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Closing the global ozone yield gap: Quantification and co-benefits for multi-stress tolerance

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Indonesia, and for maize in China and the USA. Crucially, we also show that the same areas are often also at risk of high losses from pests and diseases, heat stress and to a lesser extent aridity and nutrient stress. In a solution-focussed analysis of these results, we provide a crop ideotype with tolerance of multiple stresses (including ozone) and describe how ozone effects could be included in crop breeding programmes. We also discuss altered crop management approaches that could be applied to reduce ozone impacts in the shorter-term. Given the severity of ozone effects on staple food crops in areas of the world that are also challenged by other stresses, we recommend increased attention to the benefits that could be gained from addressing the ozone yield gap.

Closing the global ozone yield gap: Quantification and co-benefits for multi-stress tolerance

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Key words: Ozone, wheat, soybean, maize, rice, pests and diseases, aridity, nutrient stress, heat stress, stress tolerant ideotype.

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Abstract

Increasing both crop productivity and the tolerance of crops to abiotic and biotic stresses are major challenges for global food security in our rapidly changing climate. For the first time, we show how the spatial variation and severity of tropospheric ozone effects on yield compare with effects of other stresses on a global scale, and discuss mitigating actions against the negative effects of ozone. We show that the sensitivity to ozone declines in the order soybean > wheat > maize > rice, with genotypic variation in response being most pronounced for soybean and rice. Based on stomatal uptake, we estimate that ozone (mean of 2010 - 2012) reduces global yield annually by 12.4%, 7.1%, 4.4% and 6.1% for soybean, wheat, rice and maize, respectively (the 'ozone yield gaps'), adding up to 227 Tg of lost yield. Our modelling shows that the highest ozone-induced production losses for soybean are in North and South America whilst for wheat they are in India and China, for rice in parts of India, Bangladesh, China and Indonesia, and for maize in China and the USA. Crucially, we also show that the same areas are often also at risk of high losses from pests and diseases, heat stress and to a lesser extent aridity and nutrient stress. In a solution-focussed analysis of these results, we provide a crop ideotype with tolerance of multiple stresses (including ozone) and describe how ozone effects could be included in crop breeding programmes. We also

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1. Introduction

To feed the rapidly growing global population, we need to develop a new generation of crop cultivars or varieties that will have both high productivity in future climates and high tolerance of the biotic and abiotic stresses that are likely to become more prevalent in the future (Gilliham *et al*., 2016). Candidate characteristics or traits are currently being tested in ideotype modelling (Semenov and Stratonovitch, 2013) and include improved light conversion efficiency, a longer duration of green leaf area for grain fill, a higher harvest index and optimal phenology. For example, varieties that use less water per unit of carbon fixed will have higher yield under drought conditions (Rebetzke *et al*., 2002) as will those with 'stay-green' characteristics during water stress (Jordan *et al*., 2012). Whilst it is widely recognised that rapid breeding programmes will have a vital role to play in adaptations of crops to climate change (Atlin *et al*., 2017), selection of traits for tolerance of one abiotic stress, tropospheric (ground level) ozone pollution, is currently omitted from such breeding programmes (Ainsworth, 2016; Frei *et al*., 2015). This is happening even though field experiments from nine countries representing three continents have shown that reducing ozone concentrations back to pre-industrial levels would give an average wheat yield benefit of 8.4% globally (Pleijel *et al*., 2018), a figure that is matched by modelling based on the stomatal uptake of the pollutant (Mills *et al.,* 2018a). Furthermore, an earlier meta-analysis of crop responses to ozone suggested that current ozone levels in the range 31 - 50 ppb (nmol

mol^{-1}, v/v) are reducing the yield of major food crops by 5.3 to 19% (Feng & Kobayashi, 2009). We undertook this new study to build a case for improving crop yields in our changing climate by closing the ozone-induced yield gap via the inclusion of ozone tolerance in crop breeding programmes, altered crop management and more stringent ozone precursor emission controls.

Tropospheric ozone pollution is formed from photochemical reactions involving anthropogenic and biogenic emissions and is involved in a complex web of interactions with ecosystems (Simpson et al., 2014). Whilst concentrations have been beginning to decrease in eastern USA and parts of Europe (2000 – 2014) due to precursor emission controls, they have been increasing rapidly in south (S) and east (E) Asia (Chang *et al*., 2017). Ozone is a powerful oxidant that is absorbed into leaves via open stomatal pores. Once inside the leaves, ozone reacts with biomolecules to form reactive oxygen species, triggering defence mechanisms that if overwhelmed lead to programmed cell death and a reduced extent and duration of functional green leaf area producing less photosynthate for seed fill (e.g. Ainsworth, 2016). Since pests and diseases (e.g. Oerke, 2006; Huysmanss *et al*., 2017), heat stress (e.g. Driedonks, *et al*., 2016), drought (e.g. Farooq *et al*., 2017) or reduced nutrient availability (e.g. Gastal & Lemaire, 2002) usually also reduce the extent and duration of the functional green leaf area, then in simple terms, each of these biotic and abiotic stresses result in the same endpoints – reduced yield quantity that is often associated with reduced quality.

So far, most crop breeding programmes have been targeted at increasing or maintaining the yield rather than increasing stability of yield under stress (Gilliham *et al.*, 2017). Because ozone concentrations tend to be very heterogeneous across natural and agricultural regions (Klingberg *et al*., 2012) as well as over seasons and years, it is not likely that traditional selection would unintentionally favour ozone tolerant crop genotypes. The reverse seems to be the case. For example, an analysis of ozone-exposure yield data for 49 soybean varieties from 28 field exposure studies showed that ozone sensitivity has increased by an average of 33% between 1960 and 2000 (Osborne *et al*., 2016). Similarly, modern wheat varieties are more sensitive than older varieties (Biswas *et al.*, 2008; Pleijel *et al*., 2006). Potentially, this increased sensitivity to ozone over recent decades is related to selective breeding for higher stomatal conductance (Roche, 2015) that inadvertently has increased the ingress of ozone into crops (Biswas *et al*., 2008; Osborne *et al*., 2016); further study is required to fully understand the mechanistic basis of this increasing sensitivity with time.

As with many abiotic and biotic stresses, genetic variation in plant response to ozone has been found for every species that has been tested. For the major grain crops, genetic variation in ozone response has been reported for wheat (Zhu *et al.*, 2011), rice (Frei *et al*., 2008; Shi *et al.*, 2009), soybean (Mulchi *et al*., 1988; Burkey & Carter, 2009; Jiang *et al.*, 2018), and maize (Yendrek *et al.*, 2017). Variation has also been reported for other crops including snap bean (Burkey *et al*., 2005; Yuan *et al.,* 2015) and tobacco (Heggestad, 1991). These assessments are based on different criteria including foliar injury and impacts on growth and yield parameters. Taken together, the evidence suggests that sufficient natural genetic variation exists to support improvement in crop stress tolerance either as sources of ozone tolerance genes or providing contrasting genotypes for mechanism studies to identify targets for molecular manipulation. Potential targets for breeding of ozone tolerance that have the greatest likelihood of success include reducing the stomatal uptake of ozone into the leaf and increasing its detoxification once inside the leaf (Feng *et al*., 2016; Frei *et al*., 2015).

To target the regions of the world where ozone tolerant crop varieties are most required, we need to understand which crops are most at risk and where they are growing in relation to current high-risk areas for ozone. We know from a recent analysis of ozone concentrations at over 3000 rural sites that the highest ozone values are in many of the world's important crop growing regions, including parts of the USA, Europe, India and China (Mills *et al*., 2018b).

Overall, the latter study showed that the global mean cumulative ozone exposure is double the critical level set by the United Nations as a target for ozone pollution control, above which direct adverse effects on sensitive vegetation may occur according to present knowledge (CLRTAP, 2017). Several studies have modelled ozone concentrations and predicted yield effects using concentration-based yield response functions applied at a range of scales from local (e.g. for India, Lal *et al*., 2017) to global (e.g. Avnery *et al*., 2011a,b; Van Dingenen *et al*., 2009). Whilst these studies indicate effects in the highest ozone areas, they do not take into account the constantly varying effects of soil moisture, air temperature, light and humidity on the uptake of the pollutant via the stomata. In Europe, field evidence for effects of ozone on crops and other types of vegetation shows that risk assessments based on modelled stomatal uptake or flux (Emberson *et al*., 2000, Simpson *et al*., 2007) provide a stronger indication of ozone effects than those based on concentration (Mills *et al*., 2011). Furthermore, dose-response functions for crops that are based on stomatal uptake are better correlated with yield effects than those based on concentration (Pleijel *et al*., 2000, 2007), providing additional support for their use.

With ozone concentrations increasing in rapidly developing regions and predicted to continue to increase in coming decades (Wild *et al*., 2012), it is timely to consider the options for increasing the tolerance of crops to this abiotic stress. In this study, our analysis included a two-step approach to addressing the ozone problem in crops: (i) a quantitative spatial analysis of the impacts of ozone on crop yield relative to impacts of other abiotic and biotic stresses and (ii) a qualitative analysis of crop traits, including defining an ideotype with multiple stress tolerance. As an initial step, we compiled dose-response data from experiments conducted around the world to determine the scope for breeding ozone tolerant varieties by showing the genotypic range in sensitivity for four staple crops: soybean, wheat, rice and maize. We then used the response functions to model the current impacts of ozone on each crop, showing the regions where the greatest production losses are likely to be occurring. Whilst we wait for ozone effects to be included in predictive crop yield modelling (as suggested by, for example, Challinor *et al*., 2009; Emberson *et al*., 2018; Lobell & Asseng, 2017), we sought to compare on a global scale the impacts of ozone on yield with the influence of other biotic and abiotic stress. Those selected were: pests and diseases (Oerke *et al*., 2006); aridity (Trabucco & Zomer, 2009); heat stress (developed from Deryng *et al.* (2014) and Teixeira *et al*. (2013)); and soil nutrient stress (GAEZ). The effects of all five stresses were considered in more detail for India where there are major challenges for crop production and food security (Jaswal, 2014) and where global assessments consistently predict high risk from elevated ozone (e.g. Avnery *et al*., 2011a,b; Van Dingenen *et al*., 2009). In the second part of the study, we conducted an analysis of the plant traits associated with multiple stress tolerance, and considered the trade-offs and benefits of introducing ozone tolerance in crops for cross-tolerance of other biotic and abiotic stresses. This part of the study culminated in the design of an ideotype for an ozone tolerant crop that would also provide tolerance of co-occurring stresses. In an extended discussion, we assess the results from the two parts of the study and consider viable options for reducing the negative effects of ozone on yield, including crop management, breeding and global efforts to reduce ozone pollution.

2. Materials and Methods

2.1 Global spatial analysis of crop yield constraints caused by ozone

2.1.1 Crop production

Global modelled crop production data (year 2000, 0.0833° (5 arc minute) resolution) was downloaded from the GAEZ (Global Agro-Ecological Zones, v. 3) data portal (http://www.fao.org/nr/gaez/en/) for soybean, wheat, rice and maize. Irrigated and rain-fed production data was collected for each crop. Using ArcMap v. 10.3 (Environmental Systems Research Institute, Redlands, CA, USA), a 1° by 1 ° global grid was created. For each crop, production was summed per grid cell. Each cell was classed as irrigated or non-irrigated based on the percentage of irrigated crop production per cell. To define a threshold for irrigated versus non-irrigated, we first produced frequency distributions of the percentage of irrigated production for each crop (Supporting Information, Fig. S1). These showed that the majority of cells for each crop were either fully irrigated or fully rain-fed. A threshold of 75% irrigated was used to identify those cells where the majority of the production was on irrigated land. Production for the period 2010-12 was estimated per grid cell by applying a conversion factor from FAOSTAT national production data available, averaged for the years 1999 – 2001 (average production for 2010-12/average production for 1999-2001). Only cells with > 500 tonnes (0.0005 Tg) crop production in 2010-12 were included in the analysis.

As discussed in Mills *et al*. (2018a), each 1° by 1° grid cell was assigned to a climatic zone, using the global 'Climatic Zone' GIS raster layer produced by the European Soil Data Centre (ESDAC) at JRC (Joint Research Centre). For each climatic zone, a 90 day growing period was derived per crop (Table S1), with climatic zones illustrated in Fig. S2. Data sources for assigning crop timings are provided with Table S1. For ease of comparison of effects between crops, only the main growing season per year was used for each crop.

2.1.2 Intra- and inter-specific sensitivity of crops to ozone

To determine the relative sensitivity of the four crops to ozone together with the between-variety variation in response to the pollutant, it was necessary to update existing response functions based on ozone concentration as stomatal uptake-based functions are currently only available for wheat. We collated dose-response data from the scientific literature using the method developed by Osborne *et al.* (2016) for soybean and the commonly reported ozone metric, M7 (7 hour mean, averaged from 09:00 to 15:59). The soybean dose response relationship from Osborne *et al.* (2016) was included in our analysis, whilst response functions for wheat, rice and maize provided in Mills & Harmens (2011) were updated with more recent published data (Web of Science and Google Scholar searches conducted between April and October 2017 using the search terms "ozone and yield and *crop name*"). Studies were only included if they met a number of selection criteria. The duration of ozone exposure must have spanned at least 60% of the 90 day growing season for each crop and ozone levels during exposure were up to 100 ppb for wheat and 170 ppb for other crops. Experiments were included if carried out in Open Top Chambers (OTCs), ambient air or large closed chambers/greenhouses (with the air stirred by fans, minimum size 2.6 by 2.2m). Data from both container and field-sown experiments were used to ensure a wide variety of points from different varieties were included. If the seasonal M7 was not given in the text, this was calculated either using the conversion equations provided in Osborne *et al.* 2016 (e.g. for 24 hour mean to M7) or information contained in the experimental methodology of the study. As there was no new published data available at the time of analysis for maize, the response function from Mills & Harmens (2011) was used. Yield data from different experiments were standardised as first described by Fuhrer (1997) and recently re-described by Osborne *et al.* 2016. Thus, for each set of experimental data, linear regression was used to determine the yield at 0 ppb of ozone (the intercept of the line); this value was the reference for calculating the relative yield (i.e. relative yield = actual yield/yield at 0 ppb).

Individual variety dose-response functions were derived for wheat and rice for the four varieties with the most data points. Following Osborne *et al.* 2016, yield reduction estimates ($RYL_{c,p}$) were then calculated for varieties showing statistically significant declines in yield with increasing ozone by calculating the difference in percentage yield loss at 55ppb (representing current M7) relative to that at 23 ppb (representing pre-industrial M7).

2.1.3 Yield constraints caused by ozone

The EMEP MSC-W (European Monitoring and Evaluation Programme, Metereological Synthesising Centre-West) chemical transport model (version 4.16, Simpson *et al*., 2012, 2017) was used to derive daily POD_3IAM (Phytotoxic Ozone Dose above 3 nmol m^{-2} s^{-1}, parameterised for Integrated Assessment Modelling, CLRTAP, 2017) values for the years 2010 to 2012 per 1° by 1° grid cell as described by Mills *et al*. (2018a). POD_3IAM is parameterised for a generic crop represented by wheat (CLRTAP, 2017) and represents the accumulated stomatal uptake of ozone, modelled from the hourly mean values for ozone, temperature, vapour pressure deficit, irradiance and soil moisture (Mills *et al.* 2018a). Evaluation of the EMEP model performance is also presented in Mills *et al*., 2018a, and is summarised in the Supporting Information for the current paper (T1).

For each crop, the accumulated 90 day POD_3IAM was then calculated per cell using appropriate climate-specific 90 day growing periods (Table S1, Fig. S2), and an average calculated for the period 2010-2012. For example, for soybean in warm temperate climates in the Northern Hemisphere, the time interval was day 182 to day 271. The EMEP model generated irrigated (without soil water limitation) and non-irrigated (rain limited) POD_3IAM values. For grid cells classed as irrigated for each crop (See Section 2.1.1), the irrigated POD_3IAM value was used to calculate percentage yield loss, otherwise the non-irrigated POD_3IAM was used. This approach allowed crop-specific irrigation usage to be taken into account, and was different to Mills *et al*. (2018a) where POD_3IAM values were weighted by the proportion of irrigation use within a 1 x 1° cell. The global distribution of POD_3IAM for each crop is provided in Fig. S3.

Yield loss due to ozone was first calculated for wheat using the most recent methodology adopted by CLRTAP, 2017. This method also differed slightly from that used in our earlier study (Mills *et al*., 2018a) in that a reference POD_3IAM value to represent ozone uptake at pre-industrial or natural ozone levels was subtracted before crop loss was calculated (CLRTAP, 2017). This value (0.1 mmol m^{-2}, Equ. 1) was the mean POD_3IAM for the experimental conditions included in the dose-response relationship, assuming constant 10 ppb ozone throughout the 90d period. The equation used to determine percentage yield loss was:

$$\% \text{ Yield loss} = (POD_3IAM - 0.1) * 0.64 \qquad \text{[Equ. 1]}$$

Where 0.64 is the slope of the relationship between POD_3IAM and percentage yield reduction (Mills *et al*., 2018a) and represents the percentage reduction per mmol m^{-2} POD_3IAM.

For soybean, maize and rice, the climate-specific grid square POD_3IAM values were first used to calculate yield loss using the wheat equation (Equ. 1), and the resultant value was then multiplied by the relative sensitivity of the crop compared to wheat, RS_w. The latter was derived by dividing the slope of the M7 response function for the crop (Fig. 1) by that for wheat. Production loss per crop was calculated per grid square using the following equation:

$$\text{Production loss (tonnes)} = \text{Crop production} * (\% \text{ yield loss}/100) \qquad \text{[Equ. 2]}$$

2.2 Global spatial analysis of yield constraints caused by other stresses

2.2.1 Yield constraints caused by pests and diseases

Oerke *et al.* (1994; 2006) provide estimates for pre-harvest crop losses due to weeds, animal pests, (arthropods, nematodes, mammals, slugs and snails, birds), pathogens and viruses for several major global crops, using data compiled from the literature. This database provides regional percentage yield loss estimates up to 2004 for 11 crops, including soybean, wheat, rice and maize, and is available from the Centre for Agriculture and Biosciences International

(CABI) Crop Protection Compendium (CABI, 2005). A value for mean percentage yield loss due to pests and diseases for the period 2002-04 was assigned to each 1° by 1 ° grid cell, based on the country and region of the world the cell was located in. If a cell contained land from more than one country, it was assigned to a country based on where the majority of the crop was growing in the cell. Data was available for 19 global regions (Oerke *et al*., 2006). In this study, data were used that represented the remaining crop yield losses after crop protection practices had been applied.

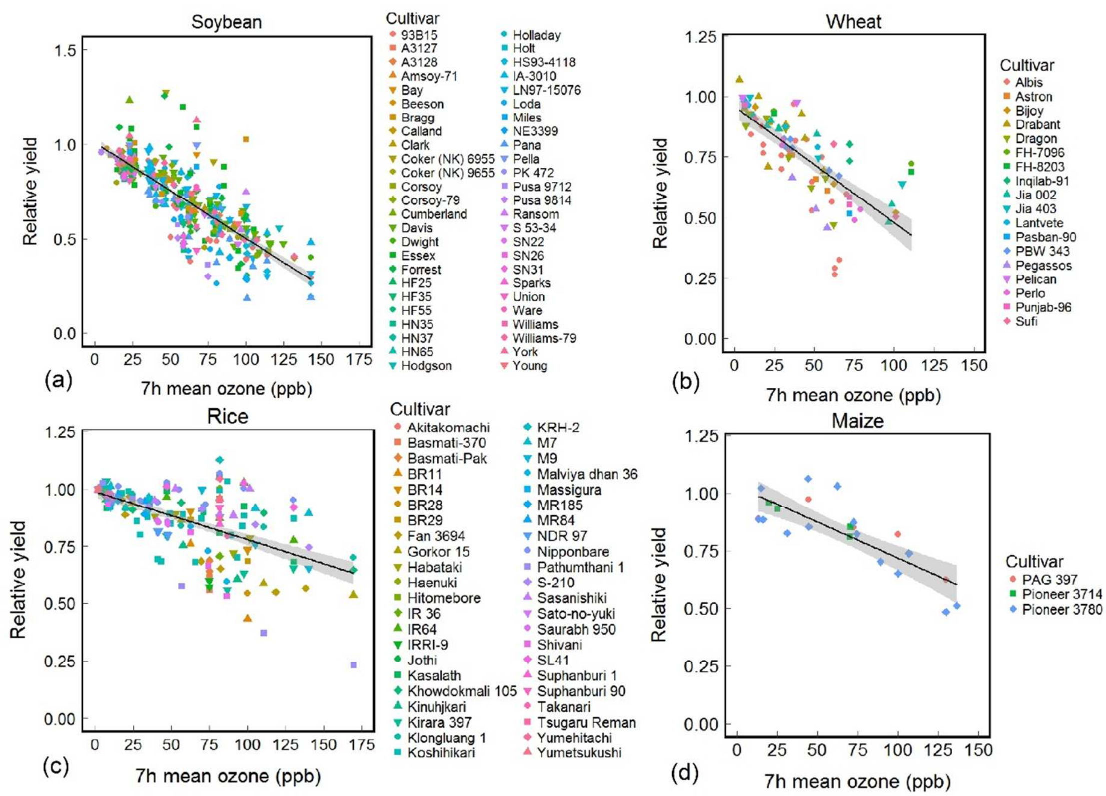


Fig. 1. Response functions for (a) soybean, (b) wheat, (c) rice and (d) maize derived from published data using the growing season ozone (7h mean, M7 in ppb) in the experiments. Data points are presented per cultivar/variety, with sources of data provided in the Supporting Information (Table S3). The response functions are: Soybean, RY = -0.0050x + 1.001 (r^2 (adj) = 0.625, p<0.001); Wheat, RY = -0.0048x +0.96 (r^2 (adj) = 0.547, p<0.001); Rice, RY= -0.0021x +0.987 (r^2 (adj) =0.347, p <0.001); and Maize, RY = -0.0031x + 1.03 (r^2 (adj) = 0.617, p <0.001).

2.2.2 Yield constraints caused by soil nutrients

Soil nutrient classifications (nutrient availability and nutrient retention) at 0.083° by 0.083° resolution were downloaded from the GAEZ (v. 3) data portal (http://www.fao.org/nr/gaez/en/) in June, 2017. The soil qualities (nutrient availability and retention) in the GAEZ dataset have been derived from combinations of soil attributes, using data in the Harmonized World Soil Database (HWSD, v. 1.1, FAO/IIASA/ISRIC/ISS CAS/JRC 2009). Nutrient availability refers to soil fertility, and classification is based on soil texture, soil organic carbon, soil pH and total exchangeable bases. The nutrient retention capacity of soil is based on the ability of soil to retain added nutrients against losses due to leaching. Classification of nutrient retention has been derived from soil texture, base saturation, cation exchange capacity of the soil and of the clay fraction and soil pH. In the GAEZ dataset, nutrient availability and retention are classed separately for topsoil (0-30cm) and subsoil (30-100cm) and then combined by weighting based on the prevalence of active roots (Fischer *et al.*, 2012). The GAEZ classes for soil nutrient availability and nutrient retention were combined in this study to produce five soil nutrient stress classes (summarised in Table 1, further details provided in Table S2). The soil nutrient class making up the majority of each 1° by 1° grid cell in areas where crops were growing, was used to represent the class for each cell.

2.2.3 Yield constraints caused by heat stress

Following the methods of Challinor *et al.* (2005), subsequently used by a number of other studies (e.g. Deryng *et al.*, 2014; Teixeira *et al.*, 2013), a heat stress index was calculated per grid cell for each crop to determine if the daily temperature within a 30 day thermal sensitive period (TSP) exceeded the tolerance thresholds for each crop. This method assumes that damage to crops occurs when daily temperatures exceed a critical temperature (T_{crit}, °C) and maximum damage occurs when temperatures exceed the limit temperature (T_{lim} °C). Using information on the reproductive phase for each crop (FAO), the thermal sensitive period was designated as days 40-70 of the 90 day growing period (which varies with climate zone for each crop, Table S1). Following Deryng *et al.*, (2014), the daily effective temperature (T_{eff}, °C, (daily mean temp + daily max temp)/2), used as a measure of the daily temperature when photosynthesis is taking place, was calculated per grid cell using global hourly temperature data for the period 1990-2014 at 0.5° by 0.5° resolution. The temperature data were from the European Centre for Medium-Range Weather Forecasts Integrated Forecasting System (ECMWF-IFS, www.emwf.int/research/ifsdocs/), as prepared for use by the EMEP model.

For each crop, a daily heat stress value (f HSd) was then calculated for each day within the TSP, per grid cell. As we required an index that could be used to detect increasing levels of stress (i.e. an index scaled from 0 to 1), heat stress was calculated following Teixeira *et al.* (2013) (Eqn. 3).

$$f\,\mathrm{HSd} = \begin{cases} 0 & \text{for } T_{eff} < T_{crit} \\ \dfrac{T_{eff} - T_{crit}}{T_{lim} - T_{crit}} & \text{for } T_{crit} \leq T_{eff} < T_{lim} \\ 1 & \text{for } T_{eff} \geq T_{lim} \end{cases} \qquad \text{[Equ. 3]}$$

An average value was then calculated across the 30 day TSP to give the final heat stress index value, (f HS) per grid cell (Eqn. 4).

$$f\,\mathrm{HS} = \frac{\sum_{j=1}^{TSP} (f\,\mathrm{HSd})}{TSP} \qquad \text{[Equ. 4]}$$

Critical and limiting temperatures per crop were taken from Deryng *et al.* (2014) (maize, wheat and soybean) and Teixeira *et al*. (2013) (rice). These were: Soybean (35°C for T_{crit} and 40°C for T_{lim}), wheat (25 and 35 °C), rice (35 and 45°C) and maize (32 and 45°C).

2.2.4 Yield constraints caused by aridity

Global Aridity Index data (Trabucco & Zomer 2009) were downloaded from the CGIAR-CSI GeoPortal (http://www.csi.cgiar.org). The mean Aridity Index for the period 1950-2000 (0.0083° by 0.0083° resolution) was calculated as:

$$\text{Aridity Index (AI)} = MAP / MAE \qquad \text{[Equ. 5]}$$

Where MAP is the Mean Annual Precipitation and MAE is the Mean Annual Potential Evapo-Transpiration.

Mean annual precipitation values were obtained from the WorldClim Global Climate Data (Hijmans *et al.* 2004), for years 1950-2000, while mean annual values of Potential Evapo-Transpiration (PET) were calculated using the average monthly PET values from the Global-PET model (Trabucco & Zomer, 2009). The mean Aridity Index per cell was calculated for each 1° by 1° grid cell where there is production for the crop.

2.3 Comparative analysis of effects of five stresses using a Yield Constraint Score (YCS)

As percentage yield loss data was only available for ozone and pests and diseases data, a percentage scale could not be used for all stresses. To overcome this problem, a yield constraint score (YCS) on a scale of 1 – 5 was developed for each abiotic and biotic stress to show spatially where each constraint is predicted to be impacting on yield and to provide some indication of the magnitude of the effect (Table 1). Yield loss was split into the same five percentage yield loss classes for ozone and pests and diseases, with the highest class being >40% and expected to be comparable to severe stress for all yield constraints. Soil nutrient retention and availability were combined to give five overall classes (Table S2). The aridity climate classes used were from the Generalized Climate Classification Scheme (UNEP 1997), while the heat stress index was classified following the methods of Teixeira *et al*. (2013). To identify those areas of the world with the highest combined stresses, the YCS for all five stresses were summed (YCS_{all}).

Table 1: **Categories of Yield Constraint Score (YCS) for ozone, pests and diseases, soil nutrients, heat and aridity** (See text for explanations and justifications of categories).

Stress	Attribute	Year(s) of data	Yield constraint score (YCS)				
			1	2	3	4	5
Ozone	% Yield Loss	Mean of 2010-2012	0 to 5	5 to 10	10 to 25	25 to 40	> 40
Pests and diseases	% Yield Loss	Mean of 2002 - 2004	0 to 5	5 to 10	10 to 25	25 to 40	> 40
Soil Nutrients	Retention	HWSD data, 2009, downloaded in June, 2017	None or slight	Slight to moderate	Slight to severe	Moderate to severe	Moderate to very severe
	Availability		None	Slight to moderate	Moderate to severe	Severe	Severe to very severe
	Overall		None	Slight	Moderate	Severe	Very severe
Heat	Index	Mean of 1990 - 2014	0	<0.05	0.05 to 0.15	0.15 to 0.3	> 0.3
Aridity	Index	Mean of 1950 - 2000	>0.65	0.5 to 0.65	0.2 to 0.5	0.03 to 0.2	<0.03
	Climate class		Humid	Dry sub-humid	Semi-arid	Arid	Hyper arid

For description of effects of the five stresses, results are described as regional and national averages, with the mean YCS and YCS_{all} rounded to the nearest integer, reflecting their categorical nature. The regional classification of countries used is that adopted by the Task Force on Hemispheric Transport of Air Pollutants (HTAP) of the Convention on Long-range Transboundary Air Pollution (LRTAP, Dentener & Guizzardi, 2013). Region names are provided in full in the text the first time they are used and thereafter are referred to by the HTAP three letter codes. The region names, three letter codes and a map illustrating the countries included per region are provided in Fig. S4.

