Leisner and Ainsworth, 2012) (Figure 1). Increased ozone concentration in the air (e.g. 80-120 ppb, frequently found near urban areas; Paoletti et al., 2014) can also change the concentration and emission distance of floral volatile organic compounds (Farré-Armengol et al., 2016; Fuentes et al., 2016; Heiden et al., 1999; Jürgens and Bischoff, 2017; McFrederick et al., 2008) and, consequently, affect pollinator olfaction and foraging behaviour (Farré-Armengol et al., 2016; Fuentes et al., 2016; Vanderplanck et al., 2021) (see Fig. 1). These effects on plant pollinator interactions may partly explain the reported negative effects of ozone on seed and fruit production detected in previous studies (Farré-Armengol et al., 2016; Fuhrer et al., 2016; Mills et al., 2013). Yet, few studies have explored the effects of air pollution (e.g., nitrogen oxides and ozone) on pollinator foraging patterns and efficiency, and if the strength and direction of such effects depends on other important environment drivers, such as pesticide use (Walker and Wu, 2017) or land use (Mazor et al., 2018; Sala et al., 2000).

Taking into account potential interactive effects with landscape quality for pollinators (i.e., natural and semi-natural vegetation composition) and pesticide exposure, we investigated how air pollution by ozone and different sources of nitrogen compromise pollinator visitation rates and their contribution to crop production (apple, blueberry, fava bean, oilseed rape). Given the negative effects on flower abundance and odours described above, and the fact that previous studies detected greater benefit from pollination under lower N availability (Marini et al., 2015; Ramos et al., 2018), we expect that increased ozone and nitrogen will lead to declines in crop pollinator visitation rates and pollination service delivery. However, availability of non-crop habitats is an important determinant of pollinator abundance, richness and pollination services (Dainese et al., 2019; Kennedy et al., 2013). We also expect that the effect of ozone and nitrogen on pollinators and pollination will be weaker in structurally more simple landscapes (less semi-natural habitat and greater risk of exposure to pesticides), where the only potential pollinators would be species with greater resilience to land use intensification (Williams et al., 2010; Bartomeus et al., 2013; Kremen and M'Gonigle, 2015; Kleijn et al., 2015).

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

2. Material and Methods

2.1. Pollinator and crop production data

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

Whenever studies provided more than one crop production metric, we selected the most pertinent variable to define crop production: seed set for oilseed rape, pod set for fava bean (see Garratt et al., 2014b) and fruit set for blueberry (see Kendall et al., 2020). For apple, studies conducted in the Netherlands gave information on fruit quality, i.e., fruit weight. For studies in apple orchards in the UK, data available concerned final fruit set at harvest. For each experimental branch, the number of apples which had developed on experimental inflorescences was recorded (see Garratt et al., 2014a, 2014b, 2016).

As data from different studies applied different methodologies to extract information on pollinators and pollination, we calculated z-scores within each study for crop pollination (i.e., contribution of pollinators to crop production) and pollinator abundances (*Apis* and non-*Apis* pollinators separately). This measure allows for the standardisation of scores with respect to the other scores into the same group (crop/year) (Garibaldi et al., 2011, 2015).

2.2. Ozone and NO_x data

Information on atmospheric nitrogen (NO_x) and ozone (O_3) were obtained from the Tropospheric Monitoring Instrument (TROPOMI), hosted by the European Space Agency's (ESA) Sentinel-5P satellite under the Copernicus programme (https://sentinel.esa.int/web/sentinel/missions/sentinel-5p). The Sentinel-5 Precursor mission is the first Copernicus mission dedicated to monitoring our atmosphere and provides information and services on air quality, climate, ozone (O_3) and Nitrogen dioxide (NO_2) between the surface and the top of the troposphere and the ozone layer. The spatial resolution of the Sentinel-5P is 7×3.5 km. Data of O_3 and NO_2 were extracted using the NASA Panoply 4.11.1 software (NASA, 2020) (Fig. 2).

