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Ozone pollution will compromise efforts to increase global wheat production

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Running head: Ozone compromises wheat yield goals

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Abstract

Introduction of high-performing crop cultivars and crop/soil water management practices that increase the stomatal uptake of carbon dioxide and photosynthesis will be instrumental in realizing the United Nations Sustainable Development Goal (SDG) of achieving food security. To date, however, global assessments of how to increase crop yield have failed to consider the negative effects of tropospheric ozone, a gaseous pollutant that enters the leaf stomatal pores of plants along with carbon dioxide, and is increasing in concentration globally, particularly in rapidly developing countries. Earlier studies have simply estimated that the largest effects are in the areas with the highest ozone concentrations. Using a modelling method that accounts for the effects of soil moisture deficit and meteorological factors on the stomatal uptake of ozone, we show for the first time that ozone impacts on wheat yield are particularly large in humid rain-fed and irrigated areas of major wheat-producing countries (e.g. USA, France, India, China and Russia). Averaged over 2010-2012, we estimate that ozone reduces wheat yields by a mean 9.9% in the northern hemisphere and 6.2% in the southern hemisphere, corresponding to some 85 Tg (million tonnes) of lost grain. Total production losses in developing countries receiving Official Development Assistance are 50% higher than those in developed countries, potentially reducing the possibility of achieving UN SDG2. Crucially, our analysis shows that ozone could reduce the potential yield benefits of increasing irrigation usage in response to climate change because added irrigation increases the uptake and subsequent negative effects of the pollutant. We show that mitigation of air pollution in a changing climate could play a vital role in achieving the above-mentioned UN SDG, whilst also contributing to other SDGs related to human health and wellbeing, ecosystems and climate change.

Introduction

Tropospheric (or ground-level) concentrations of ozone, a photochemically-produced secondary pollutant for which the precursors include oxides of nitrogen, volatile organic compounds and carbon monoxide (Simpson *et al.*, 2014), are already high in many crop-growing areas of the world, e.g. in North America, Europe, and South and East Asia (Cooper *et al.*, 2014; Mills *et al.*, submitted; Wild *et al.*, 2012). Concentrations are increasing rapidly in developing countries and are predicted to continue to increase in coming decades unless suitably ambitious measures are taken to cut precursor emissions (Cooper *et al.*, 2014; Wild *et al.*, 2012). Ozone damages sensitive crops such as wheat, rice and soybean (Grünhage *et al.*, 2012; Mills *et al.*, 2007) by diffusing into the leaves through the stomatal pores and reacting with biomolecules inside the leaf to form reactive oxygen species, thereby triggering metabolically expensive defense mechanisms, promoting leaf senescence and diverting resources away from growth and seed production (Ainsworth, 2016). Despite this, developing ways to mitigate the damaging effects of ozone pollution has attracted little attention from agronomists and policy makers investigating ways to increase yields in our changing climate. As well as quantifying the effects of ozone on yield in different climatic regions of the world, we investigate how some management and breeding approaches may actually exacerbate the effects of ozone by promoting greater ingress of the pollutant into leaves, thus leading to increased yield losses on a global scale. We focus on quantifying the impacts of ozone on the yields of wheat, a crop that provides 20% of human dietary protein and calorific intake globally (Shiferaw *et al.*, 2013).

In striving to attain the United Nations Sustainable Development Goal 2 (UN SDG2) of “ending hunger, achieving food security and improved nutrition, and promoting sustainable agriculture” by 2030 (UN SDG, 2016), it is anticipated that considerably greater production of grain crops such as wheat will be required (International Food Policy Research Institute, 2016). With global wheat demand doubling since 1980 and yields stabilizing in many wheat producing countries (Brisson *et al.*, 2010; Shiferaw *et al.*, 2013), a multi-disciplinary approach is needed if substantial increases in production are to be achieved in coming decades (Hawkesford *et al.*, 2013). Changes in crop management such as increasing water availability through water harvesting, improved irrigation efficiency and expanding irrigation infrastructure (Balkovič *et al.*, 2014; Jägermeyr, *et al.*; 2016; Mueller *et al.*, 2012) could provide substantial progress towards closing the gap between maximum possible yields and actual yields in the field. Whilst such approaches have the benefit of increasing stomatal uptake of CO₂ thereby enhancing photosynthesis (Roche, 2015), a simultaneous increase in the uptake of ozone (Ainsworth, 2016; Wilkinson *et al.*, 2012) might result in greater negative impacts of ozone and counteract some of the benefits. Others are focusing on increasing wheat yield via breeding programmes targeted at desirable traits such as higher photosynthesis, greater photosynthate partitioning to the seeds and higher nutrient use efficiency, or increased tolerance to biotic and abiotic stresses such as fungal diseases, heat and drought stress (Hawkesford *et al.*, 2013; International Food Policy Research Institute, 2016; Shiferaw *et al.*, 2013). Currently, ozone tolerance is not yet included as a desirable

trait in crop breeding schemes (Ainsworth, 2016; Frei, 2015; Lobell & Gourdj, 2012). Indeed, recently released high-yielding wheat varieties tend to be more sensitive to ozone than the older varieties, in part explained by their higher stomatal conductance (Biswas *et al.*, 2009).