2.4 Qualitative analysis of plant traits associated with multiple stress tolerance

The scientific literature on crop stress tolerance was reviewed between June and December, 2017, with the aim of developing an ideotype for an ozone- and multi-stress tolerant crop. This analysis identified target traits to induce ozone tolerance, including reducing the effects on panicles, leaves and roots. It also considered the benefits and trade-offs for tolerance of other stresses, of introducing ozone tolerance into crops.

3. Results

3.1 Quantification of the global impacts of ozone and other stresses on crop yield

3.1.1 Intra-specific sensitivity to ozone

A comprehensive collation of published data on the yield responses of soybean, wheat and rice to ozone resulted in a database representing 52, 18 and 44 varieties, respectively (Fig. 1a-c, with data sources in Table S3). Ozone-response data for these three crops provides good representation of the areas where the crops were grown: Soybean (East Asia (EAS), North America (NAM) and South Asia (SAS)); wheat (Europe (EUR), EAS, SAS) and rice (EAS and SAS). In contrast, only three varieties have been tested to date for yield responses in maize (Fig. 1d), with these experiments being conducted in the USA during the 1980s and early 1990s. For each crop, there was a significant negative response to ozone ($p<0.001$), with the slope of the negative relationships declining in the order soybean (-0.0050) > wheat (-0.0048) > maize (-0.0031) > rice (-0.0021). Within each response function, variation in ozone sensitivity due to variety provided scatter in the range of sensitivity.

For each crop, some varieties were more tolerant to ozone than others, indicating that there is scope for selecting more tolerant varieties for immediate use or as part of a breeding programme for new varieties. For soybean, $RYL_{c,p}$ ranged from 13.3% to 37.9%, with the three most sensitive varieties being the Indian varieties 'PK472', 'Pusa 9712' and 'Pusa 9814' (Osborne *et al.* 2016). For wheat, the RYLc,p for the four varieties with the most data was 16.4% ('Drabant'), 19.3% ('PBW 343'), 26.4% ('Dragon') and 32.5% ('Albis'). While the 44 rice varieties showed a range of sensitivities to ozone (Fig. 1c), overall, rice was the least sensitive of the four crops investigated. Of the four rice varieties with the most data, 'Koshihikari' showed a $RYL_{c,p}$ of 4.7%, 'Nipponbare' showed no significant negative relationship between relative yield and M7 ($p>0.05$), while 'Kasalath' and 'Kirara 397' had a RYLc,p of 6% and 11.1 % respectively. The rice variety 'Pathumthani-1' showed a higher $RYL_{c,p}$ of 18.1%, however only 5 data points were available, therefore further study may be required to confirm this result. For maize, the r^2(adj) was 0.62 for the response function ($p<0.001$, Fig. 1d), with a $RYL_{c,p}$ for all three varieties of 10%. The RYLc,p for 'Pioneer 3780' with 14 data points was 15.7% whilst RYLc,p was not calculated for 'PAG 397' as the response function for the 5 data points for this variety was not significant ($p=0.08$).

3.1.2 Spatial analysis of the global impacts of multiple stresses on crop yield

<u>Soybean</u>

The highest ozone-associated production losses (in Tg per 1 x 1° grid square) for soybean are predicted to be in NAM and South America (SAM) (Table 2), particularly in central and E

USA, S Brazil and N Argentina (Fig. 2, Table S4). However, the percentage yield losses are predicted to be lower for Brazil (12.5 – 15%) and Argentina (7.5- 10%) than for the USA (> 20% in large areas), showing that in high producing areas where ozone concentrations are more moderate (as indicated by the percentage losses), high total production losses can still be expected. In the rest of the world, production losses due to ozone in excess of 0.01 Tg per 1 x 1 ° grid square are predicted for parts of China, India and S and E Europe. In each of these areas, the production loss was not as high as expected from percentage yield losses in excess of 20%, because soybean is not widely grown.

The YCSs for ozone were mainly score 3 for the highest producing regions, with some areas with a score of 4 in E USA, NE India and China (Fig. 2, Tables 2 and S4). There is overlap between these areas and the areas with the highest YCS_{all}. Other areas with a relatively high YCS_{all} such as parts of Sub-Saharan Africa (SSA) and SEA are not predicted to have high production losses due to ozone because of lower percentage yield losses and/or low production totals per region. For soybean, the YCS for pests and diseases is 3 or more over most of the growing area, and particularly high (score of 5) in parts of SSA, SAS and SEA. The largest YCS values for nutrient availability (scores of 4 and 5) are in areas of SE USA, S Brazil and SEA including Thailand, Malaysia and Indonesia. Whilst heat stress YCSs are lower than those for aridity, the areas affected by both stresses largely coincided in soybean growing areas.

Overall, for soybean, the global mean YCS for ozone of 3 is one category below that for pests and diseases, and one higher than that for nutrients and aridity (Table 2). The mean YCSs for ozone were in the range 2 - 3 for the five highest producing regions (SAM, NAM, EAS, SAS, Russia (RBU)), with YCSs being in the range 3 – 5, 1 – 3, 1 - 2 and 1 - 2 for pests and diseases, nutrients, heat and aridity, respectively (Table 2).

Table 2: **Production and regional mean Yield Constraint Score (YCS, rounded to nearest integer) for the five highest producing regions for soybean, wheat, rice and maize.**

Soybean	**Production (Tg)**	**Ozone**	**Pests & diseases**	**Nutrients**	**Heat**	**Aridity**
Global	253.6	3	4	2	2	2
South America	125.8	2	4	3	2	1
N America	90.3	3	3	2	2	1
East Asia	14.9	3	4	2	1	2
South Asia	13.2	3	5	1	2	2
Russia	3.6	2	4	1	1	2
Wheat	**Production (Tg)**	**Ozone**	**Pests & diseases**	**Nutrients**	**Heat**	**Aridity**
Global	673.3	2	4	2	3	2
Europe	163.8	2	3	2	2	1
East Asia	118.9	2	3	2	2	2
South Asia	118.1	2	4	2	4	2
N America	84.2	2	4	2	2	2
Russia	66.1	2	4	2	2	1
Rice	**Production (Tg)**	**Ozone**	**Pests & diseases**	**Nutrients**	**Heat**	**Aridity**
Global	716.8	1	4	2	1	2
East Asia	221.2	2	4	2	1	2
South Asia	219.2	2	5	2	2	2
South East Asia	204.5	1	4	3	1	1
South America	22.7	1	4	3	1	1
Sub Saharan Africa	21.1	1	5	2	1	2
Maize	**Production (Tg)**	**Ozone**	**Pests & diseases**	**Nutrients**	**Heat**	**Aridity**
Global	869.1	2	4	2	2	2
N America	313.1	2	3	2	2	2
East Asia	194.4	2	4	2	2	2
South America	91.7	2	4	3	2	2
Europe	75.6	2	3	1	2	1
Sub Saharan Africa	60.0	1	5	2	2	2

<u>Wheat</u>

By far the highest production losses due to ozone per grid square for wheat are predicted for India and China, with large areas in N India and NW China having over 15% yield losses amounting to production losses in excess of 0.1 Tg (Fig. 3, Table S5). Production losses are also predicted to be high in the highest wheat producing areas of Europe (including France and Germany) and central states of the USA. The mean YCS for ozone globally was 2, reflecting the same mean score in the 9 highest wheat producing regions (Tables 2 and S5), and matching that globally for nutrients, and aridity. The highest predicted production losses due to ozone only overlapped with areas of the highest YCS_{all} in NW India, Pakistan and S USA (Fig. 3). Scores of 3 and above coincide for ozone, pests and diseases, heat and nutrients in a wider area including parts of EAS, SAS and NAM, whilst YCSs for aridity are lower in several parts of this region than for ozone. This study also indicated that the highest YCS values for all stresses are for heat (score 5) in areas of Northern Africa (NAF), SAS, SAM, SSA and SAM (particularly Argentina). Scores for pests and diseases are 3 or more across most of the wheat growing areas. YCSs for nutrient availability are generally the lowest of the five stresses, although there are some high risk areas with values of 4 and above in, for example, NW SAS, Central Asia (CAS) and NE EUR (e.g. Finland), E NAM and SAM (e.g. Brazil). The highest YCSs for aridity are in a zone that includes parts of NAF, the Middle East (MDE) and SAS, CAS and EAS. For the five main wheat producing regions, the mean YCS for ozone was 2 representing 5 - 10% yield loss, whilst it was 3 - 4 for pests and diseases and mainly 1-2 for the other three stresses (Table 2).

Rice

Production losses due to ozone are predicted to only be in excess of 0.1 Tg per grid square in parts of India, Bangladesh, China and Indonesia (Fig. 4). In these areas, the percentage yield losses are mainly in the range 7.5 – 12.5%, resulting in grid square ozone YCSs that are usually either 2 or 3. Across all of the rice growing regions, the mean YCS for ozone per region is either 1 or 2, resulting in a global mean score of 1 and a score of 2 in the two highest producing regions (Tables 2 and S6). YCSs of 3 for ozone occurred in areas of India and China where the YCS_{all} was usually in the highest range for rice of 13 – 15. Overall, the highest mean YCS for this crop is for pests and diseases, being score 5 in most of the rice growing areas of SSA and SAS. Nutrient YCSs are 4 or more in many parts of SSA, SAM (particularly Brazil), EAS and SAS. Heat stress is predicted to be less of a problem for rice than for wheat, with few regions having a score of 3 or more. Indeed, the regional mean YCS for heat stress in rice is mostly either 1 or 2 (Tables 2 and S6). In the three highest producing rice regions, EAS, SAS and SEA, irrigation usage is 96%, 74% and 28%, respectively (Table S5). Here, the aridity score is predicted to be 3 or more only in areas of NW China and W India.

<u>Maize</u>

China and the USA are the two countries predicted to have the largest areas where production losses due to ozone for maize that exceed 0.1 Tg per 1° x 1 ° grid square (Fig. 5). In these areas, the percentage yield losses are mainly in the range 7.5 – 15% for the USA and 12.5 – 15 % for China. There are also high-risk areas in S EUR, for example, in parts of S France and N Italy and in NAF (particularly Egypt). These areas generally have a YCS_{all} for maize in the range 10 – 15, and are not in the areas with the highest YCS_{all} for maize of >15. The latter are mainly found in parts of SAS and SSA, with occasional small areas elsewhere. The mean YCS for ozone for the four highest maize producing regions (NAM, EAS, SAM and EUR) is 2 (Tables 2 and S7). For the stresses other than ozone, the highest scores for YCS are for pests and diseases, with scores of 5 predicted in most of SSA, SAS and SEA. YCSs of 4 and above are predicted for nutrients in much of SSA (particularly in western countries), SEA, large areas of SAM (particularly Brazil), parts of E USA and small areas of Europe. For maize, the YCSs for aridity are highest in eastern NAM and SAM, parts of NAF (particularly W Egypt), MDE, SAS (particularly NW India) and EAS (particularly NE China). Heat stress YCSs are lower for maize than for wheat, indicating that the main areas of concern for this crop are in W SSA, W MDE and SAS. Globally, the mean YCS for maize is 4 for pests and diseases and 2 for each of the other four stresses (Table 2).

3.1.3 Case study - India

It is clear from the results presented above that the five environmental stresses included in this study are all predicted to be having relatively high impacts on yield in several states of India. We selected this country for a more in depth analysis. Although the spatial data for India is present on the global maps in Figs 2 - 5, for ease of interpretation, we have produced additional maps for India for wheat and rice, the two most important crops by production in Fig. S5 and S6, respectively. At the national scale, the mean YCSs for the crop with the highest total production in India, wheat, are 3, 4, 2, 4 and 2 for ozone, pests and diseases, nutrients, heat and aridity, respectively (Table S5). For rice, the second most important crop by Tg produced in India, the YCSs for the same five stresses respectively are 2, 5, 1, 2 and 2 (Table S6). As the data for the risk of losses due to pests and diseases was only available at the national scale for India, with YCSs of 4 for wheat and 5 for rice, these effects were not included in this spatial analysis, conducted at the 1 x 1 ° scale.

For wheat, the highest production is in the adjacent N states of Uttar Pradesh, Madhya Pradesh, Haryana, Rajasthan and Punjab (Fig. 6). Together, these five states account for 85% of Indian wheat production. Predicted percentage yield losses due to ozone are in the range 15 – 20 % (mean of 16.4%) in most of the wheat producing areas of Uttar Pradesh, the state with the highest wheat production, resulting in a mean ozone YCS of 3 (Fig. 6 and S5). The mean YCS for ozone was 3 for Haryana, Rajasthan and Punjab and 2 for Madhya Pradesh where ozone uptake is lower (Fig. S5). Although the highest percentage yield losses due to

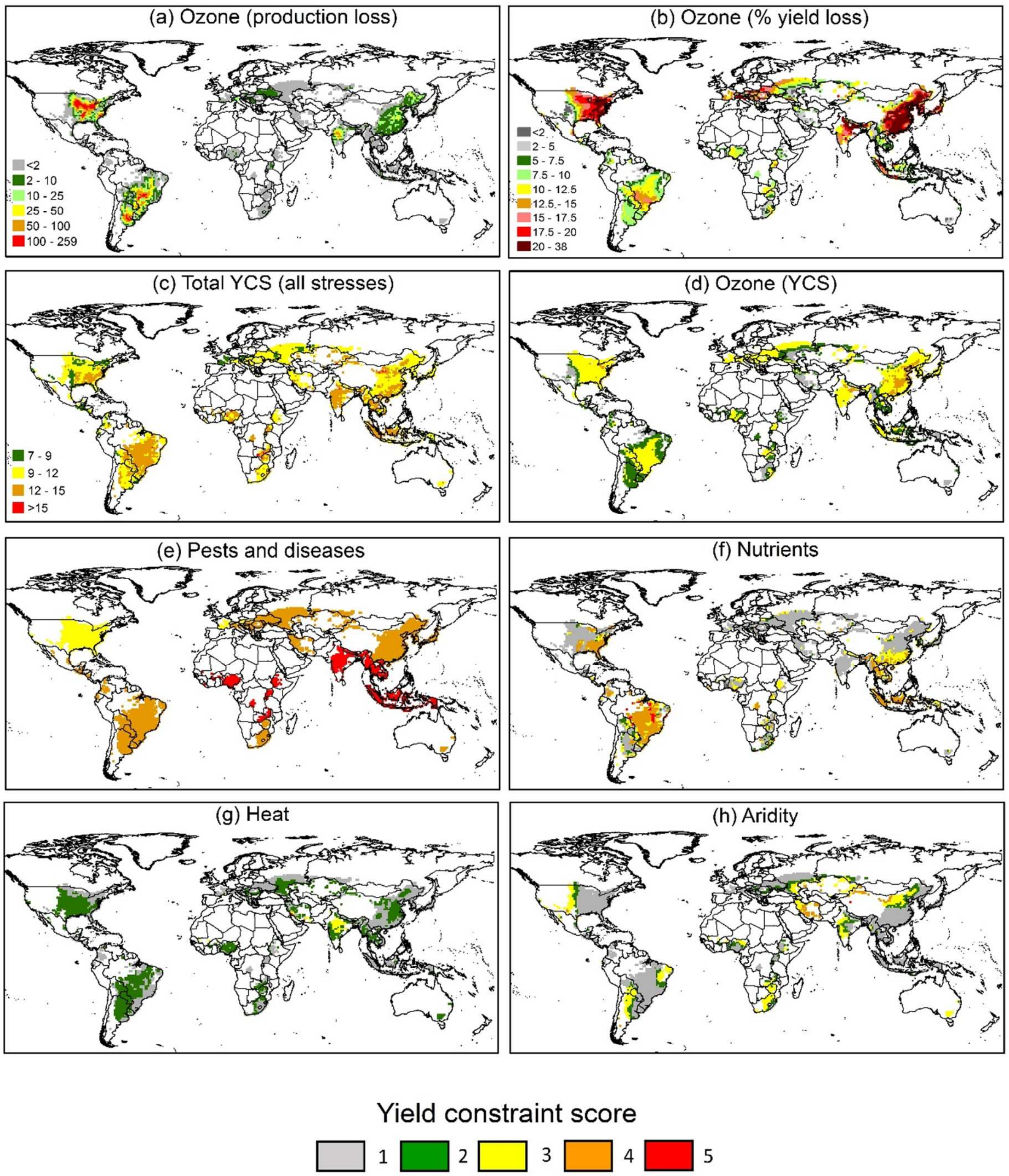


Fig. 2: The global effects of five biotic and abiotic stresses on soybean. All data are presented for the 1 x 1° grid squares where the mean production of soybean was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S4 provides all country and regional means.

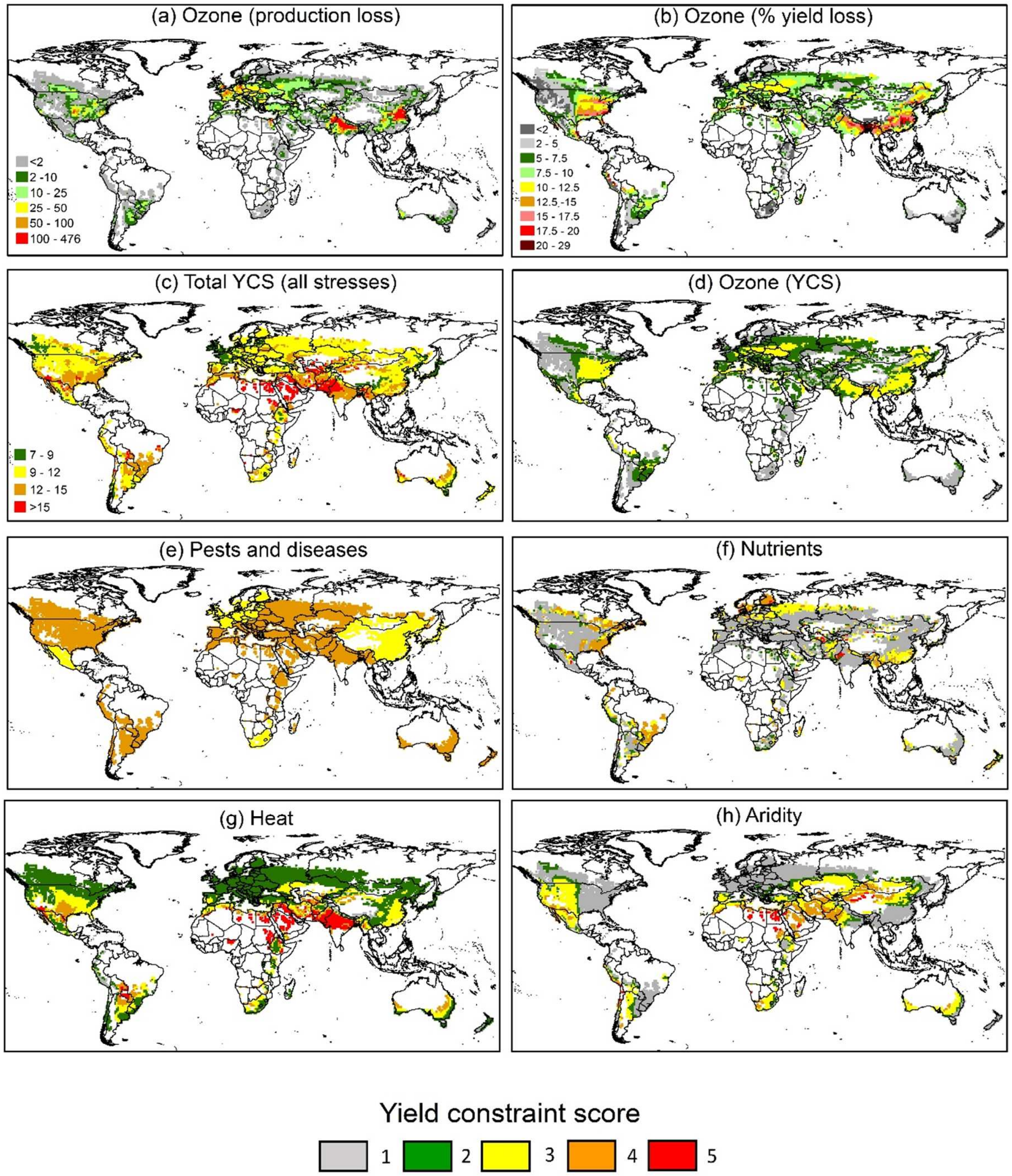


Fig. 3: The global effects of five biotic and abiotic stresses on wheat. All data are presented for the 1 x 1° grid squares where the mean production of wheat was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S5 provides all country and regional means.

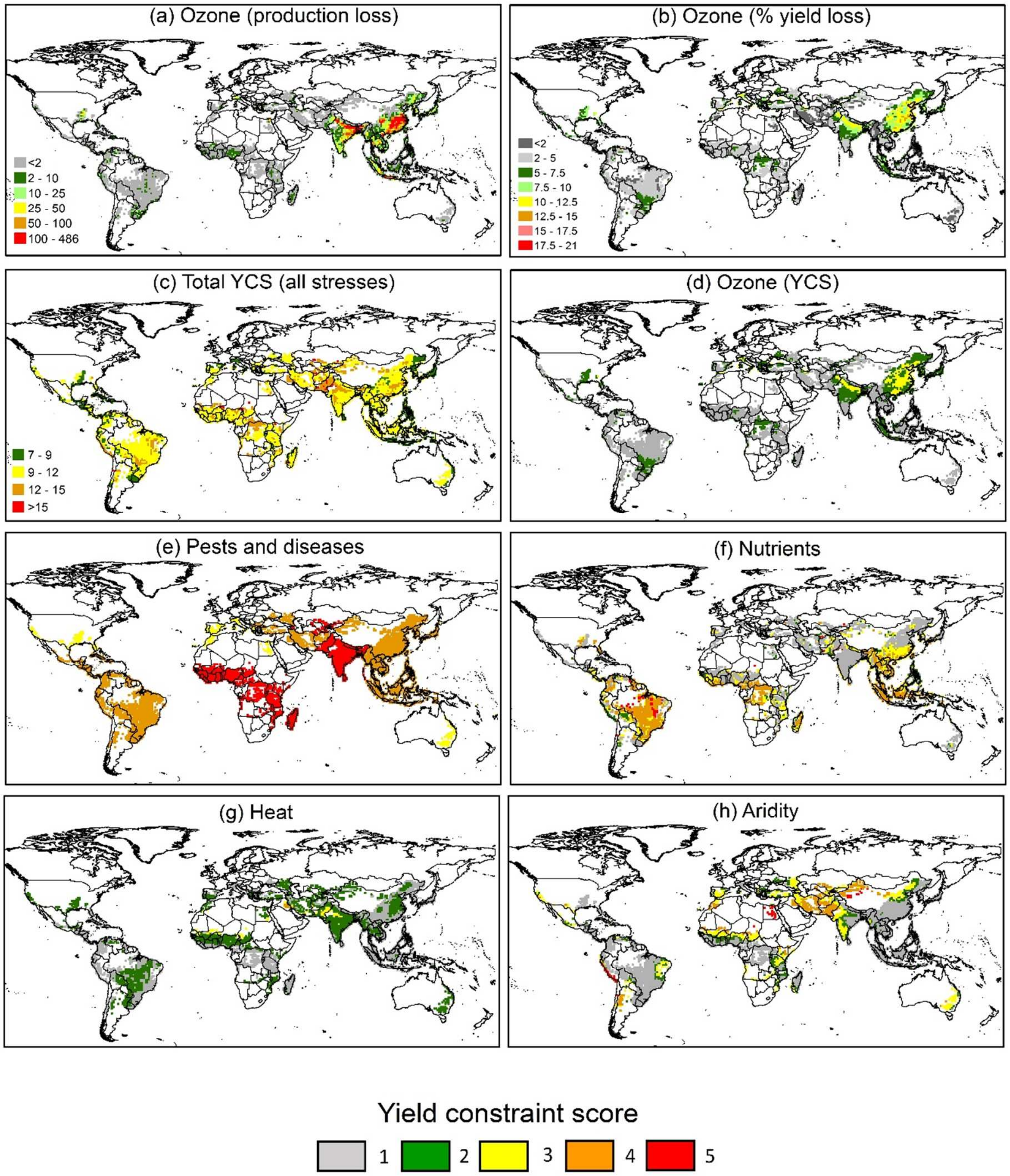


Fig. 4: The global effects of five biotic and abiotic stresses on rice. All data are presented for the 1 x 1° grid squares where the mean production of rice was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S6 provides all country and regional means.

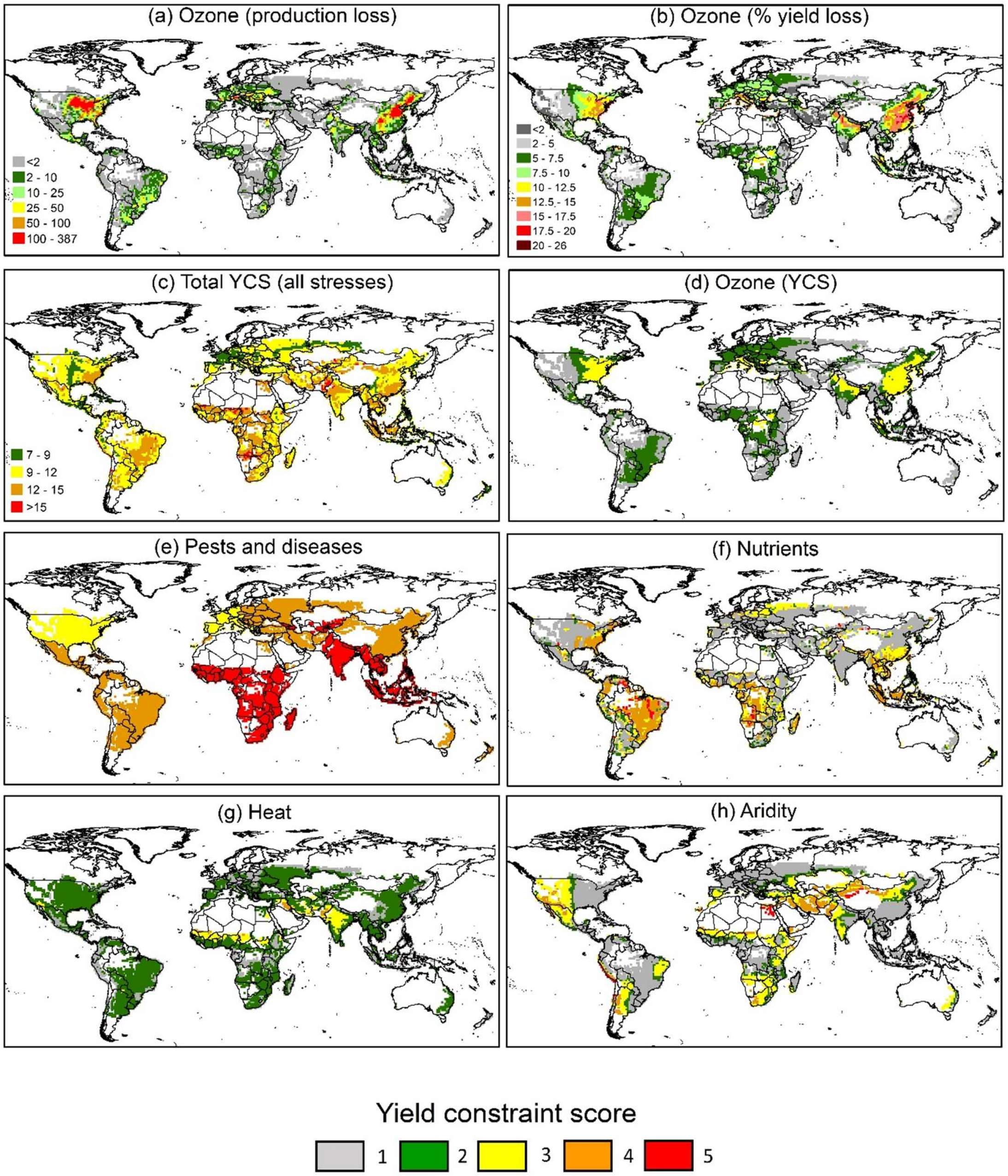


Fig. 5: The global effects of five biotic and abiotic stresses on maize. All data are presented for the 1 x 1° grid squares where the mean production of maize was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S7 provides all country and regional means.