To generate mean NO_2 and O_3 values over our specific sites, we extract daily values from TROPOMI layers between May 2019 (first of the TROPOMI-Sentinal5P products was released at the end of April 2019) and September 2019. This specific period covers the period of activity of most pollinators in the study region (Balfour et al., 2018; Peeters et al., 2012) and therefore to the period when they may be most exposed to atmospheric pollutants. We did not include data from 2020 in our mean calculation, due to the unusual change in human activity caused by covid-19 health crises. While nitrogen oxides are one of the several precursor of ozone (Mills et al., 2013), O_3 and NO_2 are not correlated (*cor* = 0.070; *p-value* = 0.421).

2.3. Agricultural nitrogen input data

Estimated average total annual application of manufactured nitrogen (1km resolution, $kg/km^2/year$) was extracted for England from the raster CEH Land Cover® plus Fertilisers (CEH, Wallingford, UK; https://www.ceh.ac.uk). CEH dataset used data from Defra British Survey of Fertiliser practice (2010-2015) to derive average annual application of manufactured fertilisers for each crop type and then derived total application at 1km resolution using crop areas from CEH Land Cover® plus: Crops (averaged 2015-2017) (Osório et al., 2019). As changes in land use intensity in the UK were limited, with trends stable overall since 1994 (Martay et al., 2018), we assume that values based on these maps are representative for the sampling years (2011 and 2012).

For the Netherlands, mean values of nitrogen fertilizer application rate by crop were extracted from the database of the Netherlands Enterprise Agency (RVO) for 2016 (*Gewascodelijst Stikstofgebruiksnormen*; https://english.rvo.nl). To estimate the mean value of nitrogen applied as fertilizer at 1km resolution (in $kg/km^2/year$), we calculated a weighted average, taking into account the proportion of each crop in the landscape. Crop coverage per site were extracted for each 1km^2 cell as an average of the BRP (*Basisregistratie gewaspercelen*) shapefiles 2015 and 2016 obtained from the RVO (https://english.rvo.nl/).

$$N\_Fer = \sum (Proportion\ of\ each\ crop\ category\ \times\ mean\ annual\ application\ rate\ for\ the\ category)$$

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

2.4. Pesticide input data

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

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

with *HQ* the hazard quotient (HQ) of each active molecule and the median lethal dose per bee (LD_{50}). The median lethal dose is one way to measure the short-term poisoning potential (acute toxicity) of a substance. The LD_{50} is the amount of a substance, given all at once, which causes the death of 50% of a group of test animals. The hazard quotient ratio gives an approximation of how close the likely exposure of bees is to a toxicologically significant level. The pesticide risk index (*RI*) was defined as the number of high risk active ingredients ($HQ>50$; see EPPO, 2010) that were applied. If $HQ<50$, the active ingredient was categorized as low risk to bees. The LD_{50} of 390 active ingredients used in the UK and the Netherlands were extracted from the "*Pesticides Properties DataBase*" (PPDB) from the University of Hertfordshire, UK (https://sitem.herts.ac.uk/aeru/ppdb/en/index.htm; Lewis et al., 2016; Lewis and Tzilivakis, 2019) (see list of active ingredients in Appendix A in Supporting Information). As proposed by EPPO (2010), the risk assessment was carried out selecting the lowest of the oral and contact LD_{50} values available across the different bee species (honey bees, bumble bees and other wild bees), to take the most conservative approach for the entire bee community (see Table S2).

However, *LD_{50}* values were mainly available for honey bees, sometimes for bumble bees, and much less frequently for other pollinators (Lewis et al., 2016; Lewis and Tzilivakis, 2019; Yasrebi-de Kom et al., 2019).