Earlier studies using dose–response relationships based only on the atmospheric concentration of ozone simply predict that the highest effects are likely in the areas with the highest ozone concentrations. Such studies indicated that in the year 2000, ozone pollution reduced global wheat yield by 12–15%, with mean losses in the range of 16–27% in countries of South and East Asia (Avnery *et al.*, 2011a; Teixeira *et al.*, 2011; Van Dingenen *et al.*, 2009). One follow-on study predicted that these global losses would increase to 25.8% by 2030, with 44.4% and 25.7% losses in South and East Asia, respectively (Avnery *et al.*, 2011b). However, these studies did not take into account factors affecting the stomatal uptake of ozone, which casts some doubt on the accuracy of their assessments of global wheat yield losses and the spatial distribution of these losses. Since stomatal conductance and ozone flux differ significantly between humid and arid climates even when concentrations are the same (being higher in the former), marked differences might be anticipated in estimates of the spatial distribution of ozone impacts derived using a concentration-based methodology versus a stomatal uptake-based methodology. We now know that the field effects of ozone are far better correlated with stomatal ozone uptake than with ozone concentration (Mills *et al.*, 2011), and a stomatal uptake–yield effect relationship has been developed for wheat using data from experiments conducted in several countries (Grünhage *et al.*, 2012).

A recent meta-analysis investigated the effects of ambient ozone on wheat by comparing yields in field-based open-top chambers (OTCs) ventilated with ambient air, with those in OTCs ventilated with filtered air that reduced the ozone concentration by on average 62%, effectively achieving pre-industrial concentrations (Pleijel *et al.*, 2018). Based on results from 33 experiments (from 9 countries, 3 continents using 17 cultivars plus one set of 4 cultivars), the average yield loss associated with reducing the mean ozone concentration from 35.6 to 13.7 ppb was 8.4%, with the starch and protein yield reduced by 10.9% and 6.2%, respectively. In the same meta-analysis, relative grain yield loss increased linearly as ozone concentration increased, with the highest losses being in the 20 - 30% range for some sites in India, China and the USA (calculated from supplementary data in Pleijel *et al.*, 2018). Despite such compelling field evidence of the negative effects of ozone on crop yields, most statistical and process-based modelling of future crop yields are not yet including the impacts of current or predicted future ozone pollution (Challinor *et al.*, 2009; Emberson *et al.*, 2018, Lobell & Asseng, 2017; Lobell & Gourdj, 2012).

We undertook this study to highlight the potential yield gains that could be made on the global scale from either reducing ozone concentrations and/or developing crop management or crop breeding approaches to reduce the negative effects of the pollutant. We consider effects in major exporters such as the USA, France, Canada and Australia as well as in countries such as India, China, and Pakistan that have a high dependency on national crop production and imports to feed rapidly expanding populations (FAOSTAT). Our results are also considered separately for developed countries (DC) and developing countries receiving Official Development Assistance (ODA) (OECD, 2017). For the first time, we quantify the potential additional negative effects of ozone that could develop if irrigation is used to offset the negative effects of current and future rainfall shortages. In so doing, we tested the hypotheses that: (i) total wheat production losses due to ozone are higher in developing than developed countries; (ii) the largest impacts of ozone in major wheat producing countries occur in humid climates or in drier regions with high irrigation usage; and (iii) future strategies to increase wheat yield and help feed the growing global population by increasing crop stomatal conductance, such as through crop breeding and additional irrigation, may exacerbate the negative effects of ozone pollution, especially in drier climates. We have used an empirical approach to test these hypotheses, involving spatial modelling of the cumulative stomatal uptake of ozone and concentration-based metrics for crop relevant time periods and application of dose-response relationships derived from field-based experiments to determine the extent of effect on yield per $1 \times 1^\circ$ grid square.

Materials and Methods

Agricultural data

Modelled global wheat production data was downloaded from the GAEZ (Global Agro-Ecological Zones) database which has been developed by the Food and Agricultural Organisation of the United Nations (FAO) and the International Institute for Applied Systems Analysis (IIASA) to assess agricultural resources. Modelled values are derived in the database from the downscaling of FAO national production data, crop distribution data and AEZ crop suitability layers, for each country. Irrigated and rain-fed wheat production data for the year 2000 in Mg (tonnes) per cell was acquired for this study from the GAEZ data portal v.3 in raster format at 5 arc minute (0.0833°) spatial resolution (Illustrated in Supporting Information, Fig. S1a). A global $1^\circ \times 1^\circ$ grid was created using ArcGIS v. 10.1 (Environmental Systems Research Institute, Redlands, CA, USA) and wheat production was summed per cell. To estimate production per grid cell in 2010 – 2012 from the year 2000 grid cell data, a conversion factor was derived for each country by dividing the average total wheat production over the period 2010-12 by that from the 1999-2001, using country totals downloaded from FAOSTAT. Only cells where the summed wheat production was >500 Mg were included.