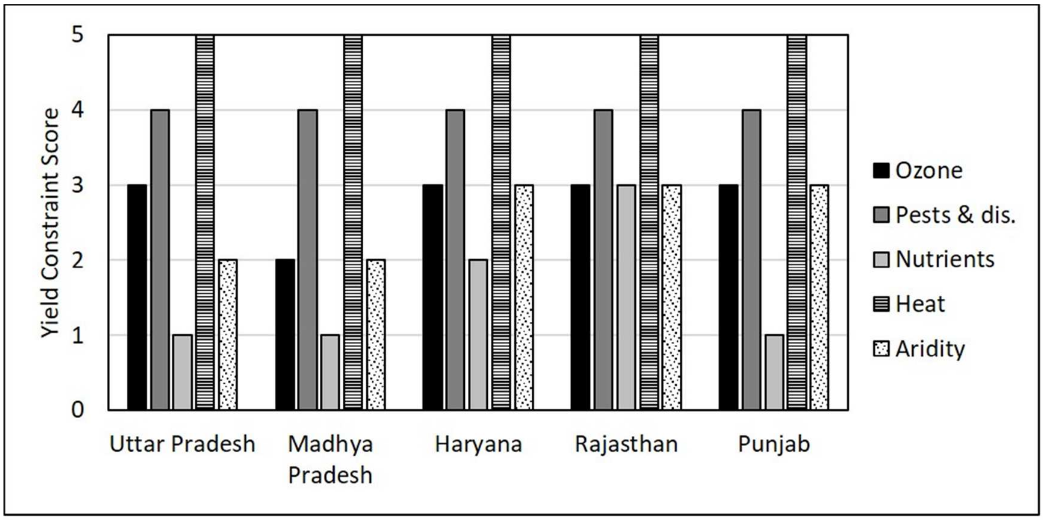
ozone were predicted for states in the far NE of India such as Assam and Manipur, total production losses there were predicted to be minimal as this is not an important wheat growing area. The area of highest ozone impacts on wheat production coincided with the area with the highest YCS for heat stress which covered most of the northern half of the country. Aridity and nutrient YCSs were highest to the W of this region, coinciding with percentage yield losses for ozone predicted to be in the range 5-15% (YCS of 2- 3). For the five highest wheat producing states, the mean YCS for heat stress was 5, with scores for aridity being 2 or 3, and nutrients being 1 – 3 (Fig. 6).

Rice growth is much more widely distributed in India than wheat growth, with the highest production being in the N, in part coinciding with wheat growing areas in states such as Uttar Pradesh, and also in E and S states such as West Bengal, West Odisha, Andhra Pradesh and Tamil Nadu (Fig. 6). Together these 5 states produce just below half of India's rice production. The highest percentage yield losses for ozone are predicted to be in the range 10 – 15% in the N of the country, including in Uttar Pradesh (mean ozone YCS of 3, Fig. 6 and S6). Lower effects were predicted for Odisha and West Bengal (mean YCS of 2) and the least ozone effects were predicted for rice producing areas in the southern states of Tamil Nadu and Andhra Pradesh (mean YCS of 1), where percentage losses were frequently less than 5%. Heat stress is less of a concern for rice, with a mean YCS of 2 predicted for each of the 5 most important rice producing states. Nutrient stress is predicted to only be important in the far NE states and in isolated grid squares in Rajasthan and along the W coast of India. The mean YCSs for nutrients and aridity for the five highest producing states are either 1 or 2 (Fig. 6).

3.2. Plant traits associated with tolerance of ozone and associated stresses in crops

The derivation of dose-response relationships for 52, 18 and 44 genotypes of soybean, wheat and rice respectively (Fig. 1) has shown that there is clearly scope for the breeding of ozone

(a)



(b)

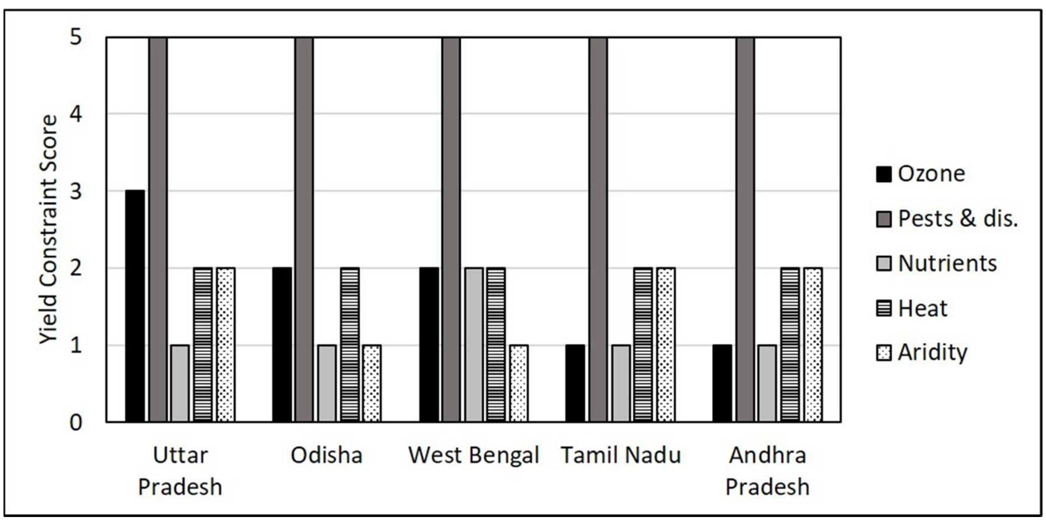
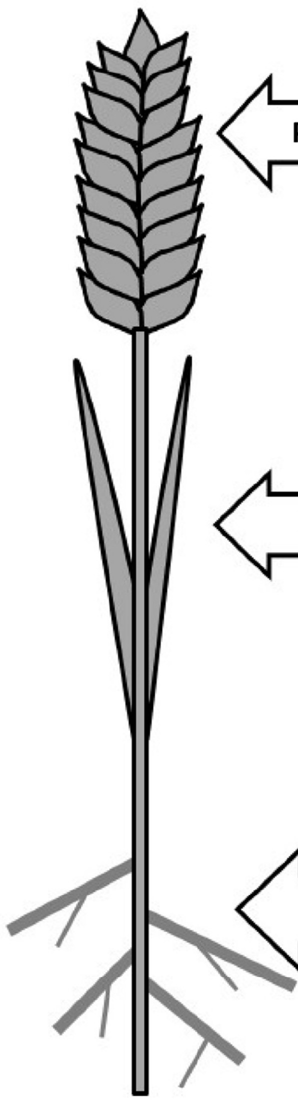


Fig. 6: Yield Constraint Score (YCS) for five constraints on the yield of (a) wheat and (b) rice in the five Indian states with the highest production per crop. The bars represent the mean YCS per 1 x 1°grid square per state on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1), rounded to the nearest integer. Note: The YCS for pests and disease is only available at the National Scale for India (score 4 for wheat and 5 for rice) and is presented here for information.

tolerant varieties, as many varieties had responses that are above the regression line. As part of this study, we identified a number of traits that could contribute to improved ozone tolerance and have summarized these in an ozone-tolerant crop ideotype, including potential trade-offs and synergies for effects of other stresses that can co-occur with ozone (Fig. 7).

Leaf traits for ozone tolerance fall into two categories, the first being processes that limit ozone entry. These include stomatal conductance, and the related trait of water use efficiency (WUE), that reduce ozone uptake while maintaining high rates of photosynthesis. These traits are associated with reduced leaf transpiration and whilst they would be beneficial for water conservation under drought conditions, they may reduce yield and could be potentially deleterious under heat stress by limiting evaporative cooling (Reynolds *et al*., 2007). Similarly, reduced water uptake associated with lower stomatal conductance has the potential to limit uptake of nutrients such as N from the soil (Zhou *et al*., 2016). While pathogens are known to have negative effects on leaf gas exchange (Debona *et al*., 2014), the impact of inherently lower stomatal conductance on disease establishment is less clear although it could be expected that ingress of leaf pathogens that access leaves through the stomatal pores would be reduced.

A second category of favourable leaf traits includes antioxidant metabolism and pathways involved in programmed cell death (PCD). Ozone is decomposed into reactive oxygen species (ROS) in the plant apoplast, which either cause direct oxidative damage, or induce signalling cascades similar to a pathogen response, ultimately leading to PCD (Kangasjärvi *et al*., 2005). Thus, balancing the interplay of redox homeostasis and PCD pathways is essential for the breeding of ozone tolerant crop plants. As a first line of defence against ozone stress, high levels of apoplastic antioxidants such as ascorbate may mitigate ROS formation, a concept that has been confirmed in crop plants such as wheat (Feng *et al*., 2010) and legumes (Yendrek *et al*., 2015). Breeding for high levels of antioxidants is also assumed to cause



Panicles

Target traits for ozone tolerance	Synergies / trade offs: Drought	Heat	Nutrients	Pests & dis.
Seed yield	0	0	0	0
Seed quality	+	+	+	+
Days to maturity	+	+	+	+

Leaves

Target traits for ozone tolerance	Synergies / trade offs: Drought	Heat	Nutrients	Pests & dis.
Water use efficieny	+	-	-	?
Low stomatal conductance	+	-	-	+?
High antioxidant capacity	+	+	+/-	+/-
Containing PCD	+/-	+/-	+/-	+/-

Roots

Target traits for ozone tolerance	Synergies / trade offs: Drought	Heat	Nutrients	Pests & dis.
Root allocation efficiency	+	+	+	0
Root dynamics	+	+	+	0

Fig. 7: An ideotype for an ozone-tolerant crop. '+' indicates where there would be a benefit for other stresses of improving tolerance to ozone for the trait, whilst '-' indicates a trade-off, and '0' is no effect.

synergies with other types of abiotic stress tolerance, including for drought and heat, both of which are associated with oxidative stress (Gill & Tuteja, 2010). In the case of some nutrient disorders and biotic stresses, functional redox balance rather than high antioxidant levels *per* *se* are considered as important (Munné-Bosch *et al*., 2013; Suzuki *et al*., 2012; Wu *et al*., 2017).

PCD is an important pathway of pathogen response in plant leaves (Huysmans *et al.,* 2017), which is controlled by the interplay of ROS, signalling cascades and plant hormones (Kangasjärvi *et al*., 2005). Breeding for ozone tolerance could thus keep plants from inducing PCD despite the presence of apoplastic ROS. This idea is supported by a study in rice, in which the disruption of the pathogen and ozone responsive apoplastic protein *OsORAP1*, which is involved in cell death, led to enhanced ozone tolerance (Ueda *et al*., 2015). The potential interference of this strategy with pathogen tolerance in crops is obvious, but it is currently unclear whether a synergistic or rather antagonistic relationship would occur with different classes of pathogens i.e. biotrophic versus necrotrophic ones (Huymans *et al.*, 2017). Implications of PCD in other stress types such as heat (Locato *et al*., 2008), drought (van Doorn 2011) and nutrient deficiency (Siyiannis *et al*., 2012) have also been reported but the implications for ozone tolerance breeding remain unclear.

Root traits that support ozone stress tolerance would include the capacity to efficiently acquire water and nutrient resources under stress environments (Resource Acquisition Efficiency in Fig. 7). Ozone is known to have a greater negative impact on roots than shoots, resulting in the decline in the root/shoot ratio commonly observed (Fiscus *et al.*, 2005). There is evidence that ozone may have an even greater impact on fine roots that acquire water and nutrients from the soil (Vollsnes *et al.*, 2010). Fine roots are the new frontier of future root research. The framework for describing fine root architecture is being refined (McCormack *et al.*, 2015; Zobel, 2016), and new techniques are now available to assess fine root dynamics (e.g. measurements of root diameter, Zobel *et al.*, 2007). The challenge ahead is to define and measure fine root traits that contribute to ozone tolerance, and then determine how these traits affect plant response to other stress factors. Presumably, traits that contribute to robust root systems will be of benefit across a range of abiotic stresses.

Traits associated with reproductive organs such as panicles or pods are of primary importance in breeding, although the effects of ozone on these organs may be rather secondary, i.e. caused by foliar responses that limit assimilate acquisition (described above) or effects on flowering and pollen viability (Black *et al.*, 2000). Yield losses due to ozone have been ascribed to various yield components in different crops, including reductions in individual seed weight, reduced spikelet number, enhanced spikelet fertility, and reduced panicle or pod number (Ainsworth, 2008; Feng *et al*., 2008; Morgan *et al*., 2003), with associated reductions in harvest index (e.g. for wheat, Pleijel *et al*., 2014). Maintaining high values in these harvest fractions despite ozone stress forms an important breeding target, but synergies or trade-offs with other types of stress would be complex and little information is available to date.

Maintaining high crop quality despite ozone stress represents another important breeding goal. Ozone can affect multiple quality traits in seed crops, including protein and starch concentration, as well as visual appearance (Broberg *et al*., 2015; Wang & Frei, 2011). In many cases, increases in seed protein concentration despite losses in protein yield are observed. This apparent beneficial effect is offset by the negative effects of ozone on seed weight (e.g. for wheat, Broberg *et al*., 2015). Another quality trait that has been affected in rice by ozone is grain chalkiness, i.e. the formation of milky patches on grains due to inhibited starch loading (Jing *et al*., 2016). Chalkiness was first described as a typical symptom of heat and drought stress (Wassmann et al., 2009), and lowering plant susceptibility to chalkiness *via* breeding may thus have potential co-benefits with regards to these stresses.

A further category of traits that could be targeted by breeders are phenological characteristics. Plants that have a shorter maturity period by entering earlier into reproductive phases might be more tolerant, as they would receive a lower cumulative ozone dose, and might avoid high ozone episodes occurring late in the cropping season. This principle was confirmed in a study by Ueda *et al*. (2015), in which more than 300 genotypes of rice were screened for ozone response, and yield losses were positively correlated with the number of days to maturity. In general, breeding fast-maturing crop varieties may produce substantial synergies, reducing the impacts of growing seasons characterized by high incidence of other stresses, such as drought, heat, nutrient, or biotic stresses.

4. Discussion

In bringing together these datasets and modelling methods to derive YCSs for five stresses and four key crops, we have conducted the first global assessment of the magnitude of ozone stress in relation to other stresses for four staple crops. We have also derived an ideotype for an ozone- and multi-stress tolerant crop. We provide an extended discussion here that first considers the results presented and then considers potential solutions for increasing crop tolerance of ozone, including crop management and breeding approaches.

4.1 The global scale of ozone impacts on crops relative to impacts of other stresses

An in depth evaluation of the spatial analysis conducted here is presented in the Supporting Information (T1) and summarised here. The benefits of impacts modelling based on the stomatal uptake of ozone rather than the concentration above the leaf, together with an evaluation of global modelling of POD_3IAM, are discussed by Mills *et al.* (2018a). In the absence of suitable stomatal uptake dose response relationships for soybean, rice and maize, the RS_w method was developed whereby the effects of ozone on these crops was determined from the POD_3IAM response of wheat. We had to assume that the differences in ozone concentration and sensitivity were a greater driver of response than differences in stomatal uptake and are unable to quantify the uncertainty introduced by this assumption. Whilst experimental data in the M7 response functions used in the RS_w method represented the major crop growing regions for soybean, wheat and rice, the function for maize was limited to relatively old data from NAM only. Thus, the data analysis presented here for maize is likely to be the most relevant for effects described in NAM, and is less certain when applied to other maize-growing regions of the world. For each crop, we assumed only one crop growth period per year. Thus, for those crops such as rice where two or three crop growth cycles may occur per year in major growing areas, assessments based on the main growth period will have an added level of uncertainty. The abiotic and biotic stresses included here were selected as examples for comparison with ozone effects, with heat stress being chosen as representative of effects of extreme climatic events associated with climate change. We acknowledge that other stresses such as flooding may also have catastrophic local effects on yield (e.g. in China, Tao *et al*., 2017), but have focussed on example stresses for which global data is readily available. Furthermore, global warming impacts on yield could be in a similar range to ozone (e.g. Challinor *et al*., 2009; Lobell & Asseng, 2017) but have not been considered here. Scores for YCS for pests and diseases may have overestimated current losses as advances in pesticide usage since the 2002-04 dataset was compiled may have reduced total impacts. As it was not possible to base all YCSs on percentage yield loss, uncertainty will have been introduced by comparing effects across stresses. We have acknowledged this uncertainty by using the YCSs to indicate the location of the largest effects rather than to quantify the extent of effects. Lastly, YCS_{all} simply summed all YCSs and provides an indication of where multiple stresses co-occur, without taking into account any interactions that may occur that might lessen or increase the combined effects on yield. Taking into account all of these caveats, this study is the first to present ozone impacts on the global scale together with impacts of other biotic and abiotic stresses, and show spatially where such stresses are likely to co-occur for four major staple crops.

At the national scale, the countries identified as having the largest potential effects of ozone (e.g. USA, India and China) match those with the highest monitored ozone concentrations (Mills *et al*., 2018b) as well as those predicted using concentration-based approaches to have the highest potential yield losses (Avnery *et al*., 2011a,b; Van Dingenen *et al*., 2009). At the sub-national scale, however, there were some differences in areas predicted to be at risk, where our stomatal uptake modelling method took into account the modifying effects of climate and soil moisture on ozone uptake rather than simply predicting the largest effects in the areas with the highest ozone concentrations. For example, in India, this study predicts the largest effects on wheat and rice in the northern areas, south of the Himalayas where ozone levels, climatic conditions and irrigation usage promote ozone uptake and subsequent effect. In contrast, an earlier concentration-based study provided little spatial differentiation in effects, predicting widespread and similar effects of ozone in the northern half of India for wheat and across most of India for rice (Van Dingenen *et al*., 2009). On a global scale, we predict that ozone (mean of 2010 - 2012) reduces soybean yield by 12.4%, wheat yield by 7.1%, rice yield by 4.4% and maize yield by 6.1%, adding up to a total of 227 Tg of lost yield. These mean percentage losses are different to those predicted by Avnery *et al*. (2011a) and Van Dingenen *et al*. (2009) using concentration based metrics. Their studies predicted higher losses for wheat (15.4% and 12.3%, respectively) and lower losses for soybean (8.5% and 5.4%, respectively) using AOT40 (Accumulated hourly mean ozone above 40 ppb during daylight hours) for the year 2000.

Multivariate analysis of trends in soybean and maize yields in the USA that included a concentration-based ozone metric indicated that the ozone effect is dependent upon temperature and water availability (McGrath *et al*., 2015). This fits with our earlier conclusion that stomatal uptake-based risk assessment provides a better indication of ozone effects on yield than concentration-based assessments (Mills *et al*., 2018a). The McGrath *et al.* (2015) study indicated a greater sensitivity of maize to ozone than soybean, with maize and soybean yield losses due to ozone over a 31-year period averaging 10% and 5%, respectively. It is possible that our analysis under-estimated the effects of ozone on maize as our analysis was based on experimental data from 1981, 1985, 1991 and 1992. Since newer varieties of wheat and soybean are more sensitive to ozone than older varieties (Biswas *et al*., 2009; Osborne *et al*., 2016), then newer maize varieties may also be more ozone sensitive leading to larger effects. Partial derivative-linear regression analysis of heat and concentration-based ozone stress impacts on yield data in the USA and Europe indicated similar areas at risk from ozone for maize and soybean to our study, but fewer areas at risk for wheat (Tai & Val Martin, 2017). The latter may reflect that their study omitted soil moisture as a confounding factor, which our earlier modelling study indicated is a particularly important factor in modifying ozone uptake (Mills *et al*., 2018a). These two statistical studies have confirmed that factors other than ozone concentration need to be taken into account in analysing ozone effects on yield and have drawn attention to potential co-occurrence of heat and ozone stress effects on crop yield.

It is clear from our analysis that yield effects due to ozone are within the range of concern for other biotic and abiotic stresses. For example, extreme heat was estimated to have reduced national cereal production by 9-10% (1964 - 2007), with later droughts reducing yields by more than earlier droughts (13.7% for 1985 – 2007 compared to 6.7% for 1964 – 1984, Lesk *et al*., 2016). If we applied the percentage yield loss ranges used for ozone in YCS (Table 1) to these drought-induced yield losses, the YCS would be 2 or 3 depending on the time period used in the analysis. The YCSs for ozone are mainly in the same range as those predicted for wheat, maize and rice for global impacts of heat stress and aridity (scores 2-3, with rice-heat having a YCS of 1). Across all four crops, the areas predicted to be at the greatest risk of ozone effects on yield are predicted to be: SE NAM, S EUR, N SAS and E EAS, with parts of SAM also predicted to be at risk of yield loss for soybean. In some of these areas, ozone effects are predicted in areas also at risk from heat stress and to a lesser extent aridity, whilst co-occurrence with nutrient stress depended on the crop and tended to be most common in parts of EAS and SAM. Potential impacts on yield due to pests and diseases were predicted to be relatively high in many areas of the world, particularly in those at risk from ozone impacts in SAS and EAS.

Ozone impacts are predicted in areas where the largest gaps occur between actual and estimated potential yield, such as parts of SAM, SSA, EAS, SEA and SAS (Neumann *et al*., 2010). Here, yield gaps are already known to be widened by limitations in nutrient and/or irrigation availability (Mueller *et al*., 2012) and may be further widened by negative effects of ozone pollution. Indeed, in the same regions there has been a plateauing or decrease in the rate of yield increase in recent decades (Grassini *et al*., 2013; Ray *et al*., 2012). We suggest that ozone pollution could be contributing to this stagnation, and suggest below how crop tolerance of the pollutant could be improved by breeding or management (Section 4.2.3).

India was selected as a case study, as our analysis indicated that ozone pollution may be a particular problem in this country, adding to the existing multi-stress constraints on crop yield (Jaswal, 2014). National mean yield losses due to ozone were predicted to be 15.8% (soybean), 12.6% (wheat), 6.2% (rice) and 7.5% (maize) amounting to 12.6 Tg of lost yield. For wheat, our predicted mean yield loss in Uttar Pradesh of 16% was comparable to a mean 17% yield benefit from reducing the ambient ozone from 46 to 5 ppb (M7) by air filtration in field studies conducted from 2004 to 2008 at Varanasi in Uttar Pradesh (Rai *et al.*, 2007, Sarkar *et al.*, 2010). Similarly, at a field site in Haryana, reduction in the M7 by filtration from 37 ppb to 6 ppb, resulted in a 16% yield benefit for wheat (Bhatia *et al*., 2011), which was similar to our state mean of a 15% yield reduction due to ozone.

Wheat yield losses were predicted to be highest in this study in the same regions of India as those predicted by Tang *et al*. (2013) in the first stomatal uptake-based risk assessment for the country. Our analysis, taking into account the added effects of soil moisture and irrigation usage, extended the region of highest ozone effects across the Indo-Gangetic Plain and including Uttar Pradesh and Bihar. Together with Haryana and Punjab, these states are considered to have the highest reductions in yield due to the combined effects of climate change and air pollution, with reductions as high as 50% being predicted in one concentration-based study (Burney & Ramanathan, 2013). Our multi-stress analysis confirmed that heat stress is particularly important in this region (Lobell *et al*., 2012). Site-specific analysis of the effects of future increases in temperature in 2030-2040, indicated that heat stress is likely to continue to reduce yields in the Indo-Gangetic Plain, especially under climate change (Asseng *et al*., 2017). Given that from a food security perspective, it is crucial to reduce yield gaps in India, reducing ozone pollution and/or its effects could potentially provide beneficial additional yield in future climates.

In considering these comparisons, we are aware that in reality ozone will interact with the other stresses considered and integrated responses in growth and yield will occur. These interactions are generally thought to be determined by factors that might affect gas exchange or metabolic responses to stress. For example, limited water stress may reduce ozone uptake but as water stress becomes more severe, any protection afforded by reduced ozone uptake may be outweighed by drought-induced yield reductions. Additionally, these stresses are thought to impart similar defence mechanisms (Huymans *et al.*, 2017; Kangasjärvi *et al*., 2005; Locato *et al*., 2008). Whether multiple stresses induce additive or synergistic metabolic responses is open to question and we do not yet have the understanding or tools to be able to quantify these interactions. Nevertheless, through providing a first global assessment of where these stresses co-occur we have identified which stresses are most important across different global regions. This will help other researchers to identify threats and target future research needs to improve our understanding of responses to multiple stress conditions.

4.2 Options for reducing ozone impacts on crops

The analysis presented here has clearly shown that ozone impacts on yield are occurring in many areas of the world for four staple crops, and that in some regions the YCSs for ozone are as high or higher than for other biotic and abiotic stresses. Whilst these results highlight the ozone problem, we offer here some possible options for reducing ozone effects on crops that might help in closing the ozone yield gap.

<u>4.2.1 Global effort to reduce ozone precursor emissions</u>

The most obvious way of closing the ozone yield gap for crops is to substantially lower the anthropogenic emissions that lead to ozone pollution. Since ozone is a transboundary air pollutant – impacts of emissions in one country can impact on crops grown in countries many 100s and even 1000s of km away – efforts to reduce ozone need to be taken at both local and global scales. One study, using ozone concentration-based metrics, indicated that 100% reductions in anthropogenic precursor emissions from NAM would reduce global yield losses due to ozone for the four crops in our study by between ca. 5% (rice) to ca. 80% (soybean), whilst a complete cut in precursor emissions from SEA would reduce global yield losses by between ca. 20% (soybean) and ca. 95% (rice) (Holloway *et al.*, 2012). Whilst such dramatic cuts in ozone precursor emissions are highly unlikely for the foreseeable future, progress has been made in EUR and NAM, with emission cuts of ca. 40% for major ozone precursors such as NOx, VOC and CO being made between 1990 and 2013 (Maas & Grennfelt, 2016). These cuts have been associated with significant decreasing trends in the concentration-based metric

AOT40 at 26% and 11% of monitoring sites in wheat growing areas of NAM and EUR, respectively over the period 1995-2014 (Mills *et al*., 2018b), although the dominant trend for EUR remains "no change". Over the same time period, increases in precursor emissions of 20-30% in other areas of the world, including by 50% in India and China have led to increases in ozone concentration in these regions (Maas & Grennfelt, 2016). For example, there has been a significant increase in ozone concentration at nearly 50% of wheat growing monitoring sites in EAS, with average annual increases in AOT40 at these sites being in the range 300–700 ppb h y^{-1} over the period 1995-2014 (Mills *et al*., 2018b).

Our modelling results suggest that even with declining emissions in NAM, current yield losses due to ozone are in the range 5.3% (rice) to 15.5% (soybean), whilst for EAS with rising emissions, current yield losses are in the range 7.9% (rice) to 19.1% (soybean). With ozone concentrations predicted to continue to rise in EAS and SEA for at least the next 2-3 decades even with the most optimistic scenarios (Wild et al., 2012), and as these two regions are predicted to produce 80% of all global ozone precursor emissions by 2050 (Maas & Grennfelt, 2016), there would be considerable benefit for crop yield in the implementation of a concerted effort to reduce precursor emissions in these rapidly developing regions. Actions to reduce ozone are already being considered in some countries. For example, in China, three approaches are being introduced to reduce ozone concentrations: enforcing the European standard V for diesel vehicle emissions; encouraging widespread use of electric vehicles; and discouraging private car use by improving public transport (Feng *et al*., 2015). Continued effort to reduce ozone is also needed in developed regions such as NAM and EUR as models predict that whilst efforts to reduce peak concentrations have been partially successful in reducing ozone concentrations in recent decades, a stabilisation in ozone concentrations in the next decade or two is likely to be followed by further rises in global background ozone concentration by 2050, primarily driven by increasing CH_4 emissions (Maas & Grennfelt, 2016).

Whilst reducing global ambient ozone concentrations remains a crucial long-term goal for reducing the ozone yield gap, approaches described below based on crop management and breeding are more likely to provide shorter-term solutions, with some having potential for implementation in the near future.

4.2.2 Exploiting existing varietal differences in ozone sensitivity

Whilst the analysis presented here has confirmed that intraspecific variation in ozone sensitivity is clearly present for wheat, rice and soybean in experiments conducted over the last 30 - 40 years (Fig. 1, Table S3), of larger importance in the context of closing the ozone yield gap is the potential for selecting ozone tolerance amongst currently grown varieties. To assess this, ideally, varieties should be exposed to ozone under the same environmental conditions, allowing for realistic comparisons of effects on yield and assessments of variety by ozone interactions. Unfortunately, relatively few such experiments have been conducted with two or more varieties in the last decade. Those recent studies showing significant variety by ozone interactions, indicating scope for selecting the more ozone tolerant variety, include examples from SAS and EAS for rice (Akthar *et al*., 2010; Shi *et al*., 2009) and wheat (Feng *et al*., 2010, 2016; Singh *et al*., 2017; Zhu *et al*., 2011;), NAM for soybean (Betzelberger *et al*., 2010; Jiang *et al*., 2018) and maize (Yendrek *et al*., 2017); and EUR for wheat (Harmens *et al*., 2018). Further support for the potential benefits of selecting tolerant varieties is also provided by comparisons of yield in filtered air versus non-filtered air (Osborne *et al*., 2016; Pleijel *et al*., 2018). For example, in recent studies, reductions of ambient ozone concentration by filtration significantly increased the yield of ozone-sensitive soybean cultivars (PUSA 9712, PUSA 9814) by over 40% (Singh & Agrawal, 2011), rice cultivar Kirara 397 by over 20% (Frei *et al*., 2012) and wheat cv PBW 343 by 18-20% (Tomer *et al*., 2015).