For the UK, the average annual application rate at 1km resolution (in kg/km^2/year) was obtained for 130 pesticide active ingredients from the *"CEH Land Cover® plus: Pesticides 2012-2016"* (Jarvis et al., 2019) of the Centre for Ecology and Hydrology (CEH, Wallingford, UK; https://www.ceh.ac.uk), across a four-year period (from 2012, 2013, 2014 and 2016). For the Netherlands, we created the average annual allowed application rate at 1km^2 (in kg/km^2/year) across a two-year period (2015 and 2016) for 179 pesticide active ingredients, combining allowed application rates produced by Yasrebi-de-Kom et al. (2019) and the BRP shapefiles for crops in 2015 and 2016 obtained from the RVO (https://english.rvo.nl).

2.5. Land cover composition data

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

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

2.6. Statistical analyses

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

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

The local abundance of honey bees is primarily determined by beekeeper behaviour rather than local effects of habitats (Büchler et al., 2014; Wood et al., 2020). As managed species they are influenced differently by environmental pressures compared to wild pollinators, and we therefore analysed *Apis mellifera* separately from non-*Apis* pollinators (i.e., other bees and hoverflies).

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

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

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

We used Variance Inflation Factors (VIF) to detect potential multicollinearity between variables in our selected statistical models, and no correlation was detected. Indeed, all VIF values were less than 1.5. In the selected models of crop pollinator abundance (*Apis* and non-*Apis*), VIF values ranged between 1.176 and 1.467, and in for the selected model of pollinator contribution to crop production, VIF values ranged between 1.001 and 1.114.

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

3. Results

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

Mean ozone value per study site varied from 0.140 to 0.144 mol.m^{-2} in the Netherlands and from 0.142 to 0.147 mol.m^{-2} in UK. These values represent ca. 7% of the full range of ozone values throughout the world (0.079 and 0.222 mol.m^{-2}, including areas of high ozone pollution as well as areas of low tropospheric ozone concentration). Previous studies that also used remote sensing data Sentinel-5P (e.g., Naqvi et al., 2022; Roman-Gonzalez et al., 2020), show a similar ranges of tropospheric ozone (and NO2) concentration for areas of comparable size (i.e., country scale: Peru, India). The mean tropospheric NO_2 per study site ranged from 27.8 to 76.5 µmol.m^{-2} (from 0 to 2.14 mmol.m^{-2} worldwide) and the gradient of fertilizer N input varied 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 index of pesticide exposure varied between 2 to 8 in the Netherlands and between 2 to 10 in UK, (i.e., between 2 and 10 high risk active molecules were applied in the 1km buffer around study sites). The proportion of natural and semi-natural habitats in the 1km^2 surrounding buffer varied from 0.1 to 37% in UK (with a mean=8.9 and median=4.4) and from 0 to 47% in the Netherlands (with mean=7.0 and median=1.8).

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

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

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

4. Discussion

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

4.1. Interacting effect of ozone with pesticide exposure

As expected (Hayes et al., 2012; Leisner and Ainsworth, 2012; Mills et al., 2013), ozone levels were negatively correlated to crop pollination. Recent studies have estimated that global agricultural losses due to high ozone levels totalled 79–121 million metric tons in 2000 with global economic losses ranging from $11 to $26 billion (Van Dingenen et al., 2009; Avnery et al., 2011a) and predicted increases of between $17 and $35 billion annually by 2030 (Van Dingenen et al., 2009; Avnery et al., 2011a). Such effects may be partly related to a reduction in pollen germination (Leisner and Ainsworth, 2012; Taia et al., 2013; Gillespie et al., 2015). Our results suggest that changes in pollination by insects (due to changes in flower visitation patterns) may also play an important role.

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

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

The negative relationship between ozone pollution and flower visitor abundance could be due to changes in plant-pollinator communication and flower attractiveness affecting crop pollinator foraging behaviour. Previous studies have shown that ozone induces changes in availability of floral resources by modifying flowering time and number of flowers, with some plant species being particularly sensitive (Hayes et al., 2012; Leisner and Ainsworth, 2012; Mills et al., 2013). Ozone also alters pollinator decision-making, modifying and reducing the volatile floral scents (Farré-Armengol et al., 2016; Fuentes et al., 2016; Saunier and Blande, 2019; Vanderplanck et al., 2021) and damaging pollinators olfactory organs (Dötterl et al., 2016; Vanderplanck et al., 2021).