Global ozone concentration and stomatal uptake

The EMEP MSC-W (European Monitoring and Evaluation Programme, Meteorological Synthesising Centre – West) chemical transport model, version 4.8 (Simpson *et al.*, 2007, 2012, 2015) was used to model ozone concentration and stomatal uptake on a global scale at a $1^\circ \times 1^\circ$ resolution. This 3-D model is used for air quality assessments and has 20 vertical layers extending from the ground to 100 hPa, using terrain-following coordinates, and makes use of a chemical scheme involving 137 reactions. Meteorological data, including a soil-moisture index (SMI), are taken from the European Centre for Medium Range Weather Forecasting Integrated Forecasting System (ECMWF-IFS) model. The SMI is calculated from the modelled soil water content and the difference between the field capacity (FC) and permanent wilting point (PWP) of the vegetation at a soil depth of 0.28 to 1 m (Simpson *et al.*, 2012).

Calculations of stomatal ozone-uptake make use of the DO₃SE (Deposition of Ozone for Stomatal Exchange) stomatal conductance algorithm that is dependent on variables such as temperature, irradiance, vapour pressure deficit, and soil moisture (Büker *et al.*, 2012; Emberson *et al.*, 2001; Simpson *et al.*, 2007, 2012, 2015) in conjunction with an ecosystem specific calculation of vertical ozone profiles down to canopy surface (Simpson *et al.*, 2012, 2015). The 90 day (d) 7h mean ozone concentration (M7, mean of 0900 to 1600), 90 d AOT40 (accumulation of ozone hourly mean concentration over 40 ppb during daylight hours) and 90 d POD₃IAM (Phytotoxic Ozone Dose above $3 \text{ nmol m}^{-2} \text{ s}^{-1}$, model adapted for large-scale Integrated Assessment Modelling) was calculated from modelled hourly stomatal uptake and accumulated during daylight hours. The latter using the parameterisations defined in the Long-range Transboundary Air Pollution (LRTAP) Convention's Modelling and Mapping Manual (LRTAP Convention, 2017). POD₃IAM, M7 and AOT40 were modelled for climatic zone specific 90 d periods (described later) for 2010, 2011 and 2012 and averaged for this study. M7 and AOT40 were chosen for comparison with POD₃IAM because they have been widely used in previous concentration-based global and regional impact assessments (e.g. Avnery *et al.*, 2011 a, b; Tang *et al.*, 2013; Van Dingenen *et al.*, 2009) and AOT40-based critical levels are applied in the 51 Parties (countries) of the LRTAP Convention (LRTAP Convention, 2017).

Dose-response relationships

To determine the effects of POD₃IAM, M7 and AOT40 on wheat productivity, response functions were derived for the effect of each metric on wheat yield based on experimental data from open-top chamber experiments conducted in the field in Finland, Sweden, Belgium and Italy (described in Grünhage *et al.*, 2012) (Fig. S2). Ozone concentration metrics were derived for the hourly mean data for each experiment, whilst POD₃IAM was estimated from air humidity (vapour pressure deficit), temperature, solar radiation and soil moisture content using the DO₃SE model stomatal algorithm (LRTAP Convention, 2017). The parametrization

of the wheat stomatal uptake model was tested with respect to observed data (Grünhage *et al.*, 2012). To derive the response functions, data were first standardized for each set of experimental data by linear regression of the yield against the metric used; this function was then used to determine the yield at zero value for the metric (Fuhrer, 1994). The latter was then used to calculate relative yield (RY), with zero POD₃IAM, AOT40 or M7 having an RY of 1, and negative effects of ozone treatments having an RY of <1. Datasets were combined across experiments by plotting all RY against the treatment values for each metric, and linear regression was used to derive yield-response functions for POD₃IAM, AOT40 and M7 (Fig. S2).

Global maps of wheat yield loss

Using the global 1° x 1° grid wheat production map aligned with the EMEP grid, each 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). These climatic zones are based on the classification of the Intergovernmental Panel on Climate Change (IPCC, 2006), with zones defined using a set of rules, which includes annual mean daily temperature, total annual precipitation, total annual potential evapotranspiration and elevation. Cells including more than one climatic zone were assigned to the zone covering the majority of the cell. The spatial distribution of the climatic zones is illustrated in Fig. S1b.