A modelling study has been conducted to highlight the potential for avoiding production loss in global wheat, maize and soybean by selecting crop varieties with lower than average sensitivity to ozone (Avnery *et al*., 2013). The variation in sensitivity among varieties was based on the experimental evidence from the large-scale US National Crop Loss Assessment Network (NCLAN) field studies conducted mainly during the 1980s (Heagle, 1989; Heck *et al*., 2013; Heck, 1989). Using a concentration-based method, the study showed that choosing crop varieties with ozone tolerance could improve global crop production by over 140 Tg in 2030, equivalent to a 12% increase. Although the older North American varieties may not represent current global variation, and some of the 1980s varieties are no longer used, the approach of Avnery *et al*. (2013) could be extended by conducting new screening experiments with a regional focus to inform farmer choice, modelling and breeding programmes and by using a stomatal uptake based modelling approach.

4.2.3 Breeding new varieties with multiple stress tolerance, including ozone

The heterogeneity in variety response to ozone for soybean, wheat and rice (Fig. 1) has clearly shown the scope for breeding ozone-tolerant varieties, and an ideotype for an ozone tolerant crop has been defined here (Fig.7). Ideally, the improved ozone response must not compromise the yield potential or other required agronomic characteristics (e.g. resistance to diseases, shattering, and lodging). Since this study has also shown that ozone stress can typically co-occur with stress caused by heat, pests and diseases, and to a lesser extent aridity and nutrients, the breeding for ozone tolerance traits may cause potential synergies or trade-offs that also need to be considered (Fig. 7). Candidate traits for ozone tolerance were described in Section 3.2.

Traditional breeding approaches such as pedigree selection require extensive screening of a large number of plants in multiple locations over extended periods of time (Frei, 2015). Whilst feasible, experimentally maintaining designated ozone concentrations on a sufficiently large scale required for breeding (e.g. in large scale FACE (free air concentration exposure) experiments) seems economically unviable. Therefore, molecular breeding approaches such as marker assisted selection (MAS) appear to be more promising. Phenotypic variation in traits associated with ozone tolerance can be evaluated in smaller-scale controlled ozone fumigation experiments and linked to genetic markers using mapping approaches, including bi-parental quantitative trait locus (QTL) mapping (Frei *at al*., 2008) and Genome-Wide Association Study (GWAS, Ueda *et al*., 2015). Theoretically, chromosomal fragments associated with ozone tolerance traits can then be introgressed into recipient varieties using marker assisted backcrossing without the need for large-scale fumigation experiments.

Although no large-scale marker-assisted breeding programs for ozone tolerance in crops have been conducted to date, proof of concept has been shown for ozone tolerant rice breeding lines carrying QTL for ozone tolerance (Chen *et al*., 2011; Frei *et al.*, 2008, 2010) that have a superior performance to the recipient varieties in terms of yield components (Wang *et al*., 2014) and grain quality (Jing *et al*., 2016). This example should encourage further breeding efforts in rice and other crop species, specifically targeting widely grown mega-varieties of crops grown in ozone-affected parts in the world. As an alternative strategy, traits contributing to ozone tolerance could be incorporated into existing crop varieties through genetic engineering. For example, rice and barley varieties engineered to contain enhanced levels of ascorbate have been engineered and showed enhanced tolerance to a variety of environmental stresses (Ali *et al*., 2018).

Physiological trait modelling could also be used to understand how different traits intended to confer tolerance for ozone, might influence crop physiology, growth and yield response under a range of environmental conditions and stresses.

4.2.4 Reducing ozone uptake by strategic limitation of irrigation application

Ozone impacts on crops could be reduced by partial stomatal closure induced by reduced irrigation, which could also save water use for irrigated crop production. In the rice growing countries, in response to the increasing water demands by other sectors than agriculture, alternate wetting and drying irrigation (AWD) has become popular in an attempt to reduce water usage and methane emissions (Bouman *et al*., 2007; Carrijo *et al*., 2017). This approach could also potentially be exploited to reduce ozone impacts on rice or other crops. A comparison of two studies conducted about 30 km apart in the same city of China suggests such a possibility. In Zhang *et al*. (2009), AWD with moderate water stress increased the growth and yield of rice while reducing stomatal conductance compared to continuously flooded crops, mostly resulting from a greater number of rice grains per panicle under AWD. Interestingly, at a nearby site, elevated ozone reduced rice yield arising from a decrease in the number of grains per panicle in two of the four varieties tested (Shi *et al*., 2009). This suggests that reduced ozone uptake could be an additional and unintended benefit of AWD for farmers. The potential benefits of the AWD approach require further study.

4.2.5 Fertilizer application to compensate for crop yield losses

Crop loss from ozone exposure could potentially be counteracted by increasing the fertilizer application rate (Cardoso-Vilhena & Barnes, 2001; Chen *et al*., 2011). However, in addition to the cost of fertilizer, recent analysis has indicated that this mitigation approach may be associated with an aggravation of other environmental problems. It has been shown that the nitrogen/protein yield of wheat is reduced by ozone at a certain level of nitrogen application and this applies also to other nutrients like phosphorus and potassium (Broberg *et al*., 2015; 2017). This means that the fraction of nitrogen applied which does not end up in the grain could enhance other environmental problems (Di & Cameron 2002; Mosier *et al.* 1998) such as nitrate leaching, conversion of fertilizer to N_2, emissions of N_2O and even NO, which promotes further ozone formation, as shown for pasture (Sánchez-Martín *et al*., 2017). Adding nitrogen fertiliser to compensate for reductions in yield may also inadvertently increase the stomatal conductance of leaves of crop plants thereby increasing ozone uptake and subsequent damage (Mills *et al*., 2016).

4.2.6 Chemical protection against ozone damage

There is scope for investigating the benefits of chemical protection against ozone damage. The most successful antiozonant applied so far has been ethylenediurea (N-[2-(2-oxo-1-imidazolidinyl)ethyl]-N'- phenylurea), abbreviated to EDU, first described by Carnahan *et al.* (1978). This chemical is usually applied as a foliar spray or soil drench, and has been used extensively in experiments and biomonitoring programmes to reduce the effects of ozone pollution, including preventing visible ozone injury on the leaves and growth and yield reductions (Agathokleous *et al*., 2016; Feng *et al*., 2010; Jiang *et al*., 2018; Manning *et al*., 2011; Pandey *et al*., 2015; Rai *et al*., 2015). A meta-analysis suggested that the antiozonant activity of EDU is biochemical rather than biophysical (Feng *et al*., 2010). Recent results showed that EDU has no negative effects on plants at low O_3 concentration, but increases the crop yield at high O_3 concentration (Ashrafuzzaman *et al*., 2017). Whilst EDU has not yet been evaluated for application at field scale, concerns have been raised about potential toxicity to aquatic plants (Agathokleous *et al.,* 2016) and more research is needed to determine if this chemical could be extensively used.

Other chemical protectants against ozone could be developed from a knowledge of plant hormonal control of stomatal functioning and stress perception (Wilkinson *et al*., 2011), and could potentially provide multi-stress tolerance such as combined tolerance of ozone, heat and drought stress. All three of these stresses induce synthesis of the crop stress hormone, ethylene, and chemicals that inhibit ethylene perception such as 1-MCP (1-methylcyclopropene) have the potential to reduce their effects (Wilkinson & Davies, 2010; Wagg, 2012). Anti-transpirants that reduce stomatal aperture could also reduce ozone effects by reducing ozone uptake in some species. However, there is a growing body of knowledge that chronic exposure to ozone reduces the ability of stomata to respond to abscisic acid under drought conditions, potentially leading to more rather than less ozone uptake (Mills *et al*., 2016; Wilkinson and Davies, 2009, 2010). An alternative chemical protection approach has also been explored experimentally. Di-1-*p*-methene, a natural terpenic polymer derived from the resin of pine trees that mimics isoprene emissions from plants, has been shown to reduce visible injury in Pinto beans after exposure to 150 ppb of ozone for 4h (Francini *et al*., 2010).

So far, chemical protection has only been explored at the experimental scale. Given the growing evidence presented here and elsewhere of the negative effects of the pollutant at the global scale, there is considerable scope for developing a chemical protectant against ozone damage, especially if it provides cross-tolerance against other co-occurring stresses.

5. Conclusions

This global-scale study shows that ozone is a very important stress, limiting yields of key crops and comparing in importance with other key stresses. For example in India, where food security concerns are particularly pressing, the mean YCS for effects of ozone on wheat of 3 falls in between those for nutrients and aridity (score 2) and for pests and diseases and heat stress (score 4). Globally, we show that the largest effects of ozone are often in areas already challenged by other stresses such as pests and diseases and heat, particularly in EAS, SAS and SEA. The global mean ozone yield gaps of 4.4 – 12.4 % identified here add up to 227 Tg of lost yield for soybean, wheat, rice and maize. We speculate that, ozone could at least partially, account for the unexplained yield gaps and stagnation in yield improvement seen in many areas of the world in recent years. Thus, international effort to reduce ozone pollution on a global scale would bring clear benefits for agriculture as well as for other types of vegetation, health, materials and climate change (Simpson *et al.*, 2014). However, it is likely to take many decades to achieve the required emission reductions, which is the only long-term solution for reducing the problems caused by tropospheric ozone. Meanwhile, the global population is expected to grow significantly, which together with increasing real income levels, will see increasing demands placed on food production (Tilman *et al.*, 2011).

Several interim solutions for closing the ozone yield gap have been outlined in this paper. These include: testing of current varieties for ozone sensitivity and selection of the most tolerant; crop breeding for multiple stress tolerance, including ozone; implementation of protective watering regimes such as AWD; and the development of chemical protection against ozone damage. Given the severity of ozone effects on staple food crops in areas of the world that are also challenged by other stresses, we recommend increased attention to the benefits that could gained from taking mitigating action to reduce the ozone yield gap.

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Data Accessibility Statement:

National-scale data are provided in the Supporting Information. Grid square values for ozone metrics for the LRTAP region can be downloaded from http://www.emep.int/mscw/mscw_data.html.

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Figure legends

Fig. 1. Response functions for (a) soybean, (b) wheat, (c) rice and (d) maize derived from published data using the growing season ozone (7h mean, M7 in ppb) in the experiments. Data points are presented per cultivar/variety, with sources of data provided in the Supporting Information (Table S3). The response functions are: Soybean, RY = -0.0050x + 1.001 (r^2 (adj) = 0.625, p<0.001); Wheat, RY = -0.0048x +0.96 (r^2 (adj) = 0.547, p<0.001); Rice, RY= -0.0021x +0.987 (r^2 (adj) =0.347, p <0.001); and Maize, RY = -0.0031x + 1.03 (r^2 (adj) = 0.617, p <0.001).

Fig. 2: The global effects of five biotic and abiotic stresses on soybean. All data are presented for the 1 x 1° grid squares where the mean production of soybean was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S4 provides all country and regional means.

Fig. 3: The global effects of five biotic and abiotic stresses on wheat. All data are presented for the 1 x 1° grid squares where the mean production of wheat was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S5 provides all country and regional means.

Fig. 4: The global effects of five biotic and abiotic stresses on rice. All data are presented for the 1 x 1° grid squares where the mean production of rice was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S6 provides all country and regional means.

Fig. 5: The global effects of five biotic and abiotic stresses on maize. All data are presented for the 1 x 1° grid squares where the mean production of maize was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S7 provides all country and regional means.

Fig. 6: Yield Constraint Score (YCS) for five constraints on the yield of (a) wheat and (b) rice in the five Indian states with the highest production per crop. The bars represent the mean YCS per 1 x 1°grid square per state on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1), rounded to the nearest integer. Note: The YCS for pests and disease is only available at the National Scale for India (score 4 for wheat and 5 for rice) and is presented here for information.

Fig. 7: An ideotype for an ozone-tolerant crop. '+' indicates where there would be a benefit for other stresses of improving tolerance to ozone for the trait, whilst '-' indicates a trade-off, and '0' is no effect.

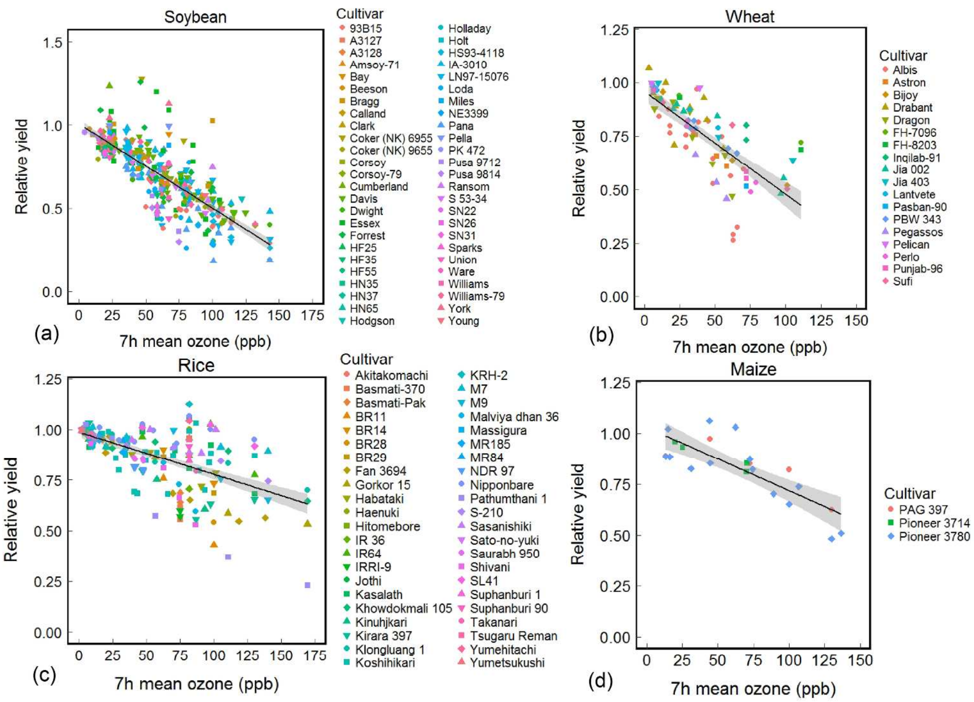

Fig. 1. Response functions for (a) soybean, (b) wheat, (c) rice and (d) maize derived from published data using the growing season ozone (7h mean, M7 in ppb) in the experiments. Data points are presented per cultivar/variety, with sources of data provided in the Supporting Information (Table S3). The response functions are: Soybean, RY = -0.0050x + 1.001 (r2 (adj) = 0.625, p<0.001); Wheat, RY = -0.0048x +0.96 (r2 (adj) = 0.547, p<0.001); Rice, RY= -0.0021x +0.987 (r2 (adj) =0.347, p <0.001); and Maize, RY = -0.0031x + 1.03 (r2 (adj) = 0.617, p <0.001).

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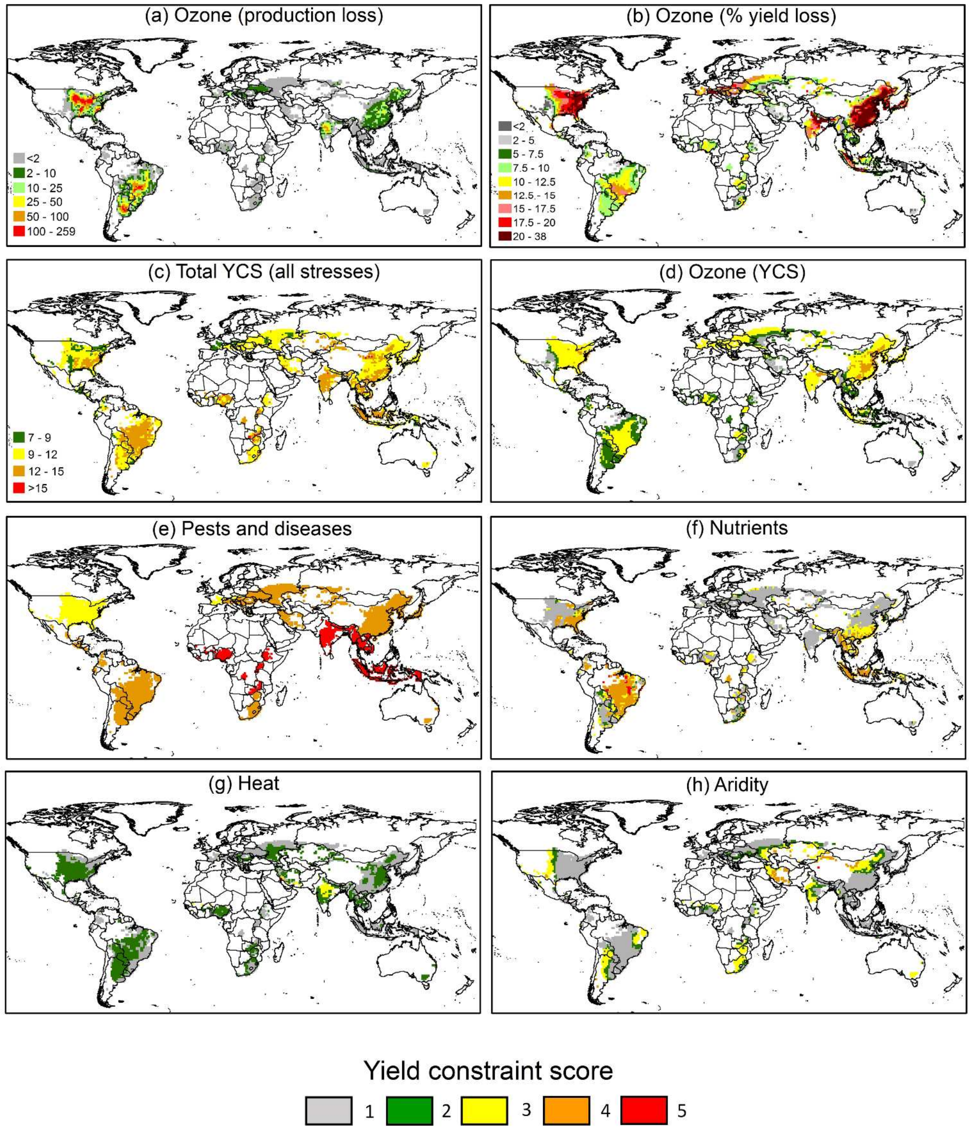

Fig. 2: The global effects of five biotic and abiotic stresses on soybean. All data are presented for the 1 x 1° grid squares where the mean production of soybean was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCSall) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S4 provides all country and regional means.

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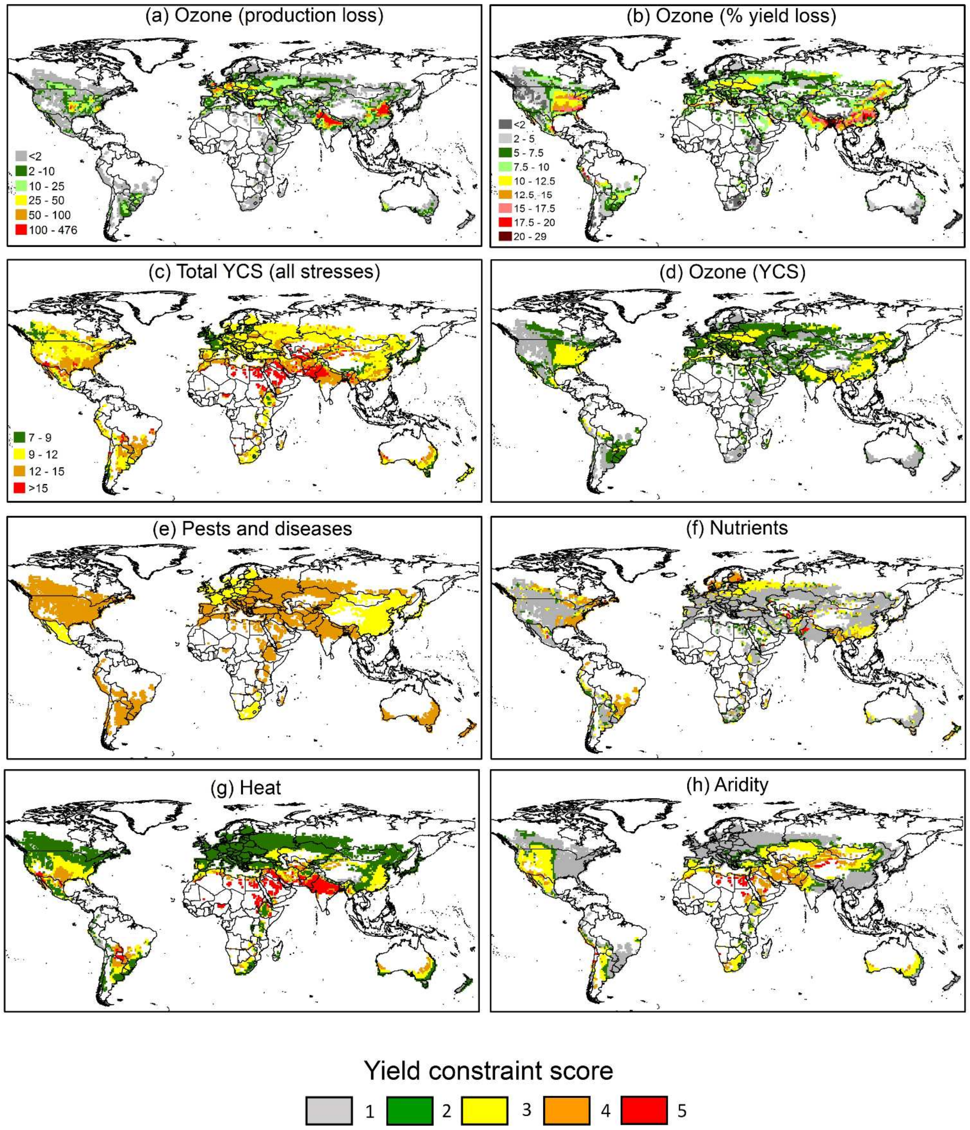

Fig. 3: The global effects of five biotic and abiotic stresses on wheat. All data are presented for the 1 x 1° grid squares where the mean production of wheat was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCSall) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S5 provides all country and regional means.

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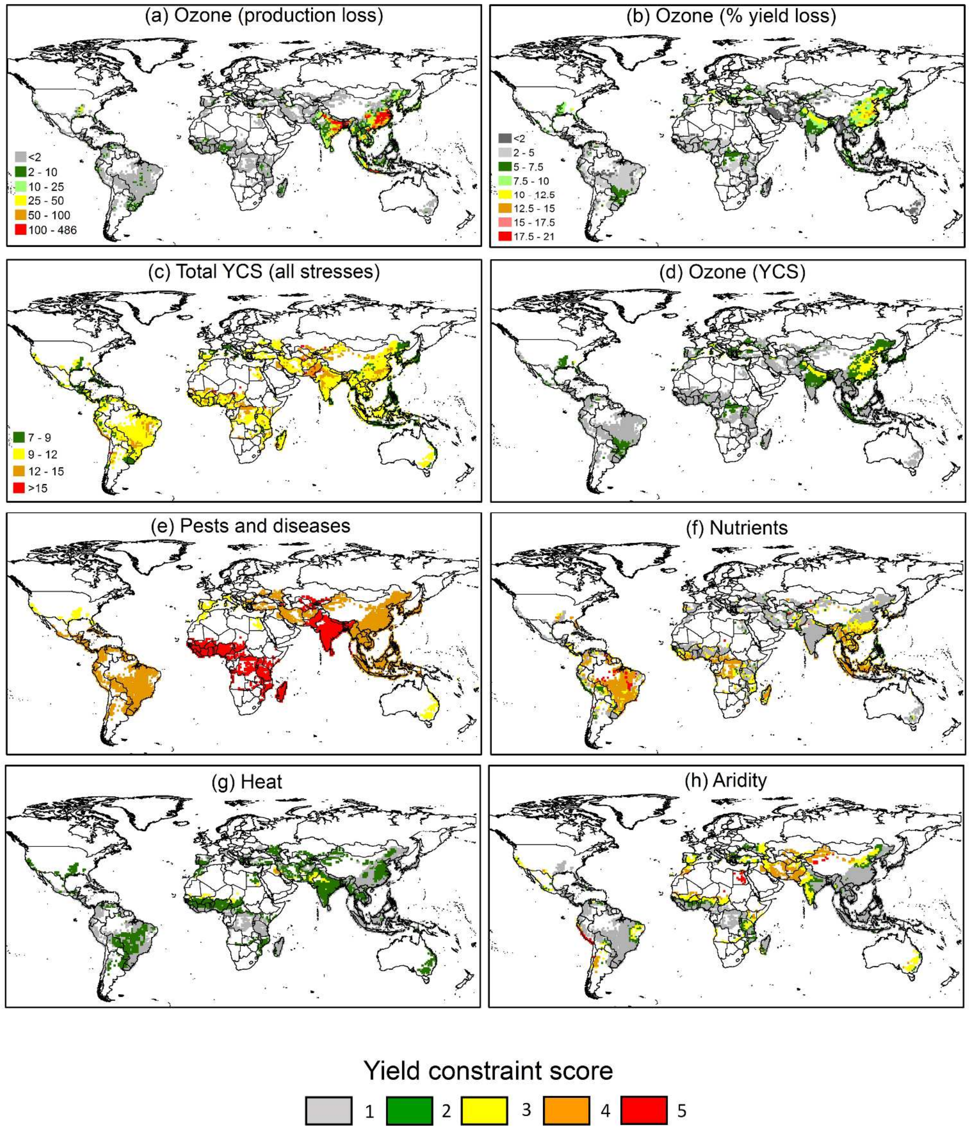

Fig. 4: The global effects of five biotic and abiotic stresses on rice. All data are presented for the 1 x 1° grid squares where the mean production of rice was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCSall) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S6 provides all country and regional means.

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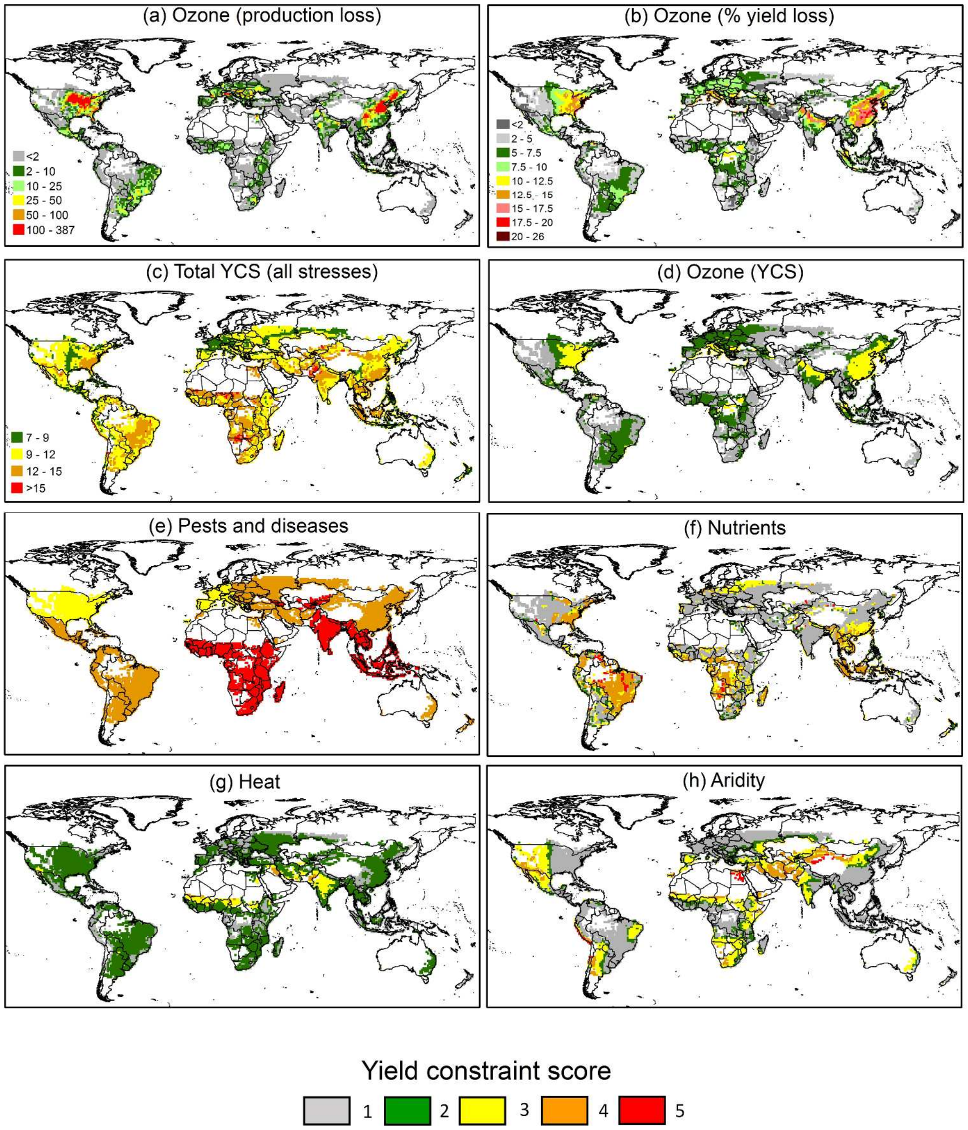

Fig. 5: The global effects of five biotic and abiotic stresses on maize. All data are presented for the 1 x 1° grid squares where the mean production of maize was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCSall) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S7 provides all country and regional means.