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

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

4.2. Effect of the temperature on pollinators

The negative effect of the increase in mean annual temperature on the abundance of wild pollinators are in agreement with recent studies showing that climate change characterised by events of extreme heat, shifts European pollinator phenology with consequences on pollinators assemblages (Bartomeus et al., 2011; Duchenne et al., 2020; Vasiliev and Greenwood, 2021). The decade 2011-2020 (during which our studies were carried out) has been the hottest on record since the preindustrial period, with several summer heat waves being recorded in Northern and Western Europe (see WMO; https://public.wmo.int/en). Therefore, regions with slightly milder temperatures might be preferable for native pollinators. The fact that the negative effect of annual temperature on the abundance of non-*Apis* pollinators was reduced or became null at higher ozone concentration, could be due to the nature of pollinator species assemblages in regions with higher ozone levels. Further studies are needed to better understand if species persisting in regions of higher ozone levels have traits that affect their response to temperature. Nevertheless, this result is consistent with other studies that have found that the impact of temperature on pollination and crop production was less accentuated under high ground-level ozone concentrations (Mahmood et al., 2020). Yet, some species with a broad temperature range are likely to cope well with higher temperatures, since higher temperatures reduces the energy cost necessary to maintain optimum body temperature improving bee foraging activity (Abou-Shaara et al., 2017; Corbet et al., 1993; Kwon and Saeed, 2003). This could explain the positive effect of temperature on honey bee abundance observed here (foraging temperature ranging from 21.0 to 33.5 °C, Usha et al., 2020). Moreover, as it is a managed species, abundance of honey bees is primarily determined by beekeeper behaviour and do not have to adapt in the same way to environmental changes (Büchler et al., 2014; Wood et al., 2020).

4.3. Effect of nitrogen enrichment

Contrary to our expectations, we did not observe effects of nitrogen enrichment sources on crop pollinators and pollination. It is possible that the proxies used in our study do not adequately represent the real nitrogen exposure levels in our study fields. Indeed, while pollinators can be affected by local (i.e., within field) changes in nutrient availability (David et al., 2019), our proxies for nitrogen levels are taken at much broader scales. The amount of nitrogen that is in reality deposited in a specific location of the biosphere may not be well represented by the NO_2 levels measured in the troposphere at much larger spatial resolution of the available data from the Sentinel-5 satellite (i.e., 7×3.5 km). Similarly, the estimated mean application rate of fertilizers at each study region (which is based on average application levels for each crop at country level, and do not consider personal decisions of landowners) may not be of a sufficient resolution to detect changes in fertilization rate and its effects at the local scale. For example, the presence of (semi-)natural habitats in the landscape will inevitably decrease the estimated average fertilizer application rate at $1km^2$ resolution, while a high proportion of highly enriched crops, such as cereals, maize or fertilised grasslands (e.g., ray grass) will tend to increase the estimated average application rate. Future work involving farmer interviews asking for the actual amount of fertilizer applied to better characterize nutrient availability would be important. Moreover, although we had a clear gradient of N fertilizer input across sites, all study sites were located in landscapes with a critical positive surplus of nitrogen inputs (that goes up to 20 $t.km^{-2}$ for the year 2010) (European Environment Agency, 2020). Consequently, it is possible that throughout the study region pollinator communities are dominated by nitrophilous species (Carvalheiro et al., 2020) well adapted to high nitrogen conditions and the negative effects of nitrogen on pollinators and their contribution to crop production are no longer detectable in our specific study sites.

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

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

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

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

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

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

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Statement of authorship:

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

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Table 1. Sources of data for crop production and pollinator abundance included in the analyses. UK: United Kingdom; NL: Netherlands.