A 90 d period for accumulation for each metric was derived for each climatic zone and hemisphere combination and assigned to each 1° x 1° cell (Table S1). The time period was back projected from two weeks before harvest and includes the ozone sensitive period between anthesis and end of grain fill (Soja *et al.*, 2000) with the time period reflecting the most common growth cycle for that climate (e.g. for winter rather than spring wheat in NW Europe). The harvest date per climatic zone was established by overlaying maps produced by the USDA Major World Crop Areas and Climate Profiles (MWCACP) with climatic zones, and checking data with a previous study (Sacks *et al.*, 2010) and web-based national sources for representative countries per climatic zone. Some climatic zones were merged for simplification and sub-divisions were added for cool temperate climates 0 – 30° south and > 30° south to account for variations in growth cycles for climates that span a large geographical area.

Percentage yield loss and production loss was calculated per grid cell relative to zero POD₃IAM or AOT40 to represent a clean air situation with ozone concentrations reduced to close to pre-industrial levels (Cooper *et al.*, 2014; Royal Society, 2008), or 25 ppb M7 to facilitate comparison with an M7-based earlier study (Van Dingenen *et al.*, 2009). For POD₃IAM, effects were determined separately per 1° x 1° grid cell using (i) POD₃IAM values that were weighted by the proportion of irrigated and rain-fed wheat production in the cell and (ii) POD₃IAM values representing full use of irrigation by assuming that soil moisture was not limiting to ozone uptake.

The impacts of ozone were analyzed by country, climatic region and countries assigned by the Organisation for Economic Co-operation and Development (OECD) Development Assistance Committee (DAC) into the following categories receiving Official Development Assistance (ODA): Least Developed Countries (LDC); Other Low Income Countries (OLIC), Lower Middle Income Countries and Territories (LMIC); and Upper Middle Income Countries and Territories (UMIC). The countries included in these categories are listed in OECD (2017), with their designation indicated in Table S2. For comparison, effects of ozone on wheat yield are also described for developed countries (DC), assumed here to be all other countries that are not included in the ODA recipients list.

Evaluation of modelled ozone concentration and stomatal uptake

EMEP model results were evaluated against observational data from the Global Atmosphere Watch (GAW) stations (Schultz *et al.*, 2015). In order to obtain relevant comparisons for ozone at rural locations (and not mountain tops), the data were tested by two criteria: (i) relative height (hRel), and (ii) a diurnal variation index (DVI). The hRel calculation used an estimate of the height of the station relative to the height of the lowest terrain within 5 km radius (following Loibl *et al.*, 1994; Klingberg *et al.*, 2012) using global topographic data from the ETOPO1 database (Amante & Eakins, 2009). The DVI reflects the fact that mountain-top stations show hardly any variation from day to night (Klingberg *et al.*, 2012) and was derived by first calculating the average concentration for each hour of the day over the year, and then calculating the ratio of the maximum mean-hourly value to the daily means. We used the criteria that hRel should be less than 100 m, and DVI greater than 1.04. Modelled ozone concentrations were also tested against time series for measured concentrations at representative GAW stations from sites in Argentina, Canada, Germany, Ireland, Japan and the USA. We were unable to test our analysis for countries such as China or India because of the lack of suitable publically available climate and ozone data.

To test the validity of the stomatal uptake component of the model, spatial variation in POD_3IAM was compared with variation in the ratio of actual evapotranspiration (AET) to potential evapotranspiration (PET) across the US. The rationale for this is that stomatal conductance is a key regulator of land–atmosphere gas exchange, controlling fluxes of both ozone and water vapor (and thus AET). Satellite data was compiled using the annual raster global datasets of AET and PET from the MODIS Evapotranspiration Data Product MOD16A3 (Mu *et al.*, 2011) available from the NASA’s Numerical Terradynamic Simulation Group (NTSG) server. These data were calculated using an algorithm based on the Penman-Monteith equation (Monteith, 1965). AET was divided by the corresponding PET value for each cell of the 1 km²-grid for each of the years 2010, 2011 and 2012 where the land area dedicated to wheat production exceeded 10%. Thereafter, we obtained the arithmetic annual mean of the three years per cell. The area-weighted mean AET/PET for each of the 1° x 1° cells previously created as described above for wheat production was calculated. This was plotted against the three year mean POD_3IAM (including soil moisture

limitation) divided by M7 for the 90 d accumulation period to provide an indication of the integrated growing season stomatal conductance, without the confounding influence of the ozone concentration. A similar analysis was conducted using AET/PET divided by leaf area index (LAI), with spatial LAI data taken from the MODIS Leaf Area Index/FPAR product (Myneni *et al.*, 2015). This was done to separate the influence of LAI from that of stomatal conductance, since both control AET.