249x290mm (600 x 600 DPI)

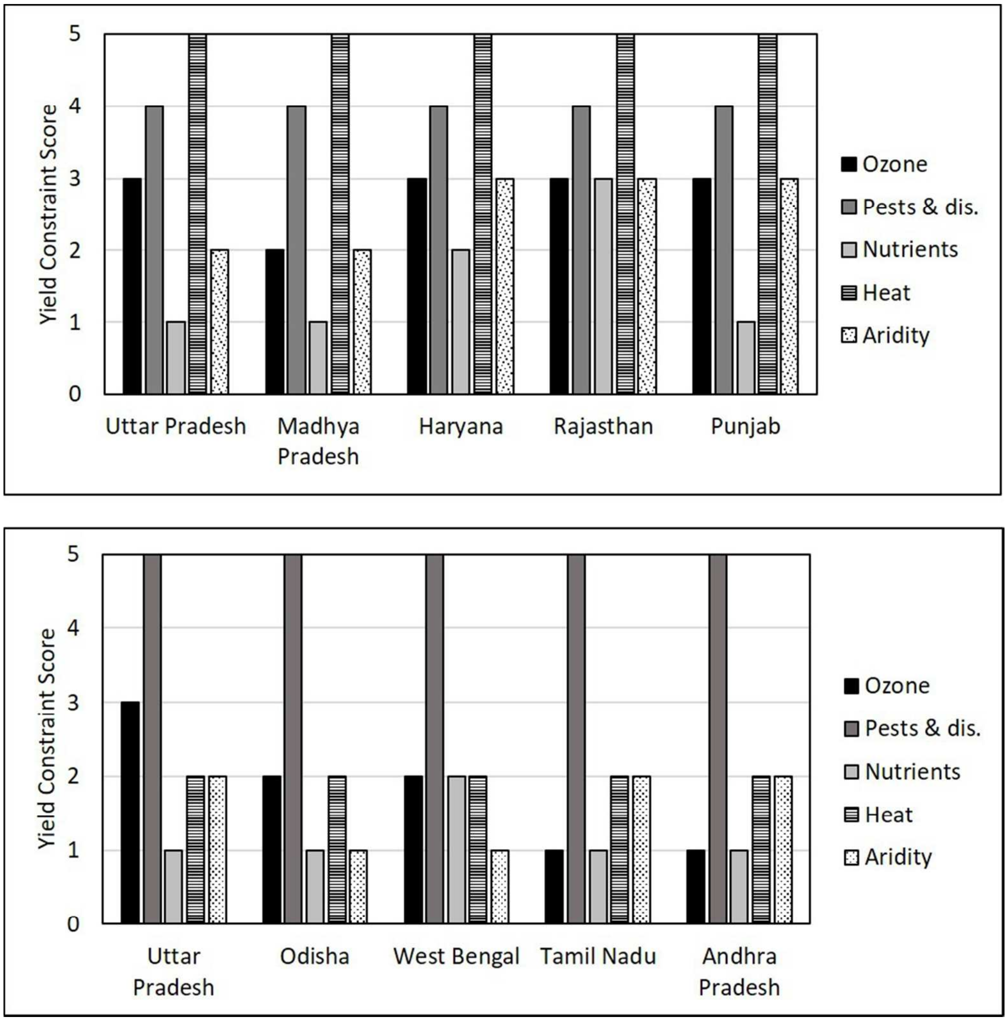

Fig. 6: Yield Constraint Score (YCS) for five constraints on the yield of (a) wheat and (b) rice in the five Indian states with the highest production per crop. The bars represent the mean YCS per 1 x 1°grid square per state on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1), rounded to the nearest integer. Note: The YCS for pests and disease is only available at the National Scale for India (score 4 for wheat and 5 for rice) and is presented here for information.

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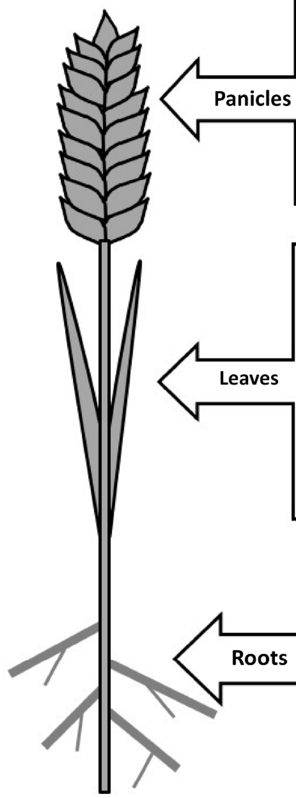

Target traits for ozone tolerance	Synergies / trade offs: Drought	Heat	Nutrients	Pests & dis.
Seed yield	0	0	0	0
Seed quality	+	+	+	+
Days to maturity	+	+	+	+

Target traits for ozone tolerance	Synergies / trade offs: Drought	Heat	Nutrients	Pests & dis.
Water use efficieny	+	-	-	?
Low stomatal conductance	+	-	-	+?
High antioxidant capacity	+	+	+/-	+/-
Containing PCD	+/-	+/-	+/-	+/-

Target traits for ozone tolerance	Synergies / trade offs: Drought	Heat	Nutrients	Pests & dis.
Root allocation efficiency	+	+	+	0
Root dynamics	+	+	+	0

Fig. 7: An ideotype for an ozone-tolerant crop. '+' indicates where there would be a benefit for other stresses of improving tolerance to ozone for the trait, whilst '-' indicates a trade-off, and '0' is no effect.

249x367mm (600 x 600 DPI)

GCB-18-0501: Response to Reviewers

Closing the global ozone yield gap: Quantification and co-benefits for multi-stress tolerance

Gina Mills, Katrina Sharps, David Simpson, Håkan Pleijel, Michael Frei, Kent Burkey, Lisa Emberson, Johan Uddling, Malin Broberg, Zhaozhong Feng, Kazuhiko Kobayashi and Madhoolika Agrawal

Note: All page and line numbers refer to the manuscript with changes accepted. An additional version of the manuscript showing all changes made is also provided, for information.

Changes made to improve the integration of the difference components of the paper

Having read the editor's and reviewer's comments, we have made some small changes to the text and structure to improve the integration of the different components of the paper for the reader. Excluding the introduction, for each section, we have introduced a higher-level sub-division that distinguishes between text related to quantifying the problem and text related to solutions for the problem. The structural changes made are:

<u>Introduction</u>

To ensure that the flow of the paper is clearer for the reader, we have modified:

(i) The last two sentences of the first paragraph to refer back to the title of the paper and introduce the idea that the paper both defines the problem and suggests solutions (P4, L 2- 5).

(ii) The last paragraph to reflect the (revised) structure of the paper, including specifically mentioning that the paper describes results from two analytical approaches (quantitative spatial analysis and a qualitative analysis leading to an ozone tolerant crop ideotype), and also includes an extended discussion considering options for reducing the negative effects of ozone on yield (P6, L16 to P7, L18).

<u>Methods</u>

The different sub-sections have been re-organised as follows:

2.1 Global spatial analysis of crop yield constraints caused by ozone

2.1.1 Crop production
2.1.2 Intra- and inter-specific sensitivity of crops to ozone
2.1.3 Yield constraints caused by ozone

2.2 Global spatial analysis of yield constraints caused by other stresses

2.2.1Yield constraints caused by pests and diseases
2.2.2 Yield constraints caused by soil nutrients
2.2.3 Yield constraints caused by heat stress
2.2.4 Yield constraints caused by aridity

2.3 Comparative analysis of effects of five stresses using a Yield Constraint Score (YCS)

2.4. Qualitative analysis of plant traits associated with multiple stress tolerance

Results

The different sub-sections have been re-organised as follows:

3.1 Quantification of the global impacts of ozone and other stresses on crop yield

3.1.1 Intra-specific sensitivity to ozone
3.1.2 Spatial analysis of the global impacts of multiple stresses on crop yield
3.1.3 Case study – India

3.2. Plant traits associated with tolerance of ozone and associated stresses in crops

Discussion

The different sub-sections have been re-organised as follows:

4.1 The global scale of ozone impacts on crops relative to impacts of other stresses

4.2 Options for reducing ozone impacts on crops

4.2.1 Global effort to reduce ozone precursor emissions [*Note: new section added, see response to Reviewer 1*]
4.2.2 Exploiting existing varietal differences in ozone sensitivity
4.2.3 Breeding new varieties with multiple stress tolerance, including of ozone
4.2.4 Reducing ozone uptake by strategic limitation of irrigation application
4.2.5 Fertilizer application to compensate for crop yield losses
4.2.6 Chemical protection against ozone damage

Conclusions
Structure unchanged, 2 paragraphs

Reviewer 1 (Reviewer's comments in black font, our response in blue font)

This is very important contribution to the Scientific Community in particular on the relationship between ozone pollution and its impacts on crop production. I think the review paper has summarized almost all recently progress in the area.

We wish to thank this reviewer for their very supportive comments about the importance of our manuscript for the scientific community and for making suggestions for further improvements.

I have only two minor suggestions: (1) Discussion sector: The options (as a seperate paragraph) for reducing surface ozone concentration or pollution should be considered (which can be regarded as the most important option) to reduce ozone impacts crops;

Thank you for this suggestion. We have added a new section, 4.2.1 on "Global effort to reduce ozone precursor emissions" encompassing this suggestion (P40, L11 to P42, L6).

(2) One additional reference which has also addressed this issue could be added in the revision: Feng Z, Liu X, Zhang F. (2015) Air pollution effects on food security in China: Taking ozone as an example. Front. Agr. Sci. Eng. 2(2): 152-158.

We have included reference to this paper in the new section 4.2.1 (P41, L17 - 20).

Reviewer 2 (Reviewer's comments in black font, our response in blue font)

Comments to the Author
Based on published data in some literatures, the authors made a comprehensive prediction of ozone impacts on major grain crop yield in the world by using models. They also compared the impacts of ozone with those of other stresses (pest, heat, etc.). They found the larges loss in yield was soybean with significant spatial differences. To my knowledge, this is the first comprehensive report about ozone impacts on the four major grain crops globally. They authors also provided some adaptation strategies in crop improvement and agronomic innovations for ozone impact mitigation for food security.

We wish to thank reviewer 2 for also recognising the importance of our study including its novelty, and for making suggestions for further improvements.

However, there are still some limitations:
(1) The available published data is limited, especially about ozone impacts on maize, the average response functions (Fig. 1) might not represent the specific cropping regions or countries. Moreover, the experimental results from a specific location were not only due to ozone elevation, the results might also due to the interaction between ozone and the local temperature and other environmental factors. Thus the experimental locations in the data base should be reprehensive of the major cropping regions of each grain crop (Soybean, wheat, maize, and rice), especially for the prediction of total production loss.

In producing the response functions, we sought to include all available experimental data that met the inclusion criteria listed in Section 2.1.2. For soybean, wheat and rice, the experimental data provided good representation of the major growing areas for these crops. A new sentence has been added to Section 3.1.1 that lists the HTAP regions represented in the response functions for these three crops (P17, L20 to P18, L1) and reference is now made to this point in the discussion (P35, L2 – 7). Unfortunately, for maize we were limited to data for three cultivars from experiments conducted in the USA during the late 1980s/early 1990s. Thus, the analysis conducted for maize has the highest uncertainty. However, as the highest maize production losses were predicted for the USA where the experiments had been conducted, we considered it relevant to include the analysis for maize within the paper to ensure that we covered all of the main grain crops. We have added new text on the uncertainty associated with the maize analysis in the discussion (Section 4.1, P35, L5 - 8), whereas previously this uncertainty was only mentioned in T1 of the Supporting Information.

(2) In this manuscript, the authors only compared ozone with pest and disease, soil nutrient, heat and aridity. In fact, there are other important stresses, such as global warming, extreme climate events, flooded, etc. These stresses might have more serious impacts on crop production.

We can fully understand the reviewer's point, but unfortunately it was not feasible to consider every stress that could have a potentially large effect on yield in the paper. The stresses chosen were for illustration purposes – to place ozone effects within the context of effects of example other common stresses that also reduce yield. A detailed justification for the stresses chosen, including reference to other studies using these datasets or approaches is provided in the Supporting Information, Text 1. We thought that inclusion of this text within the discussion of the main paper would have made the discussion too long both for the reader and the journal. When choosing stresses, we were largely restricted by availability of datasets and methods suitable for quantifying impacts of current levels of stress. Heat stress was included as an example of an effect of extreme climatic events, with the heat stress index being determined for crop-specific thermal sensitive time-periods over the period 1990-2014.

To address the reviewer's concerns, we have added in the word "example" where we first mention comparison with other abiotic and biotic stress in the Introduction (P7, L5) and discussion (P35, L10 - 12) to show that we are not covering all such stresses. We have also added new text to Section 4.1 of the discussion pointing out that other detrimental stresses such as flooding and global warming have been excluded from this study but their effects can be significant (P35, L12 - 15).

(3) For each prediction, the authors need to give the scenario time and the baseline time. For example, the readers cannot get the information about the prediction years for each Fig. of the manuscript.

Thank you for pointing this out. Whilst this information is included in the methods section, we agree that it was not easy to find quickly in the original manuscript. To avoid overly-long repetitive legends for each figure, we have added a new column to Table 1 'Year(s) of data' (see P16) to facilitate comparisons across stresses and also to make this information easy to find.

(4) About the section of crop ideotype, it would be better more it to the section of discussion.

Whilst writing the paper and after receiving this comment, we discussed at length the best location for this part of the paper. The ideotype is a result in that it is based on a synthesis of information from the scientific literature. Conversely, the associated text explaining the ideotype reads more like a discussion. We are concerned that placing both in the discussion might make the discussion overly long. After further consideration, we think it is better to retain this figure and text in the results (Section 3.2), with a description of how the ideotype could be included in crop breeding located in the discussion (Section 4.2.3). As described above, we have added higher level headings throughout the methods, results and discussion sections to make it clearer when we are quantifying the ozone problem in the context of other stresses, and when we are reporting on options for reducing ozone impacts. We believe that this insertion of extra sub-headings will make the paper more streamlined for the reader and easier to follow.

(5) About the differences in ozone response between crop cultivars, can the authors get the corresponding yield of each cultivar? So the authors can compare the differences the ozone response and the crop yield. Did the higher sensitive cultivars have lower yield under an ambient ozone level? If it is true, this really indicates that there is a potential to enhance crop tolerance to ozone with high yield through cultivar improvement.

This is an important point that we have now added to the discussion (P42, L20 to P43, L2). Because of the different growing and environmental conditions in each experiment it is not possible to compare the actual yield directly across cultivars unless the cultivars have been exposed to ozone at the same location and in the same year. For this reason, the Fuhrer method was developed for international risk assessment. This approach allows cultivars to be compared in response functions based on their relative yield, and has been used in this study to prepare Figure 1. In revising the manuscript, we have inserted new text to provide examples from experiments conducted in the last decade where ozone sensitive varieties of rice, wheat and soybean have significantly higher yield in filtered air with reduced ozone compared to ambient air treatments (P42, L22 to P43, L2).

Closing the global ozone yield gap: Quantification and co-benefits for multi-stress tolerance

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Running head: Global benefits of closing the O_3 yield gap

Key words: Ozone, wheat, soybean, maize, rice, pests and diseases, aridity, nutrient stress, heat stress, stress tolerant ideotype.

Paper type: Primary research

Abstract

Increasing both crop productivity and the tolerance of crops to abiotic and biotic stresses are major challenges for global food security in our rapidly changing climate. For the first time, we show how the spatial variation and severity of tropospheric ozone effects on yield compare with effects of other stresses on a global scale, and discuss mitigating actions against the negative effects of ozone. We show that the sensitivity to ozone declines in the order soybean > wheat > maize > rice, with genotypic variation in response being most pronounced for soybean and rice. Based on stomatal uptake, we estimate that ozone (mean of 2010 - 2012) reduces global yield annually by 12.4%, 7.1%, 4.4% and 6.1% for soybean, wheat, rice and maize, respectively (the 'ozone yield gaps'), adding up to 227 Tg of lost yield. Our modelling shows that the highest ozone-induced production losses for soybean are in North and South America whilst for wheat they are in India and China, for rice in parts of India, Bangladesh, China and Indonesia, and for maize in China and the USA. Crucially, we also show that the same areas are often also at risk of high losses from pests and diseases, heat stress and to a lesser extent aridity and nutrient stress. In a solution-focussed analysis of these results, we provide a crop ideotype with tolerance of multiple stresses (including ozone) and describe how ozone effects could be included in crop breeding programmes. We also

discuss altered crop management approaches that could be applied to reduce ozone impacts in the shorter-term. Given the severity of ozone effects on staple food crops in areas of the world that are also challenged by other stresses, we recommend increased attention to the benefits that could be gained from addressing the ozone yield gap.

1. Introduction

To feed the rapidly growing global population, we need to develop a new generation of crop cultivars or varieties that will have both high productivity in future climates and high tolerance of the biotic and abiotic stresses that are likely to become more prevalent in the future (Gilliham *et al*., 2016). Candidate characteristics or traits are currently being tested in ideotype modelling (Semenov and Stratonovitch, 2013) and include improved light conversion efficiency, a longer duration of green leaf area for grain fill, a higher harvest index and optimal phenology. For example, varieties that use less water per unit of carbon fixed will have higher yield under drought conditions (Rebetzke *et al*., 2002) as will those with 'stay-green' characteristics during water stress (Jordan *et al*., 2012). Whilst it is widely recognised that rapid breeding programmes will have a vital role to play in adaptations of crops to climate change (Atlin *et al*., 2017), selection of traits for tolerance of one abiotic stress, tropospheric (ground level) ozone pollution, is currently omitted from such breeding programmes (Ainsworth, 2016; Frei *et al*., 2015). This is happening even though field experiments from nine countries representing three continents have shown that reducing ozone concentrations back to pre-industrial levels would give an average wheat yield benefit of 8.4% globally (Pleijel *et al*., 2018), a figure that is matched by modelling based on the stomatal uptake of the pollutant (Mills *et al.,* 2018a).- Furthermore, an earlier meta-analysis of crop responses to ozone suggested that current ozone levels in the range 31 - 50 ppb (nmol

mol^{-1}, v/v) are reducing the yield of major food crops by 5.3 to 19% (Feng & Kobayashi, 2009). We undertook this new study to build a case for improving crop yields in our changing climate by closing the ozone-induced yield gap via the inclusion of ozone tolerance in crop breeding programmes, altered crop management and more stringent ozone precursor emission controls. ~~We undertook this new study to build a case for including ozone tolerance in crop breeding programmes. We took a multi-faceted approach that includes a spatial analysis of the impacts of ozone on yield relative to impacts of other abiotic and biotic stresses, defining an ideotype for an ozone tolerant crop and considering options for reducing the negative effects of the pollutant by altered crop management.~~

Tropospheric ozone pollution is formed from photochemical reactions involving anthropogenic and biogenic emissions and is involved in a complex web of interactions with ecosystems (Simpson et al., 2014). Whilst concentrations have been beginning to decrease in eastern USA and parts of Europe (2000 – 2014) due to precursor emission controls, they have been increasing rapidly in south (S) and east (E) Asia (Chang *et al*., 2017). Ozone is a powerful oxidant that is absorbed into leaves via open stomatal pores. Once inside the leaves, ozone reacts with biomolecules to form reactive oxygen species, triggering defence mechanisms that if overwhelmed lead to programmed cell death and a reduced extent and duration of functional green leaf area producing less photosynthate for seed fill (e.g. Ainsworth, 2016). Since pests and diseases (e.g. Oerke, 2006; Huysmanss *et al*., 2017), heat stress (e.g. Driedonks, *et al*., 2016), drought (e.g.Farooq *et al*., 2017) or reduced nutrient availability (e.g. Gastal & Lemaire, 2002) usually also reduce the extent and duration of the functional green leaf area, then in simple terms, each of these biotic and abiotic stresses result in the same endpoints – reduced yield quantity that is often associated with reduced quality.

So far, most crop breeding programmes have been targeted at increasing or maintaining the yield rather than increasing stability of yield under stress (Gilliham *et al.*, 2017). Because ozone concentrations tend to be very ~~heterogenous~~heterogeneous across natural and agricultural regions (Klingberg *et al*., 2012) as well as over seasons and years, it is not likely that traditional selection would unintentionally favour ozone tolerant crop genotypes. The reverse seems to be the case. For example, an analysis of ozone-exposure yield data for 49 soybean varieties from 28 field exposure studies showed that ozone sensitivity has increased by an average of 33% between 1960 and 2000 (Osborne *et al*., 2016). Similarly, modern wheat varieties are more sensitive than older varieties (Biswas *et al.*, 2008; Pleijel *et al*., 2006). Potentially, this increased sensitivity to ozone over recent decades is related to selective breeding for higher stomatal conductance (Roche, 2015) that inadvertently has increased the ingress of ozone into crops (Biswas *et al*., 2008; Osborne *et al*., 2016); further study is required to fully understand the mechanistic basis of this increasing sensitivity with time.

As with many abiotic and biotic stresses, genetic variation in plant response to ozone has been found for every species that has been tested. For the major grain crops, genetic variation in ozone response has been reported for wheat (Zhu *et al.*, 2011), rice (Frei *et al*., 2008; Shi *et al.*, 2009), soybean (Mulchi *et al*., 1988; Burkey & Carter, 2009; Jiang *et al.*, 2018), and maize (Yendrek *et al.*, 2017). Variation has also been reported for other crops including snap bean (Burkey *et al*., 2005; Yuan *et al.,* 2015) and tobacco (Heggestad, 1991). These assessments are based on different criteria including foliar injury and impacts on growth and yield parameters. Taken together, the evidence suggests that sufficient natural genetic variation exists to support improvement in crop stress tolerance either as sources of ozone tolerance genes or providing contrasting genotypes for mechanism studies to identify targets for molecular manipulation. Potential targets for breeding of ozone tolerance that have the greatest likelihood of success include reducing the stomatal uptake of ozone into the leaf and increasing its detoxification once inside the leaf (Feng *et al*., 2016; Frei *et al*., 2015).

To target the regions of the world where ozone tolerant crop varieties are most required, we need to understand which crops are most at risk and where they are growing in relation to current high-risk areas for ozone. We know from a recent analysis of ozone concentrations at over 3000 rural sites that the highest ozone values are in many of the world's important crop growing regions, including parts of the USA, Europe, India and China (Mills *et al*., 2018b~~submitted~~). Overall, the latter study showed that the global mean cumulative ozone exposure is double the critical level set by the United Nations as a target for ozone pollution control, above which direct adverse effects on sensitive vegetation may occur according to present knowledge (CLRTAP, 2017). Several studies have modelled ozone concentrations and predicted yield effects using concentration-based yield response functions applied at a range of scales from local (e.g. for India, Lal *et al*., 2017) to global (e.g. Avnery *et al*., 2011a,b; Van Dingenen *et al*., 2009). Whilst these studies indicate effects in the highest ozone areas, they do not take into account the constantly varying effects of soil moisture, air temperature, light and humidity on the uptake of the pollutant via the stomata. In Europe, field evidence for effects of ozone on crops and other types of vegetation shows that risk assessments based on modelled stomatal uptake or flux (Emberson *et al*., 2000, Simpson *et al*., 2007) provide a stronger indication of ozone effects than those based on concentration (Mills *et al*., 2011). Furthermore, dose-response functions for crops that are based on stomatal uptake are better correlated with yield effects than those based on concentration (Pleijel *et al*., 2000, 2007), providing additional support for their use.

With ozone concentrations increasing in rapidly developing regions and predicted to continue to increase in coming decades (Wild *et al*., 2012), it is timely to consider the options for increasing the tolerance of crops to this abiotic stress. In this study, our analysis included a two-step approach to addressing the ozone problem in crops: (i) a quantitative spatial analysis of the impacts of ozone on crop yield relative to impacts of other abiotic and biotic stresses and (ii) a qualitative analysis of crop traits, including defining an ideotype with multiple stress tolerance. As an initial step, we compiled dose-response data from experiments conducted around the world to determine the scope for breeding ozone tolerant varieties by showing the genotypic range in sensitivity for four staple crops: soybean, wheat, rice and maize. We then used the response functions to modell~~ed~~ the current impacts of ozone on each crop, showing the regions where the greatest production losses are likely to be occurring. Whilst we wait for ozone effects to be included in predictive crop yield modelling (as suggested by, for example, Challinor *et al.*, 2009; Emberson *et al.*, 2018; Lobell & Asseng, 2017), we sought to compare on a global scale the impacts of ozone on yield with the influence of other example biotic and abiotic stresses. Those selected were: pests and diseases (Oerke *et al.*, 2006); aridity (Trabucco & Zomer, 2009); heat stress (developed from Deryng *et al.* (2014) and Teixeira *et al.* (2013)); and soil nutrient ~~status~~ stress (GAEZ). The effects of all five stresses were considered ~~this~~ in more detail for India where there are major challenges for crop production and food security (Jaswal, 2014) and where global assessments consistently predict high risk from elevated ozone (e.g. Avnery *et al.*, 2011a,b; Van Dingenen *et al.*, 2009). In the second part of the study, we conducted an

~~We also~~ analysis of the plant traits associated with multiple stress tolerance, and considered the trade-offs and benefits of introducing ozone tolerance in crops for cross-tolerance of other biotic and abiotic stresses. This part of the study culminated in the design of ~~ed~~ an ideotype for an ozone tolerant crop that would also provide tolerance of ~~against the~~ co-occurring stresses. In an extended discussion, we assess the results from the two parts of the study and consider ~~address~~ viable ~~approaches to~~ options for reducing ~~reduce~~ the negative effects of ozone on yield, including ~~options for~~ crop management, breeding and global efforts to reduce~~ing~~ ozone pollution.

2. Materials and Methods

2.1 Global spatial analysis of crop yield constraints caused by ozone

2.1.1 Crop production

Global modelled crop production data (year 2000, 0.0833° (5 arc minute) resolution) was downloaded from the GAEZ (Global Agro-Ecological Zones, v. 3) data portal (http://www.fao.org/nr/gaez/en/) for soybean, wheat, rice and maize. Irrigated and rain-fed production data was collected for each crop. Using ArcMap v. 10.3 (Environmental Systems Research Institute, Redlands, CA, USA), a 1° by 1 ° global grid was created. For each crop, production was summed per grid cell. Each cell was classed as irrigated or non-irrigated based on the percentage of irrigated crop production per cell. To define a threshold for irrigated versus non-irrigated, we first produced frequency distributions of the percentage of irrigated production for each crop (Supporting Information, Fig. S1). These showed that the majority of cells for each crop were either fully irrigated or fully rain-fed. A threshold of 75% irrigated was used to identify those cells where the majority of the production was on irrigated land. Production for the period 2010-12 was estimated per grid cell by applying a conversion factor from FAOSTAT national production data available, averaged for the years 1999 – 2001 (average production for 2010-12/average production for 1999-2001). Only cells with > 500 tonnes (0.0005 Tg) crop production in 2010-12 were included in the analysis.

As discussed in Mills *et al*. (2018a), each 1° by 1° grid cell was assigned to a climatic zone, using the global 'Climatic Zone' GIS raster layer produced by the European Soil Data Centre (ESDAC) at JRC (Joint Research Centre). For each climatic zone, a 90 day growing period was derived per crop (Table S1), with climatic zones illustrated in Fig. S2. Data sources for assigning crop timings are provided with Table S1. For ease of comparison of effects between crops, only the main growing season per year was used for each crop.