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

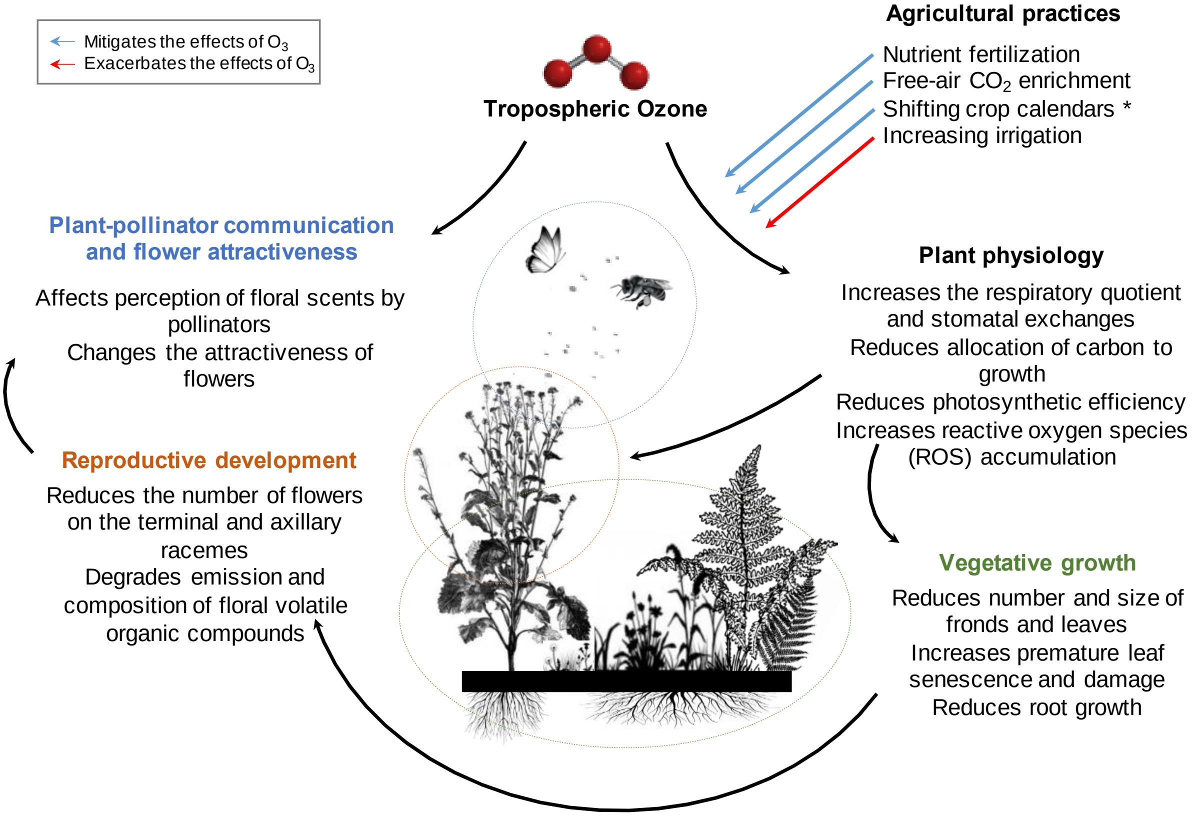

Figure 1. Review of known effects of tropospheric ozone on plants and plant-pollinator interactions. Blue and red arrows indicate agricultural practices that can respectively, mitigate or exacerbate effects of ozone on plant physiology (**Shifting crop calendars consists of a change in the sowing period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