Results

Model validation

Before consideration of the results of this analysis, three approaches were used to test the validity of the stomatal uptake modelling. Firstly, time-series of the daily maximum hourly mean ozone concentration (Dmax) data were compared with modelled data for sites from the global GAW network to illustrate the model performance during the times of peak ozone each day. Over the course of a year, the EMEP model captured the spatial and temporal variations across the regions, including the seasons with higher concentrations and longer ozone episodes (Fig. S3). Secondly, we tested how well the model estimated Dmax and M7 at all of the GAW stations for 2012, which satisfied the hRel and DVI criteria (Fig. S4). The red points and lines are for the complete data-set whilst the black data and lines are for the same data after exclusion of outliers. This procedure was followed since all global data-sets are subject to errors in the data reporting and collection, and not all sites are equally representative for their surrounding area. Fig. S4 shows a very good correlation between the modelled and observed values (r^2 ranges from 0.88 to 0.95), with rather little scatter considering the large geographical network. Thirdly, we tested how well the model estimated stomatal conductance by focusing on data for the wheat growing areas of the USA for which satellite data for the ratio of AET to PET is available. We found that there is a strong correlation between AET/PET and $POD_3IAM/M7$ ($r^2 = 0.63$, $p < 0.001$, Fig. S5). Also AET/(PET x LAI) was significantly, but less strongly, correlated with $POD_3IAM/M7$ ($p < 0.039$, data not shown). The AET/(PET x LAI) index reflects stomatal control of AET since the additional influence of LAI has been accounted for. From these results, we gained confidence that our new modelling approach for estimating the effects of ozone on yield based on stomatal uptake could be used at the global scale.

Comparison of current yield effects using AOT40, M7 and POD_3IAM .

For comparison with earlier studies, we first examined the extent to which the magnitude and spatial variation of yield loss estimates based on the stomatal uptake approach used here differ from those based on the concentration of ozone using AOT40 and M7, using data averaged for 2010-2012. The spatial distributions for AOT40 and M7 are fairly similar, with the highest values being in the western half of the USA, northern half of India and Pakistan, northern and western China and parts of Turkey (Fig. 1 a, b). However, the grid square

values for POD₃IAM, weighted by the proportion of irrigation in use, provide a different spatial pattern, with clearly defined areas of high POD₃IAM in the eastern half of the USA, Mediterranean areas, central Europe, parts of India, Pakistan and western and southern China (Fig. 1 c).

Globally, the mean percentage yield losses in the wheat growing areas are 13.8, 13.4 and 9.4% for AOT40, M7 and POD₃IAM, respectively (Table S2). Thus, loss estimates suggested by the older concentration-based methods are similar and more than one third higher, on average than those obtained from our stomatal uptake modelling using POD₃IAM. In the top wheat producing countries, China and India, yield loss estimations using AOT40 of 25 and 21.5% respectively are around double those estimated using POD₃IAM (11.7 and 12.2%, respectively), whilst in other high wheat producing countries such as Russia and Canada, percentage yield losses are similar using both metrics (averaging 10.6% and 7.4%, respectively).

There are also spatial differences within countries of the areas estimated to have the highest losses determined using the different approaches (Fig. 1). For example, using concentration-based approaches, estimated yield losses in the USA are 17.2–19.8% in western warm-temperate-dry climates (AOT40 of 18.5 ppm h, M7 of 58.1 ppb) but only 14.8–15.5% in eastern warm-temperate-moist climates (AOT40 13.8 ppm h, M7 53.5 ppb). In contrast, the stomatal uptake approach estimates that conditions are less conducive to ozone uptake in the hotter, drier western areas, where ozone concentrations are higher but irrigation is used on less than 50% of fields, resulting in a lower mean yield loss of 8.2%. In the wetter eastern states, where more humid conditions promote plant ozone uptake, yield loss is estimated to be 13.9%. Similarly, using concentration-based metrics, high yield losses are estimated in central states of India (range 17.5% to 55% yield loss, Fig. 1 d, e). However, estimations based on stomatal uptake shift the region of greatest risk to the northern and north-eastern states of India, such as Uttar Pradesh and Bihar, where climatic conditions and abundant ozone pollution are highly conducive to ozone uptake, inducing yield losses in the range 12.5–17.5% (Fig. 1f).

On a global scale, production losses estimated using POD₃IAM (85.2 Tg, where one Tg = 1 million tonnes) are smaller than estimated using M7 (122 Tg) or AOT40 (127 Tg) (Table S2, Fig. 2, Fig. S6). These differences are particularly large for countries such as India and China, where production losses of 13 and 18 Tg respectively are estimated using POD₃IAM, compared to losses in the range 20 – 21 Tg and 32 – 39 Tg, respectively for concentration-based indices.