2.1.2 Intra- and inter-specific sensitivity of crops to ozone

To determine the relative sensitivity of the four crops to ozone together with the between-variety variation in response to the pollutant, it was necessary to update existing response functions based on ozone concentration as stomatal uptake-based functions are currently only available for wheat. We collated dose-response data from the scientific literature using the method developed by Osborne *et al.* (2016) for soybean and the commonly reported ozone metric, M7 (7 hour mean, averaged from 09:00 to 15:59). The soybean dose response relationship from Osborne *et al.* (2016) was included in our analysis, whilst response functions for wheat, rice and maize provided in Mills & Harmens (2011) were updated with more recent published data (Web of Science and Google Scholar searches conducted between April and October 2017 using the search terms "ozone and yield and *crop name*"). Studies were only included if they met a number of selection criteria. The duration of ozone exposure must have spanned at least 60% of the 90 day growing season for each crop and ozone levels during exposure were up to 100 ppb for wheat and 170 ppb for other crops. Experiments were included if carried out in Open Top Chambers (OTCs), ambient air or large closed chambers/greenhouses (with the air stirred by fans, minimum size 2.6 by 2.2m). Data from both container and field-sown experiments were used to ensure a wide variety of points from different varieties were included. If the seasonal M7 was not given in the text, this was calculated either using the conversion equations provided in Osborne *et al.* 2016 (e.g. for 24 hour mean to M7) or information contained in the experimental methodology of the study. As there was no new published data available at the time of analysis for maize, the response function from Mills & Harmens (2011) was used. Yield data from different experiments were standardised as first described by Fuhrer (1997) and recently re-described by Osborne *et al.* 2016. Thus, for each set of experimental data, linear regression was used to determine the yield at 0 ppb of ozone (the intercept of the line); this value was the reference for calculating the relative yield (i.e. relative yield = actual yield/yield at 0 ppb).

Individual variety dose-response functions were derived for wheat and rice for the four ~~varieites~~varieties with the most data points. Following Osborne *et al.* 2016, yield reduction estimates ($RYL_{c,p}$) were then calculated for varieties showing statistically significant declines in yield with increasing ozone by calculating the difference in percentage yield loss at 55ppb (representing current M7) relative to that at 23 ppb (representing pre-industrial M7).

2.1.3 Yield constraints caused by ozone

The EMEP MSC-W (European Monitoring and Evaluation Programme, Metereological Synthesising Centre-West) chemical transport model (version 4.16, Simpson *et al.*, 2012, 2017) was used to derive daily POD_3IAM (Phytotoxic Ozone Dose above 3 nmol m^{-2} s^{-1}, parameterised for Integrated Assessment Modelling, CLRTAP, 2017) values for the years 2010 to 2012 per 1° by 1° grid cell as described by Mills *et al.* (2018a). POD_3IAM is parameterised for a generic crop represented by wheat (CLRTAP, 2017) and represents the accumulated stomatal uptake of ozone, modelled from the hourly mean values for ozone, temperature, vapour pressure deficit, irradiance and soil moisture (Mills *et al.* 2018a). Evaluation of the EMEP model performance is also presented in Mills *et al.*, 2018a, and is summarised in the Supporting Information for the current paper (T1).

For each crop, the accumulated 90 day POD_3IAM was then calculated per cell using appropriate climate-specific 90 day growing periods (Table S1, Fig. S2), and an average calculated for the period 2010-2012. For example, for soybean in warm temperate climates in the Northern Hemisphere, the time interval was day 182 to day 271. The EMEP model generated irrigated (without soil water limitation) and non-irrigated (rain limited) POD_3IAM values. For grid cells classed as irrigated for each crop (See Section 2.1.1), the irrigated POD_3IAM value was used to calculate percentage yield loss, otherwise the non-irrigated POD_3IAM was used. This approach allowed crop-specific irrigation usage to be taken into account, and was different to Mills *et al*. (2018a) where POD_3IAM values were weighted by the proportion of irrigation use within a 1 x 1° cell. The global distribution of POD_3IAM for each crop is provided in Fig. S3.

Yield loss due to ozone was first calculated for wheat using the most recent methodology adopted by CLRTAP, 2017. This method also differed slightly from that used in our earlier study (Mills *et al*., 2018a) in that a reference POD_3IAM value to represent ozone uptake at pre-industrial or natural ozone levels was subtracted before crop loss was calculated (CLRTAP, 2017). This value (0.1 mmol m^{-2}, Equ. 1) was the mean POD_3IAM for the experimental conditions included in the dose-response relationship, assuming constant 10 ppb ozone throughout the 90d period. The equation used to determine percentage yield loss was:

$$\% \text{ Yield loss} = (POD_3IAM - 0.1) * 0.64 \qquad \text{[Equ. 1]}$$

Where 0.64 is the slope of the relationship between POD_3IAM and percentage yield reduction (Mills *et al*., 2018a) and represents the percentage reduction per mmol m^{-2} POD_3IAM.

For soybean, maize and rice, the climate-specific grid square POD_3IAM values were first used to calculate yield loss using the wheat equation (Equ. 1), and the resultant value was then multiplied by the relative sensitivity of the crop compared to wheat, RS_w. The latter was derived by dividing the slope of the M7 response function for the crop (Fig. 1) by that for wheat. Production loss per crop was calculated per grid square using the following equation:

$$\text{Production loss (tonnes)} = \text{Crop production} * (\% \text{ yield loss}/100) \qquad \text{[Equ. 2]}$$

2.2 Global spatial analysis of yield constraints caused by other stresses

2.2.1~~2.4~~ Yield constraints caused by pests and diseases

Oerke *et al.* (1994; 2006) provide estimates for pre-harvest crop losses due to weeds, animal pests, (arthropods, nematodes, mammals, slugs and snails, birds), pathogens and viruses for several major global crops, using data compiled from the literature. This database provides regional percentage yield loss estimates up to 2004 for 11 crops, including soybean, wheat, rice and maize, and is available from the Centre for Agriculture and Biosciences International (CABI) Crop Protection Compendium (CABI, 2005). A value for mean percentage yield loss due to pests and diseases for the period 2002-04 was assigned to each 1° by 1 ° grid cell, based on the country and region of the world the cell was located in. If a cell contained land from more than one country, it was assigned to a country based on where the majority of the crop was growing in the cell. Data was available for 19 global regions (Oerke *et al.*, 2006). In this study, data were used that represented the remaining crop yield losses after crop protection practices had been applied.

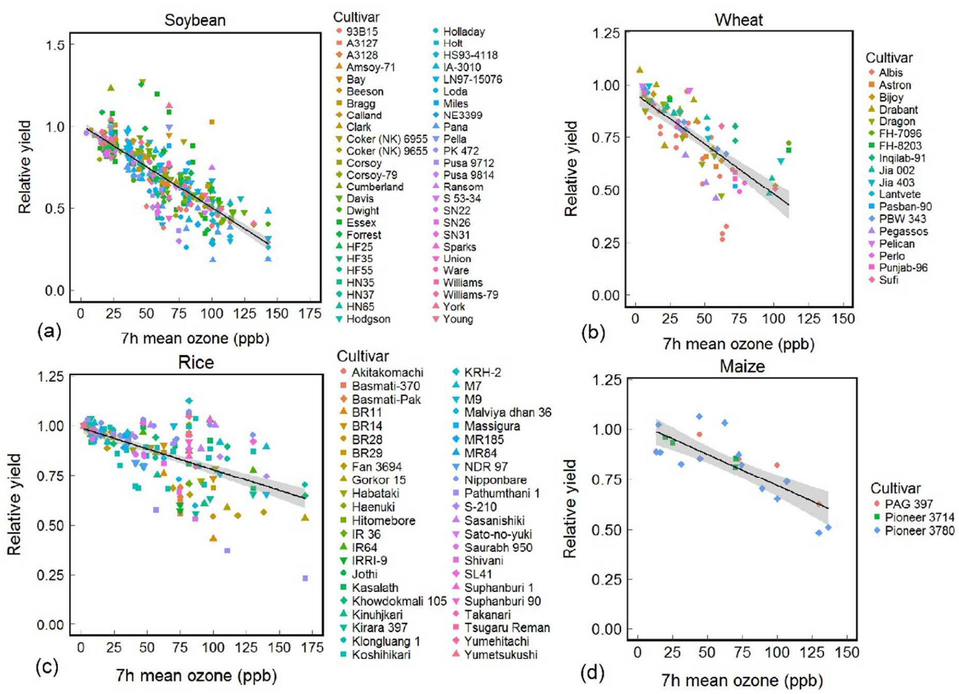

Fig. 1. Response functions for (a) soybean, (b) wheat, (c) rice and (d) maize derived from published data using the growing season ozone (7h mean, M7 in ppb) in the experiments. Data points are presented per cultivar/variety, with sources of data provided in the Supporting Information (Table S3). The response functions are: Soybean, RY = -0.0050x + 1.001 (r^2 (adj) = 0.625, p<0.001); Wheat, RY = -0.0048x +0.96 (r^2 (adj) = 0.547, p<0.001); Rice, RY= -0.0021x +0.987 (r^2 (adj) =0.347, p <0.001); and Maize, RY = -0.0031x + 1.03 (r^2 (adj) = 0.617, p <0.001).

2.2.2~~5~~ Yield constraints caused by soil nutrients

Soil nutrient classifications (nutrient availability and nutrient retention) at 0.083° by 0.083° resolution were downloaded from the GAEZ (v. 3) data portal (http://www.fao.org/nr/gaez/en/) in June, 2017. The soil qualities (nutrient availability and retention) in the GAEZ dataset have been derived from combinations of soil attributes, using

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Comment [MGE1]: Higher resolution figure inserted data in the Harmonized World Soil Database (HWSD, v. 1.1, FAO/IIASA/ISRIC/ISS CAS/JRC 2009). Nutrient availability refers to soil fertility, and classification is based on soil texture, soil organic carbon, soil pH and total exchangeable bases. The nutrient retention capacity of soil is based on the ability of soil to retain added nutrients against losses due to leaching. Classification of nutrient retention has been derived from soil texture, base saturation, cation exchange capacity of the soil and of the clay fraction and soil pH. In the GAEZ dataset, nutrient availability and retention are classed separately for topsoil (0-30cm) and subsoil (30-100cm) and then combined by weighting based on the prevalence of active roots (Fischer *et al.*, 2012). The GAEZ classes for soil nutrient availability and nutrient retention were combined in this study to produce five soil nutrient stress classes (summarised in Table 1, further details provided in Table S2). The soil nutrient class making up the majority of each 1° by 1° grid cell in areas where crops were growing, was used to represent the class for each cell.

2.2.3~~6~~ Yield constraints caused by heat stress

Following the methods of Challinor *et al.* (2005), subsequently used by a number of other studies (e.g. Deryng *et al.*, 2014; Teixeira *et al.*, 2013), a heat stress index was calculated per grid cell for each crop to determine if the daily temperature within a 30 day thermal sensitive period (TSP) exceeded the tolerance thresholds for each crop. This method assumes that damage to crops occurs when daily temperatures exceed a critical temperature (T_{crit}, °C) and maximum damage occurs when temperatures exceed the limit temperature (T_{lim} °C). Using information on the reproductive phase for each crop (FAO), the thermal sensitive period was designated as days 40-70 of the 90 day growing period (which varies with climate zone for each crop, Table S1). Following Deryng *et al.*, (2014), the daily effective temperature (T_{eff}, °C, (daily mean temp + daily max temp)/2), used as a measure of the daily temperature when photosynthesis is taking place, was calculated per grid cell using global hourly temperature data for the period 1990-2014 at 0.5° by 0.5° resolution. The temperature data were from the European Centre for Medium-Range Weather Forecasts Integrated Forecasting System (ECMWF-IFS, www.emwf.int/research/ifsdocs/), as prepared for use by the EMEP model.

For each crop, a daily heat stress value (*f* HSd) was then calculated for each day within the TSP, per grid cell. As we required an index that could be used to detect increasing levels of stress (i.e. an index scaled from 0 to 1), heat stress was calculated following Teixeira *et al.* (2013) (Eqn. 3).

$$f\,\mathrm{HSd} = \begin{cases} 0 & \text{for } T_{\mathrm{eff}} < T_{\mathrm{crit}} \\ \dfrac{T_{\mathrm{eff}} - T_{\mathrm{crit}}}{T_{\mathrm{lim}} - T_{\mathrm{crit}}} & \text{for } T_{\mathrm{crit}} \le T_{\mathrm{eff}} < T_{\mathrm{lim}} \\ 1 & \text{for } T_{\mathrm{eff}} \ge T_{\mathrm{lim}} \end{cases} \qquad \text{[Equ. 3]}$$

An average value was then calculated across the 30 day TSP to give the final heat stress index value, (*f* HS) per grid cell (Eqn. 4).

$$f\,\mathrm{HS} = \frac{\sum_{j=1}^{\mathrm{TSP}} (f\,\mathrm{HSd})}{\mathrm{TSP}} \qquad \text{[Equ. 4]}$$

Critical and limiting temperatures per crop were taken from Deryng *et al.* (2014) (maize, wheat and soybean) and Teixeira *et al.* (2013) (rice). These were: Soybean (35°C for T_{crit} and 40°C for T_{lim}), wheat (25 and 35 °C), rice (35 and 45°C) and maize (32 and 45°C).

2.2.4~~7~~ Yield constraints caused by aridity

Global Aridity Index data (Trabucco & Zomer 2009) were downloaded from the CGIAR-CSI GeoPortal (http://www.csi.cgiar.org). The mean Aridity Index for the period 1950-2000 (0.0083° by 0.0083° resolution) was calculated as:

Aridity Index (AI) = MAP / MAE [Equ. 5]

Where MAP is the Mean Annual Precipitation and MAE is the Mean Annual Potential Evapo-Transpiration.

Mean annual precipitation values were obtained from the WorldClim Global Climate Data (Hijmans *et al.* 2004), for years 1950-2000, while mean annual values of Potential Evapo-Transpiration (PET) were calculated using the average monthly PET values from the Global-PET model (Trabucco & Zomer, 2009). The mean Aridity Index per cell was calculated for each 1° by 1° grid cell where there is production for the crop.

2.3 Comparative analysis of effects of five stresses using a ~~8~~ Yield Constraint Score (YCS)

As percentage yield loss data was only available for ozone and pests and diseases data, a percentage scale could not be used for all stresses. To overcome this problem, a yield constraint score (YCS) on a scale of 1 – 5 was developed for each abiotic and biotic stress to show spatially where each constraint is predicted to be impacting on yield and to provide some indication of the magnitude of the effect (Table 1). Yield loss was split into the same five percentage yield loss classes for ozone and pests and diseases, with the highest class being >40% and expected to be comparable to severe stress for all yield constraints. Soil nutrient retention and availability were combined to give five overall classes (Table S2). The aridity climate classes used were from the Generalized Climate Classification Scheme

(UNEP 1997), while the heat stress index was classified following the methods of Teixeira *et al*. (2013). To identify those areas of the world with the highest combined stresses, the YCS for all five stresses were summed (YCS_{all}).

Table 1: **Categories of Yield Constraint Score (YCS) for ozone, pests and diseases, soil nutrients, heat and aridity** (See text for explanations and justifications of categories).

Yield constraint score (YCS)		1	2	3	4	5
Ozone	% Yield Loss	0 to 5	5 to 10	10 to 25	25 to 40	> 40
Pests and diseases	% Yield Loss	0 to 5	5 to 10	10 to 25	25 to 40	> 40
Nutrients	Retention	None or slight	Slight to moderate	Slight to severe	Moderate to severe	Moderate to very severe
	Availability	None	Slight to moderate	Moderate to severe	Severe	Severe to very severe
	Overall	None	Slight	Moderate	Severe	Very severe
Heat	Index	0	<0.05	0.05 to 0.15	0.15 to 0.3	> 0.3
Aridity	Index	>0.65	0.5 to 0.65	0.2 to 0.5	0.03 to 0.2	<0.03
	Climate class	Humid	Dry sub-humid	Semi-arid	Arid	Hyper arid

~~**2.9 Regional classification of impacts**~~

Stress	Attribute	Year(s) of data	Yield constraint score (YCS)				
			1	2	3	4	5
Ozone	% Yield Loss	Mean of 2010-2012	0 to 5	5 to 10	10 to 25	25 to 40	> 40
Pests and diseases	% Yield Loss	Mean of 2002 - 2004	0 to 5	5 to 10	10 to 25	25 to 40	> 40
~~N~~Soil Nutrients	Retention	HWSD data, 2009, downloaded	None or slight	Slight to moderate	Slight to severe	Moderate to severe	Moderate to very severe

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	Availability	in June, 2017	None	Slight to moderate	Moderate to severe	Severe	Severe to very severe
	Overall		None	Slight	Moderate	Severe	Very severe
Heat	Index	Mean of 1990 - 2014	0	<0.05	0.05 to 0.15	0.15 to 0.3	> 0.3
Aridity	Index	Mean of 1950 - 2000	>0.65	0.5 to 0.65	0.2 to 0.5	0.03 to 0.2	<0.03
	Climate class		Humid	Dry sub-humid	Semi-arid	Arid	Hyper arid

For description of effects of the five stresses, results are described as regional and national averages, with the mean YCS and YCS_{all} rounded to the nearest integer, reflecting their categorical nature. The regional classification of countries used is that adopted by the Task Force on Hemispheric Transport of Air Pollutants (HTAP) of the Convention on Long-range Transboundary Air Pollution (LRTAP, Dentener & Guizzardi, 2013). Region names are provided in full in the text the first time they are used and thereafter are referred to by the HTAP three letter codes. The region names, three letter codes and a map illustrating the countries included per region are provided in Fig. S4.

2.4 Qualitative analysis of plant traits associated with ~~10 Defining a crop ideotype for ozone and~~ multiple stress tolerance

The scientific literature on crop stress tolerance was reviewed between June and December, 2017, with the aim of developing an ideotype for an ozone- and multi-stress tolerant crop. This analysis identified target traits to induce ozone tolerance, including reducing the effects on panicles, leaves and roots. It also considered the benefits and trade-offs for tolerance of other stresses, of introducing ozone tolerance into crops.

3. Results

3.1 Quantification of the global impacts of ozone and other stresses on crop yield

3.1.1 Intra-specific sensitivity to ozone

A comprehensive collation of published data on the yield responses of soybean, wheat and rice to ozone resulted in a database representing 52, 18 and 44 varieties, respectively (Fig. 1a-c, with data sources in Table S3). Ozone-response data for these three crops provides good representation of the areas where the crops were grown: Soybean (East Asia (EAS), North America (NAM) and South Asia (SAS)); wheat (Europe (EUR), EAS, SAS) and rice (EAS and SAS). In contrast, only three varieties have been tested to date for yield responses in maize (Fig. 1d), with these experiments being conducted in the USA during the 1980s and early 1990s. For each crop, there was a significant negative response to ozone ($p<0.001$), with the slope of the negative relationships declining in the order soybean (-0.0050) > wheat (-0.0048) > maize (-0.0031) > rice (-0.0021). Within each response function, variation in ozone sensitivity due to variety provided scatter in the range of sensitivity.

For each crop, some varieties were more tolerant to ozone than others, indicating that there is scope for selecting more tolerant varieties for immediate use or as part of a breeding programme for new varieties. For soybean, $RYL_{c,p}$ ranged from 13.3% to 37.9%, with the three most sensitive varieties being the Indian varieties 'PK472', 'Pusa 9712' and 'Pusa 9814' (Osborne *et al.* 2016). For wheat, the RYLc,p for the four varieties with the most data was 16.4% ('Drabant'), 19.3% ('PBW 343'), 26.4% ('Dragon') and 32.5% ('Albis'). While the 44 rice varieties showed a range of sensitivities to ozone (Fig. 1c), overall, ~~indicating~~ rice ~~it~~ was the least sensitive of the four crops investigated. Of the four rice varieties with the most data, 'Koshihikari' showed a $RYL_{c,p}$ of 4.7%, 'Nipponbare' showed no significant negative relationship between relative yield and M7 ($p>0.05$), while 'Kasalath' and 'Kirara 397' had a RYLc,p of 6% and 11.1 % respectively. The rice variety 'Pathumthani-1' showed a higher $RYL_{c,p}$ of 18.1%, however only 5 data points were available, therefore further study may be required to confirm this result. For maize, the r^2(adj) was 0.62 for the response function ($p<0.001$, Fig. 1d), with a $RYL_{c,p}$ for all three varieties of 10%. The RYLc,p for 'Pioneer 3780' with 14 data points was 15.7% whilst RYLc,p was not calculated for 'PAG 397' as the response function for the 5 data points for this variety was not significant ($p=0.08$).

3.1.2~~2~~ Spatial analysis of the global impacts of multiple stresses on crop yield

~~3.2.1~~ Soybean

The highest ozone-associated production losses (in Tg per 1 x 1° grid square) for soybean are predicted to be in ~~North America (~~NAM~~)~~ and South America (SAM) (Table 2), particularly in central and E USA, S Brazil and N Argentina (Fig. 2, Table S4). However, the percentage yield losses are predicted to be lower for Brazil (12.5 – 15%) and Argentina (7.5- 10%) than for the USA (> 20% in large areas), showing that in high producing areas where ozone concentrations are more moderate (as indicated by the percentage losses), high total production losses can still be expected. In the rest of the world, production losses due to ozone in excess of 0.01 Tg per 1 x 1 ° grid square are predicted for parts of China, India and S and E Europe. In each of these areas, the production loss was not as high as expected from percentage yield losses in excess of 20%, because soybean is not widely grown.

The YCSs for ozone were mainly score 3 for the highest producing regions, with some areas with a score of 4 in E USA, NE India and China (Fig. 2, Tables 2 and S4). There is overlap between these areas and the areas with the highest YCS_{all}. Other areas with a relatively high YCS_{all} such as parts of Sub-Saharan Africa (SSA) and ~~South East Asia (~~SEA~~)~~ are not predicted to have high production losses due to ozone because of lower percentage yield losses and/or low production totals per region. For soybean, the YCS for pests and diseases is 3 or more over most of the growing area, and particularly high (score of 5) in parts of SSA, ~~South Asia (~~SAS~~)~~ and SEA. The largest YCS values for nutrient availability (scores of 4 and

5) are in areas of SE USA, S Brazil and SEA including Thailand, Malaysia and Indonesia. Whilst heat stress YCSs are lower than those for aridity, the areas affected by both stresses largely coincided in soybean growing areas.

Overall, for soybean, the global mean YCS for ozone of 3 is one category below that for pests and diseases, and one higher than that for nutrients and aridity (Table 2). The mean YCSs for ozone were in the range 2 - 3 for the five highest producing regions (SAM, NAM, EAS, SAS, Russia (RBU)), with YCSs being in the range 3 – 5, 1 – 3, 1 - 2 and 1 - 2 for pests and diseases, nutrients, heat and aridity, respectively (Table 2).

Table 2: Production and regional mean Yield Constraint Score (YCS, rounded to nearest integer) for the five highest producing regions for soybean, wheat, rice and maize.

Soybean	**Production (Tg)**	**Ozone**	**Pests & diseases**	**Nutrients**	**Heat**	**Aridity**
Global	253.6	3	4	2	2	2
South America	125.8	2	4	3	2	1
N America	90.3	3	3	2	2	1
East Asia	14.9	3	4	2	1	2
South Asia	13.2	3	5	1	2	2
Russia	3.6	2	4	1	1	2
Wheat	**Production (Tg)**	**Ozone**	**Pests & diseases**	**Nutrients**	**Heat**	**Aridity**
Global	673.3	2	4	2	3	2
Europe	163.8	2	3	2	2	1
East Asia	118.9	2	3	2	2	2
South Asia	118.1	2	4	2	4	2
N America	84.2	2	4	2	2	2
Russia	66.1	2	4	2	2	1
Rice	**Production (Tg)**	**Ozone**	**Pests & diseases**	**Nutrients**	**Heat**	**Aridity**
Global	716.8	1	4	2	1	2
East Asia	221.2	2	4	2	1	2
South Asia	219.2	2	5	2	2	2
South East Asia	204.5	1	4	3	1	1

South America	22.7	1	4	3	1	1
Sub Saharan Africa	21.1	1	5	2	1	2
Maize	**Production (Tg)**	**Ozone**	**Pests & diseases**	**Nutrients**	**Heat**	**Aridity**
Global	869.1	2	4	2	2	2
N America	313.1	2	3	2	2	2
East Asia	194.4	2	4	2	2	2
South America	91.7	2	4	3	2	2
Europe	75.6	2	3	1	2	1
Sub Saharan Africa	60.0	1	5	2	2	2

~~3.2.2~~ Wheat

By far the highest production losses due to ozone per grid square for wheat are predicted for India and China, with large areas in N India and NW China having over 15% yield losses amounting to production losses in excess of 0.1 Tg (Fig. 3, Table S5). Production losses are also predicted to be high in the highest wheat producing areas of Europe (including France and Germany) and central states of the USA. The mean YCS for ozone globally was 2, reflecting the same mean score in the 9 highest wheat producing regions (Tables 2 and S5), and matching that globally for nutrients, and aridity. The highest predicted production losses due to ozone only overlapped with areas of the highest YCS_{all} in NW India, Pakistan and S USA (Fig. 3). Scores of 3 and above coincide for ozone, pests and diseases, heat and nutrients in a wider area including parts of EAS, SAS and NAM, whilst YCSs for aridity are lower in several parts of this region than for ozone. This study also indicated that the highest YCS values for all stresses are for heat (score 5) in areas of Northern Africa (NAF), SAS, SAM, SSA and SAM (particularly Argentina). Scores for pests and diseases are 3 or more across most of the wheat growing areas. YCSs for nutrient availability are generally the lowest of the five stresses, although there are some high risk areas with values of 4 and above in, for example, NW SAS, Central Asia (CAS) and NE EUR (e.g. Finland), E NAM and SAM (e.g. Brazil). The highest YCSs for aridity are in a zone that includes parts of NAF, the

Middle East (MDE) and SAS, CAS and EAS. For the five main wheat producing regions, the mean YCS for ozone was 2 representing 5 - 10% yield loss, whilst it was 3 - 4 for pests and diseases and mainly 1-2 for the other three stresses (Table 2).

~~3.2.3~~ Rice

Production losses due to ozone are predicted to only be in excess of 0.1 Tg per grid square in parts of India, Bangladesh, China and Indonesia (Fig. 4). In these areas, the percentage yield losses are mainly in the range 7.5 – 12.5%, resulting in grid square ozone YCSs that are usually either 2 or 3. Across all of the rice growing regions, the mean YCS for ozone per region is either 1 or 2, resulting in a global mean score of 1 and a score of 2 in the two highest producing regions (Tables 2 and S6). YCSs of 3 for ozone occurred in areas of India and China where the YCS_{all} was usually in the highest range for rice of 13 – 15. Overall, the highest mean YCS for this crop is for pests and diseases, being score 5 in most of the rice growing areas of SSA and SAS. Nutrient YCSs are 4 or more in many parts of SSA, SAM (particularly Brazil), EAS and SAS. Heat stress is predicted to be less of a problem for rice than for wheat, with few regions having a score of 3 or more. Indeed, the regional mean YCS for heat stress in rice is mostly either 1 or 2 (Tables 2 and S6). In the three highest producing rice regions, EAS, SAS and SEA, irrigation usage is 96%, 74% and 28%, respectively (Table S5). Here, the aridity score is predicted to ~~only~~ be 3 or more only in areas of NW China and W India.

~~3.2.4~~ Maize

China and the USA are the two countries predicted to have the largest areas where production losses due to ozone for maize that exceed 0.1 Tg per 1° x 1 ° grid square (Fig. 5). In these areas, the percentage yield losses are mainly in the range 7.5 – 15% for the USA and 12.5 – 15 % for China. There are also high-risk areas in S EUR, for example, in parts of S France and N Italy and in NAF (particularly Egypt). These areas generally have a YCS_{all} for maize in the range 10 – 15, and are not in the areas with the highest YCS_{all} for maize of >15. The latter are mainly found in parts of SAS and SSA, with occasional small areas elsewhere. The mean YCS for ozone for the four highest maize producing regions (NAM, EAS, SAM and EUR) is 2 (Tables 2 and S7). For the stresses other than ozone, the highest scores for YCS are for pests and diseases, with scores of 5 predicted in most of SSA, SAS and SEA. YCSs of 4 and above are predicted for nutrients in much of SSA (particularly in western countries), SEA, large areas of SAM (particularly Brazil), parts of E USA and small areas of Europe. For maize, the YCSs for aridity are highest in eastern NAM and SAM, parts of NAF (particularly W Egypt), MDE, SAS (particularly NW India) and EAS (particularly NE China). Heat stress YCSs are lower for maize than for wheat, indicating that the main areas of concern for this crop are in W SSA, W MDE and SAS. Globally, the mean YCS for maize is 4 for pests and diseases and 2 for each of the other four stresses (Table 2).