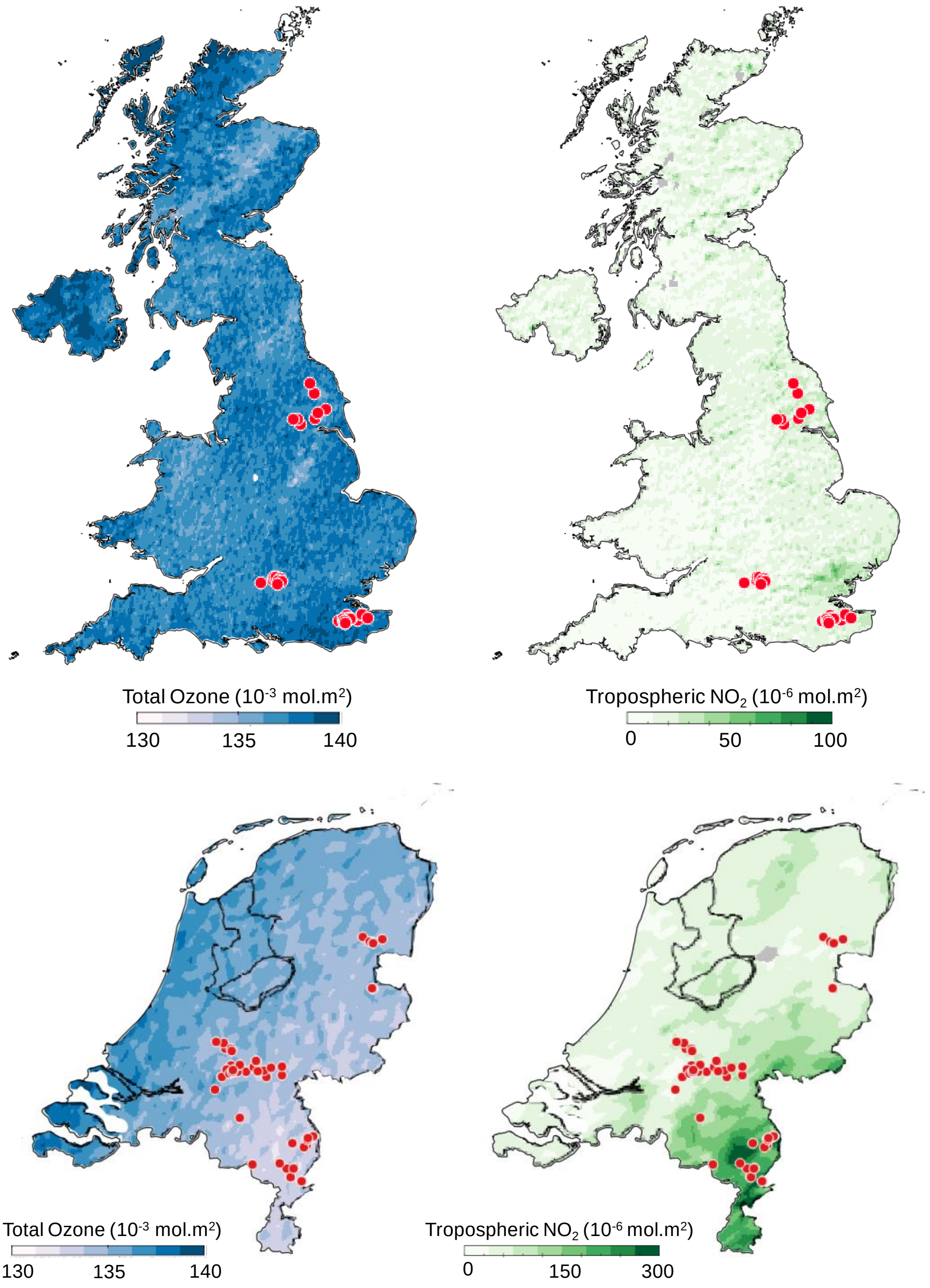

Figure 2. Sampling sites (red dots) included in the study and gradient of ozone (O_3) and dioxide nitrogen (NO_2) in the United Kingdom (UK) and the Netherlands (NL). O_3 and NO_2 gradients were mapped using the software NASA Panoply v.4.11.1 *(e.g. Sentinel-5 satellite data extraction for August 2019*)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