Yield and production losses estimated using POD₃IAM (2010-2012)

Our analysis also shows that the mean estimated yield losses in the Northern Hemisphere (NH, 9.9%) are greater than those in the Southern Hemisphere (SH, 6.2%), where ozone concentrations are lower (Fig. 1, Table S2). The highest mean percentage yield losses are associated with warm-temperate-moist (12.5%), tropical-moist (12.6%) and tropical-wet (17.2%) climates of the NH (Fig. 3a). Drier NH climates have relatively similar, but smaller yield losses, including for boreal-dry (10.1%), cool-temperate-dry (9.3%) and warm-temperate-dry (10.1%) climates. In the SH, the greatest losses are found in tropical climates, with mean yield losses being 7.1, 9.2 and 6.7 % in tropical-dry, tropical-moist and tropical-montane climates, respectively (Fig. 3b). Here, the lowest percentage yield losses are in the cool-temperate-dry and cool-temperate-moist climates (both with a mean of 2.1%), with intermediate yield losses in warm-temperate-dry and warm-temperate-moist climates of 4.9 and 6.3 %, respectively.

Globally, 39.7% of the lost production occurs in DC (Developed Countries), 31.8% in UMICs (Upper Middle Income Countries and Territories) and 26.8% in LMICs (Lower Middle Income Countries and Territories, Fig. 4). Mean percentage yield losses are highest for LMIC at 10.7%, followed by 9.5% for UMIC and 9.1% for DC. Yield losses in the highest wheat producing LMICs of India, Pakistan, Egypt and Ukraine are estimated to be 12.2, 9.5, 10.9 and 12.9 % respectively amounting to 20.4 Tg of lost grain (Table S2). Of the highest wheat producing UMICs, mean yield losses due to ozone are estimated to be 11.7%, 8.2%, 9.1% and 6.0% for China, Turkey, Kazakhstan and Argentina, respectively, totaling 22.2 Tg of lost production.

This analysis also shows that some of the highest percentage yield losses occur in the top wheat producing countries of the world such as China, India, USA and Russia (11.7%, 12.2%, 10.1% and 10.8%, respectively), which corresponds to 45 Tg of lost grain yield (Fig. 5). For these countries, the greatest within-country production losses are in the warm-temperate-dry climatic regions of China (12.8 Tg) and the tropical-dry-climates of India (8.8 Tg), where > 80% of the wheat is irrigated (Fig. S7). In the USA and France, the greatest yield losses are in areas with warm-temperate-moist climates where irrigation usage is 5.1 and 24.7% respectively, whilst in Russia and Germany the largest losses are in cool-temperate-moist climates with relatively little irrigation usage (7.7 and 7.8%, respectively).

Additional yield losses associated with increased use of irrigation

We found that the ozone penalty for yield is intensified in all climatic zones where a high proportion of wheat is currently rain fed, if fully irrigated conditions are simulated to provide non-limiting soil moisture conditions (Fig. 6). The total additional production losses would be highest in DCs and UMICs (totaling 1.8 and 1.2 Tg, respectively, Fig. 7). In LMICs, total

additional losses would be lower at 0.3 Tg, in part reflecting the already high proportion of irrigated production in the two highest wheat producing LMICs, India and Pakistan (73.8 and 86.4 %, respectively).

Additional yield losses would be greatest in drier climates where soil moisture is limiting ozone uptake, especially where irrigation use is currently < 50% (Fig. 8, S8). For example, nearly one Tg of additional yield losses would occur in the drier temperate climates of the USA (Fig. 8), three-quarters of which would be in central and western states (Fig. 6). Even a small, 15% increase in irrigation in warm temperate dry climates of China would entail a further 0.2 Tg of lost grain due to ozone (Fig. 8). Added losses associated with increased irrigation would also occur in temperate and tropical dry climates of other major producers such as Russia, Argentina, Australia, India, and Kazakhstan, where irrigation is currently used only sparingly. Furthermore, yield losses due to ozone would increase by up to 50% if irrigation use were to be expanded in countries where food demand outweighs supply and substantial yield increases will be needed to mitigate import dependence (e.g. Ethiopia, Tanzania, South Africa, Fig. 6 and 8). The intensification of ozone effects due to increased irrigation usage would be most pronounced in dry climates where less than 25% of the grid square is currently irrigated, with small added yield penalties still occurring where irrigation usage is 75 - 100% (Fig. S8). In contrast, added losses due to enhanced irrigation in moist climates would only be pronounced where less than 10% of the grid square is currently irrigated.