3.1.3 Case study - India

It is clear from the results presented above that the five environmental stresses included in this study are all predicted to be having relatively high impacts on yield in several states of India. We selected this country for a more in depth analysis. Although the spatial data for India is present on the global maps in Figs 2 - 5, for ease of interpretation, we have produced additional maps for India for wheat and rice, the two most important crops by production in Fig. S5 and S6, respectively. At the national scale, the mean YCSs for the crop with the highest total production in India, wheat, are 3, 4, 2, 4 and 2 for ozone, pests and diseases,

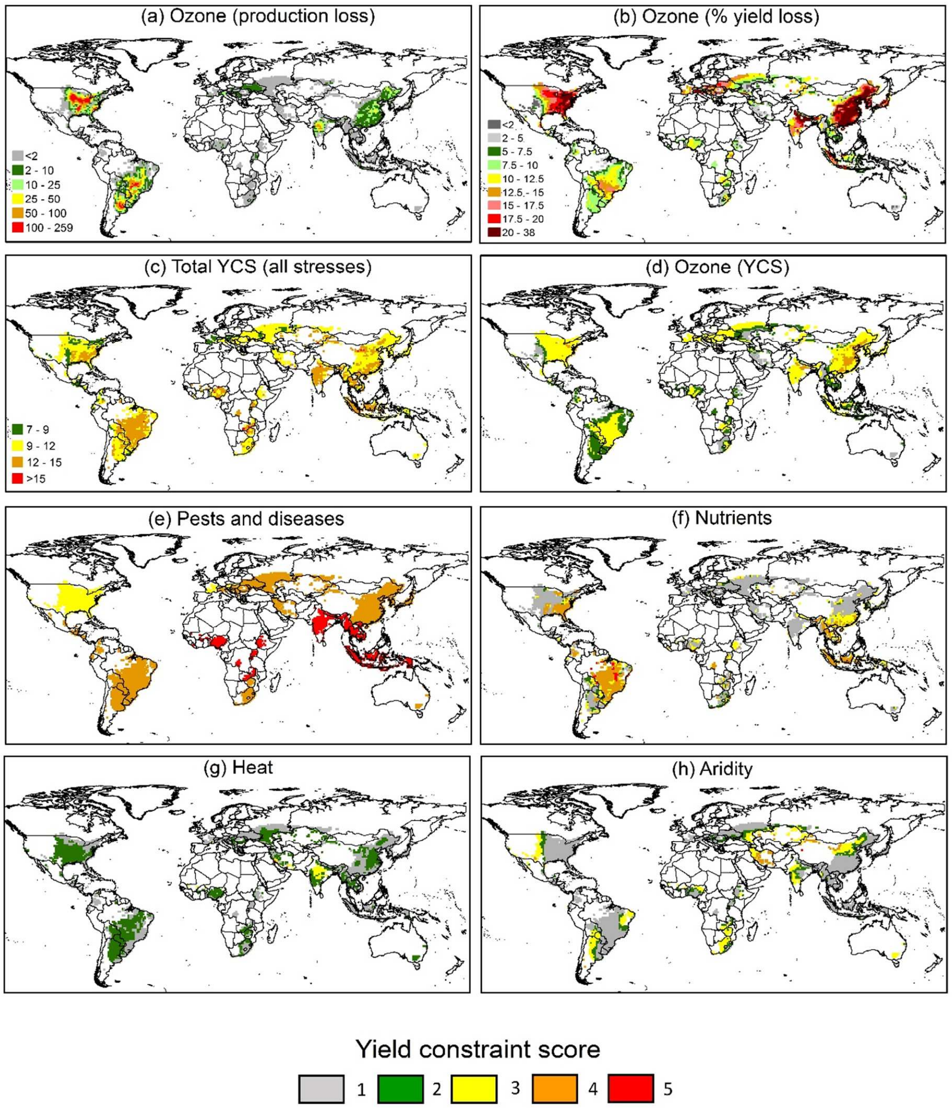

Fig. 2: The global effects of five biotic and abiotic stresses on soybean. All data are presented for the 1 x 1° grid squares where the mean production of soybean was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S4 provides all country and regional means.

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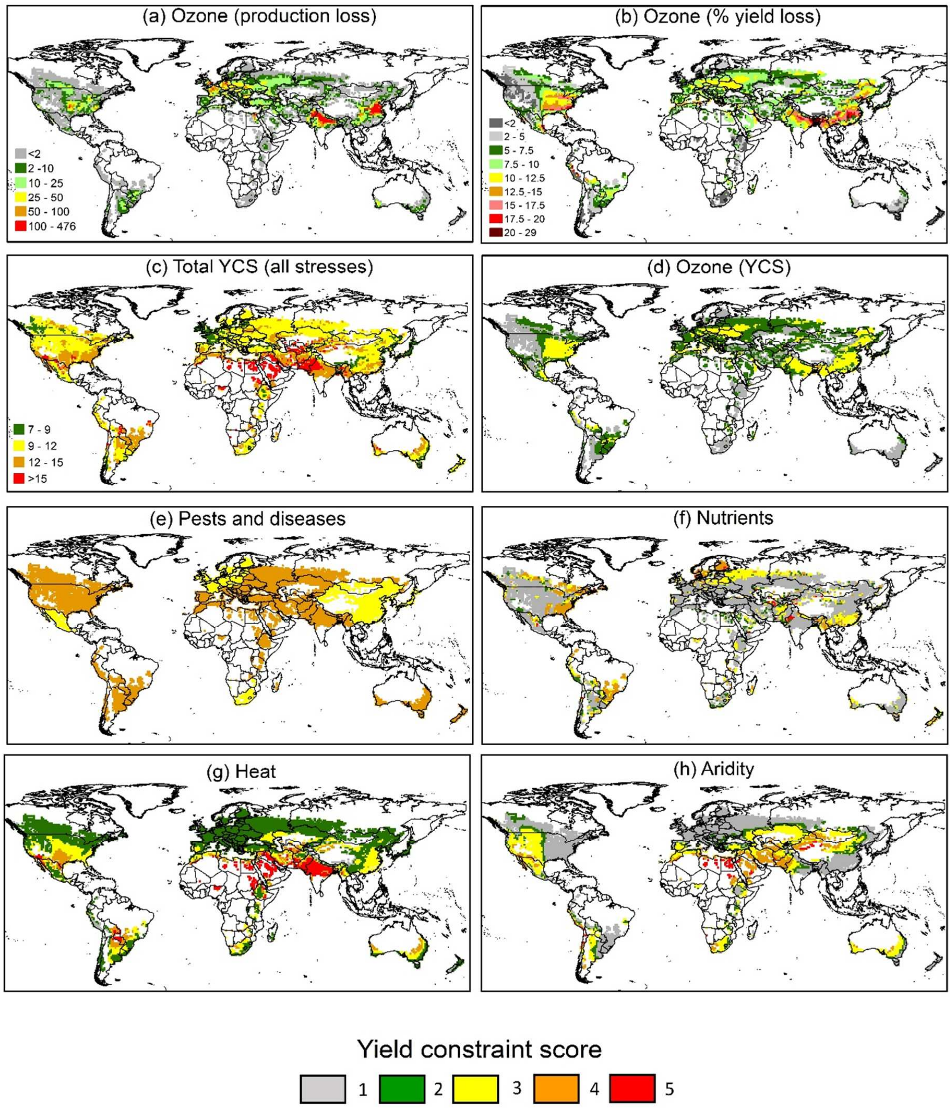

Fig. 3: The global effects of five biotic and abiotic stresses on wheat. All data are presented for the 1 x 1° grid squares where the mean production of wheat was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S5 provides all country and regional means.

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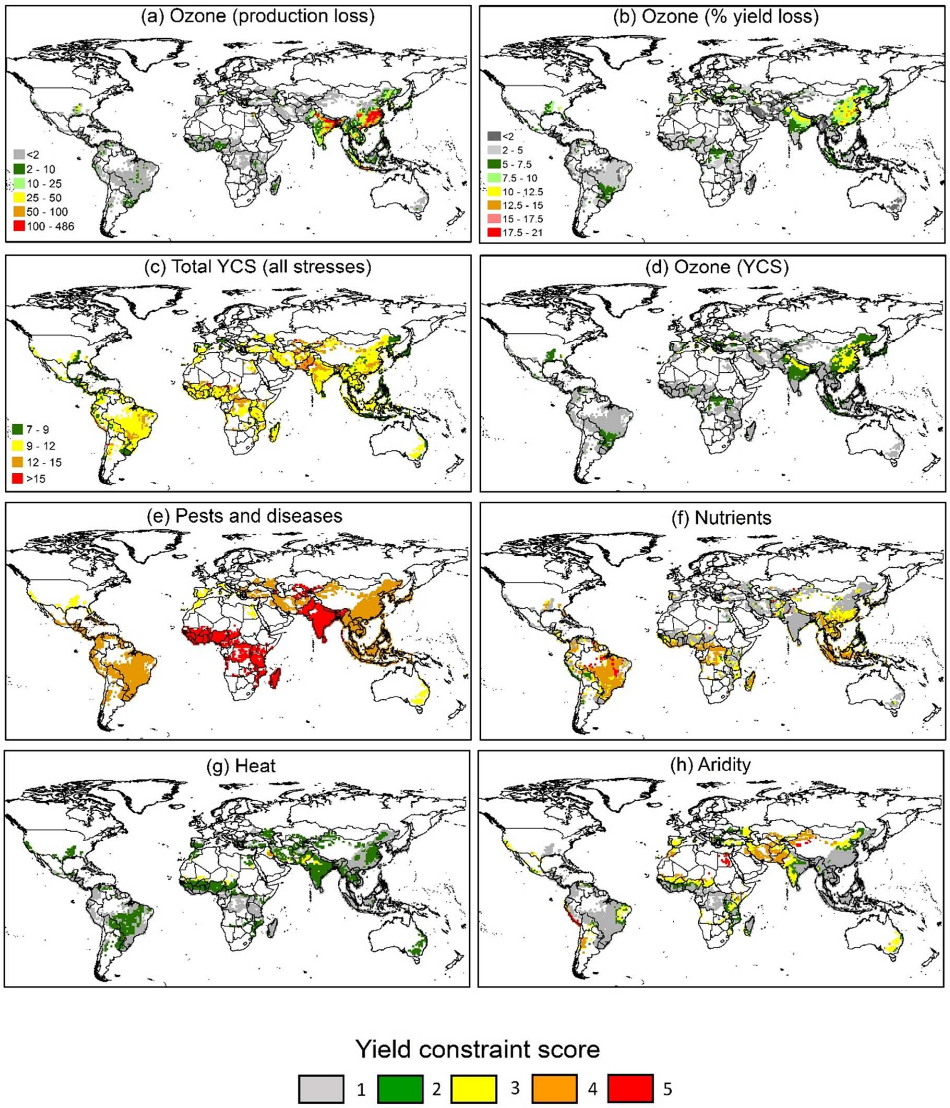

Fig. 4: The global effects of five biotic and abiotic stresses on rice. All data are presented for the 1 x 1° grid squares where the mean production of rice was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S6 provides all country and regional means.

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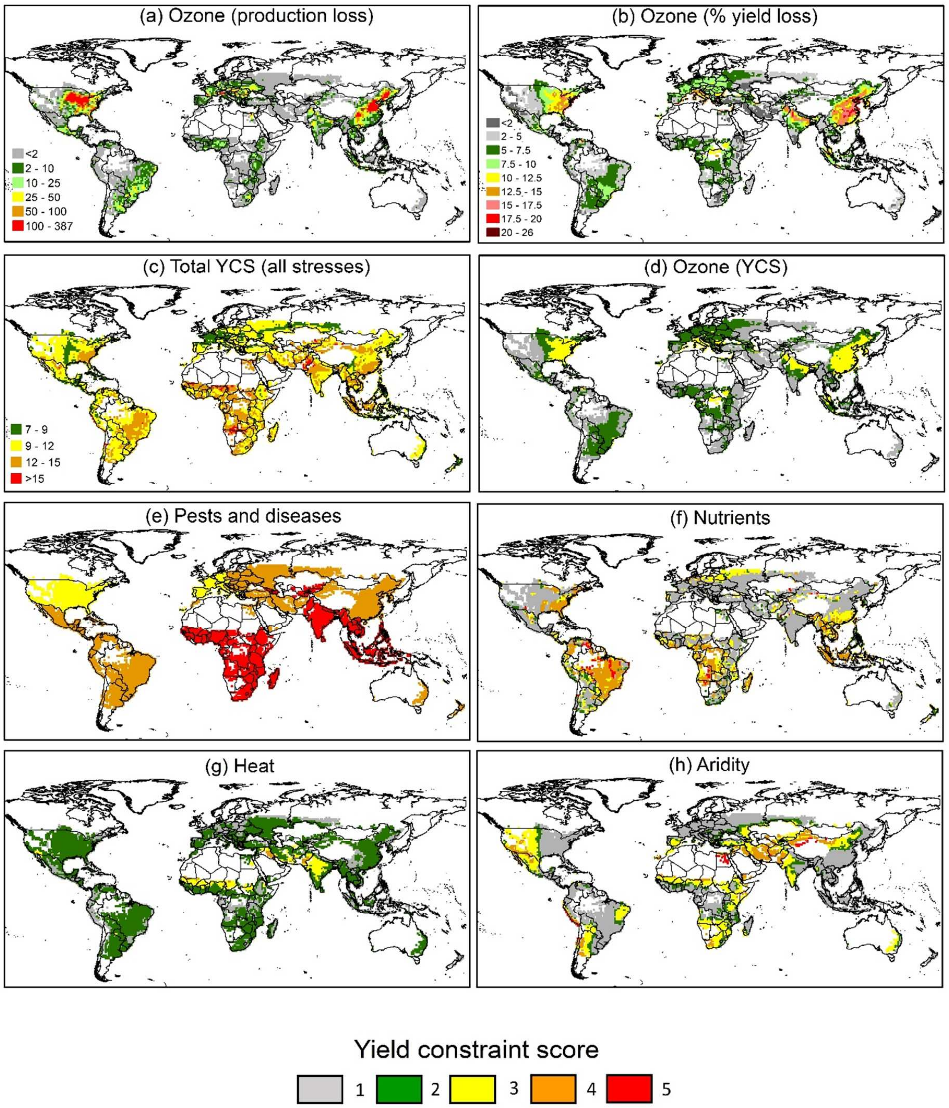


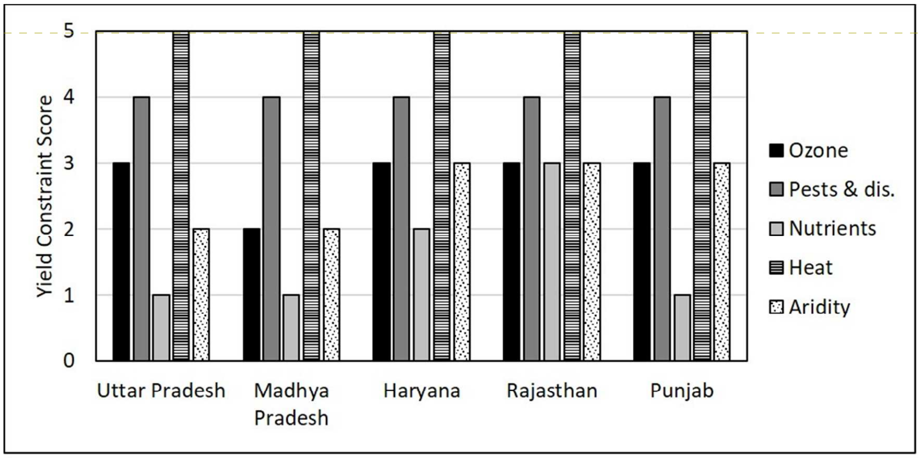
Fig. 5: The global effects of five biotic and abiotic stresses on maize. All data are presented for the 1 x 1° grid squares where the mean production of maize was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S7 provides all country and regional means.

Comment [MGE6]: Journal-ready higher resolution figure inserted nutrients, heat and aridity, respectively (Table S5). For rice, the second most important crop by Tg produced in India, the YCSs for the same five stresses respectively are 2, 5, 1, 2 and 2 (Table S6). As the data for the risk of losses due to pests and diseases was only available at the national scale for India, with YCSs of 4 for wheat and 5 for rice, these effects were not included in this spatial analysis, conducted at the 1 x 1 ° scale.

For wheat, the highest production is in the adjacent N states of Uttar Pradesh, Madhya Pradesh, Haryana, Rajasthan and Punjab (Fig. 6). Together, these five states account for 85% of Indian wheat production. Predicted percentage yield losses due to ozone are in the range 15 – 20 % (mean of 16.4%) in most of the wheat producing areas of Uttar Pradesh, the state with the highest wheat production, resulting in a mean ozone YCS of 3 (Fig. 6 and S5). The mean YCS for ozone was 3 for Haryana, Rajasthan and Punjab and 2 for Madhya Pradesh where ozone uptake is lower (Fig. S5). Although the highest percentage yield losses due to ozone were predicted for states in the far NE of India such as Assam and Manipur, total production losses there were predicted to be minimal as this is not an important wheat growing area. The area of highest ozone impacts on wheat production coincided with the area with the highest YCS for heat stress which covered most of the northern half of the country. Aridity and nutrient YCSs were highest to the W of this region, coinciding with percentage yield losses for ozone predicted to be in the range 5-15% (YCS of 2- 3). For the five highest wheat producing states, the mean YCS for heat stress was 5, with scores for aridity being 2 or 3, and nutrients being 1 – 3 (Fig. 6).

Rice growth is much more widely distributed in India than wheat growth, with the highest production being in the N, in part coinciding with wheat growing areas in states such as Uttar Pradesh, and also in E and S states such as West Bengal, West Odisha, Andhra Pradesh and Tamil Nadu (Fig. 6). Together these 5 states produce just below half of India's rice

(a)



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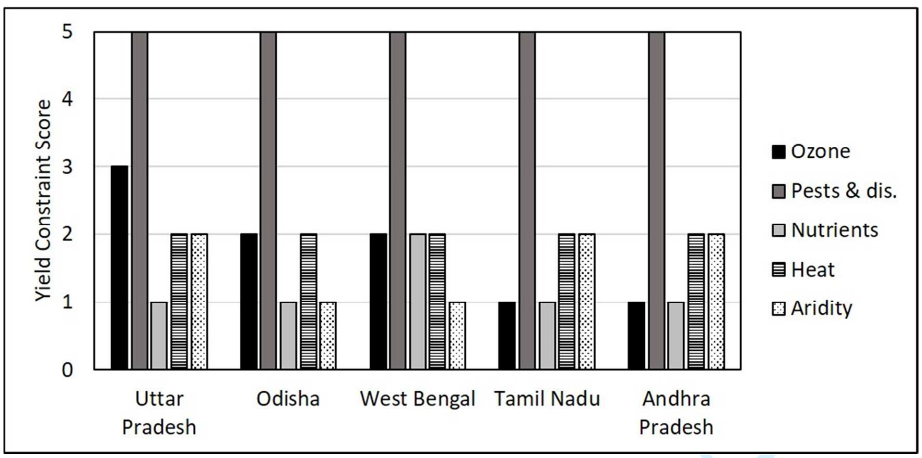

Fig. 6: Yield Constraint Score (YCS) for five constraints on the yield of (a) wheat and (b) rice in the five Indian states with the highest production per crop. The bars represent the mean YCS per 1 x 1°grid square per state on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1), rounded to the nearest integer. Note: The YCS for pests and disease is only available at the National Scale for India (score 4 for wheat and 5 for rice) and is presented here for information.

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Comment [MGE7]: Colour figure replaced with black and white figure production. The highest percentage yield losses for ozone are predicted to be in the range 10 – 15% in the N of the country, including in Uttar Pradesh (mean ozone YCS of 3, Fig. 6 and S6). Lower effects were predicted for Odisha and West Bengal (mean YCS of 2) and the least ozone effects were predicted for rice producing areas in the southern states of Tamil Nadu and Andhra Pradesh (mean YCS of 1), where percentage losses were frequently less than 5%. Heat stress is less of a concern for rice, with a mean YCS of 2 predicted for each of the 5 most important rice producing states. Nutrient stress is predicted to only be important in the far NE states and in isolated grid squares in Rajasthan and along the W coast of India. The mean YCSs for nutrients and aridity for the five highest producing states are either 1 or 2 (Fig. 6).

3.2. Plant traits associated with tolerance of ozone and associated stresses in crops

~~4 Crop ideotype for ozone and multiple stress tolerance~~

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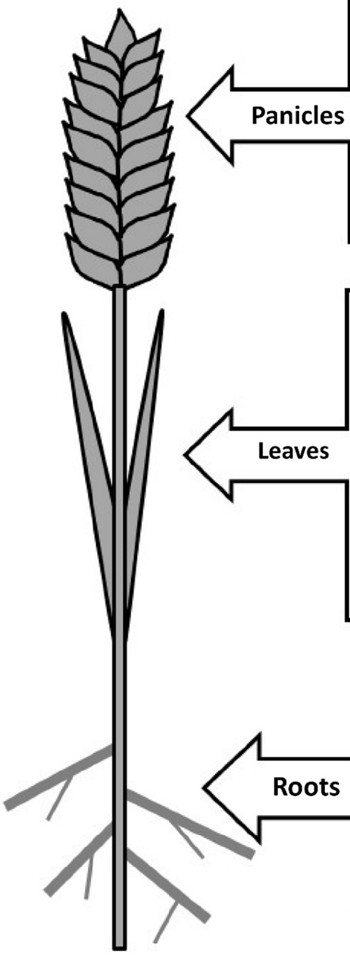
The derivation of dose-response relationships for 52, 18 and 44 genotypes of soybean, wheat and rice respectively (Fig. 1) has shown that there is clearly scope for the breeding of ozone tolerant varieties, as many varieties had responses that are above the regression line. As part of this study, we identified a number of traits that could contribute to improved ozone tolerance and have summarized these in an ozone-tolerant crop ideotype, including potential trade-offs and synergies for effects of other stresses that can co-occur with ozone (Fig. 7).

Leaf traits for ozone tolerance fall into two categories, the first being processes that limit ozone entry. These include stomatal conductance, and the related trait of water use efficiency (WUE), that reduce ozone uptake while maintaining high rates of photosynthesis. These traits are associated with reduced leaf transpiration and whilst they would be beneficial for water conservation under drought conditions, they may reduce yield and could be potentially deleterious under heat stress by limiting evaporative cooling (Reynolds *et al*., 2007).

Similarly, reduced water uptake associated with lower stomatal conductance has the potential to limit uptake of nutrients such as N from the soil (Zhou *et al*., 2016). While pathogens are



Target traits for ozone tolerance	Synergies / trade offs: Drought	Heat	Nutrients	Pests & dis.
Seed yield	0	0	0	0
Seed quality	+	+	+	+
Days to maturity	+	+	+	+

Target traits for ozone tolerance	Synergies / trade offs: Drought	Heat	Nutrients	Pests & dis.
Water use efficieny	+	-	-	?
Low stomatal conductance	+	-	-	+?
High antioxidant capacity	+	+	+/-	+/-
Containing PCD	+/-	+/-	+/-	+/-

Target traits for ozone tolerance	Synergies / trade offs: Drought	Heat	Nutrients	Pests & dis.
Root allocation efficiency	+	+	+	0
Root dynamics	+	+	+	0

Fig. 7: An ideotype for an ozone-tolerant~~resilient~~ crop. '+' indicates where there would be a benefit for other stresses of improving tolerance to ozone for the trait, whilst '-' indicates a trade-off, and '0' is no effect.

Comment [MGE8]: Colour figure replaced with black and white figure known to have negative effects on leaf gas exchange (Debona *et al*., 2014), the impact of inherently lower stomatal conductance on disease establishment is less clear although it could be expected that ingress of leaf pathogens that access leaves through the stomatal pores would be reduced.

A second category of favourable leaf traits includes antioxidant metabolism and pathways involved in programmed cell death (PCD). Ozone is decomposed into reactive oxygen species (ROS) in the plant apoplast, which either cause direct oxidative damage, or induce signalling cascades similar to a pathogen response, ultimately leading to PCD (Kangasjärvi *et al*., 2005). Thus, balancing the interplay of redox homeostasis and PCD pathways is essential for the breeding of ozone tolerant crop plants. As a first line of defence against ozone stress, high levels of apoplastic antioxidants such as ascorbate may mitigate ROS formation, a concept that has been confirmed in crop plants such as wheat (Feng *et al*., 2010) and legumes (Yendrek *et al*., 2015). Breeding for high levels of antioxidants is also assumed to cause synergies with other types of abiotic stress tolerance, including for drought and heat, both of which are associated with oxidative stress (Gill & Tuteja, 2010). In the case of some nutrient disorders and biotic stresses, functional redox balance rather than high antioxidant levels *per se* are considered as important (Munné-Bosch *et al*., 2013; Suzuki *et al*., 2012; Wu *et al*., 2017).

PCD is an important pathway of pathogen response in plant leaves (Huysmans *et al.,* 2017), which is controlled by the interplay of ROS, signalling cascades and plant hormones (Kangasjärvi *et al*., 2005). Breeding for ozone tolerance could thus keep plants from inducing PCD despite the presence of apoplastic ROS. This idea is supported by a study in rice, in which the disruption of the pathogen and ozone responsive apoplastic protein *OsORAP1*, which is involved in cell death, led to enhanced ozone tolerance (Ueda *et al*., 2015). The potential interference of this strategy with pathogen tolerance in crops is obvious, but it is currently unclear whether a synergistic or rather antagonistic relationship would occur with different classes of pathogens i.e. biotrophic versus necrotrophic ones (Huymans *et al.*, 2017). Implications of PCD in other stress types such as heat (Locato *et al*., 2008), drought (van Doorn 2011) and nutrient deficiency (Siyiannis *et al*., 2012) have also been reported but the implications for ozone tolerance breeding remain unclear.

Root traits that support ozone stress tolerance would include the capacity to efficiently acquire water and nutrient resources under stress environments (Resource Acquisition Efficiency in Fig. 7). Ozone is known to have a greater negative impact on roots than shoots, resulting in the decline in the root/shoot ratio commonly observed (Fiscus *et al.*, 2005). There is evidence that ozone may have an even greater impact on fine roots that acquire water and nutrients from the soil (Vollsnes *et al.*, 2010). Fine roots are the new frontier of future root research. The framework for describing fine root architecture is being refined (McCormack *et al.*, 2015; Zobel, 2016), and new techniques are now available to assess fine root dynamics (e.g. measurements of root diameter, Zobel *et al*., 2007). The challenge ahead is to define and measure fine root traits that contribute to ozone tolerance, and then determine how these traits affect plant response to other stress factors. Presumably, traits that contribute to robust root systems will be of benefit across a range of abiotic stresses.

Traits associated with reproductive organs such as panicles or pods are of primary importance in breeding, although the effects of ozone on these organs may be rather secondary, i.e. caused by foliar responses that limit assimilate acquisition (described above) or effects on flowering and pollen viability (Black *et al.*, 2000). Yield losses due to ozone have been ascribed to various yield components in different crops, including reductions in individual seed weight, reduced spikelet number, enhanced spikelet fertility, and reduced panicle or pod number (Ainsworth, 2008; Feng *et al*., 2008; Morgan *et al*., 2003), with associated reductions in harvest index (e.g. for wheat, Pleijel *et al*., 2014). Maintaining high values in these harvest fractions despite ozone stress forms an important breeding target, but synergies or trade-offs with other types of stress would be complex and little information is available to date.

Maintaining high crop quality despite ozone stress represents another important breeding goal. Ozone can affect multiple quality traits in seed crops, including protein and starch concentration, as well as visual appearance (Broberg *et al*., 2015; Wang & Frei, 2011). In many cases, increases in seed protein concentration despite losses in protein yield are observed. This apparent beneficial effect is offset by the negative effects of ozone on seed weight (e.g. for wheat, Broberg *et al*., 2015). Another quality trait that has been affected in rice by ozone is grain chalkiness, i.e. the formation of milky patches on grains due to inhibited starch loading (Jing *et al*., 2016). Chalkiness was first described as a typical symptom of heat and drought stress (Wassmann et al., 2009), and lowering plant susceptibility to chalkiness *via* breeding may thus have potential co-benefits with regards to these stresses.