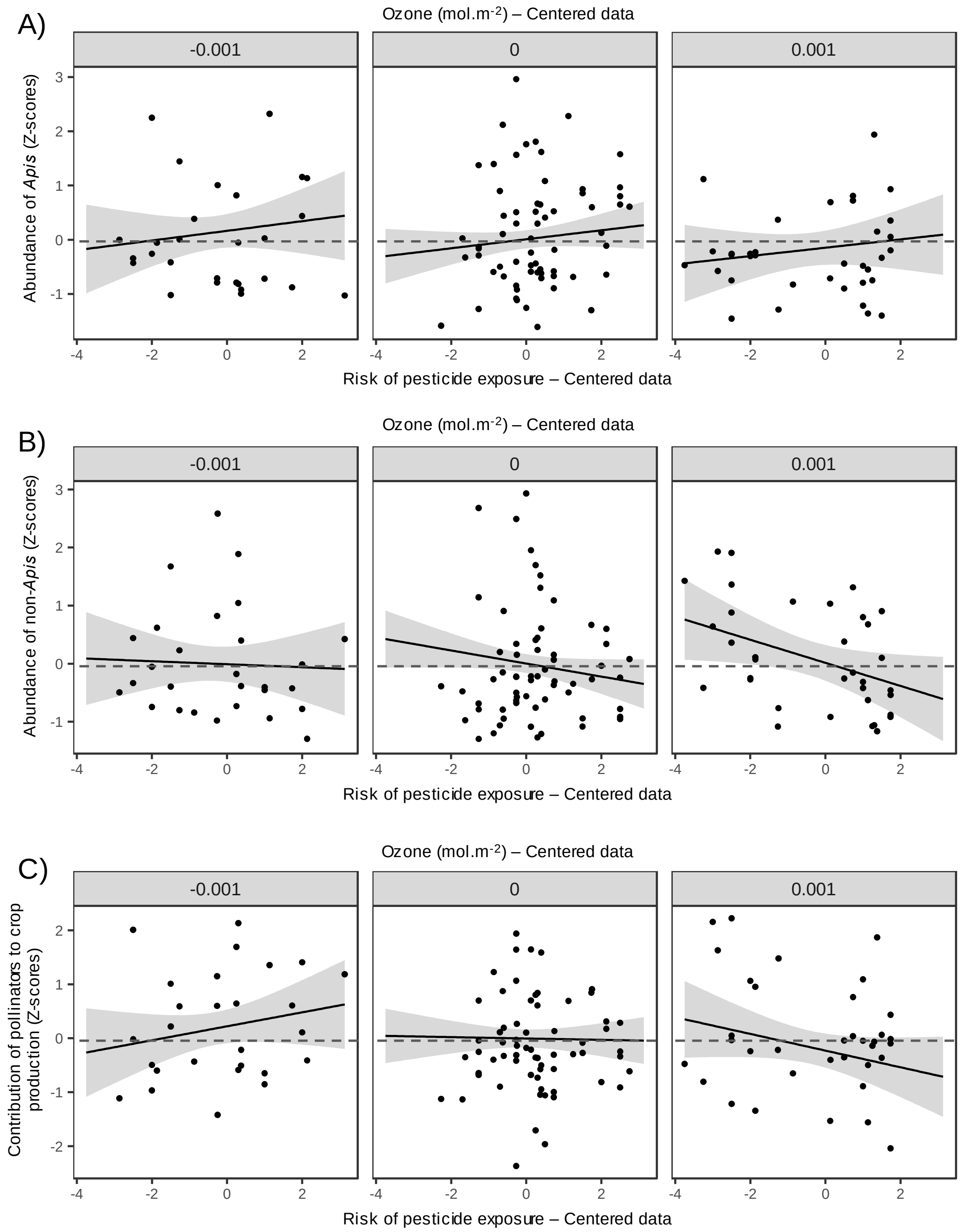

Figure 3. The increase in ozone concentration modifies the relationship between the risk of pesticide exposure and (A) the abundance of honey bees, (B) the abundance of non-*Apis* pollinators and (C) the contribution of pollinators on crop production. The dashed lines show a null difference of the response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