Discussion

By modeling the stomatal uptake of ozone on a global scale, our results estimate that the current global wheat yield penalty from ozone pollution is a mean of 9.4% for 2010-2012. This corresponds to 85 Tg of lost grain per year, or losses of \$24.2 billion dollars per year at the mean global market price of \$285 per Mg for 2010-12 (FAOSTAT). The only studies available that could be used to validate this figure show that the benefit of reducing ozone to pre-industrial levels by filtration in field-based OTCs is in the range 8 – 9 %, averaged over 9 countries (Pleijel, 2011; Pleijel *et al.*, 2018). As these countries represent North America, Europe and Asia, it gives confidence that the mean global yield loss estimated in our study was comparable to the yield benefit found by reducing the ozone concentration at crop canopy height by an average of 62% by charcoal filtration. In addition, estimated yield losses for wheat in our study were comparable to yield losses attributed to ozone for maize (10%) and higher than those for soybean (5%) in regression analysis of historical yield data (McGrath *et al.*, 2015). Further confidence in our analysis was gained from good agreement between measured and modelled ozone concentrations from sites around the world, and between our stomatal conductance modelling and annual data for the ratio of actual-to-potential evapotranspiration in wheat-growing grid cells of the USA.

For the first time, this study compared the effects of ozone in DC that export 42% of their wheat grain with effects in countries receiving ODA that rely largely on home-grown wheat, exporting only 0.7, 0.4, 6.0 and 8.9% of production for LDC, OLIC, LMIC and UMIC, respectively (Mean of 2010-2012, FAOSTAT). The combined production losses for countries receiving ODA of 51 Tg was roughly 50% higher than those estimated for DC (34 Tg), providing support for our first hypothesis. Given that the demand for wheat is increasing at twice the rate in developing than developed countries (Shiferaw *et al.*, 2013), then these losses due to ozone may reduce the likelihood of achieving the UN SDG 2 of securing food supplies and ending hunger by 2030, especially in UMICs (e.g. China) and LMICs (e.g. India and Pakistan). This situation may be further exacerbated by breeding initiatives that favour increased stomatal conductance (Lobell & Gourdj, 2012; Roche, 2015) that may be inadvertently increasing ozone uptake and effect on wheat, as newer varieties with higher stomatal conductance tend to be more sensitive to ozone, in part due to higher uptake of the pollutant (Bizwas *et al.*, 2009).

In this study, we found that the greatest yield losses are in the warm-temperate-moist, tropical-moist and tropical-wet climates of the NH and tropical-moist and –wet climates of the SH. Under these conditions, stomatal uptake of ozone is often maximized and mean yield losses per climatic zone of 12 - 17% and 9 - 11% are estimated for the NH and SH, respectively. Interestingly, we also showed that in drier climates where irrigation is in use, yield losses can be in the same range as in moist climates, highlighting the importance of soil moisture as a factor influencing the extent of ozone uptake and subsequent yield reduction. For example, 67 - 99% of the production losses estimated for India, China and Pakistan were in tropical-dry and warm-temperate-dry climates where 78 – 93% of wheat producing areas are irrigated. Of the countries with the highest production losses due to ozone, only in SW Russia was there a significant portion of a country's lost production in drier climates (43% for temperate-cool-dry) where irrigation usage was limited (13%).

Climatic zone impacts were also apparent when comparing estimations based on AOT40 or M7 with those based on POD₃IAM. For example, in the USA, the area of greatest concern switched from the temperate-dry climates of the western states that had the highest ozone concentrations to temperate-moist climates in eastern states with more moderate concentrations, but higher POD₃IAM values. Similar changes in areas of greatest concern are apparent when comparing with other spatial assessments based on ozone concentration. For example, our POD₃IAM modelling estimated that the areas of highest risk were further south in China and further north in India than those estimated using AOT40 and M7 by Van Dingenen *et al.* (2009). Whilst our concentration-based analysis showed similar areas being at risk in North America and Europe as those identified by Tai and Val Martin (2018), our stomatal uptake approach showed some spatial similarities and some differences to their Partial Derivative Linear Regression analysis of wheat yield data over an 18 year period. Their study showed high potential sensitivity of wheat to ozone along the USA-Canada

border in an area where our study also predicted relatively high production losses. Here, low-moderate ozone concentrations coincide with climatic and soil moisture conditions that are highly conducive to stomatal uptake of ozone. In most other areas of the USA, their analysis including only ozone and heat as explanatory factors, found no significant trends over an 18 year period. We suggest that their lack of significance for these areas may in part be due to the omission of soil moisture as an explanatory factor – seen in this study to have a large modifying effect on wheat yield response to ambient ozone. This study also diverged from the first stomatal uptake based assessment for India and China by Tang *et al.* (2013), whose spatial distribution of flux-based ozone risk is indeed closer to that of the concentration-based risks estimated in this study. This difference between the two studies could be attributed to the omission of soil moisture constraint on ozone uptake in Tang *et al.* (2013), and provides further support to the importance of soil water regime in the estimation of ozone risk.