A further category of traits that could be targeted by breeders are phenological characteristics. Plants that have a shorter maturity period by entering earlier into reproductive phases might be more tolerant, as they would receive a lower cumulative ozone dose, and might avoid high ozone episodes occurring late in the cropping season. This principle was confirmed in a study by Ueda *et al*. (2015), in which more than 300 genotypes of rice were screened for ozone response, and yield losses were positively correlated with the number of days to maturity. In general, breeding fast-maturing crop varieties may produce substantial synergies, reducing the impacts of growing seasons characterized by high incidence of other stresses, such as drought, heat, nutrient, or biotic stresses.

4. Discussion

In bringing together these datasets and modelling methods to derive YCSs for five stresses and four key crops, we have conducted the first global assessment of the magnitude of ozone stress in relation to other stresses for four staple crops. We have also derived an ideotype for an ozone- and multi-stress tolerant crop. We provide an extended discussion here that first considers the results presented and then consi~~u~~ders potential solutions for increasing crop tolerance of ozone, including crop management and breeding approaches.

4.~~2~~1 The global scale of ozone impacts on crops relative to impacts of other stresses

~~**Spatial analysis of the impacts of multiple stresses on crop yield**~~

An in depth evaluation of the spatial analysis conducted here is presented in the Supporting Information (T1) and summarised here. The benefits of impacts modelling based on the stomatal uptake of ozone rather than the concentration above the leaf, together with an evaluation of global modelling of POD_3IAM, are discussed by Mills *et al*. (2018a). In the absence of suitable stomatal uptake dose response relationships for soybean, rice and maize, the RS_w method was developed whereby the effects of ozone on these crops was determined from the POD_3IAM response of wheat. We had to assume that the differences in ozone concentration and sensitivity were a greater driver of response than differences in stomatal uptake and are unable to quantify the uncertainty introduced by this assumption. Whilst experimental data in the M7 response functions used in the RS_w method represented the major crop growing regions for soybean, wheat and rice, the function for maize was limited to relatively old data from NAM only. Thus, the data analysis presented here for maize is likely to be the most relevant for effects described in NAM, and is less certain when applied to other maize-growing regions of the world. For each crop, we assumed only one crop growth period per year. Thus, for those crops such as rice where two or three crop growth cycles may occur per year in major growing areas, assessments based on the main growth period will have an added level of uncertainty.~~then our estimates of total effects on~~

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~~production are likely to be underestimated~~. The abiotic and biotic stresses included here were selected as examples for comparison with ozone effects, with heat stress being chosen as representative of effects of extreme climatic events associated with climate change~~warming~~ ~~and~~ . We acknowledge that other stresses such as flooding may also have catastrophic local effects on yield (e.g. in China, Tao *et al*., 2017), but have focussed on example stresses for which global data is readily available. Furthermore, global warming impacts on yield could be in a similar range to ozone (e.g. Challinor *et al*., 2009; Lobell & Asseng, 2017) but have not been considered here. Scores for YCS for pests and diseases may have overestimated current losses as advances in pesticide usage since the 2002-04 dataset was compiled may have reduced total impacts. As it was not possible to base all YCSs on percentage yield loss, uncertainty will have been introduced by comparing effects across stresses. We have acknowledged this uncertainty by using the YCSs to indicate the location of the largest effects rather than to quantify the extent of effects. Lastly, YCS_{all} simply summed all YCSs and provides an indication of where multiple stresses co-occur, without taking into account any interactions that may occur that might lessen or increase the combined effects on yield. Taking into account all of these caveats, this study is the first to present ozone impacts on the global scale together with impacts of other biotic and abiotic stresses, and show spatially where such stresses are likely to co-occur for four major staple crops.

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At the national scale, the countries identified as having the largest potential effects of ozone (e.g. USA, India and China) match those with the highest monitored ozone concentrations (Mills *et al*., 2018b~~submitted~~) as well as those predicted using concentration-based approaches to have the highest potential yield losses (Avnery *et al*., 2011-a,b; Van Dingenen *et al*., 2009). At the sub-national scale, however, there were some differences in areas predicted to be at risk, where our stomatal uptake modelling method took into account the modifying effects of climate and soil moisture on ozone uptake rather than simply predicting the largest effects in the areas with the highest ozone concentrations. For example, in India, this study predicts the largest effects on wheat and rice in the northern areas, south of the Himalayas where ozone levels, climatic conditions and irrigation usage promote ozone uptake and subsequent effect. In contrast, an earlier concentration-based study provided little spatial differentiation in effects, predicting widespread and similar effects of ozone in the northern half of India for wheat and across most of India for rice (Van Dingenen *et al*., 2009). On a global scale, we predict that ozone (mean of 2010 - 2012) reduces soybean yield by 12.4%, wheat yield by 7.1%, rice yield by 4.4% and maize yield by 6.1%, adding up to a total of 227 Tg of lost yield. These mean percentage losses are different to those predicted by Avnery *et al*. (2011a) and Van Dingenen *et al*. (2009) using concentration based metrics. Their studies predicted higher losses for wheat (15.4% and 12.3%, respectively) and lower losses for soybean (8.5% and 5.4%, respectively) using AOT40 (Accumulated hourly mean ozone above 40 ppb during daylight hours) for the year 2000.

Multivariate analysis of trends in soybean and maize yields in the USA that included a concentration-based ozone metric indicated that the ozone effect is dependent upon temperature and water availability (McGrath *et al*., 2015). This fits with our earlier conclusion that stomatal uptake-based risk assessment provides a better indication of ozone effects on yield than concentration-based assessments (Mills *et al*., 2018a). The McGrath *et al.* (2015) study indicated a greater sensitivity of maize to ozone than soybean, with maize and soybean yield losses due to ozone over a 31-year period averaging 10% and 5%, respectively. It is possible that our analysis under-estimated the effects of ozone on maize as our analysis was based on experimental data from 1981, 1985, 1991 and 1992. Since newer varieties of wheat and soybean are more sensitive to ozone than older varieties (Biswas *et al*., 2009; Osborne *et al*., 2016), then newer maize varieties may also be more ozone sensitive leading to larger effects. Partial derivative-linear regression analysis of heat and concentration-based ozone stress impacts on yield data in the USA and Europe indicated similar areas at risk from ozone for maize and soybean to our study, but fewer areas at risk for wheat (Tai & Val Martin, 2017). The latter may reflect that their study omitted soil moisture as a confounding factor, which our earlier modelling study indicated is a particularly important factor in modifying ozone uptake (Mills *et al*., 2018a). These two statistical studies have confirmed that factors other than ozone concentration need to be taken into account in analysing ozone effects on yield and have drawn attention to potential co-occurrence of heat and ozone stress effects on crop yield.

It is clear from our analysis that yield effects due to ozone are within the range of concern for other biotic and abiotic stresses. For example, extreme heat was estimated to have reduced national cereal production by 9-10% (1964 - 2007), with later droughts reducing yields by more than earlier droughts (13.7% for 1985 – 2007 compared to 6.7% for 1964 – 1984, Lesk *et al*., 2016). If we applied the percentage yield loss ranges used for ozone in YCS (Table 1) to these drought-induced yield losses, the YCS would be 2 or 3 depending on the time period used in the analysis. The YCSs for ozone are mainly in the same range as those predicted for wheat, maize and rice for global impacts of heat stress and aridity (scores 2-3, with rice-heat having a YCS of 1). Across all four crops, the areas predicted to be at the greatest risk of ozone effects on yield are predicted to be: SE NAM, S EUR, N SAS and E EAS, with parts of SAM also predicted to be at risk of yield loss for soybean. In some of these areas, ozone effects are predicted in areas also at risk from heat stress and to a lesser extent aridity, whilst co-occurrence with nutrient stress depended on the crop and tended to be most common in parts of EAS and SAM. Potential impacts on yield due to pests and diseases were predicted to be relatively high in many areas of the world, particularly in those at risk from ozone impacts in SAS and EAS.

Ozone impacts are predicted in areas where the largest gaps occur between actual and estimated potential yield, such as parts of SAM, SSA, EAS, SEA and SAS (Neumann *et al*., 2010). Here, yield gaps are already known to be widened by limitations in nutrient and/or irrigation availability (Mueller *et al*., 2012) and may be further widened by negative effects of ozone pollution. Indeed, in the same regions there has been a plateauing or decrease in the rate of yield increase in recent decades (Grassini *et al*., 2013; Ray *et al*., 2012). We suggest that ozone pollution could be contributing to this stagnation, and suggest below how crop tolerance of the pollutant could be improved by breeding or management (Section 4.2.3).

India was ~~, was~~ selected as a case study, as our analysis~~,~~ indicated that ozone pollution may be a particular problem in this country, adding to the existing multi-stress constraints on crop yield (Jaswal, 2014). National mean yield losses due to ozone were predicted to be 15.8% (soybean), 12.6% (wheat), 6.2% (rice) and 7.5% (maize) amounting to 12.6 Tg of lost yield. For wheat, our predicted mean yield loss in Uttar Pradesh of 16% was comparable to a mean 17% yield benefit from reducing the ambient ozone from 46 to 5 ppb (M7) by air filtration in field studies conducted from 2004 to 2008 at Varanasi in Uttar Pradesh (Rai *et al*., 2007, Sarkar *et al.*, 2010). Similarly, at a field site in Haryana, reduction in the M7 by filtration from 37 ppb to 6 ppb, resulted in a 16% yield benefit for wheat (Bhatia *et al*., 2011), which was similar to our state mean of a 15% yield reduction due to ozone.

Wheat yield losses were predicted to be highest in this study in the same regions of India as those predicted by Tang *et al*. (2013) in the first stomatal uptake-based risk assessment for the country. Our analysis, taking into account the added effects of soil moisture and irrigation usage, extended the region of highest ozone effects across the Indo-Gangetic Plain and including Uttar Pradesh and Bihar. Together with Haryana and Punjab, these states are considered to have the highest reductions in yield due to the combined effects of climate change and air pollution, with reductions as high as 50% being predicted in one concentration-based study (Burney & Ramanathan, 2013). Our multi-stress analysis confirmed that heat stress is ~~potentially~~ particularly important in this region (Lobell *et al.*, 2012). Site-specific analysis of the effects of future increases in temperature in 2030-2040, indicated that heat stress is likely to continue to reduce yields in the Indo-Gangetic Plain, especially under climate change (Asseng *et al.*, 2017). Given that from a food security perspective, it is crucial to reduce yield gaps in India, reducing ozone pollution and/or its effects could potentially provide beneficial additional yield in future climates.

In considering these comparisons, we are aware that in reality ozone will interact with the other stresses considered and integrated responses in growth and yield will occur. These interactions are generally thought to be determined by factors that might affect gas exchange or metabolic responses to stress. For example, limited water stress may reduce ozone uptake but as water stress becomes more severe, any protection afforded by reduced ozone uptake may be outweighed by drought-induced yield reductions. Additionally, these stresses are thought to impart similar defence mechanisms (Huymans *et al.*, 2017; Kangasjärvi *et al.*, 2005; Locato *et al.*, 2008). Whether multiple stresses induce additive or synergistic metabolic responses is open to question and we do not yet have the understanding or tools to be able to quantify these interactions. Nevertheless, through providing a first global assessment of where these stresses co-occur we have identified which stresses are most important across different global regions. This will help other researchers to identify threats and target future research needs to improve our understanding of responses to multiple stress conditions.

4.2 Options for reducing ozone impacts on crops

The analysis presented here has clearly shown that ozone impacts on yield are occurring in many areas of the world for four staple crops, and that in some regions the YCSs for ozone are as high or higher than for other biotic and abiotic stresses. Whilst these results highlight the ozone problem, we offer here some possible options for reducing ozone effects on crops that might help in closing the ozone yield gap.

4.2.1 Global effort to reduce ozone precursor emissions

The most obvious way of closing the ozone yield gap for crops is to substantially lower the anthropogenic emissions that lead to ozone pollution. Since ozone is a transboundary air pollutant – impacts of emissions in one country can impact on crops grown in countries many 100s and even 1000s of km away – efforts to reduce ozone need to be taken at both local and global scales. One study, using~~based on~~ ozone concentration–based metrics, indicated that 100% reductions in anthropogenic precursor emissions from NAM would reduce global yield losses due to ozone for the four crops in our study by between ca. 5% (rice) to ca. 80% (soybean), whilst a complete cut in precursor emissions from SEA would reduce global yield losses by between ca. 20% (soybean) and ca. 95% (rice) (Holloway *et al*., 2012). Whilst such dramatic cuts in ozone precursor emissions are highly unlikely for the foreseeable future, progress has been made in EUR and NAM, with emission cuts of ca. 40% for major ozone precursors such as NOx, VOC and CO being made between 1990 and 2013 (Maas & Grennfelt, 2016). These cuts have been associated with significant decreasing trends in the concentration-based metric AOT40 at 26% and 11% of monitoring sites in wheat growing areas of NAM and EUR, respectively over the period 1995-2014 (Mills *et al*., 2018b), although the dominant trend for EUR remains "no change". Over the same time period, increases in precursor emissions of 20-30% in other areas of the world, including by 50% in India and China have led to increases in ozone concentration in these regions (Maas & Grennfelt, 2016). For example, there has been a significant increase in ozone concentration at nearly 50% of wheat growing monitoring sites in EAS, with average annual increases in AOT40 at these sites being in the range 300 – 700 ppb h y^{-1} over the period 1995-2014 (Mills *et al*., 2018b).

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Our modelling results suggest~~predicts~~ that even with declining emissions in NAM, current yield losses due to ozone are in the range 5.3% (rice) to 15.5% (soybean), whilst for EAS with rising emissions, current yield losses are in the range 7.9% (rice) to 19.1% (soybean). With ozone concentrations predicted to continue to rise in EAS and SEA for at least the next 2-3 decades even with the most optimistic scenarios (Wild et al., 2012), and as these two regions are predicted to produce 80% of all global ozone precursor emissions by 2050 (Maas & Grennfelt, 2016), there would be considerable benefit for crop yield in the implementation of a concerted effort to reduce precursor emissions in these rapidly developing regions. Actions to reduce ozone are already being considered in some countries. For example, in China, three approaches are being introduced to reduce ozone concentrations: enforcing European standard V for diesel vehicle emissions; encouraging widespread use of electric vehicles; and discouraging private car use by improving public transport (Feng *et al*., 2015). Continued effort to reduce ozone is also needed in developed regions such as NAM and EUR as models predict that whilst efforts to reduce peak concentrations have been partially successful in reducing ozone concentrations in recent decades, a stabilisation in ozone concentrations in the next decade or two is likely to be followed by further rises in global background ozone concentration by 2050, primarily driven by increasing CH_4 emissions (Maas & Grennfelt, 2016).

Whilst reducing global ambient ozone concentrations remains a crucial long-term goal for reducing the ozone yield gap, approaches described below based on crop management and breeding are more likely to provide shorter-term solutions, with some having potential for implementation in the near future.

4.2.2 Exploiting existing varietal differences in ozone sensitivity

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Whilst the analysis presented here has confirmed that intraspecific variation in ozone sensitivity is clearly present for wheat, rice and soybean in experiments conducted over the last 30 - 40 years (Fig. 1, Table S3), of larger importance in the context of closing the ozone yield gap is the potential for selecting ozone tolerance amongst currently grown varieties. To assess this, ideally, varieties should be exposed to ozone under the same environmental conditions, allowing for realistic comparisons of effects on yield and assessments of variety by ozone interactions. Unfortunately, relatively few such experiments have been conducted with two or more varieties in the last decade. Those recent studies showing significant variety by ozone interactions, indicating scope for selecting the more ozone tolerant variety, include examples from SAS and EAS for rice (Akthar *et al*., 2010; Shi *et al*., 2009) and wheat (Feng *et al*., 2010, 2016; Singh *et al*., 2017; Zhu *et al*., 2011;), NAM for soybean (Betzelberger *et al*., 2010; Jiang *et al*., 2018) and maize (Yendrek *et al*., 2017); and EUR for wheat (Harmens *et al*., 2018). Further support for the potential benefits of selecting tolerant varieties is also provided by comparisons of yield in filtered air versus non-filtered air (Osborne *et al*., 2016; Pleijel *et al*., 2018). For example, in recent studies, reductions of ambient ozone concentration by filtration significantly increased the yield of ozone-sensitive soybean cultivars (PUSA 9712, PUSA 9814) by over 40% (Singh & Agrawal, 2011), rice cultivar Kirara 397 by over 20% (Frei *et al*., 2012) and wheat cv PBW 343 by 18-20% (Tomer *et al*., 2015).

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A modelling study has been conducted to highlight the potential for avoiding production loss in global wheat, maize and soybean by selecting crop varieties with lower than average sensitivity to ozone (Avnery *et al*., 2013). The variation in sensitivity among varieties was based on the experimental evidence from the large-scale US National Crop Loss Assessment Network (NCLAN) field studies conducted mainly during the 1980s (Heagle, 1989; Heck *et* *al*., 2013; Heck, 1989). Using a concentration-based method, the study showed that choosing crop varieties with ozone tolerance could improve global crop production by over 140 Tg in 2030, equivalent to a 12% increase. Although the older North American varieties may not represent current global variation, and some of the 1980s varieties are no longer used, the approach of Avnery *et al*. (2013) could be extended by conducting new screening experiments with a regional focus to inform farmer choice, modelling and breeding programmes and by using a stomatal uptake based modelling approach.

4.2.3 Breeding new varieties with multiple stress tolerance, including ~~of~~ ozone

The heterogeneity in variety response to ozone for soybean, wheat and rice (Fig. 1) has clearly shown ~~that there is clearly~~ the scope for breeding ~~of~~ ozone~~-~~-tolerant varieties, and an ideotype for an ozone tolerant crop has been defined here (Fig.7). Ideally, the improved ozone response must not compromise the yield potential or other required agronomic characteristics (e.g. resistance to diseases, shattering, and lodging). Since this study has also shown that ozone stress can typically co-occur with stress caused by heat, pests and diseases, and~~,~~ to a lesser extent~~,~~ aridity and nutrients, the breeding for ozone tolerance traits may cause potential synergies or trade-offs that also need to be considered (Fig. 7). Candidate traits for ozone tolerance were described in Section 3.~~4~~2.

Traditional breeding approaches such as pedigree selection require extensive screening of a large number of plants in multiple locations over extended periods of time (Frei, 2015). Whilst feasible, experimentally maintaining designated ozone concentrations on a sufficiently large scale required for breeding (e.g. in large scale FACE (free air concentration exposure) experiments) seems economically unviable. Therefore, molecular breeding approaches such as marker assisted selection (MAS) appear to be more promising. Phenotypic variation in traits associated with ozone tolerance can be evaluated in smaller-scale controlled ozone fumigation experiments and linked to genetic markers using mapping approaches, including bi-parental quantitative trait locus (QTL) mapping (Frei *at al*., 2008) and Genome-Wide Association Study (GWAS, Ueda *et al*., 2015). Theoretically, chromosomal fragments associated with ozone tolerance traits can then be introgressed into recipient varieties using marker assisted backcrossing without the need for large-scale fumigation experiments.

Although no large-scale marker-assisted breeding programs for ozone tolerance in crops have been conducted to date, proof of concept has been shown for ozone tolerant rice breeding lines carrying QTL for ozone tolerance (Chen *et al*., 2011; Frei *et al.*, 2008, 2010) that have a superior performance to the recipient varieties in terms of yield components (Wang *et al*., 2014) and grain quality (Jing *et al*., 2016). This example should encourage further breeding efforts in rice and other crop species, specifically targeting widely grown mega-varieties of crops grown in ozone-affected parts in the world. As an alternative strategy, traits contributing to ozone tolerance could be incorporated into existing crop varieties through genetic engineering. For example, rice and barley varieties engineered to contain enhanced levels of ascorbate have been engineered and showed enhanced tolerance to a variety of environmental stresses (Ali *et al*., 2018).

Physiological trait modelling could also be used to understand how different traits intended to confer tolerance for ozone, might influence crop physiology, growth and yield response under a range of environmental conditions and stresses.

4.2.4 Reducing ozone uptake by strategic limitation of irrigation application

Ozone impacts on crops could be reduced by partial stomatal closure induced by reduced irrigation, which could also save water use for irrigated crop production. In the rice growing countries, in response to the increasing water demands by other sectors than agriculture, alternate wetting and drying irrigation (AWD) has become popular in an attempt to reduce water usage and methane emissions (Bouman *et al*., 2007; Carrijo *et al*., 2017). This approach could also potentially be exploited to reduce ozone impacts on rice or other crops. A comparison of two studies conducted about 30 km apart in the same city of China suggests such a possibility. In Zhang *et al*. (2009), AWD with moderate water stress increased the growth and yield of rice while reducing stomatal conductance compared to continuously flooded crops, mostly resulting from a greater number of rice grains per panicle under AWD. Interestingly, at a nearby site, elevated ozone reduced rice yield arising from a decrease in the number of grains per panicle in two of the four varieties tested (Shi *et al*., 2009). This suggests that reduced ozone uptake could be an additional and unintended benefit of AWD for farmers. The potential benefits of the AWD approach require further study.

4.2.5 Fertilizer application to compensate for crop yield losses

Crop loss from ozone exposure could potentially be counteracted by increasing the fertilizer application rate (Cardoso-Vilhena & Barnes, 2001; Chen *et al*., 2011). However, in addition to the cost of fertilizer, recent analysis has indicated that this mitigation approach may be associated with an aggravation of other environmental problems. It has been shown that the nitrogen/protein yield of wheat is reduced by ozone at a certain level of nitrogen application and this applies also to other nutrients like phosphorus and potassium (Broberg *et al*., 2015; 2017). This means that the fraction of nitrogen applied which does not end up in the grain could enhance other environmental problems (Di & Cameron 2002; Mosier *et al.* 1998) such as nitrate leaching, conversion of fertilizer to N_2, emissions of N_2O and even NO, which promotes further ozone formation, as shown for pasture (Sánchez-Martín *et al*., 2017). Adding nitrogen fertiliser to compensate for reductions in yield may also inadvertently increase the stomatal conductance of leaves of crop plants thereby increasing ozone uptake and subsequent damage (Mills *et al*., 2016).

4.2.6 Chemical protection against ozone damage

There is scope for investigating the benefits of chemical protection against ozone damage. The most successful antiozonant applied so far has been ethylenediurea (N-[2-(2-oxo-1-imidazolidinyl)ethyl]-N'- phenylurea), abbreviated to EDU, first described by Carnahan *et al.* (1978). This chemical is usually applied as a foliar spray or soil drench, and has been used extensively in experiments and biomonitoring programmes to reduce the effects of ozone pollution, including preventing visible ozone injury on the leaves and growth and yield reductions (Agathokleous *et al*., 2016; Feng *et al*., 2010; Jiang *et al*., 2018; Manning *et al*., 2011; Pandey *et al*., 2015; Rai *et al*., 2015). A meta-analysis suggested that the antiozonant activity of EDU is biochemical rather than biophysical (Feng *et al*., 2010). Recent results showed that EDU has no negative effects on plants at low O_3 concentration, but increases the crop yield at high O_3 concentration (Ashrafuzzaman *et al*., 2017). Whilst EDU has not yet been evaluated for application at field scale, concerns have been raised about potential toxicity to aquatic plants (Agathokleous *et al.,* 2016) and more research is needed to determine if this chemical could be extensively used.

Other chemical protectants against ozone could be developed from a knowledge of plant hormonal control of stomatal functioning and stress perception (Wilkinson *et al*., 2011), and could potentially provide multi-stress tolerance such as combined tolerance of ozone, heat and drought stress. All three of these stresses induce synthesis of the crop stress hormone, ethylene, and chemicals that inhibit ethylene perception such as 1-MCP (1-methylcyclopropene) have the potential to reduce their effects (Wilkinson & Davies, 2010; Wagg, 2012). Anti-transpirants that reduce stomatal aperture could also reduce ozone effects by reducing ozone uptake in some species. However, there is a growing body of knowledge that chronic exposure to ozone reduces the ability of stomata to respond to abscisic acid under drought conditions, potentially leading to more rather than less ozone uptake (Mills *et al*., 2016; Wilkinson and Davies, 2009, 2010). An alternative chemical protection approach has also been explored experimentally. Di-1-*p*-methene, a natural terpenic polymer derived from the resin of pine trees that mimics isoprene emissions from plants, has been shown to reduce visible injury in Pinto beans after exposure to 150 ppb of ozone for 4h (Francini *et al*., 2010).

So far, chemical protection has only been explored at the experimental scale. Given the growing evidence presented here and elsewhere of the negative effects of the pollutant at the global scale, there is considerable scope for developing a chemical protectant against ozone damage, especially if it provides cross-tolerance against other co-occurring stresses.

5. Conclusions

This global-scale study shows that ozone is a very important stress, limiting yields of key crops and comparing in importance with other key stresses. For example in India, where food security concerns are particularly pressing, the mean YCS for effects of ozone on wheat of 3 falls in between those for nutrients and aridity (score 2) and for pests and diseases and heat stress (score 4). Globally, we show that the largest effects of ozone are often in areas already challenged by other stresses such as pests and diseases and heat, particularly in EAS, SAS and SEA. The global mean ozone yield gaps of 4.4 – 12.4 % identified here add up to 227 Tg of lost yield for soybean, wheat, rice and maize. We speculate that, ozone could at least partially, account for the unexplained yield gaps and stagnation in yield improvement seen in many areas of the world in recent years. Thus, international effort to reduce ozone pollution on a global scale would bring clear benefits for agriculture as well as for other types of vegetation, health, materials and climate change (Simpson et al., 2014). However, it is likely to take many decades to achieve the required emission reductions, which is the only long-term solution for reducing the problems caused by tropospheric ozone. Meanwhile, the global population is expected to grow significantly, which together with increasing real income levels, will see increasing demands placed on food production (Tilman *et al*., 2011).

Several interim solutions for closing the ozone yield gap have been outlined in this paper. These include: testing of current varieties for ozone sensitivity and selection of the most tolerant; crop breeding for multiple stress tolerance, including ozone; implementation of protective watering regimes such as AWD; and the development of chemical protection against ozone damage. Given the severity of ozone effects on staple food crops in areas of the world that are also challenged by other stresses, we recommend increased attention to the benefits that could gained from taking mitigating action to reduce the ozone yield gap.

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Data Accessibility Statement:

National-scale data are provided in the Supporting Information. Grid square values for ozone metrics for the LRTAP region can be downloaded from http://www.emep.int/mscw/mscw_data.html.

Statement on Competing Interests: none

The authors declare no competing interests.

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Figure legends

Fig. 1. Response functions for (a) soybean, (b) wheat, (c) rice and (d) maize derived from published data using the growing season ozone (7h mean, M7 in ppb) in the experiments. Data points are presented per cultivar/variety, with sources of data provided in the Supporting Information (Table S3). The response functions are: Soybean, RY = -0.0050x + 1.001 (r^2 (adj) = 0.625, p<0.001); Wheat, RY = -0.0048x +0.96 (r^2 (adj) = 0.547, p<0.001); Rice, RY= -0.0021x +0.987 (r^2 (adj) =0.347, p <0.001); and Maize, RY = -0.0031x + 1.03 (r^2 (adj) = 0.617, p <0.001).

Fig. 2: The global effects of five biotic and abiotic stresses on soybean. All data are presented for the 1 x 1° grid squares where the mean production of soybean was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S4 provides all country and regional means.

Fig. 3: The global effects of five biotic and abiotic stresses on wheat. All data are presented for the 1 x 1° grid squares where the mean production of wheat was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S5 provides all country and regional means.

Fig. 4: The global effects of five biotic and abiotic stresses on rice. All data are presented for the 1 x 1° grid squares where the mean production of rice was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S6 provides all country and regional means.

Fig. 5: The global effects of five biotic and abiotic stresses on maize. All data are presented for the 1 x 1° grid squares where the mean production of maize was > 500 tonnes (0.0005 Tg). (a) Presents the effects of ozone on crop production (thousand tonnes or 0.001 Tg per grid square), and (b) the percentage yield loss due to ozone, averaged for the period 2010-2012. In (d) to (h), the Yield Constraint Score (YCS) is presented per grid square on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1) for ozone, pests and diseases, nutrients, heat stress and aridity, respectively, whilst (c) is the total YCS (YCS_{all}) calculated from the sum of each of these per grid square. The regional impacts are summarised in Table 2 for the five highest producing regions and Table S7 provides all country and regional means.

Fig. 6: Yield Constraint Score (YCS) for five constraints on the yield of (a) wheat and (b) rice in the five Indian states with the highest production per crop. The bars represent the mean YCS per 1 x 1°grid square per state on a scale of 1 to 5, where 5 is the highest level of stress (see Table 1), rounded to the nearest integer. Note: The YCS for pests and disease is only available at the National Scale for India (score 4 for wheat and 5 for rice) and is presented here for information.

Fig. 7: An ideotype for an ozone-tolerant crop. '+' indicates where there would be a benefit for other stresses of improving tolerance to ozone for the trait, whilst '-' indicates a trade-off, and '0' is no effect.