This study also explored the consequences of a frequently discussed adaptation pathway for maintaining or increasing wheat production in future years: expansion of irrigation infrastructure and usage (Balkovič, *et al.*, 2014; Jägermeyr, *et al.*, 2016; Tanaka *et al.*, 2015). As a first step, we determined the potential added negative effects of ozone on wheat production if irrigation water is supplied whenever needed by removing the limitations due to soil moisture on ozone uptake. This relatively simple approach did not include the effects of irrigation-induced changes in wheat microclimate such as changes in VPD, heat and ozone fluxes (Li *et al.*, 2008; Tuovinen *et al.*, 2009) on ozone uptake. On the evidence provided here, we argue that discussion of the benefits of added irrigation usage should take into account the potential negative effects of ozone uptake in countries and regions where the pollutant is prevalent. For example, in China, India, Germany and Pakistan where it is predicted that current wheat yields could be maintained in a future climate by use of new heat tolerant varieties and increasing the ratio of irrigated : unirrigated land by 50% (Tanaka *et al.*, 2015), we suggest that crop yield could be reduced by an average of 9.5 – 12.6% by ozone pollution if soil moisture is non-limiting, outweighing some of the added benefits. Our analysis also shows that expanding irrigation usage to 100% in India and China as suggested in Tanaka *et al.* (2015) as a way to maintain wheat yields may increase yield losses due to ozone from 7.5 – 15% to 11.5 to 25% in the major wheat growing areas of these countries under current ozone levels, with effects potentially even higher as concentrations rise in coming decades (Wild *et al.*, 2012). As well as reducing grain availability, such additional yield losses would also result in wasted fertilizer usage due to reduced fertilizer efficiency (Broberg *et al.*, 2017) and unproductive use of water, energy and labour, all of which are in short supply in many of these areas. In Argentina, one of the top 15 wheat producers and identified as having potential for expansion of irrigation (Jägermeyr, *et al.*, 2016), we have shown that full use of irrigation would result in a further 0.2 Tg of lost grain and increase yield loss due to ozone from 5.6 to 7.8%. In contrast, yield losses in Pakistan, where irrigation usage is already nearly 90%, would only increase by a small amount (0.01 Tg) if irrigation was used in all wheat growing areas, as currently unirrigated areas in the NW tend to have lower ozone concentrations. In summary, when we compare our results with studies

that identify regions where there is scope for increasing yield by irrigation expansion and improved water management, we predict that as much as one third of the yield benefits of added irrigation could potentially be lost due to ozone pollution.

This analysis has compared the current effects of ozone against pre-industrial ozone to give an indication of the extent of the negative effects of the pollutant under current climates. To facilitate comparisons, we have not taken into account that CO₂ concentrations were also lower in pre-industrial times which could have increased stomatal conductance for C3 plants by 20 - 30% (Lammertsma *et al.*, 2011; Purcell *et al.*, 2018). Whilst additional increases in CO₂ by the end of the century might reduce stomatal conductance further (Purcell *et al.*, 2018), complex interactions and feedbacks suggest that the predicted compensation for ozone effects is not supported by field evidence in C3 crops (Ainsworth *et al.*, 2012; Mills *et al.*, 2016). Indeed, site specific predictions for Europe indicate that the stomatal uptake based risk of ozone damage may remain more or less the same as the effects of the combination of climate change, rising CO₂ and rising O₃, balance each other out (Klingberg *et al.*, 2011). As well as increases in ozone on a global scale due to increased precursor emissions and long-range transport (Wild *et al.*, 2012), reduced stomatal uptake of ozone under rising CO₂ will also lead to increased ambient ozone concentrations, further reducing the compensatory effects of rising CO₂ (Ainsworth *et al.*, 2012).

Although recognized as a modifying factor that may be as, or more, important than effects of future changes in rainfall (Lobell & Asseng, 2017), ozone is rarely included in either statistical or process-based modelling of current or future crop yields (e.g. Challinor *et al.*, 2009; Lobell & Asseng, 2017; Lobell & Gourdjji, 2012). There are several possible reasons for this. Firstly, the negative effects of the pollutant are not being recognized outside the ozone-effects community. This is surprising given that the magnitude of effects reported here and elsewhere (e.g. Avnery *et al.*, 2011a,b; Van Dingenen *et al.*, 2009) are in the same range as losses predicted for other effects of climate change. For example, a 6% decrease in wheat yield per 1°C rise in temperature is a cause for concern at the global scale (Asseng *et al.*, 2013; Liu *et al.*, 2016), with larger detrimental effects predicted at country scales (e.g. India, Lobell *et al.*, 2012, and Australia, Asseng *et al.*, 2011). The second reason could be a lack of availability of accessible ozone or ozone stomatal uptake data, also highlighted in Lobell & Asseng (2017) as a contributing factor. This situation is now changing. A new initiative - the International Global Atmospheric Chemistry (IGAC) Global Tropospheric Ozone Assessment Report (TOAR) for the first time has brought together all publically available ozone data into one online easily searchable global database (Schultz *et al.*, 2018 and <http://www.igacproject.org/activities/TOAR>). The TOAR study also includes an analysis of vegetation metrics, including AOT40 for wheat, from over 3300 vegetated sites in 44 countries presented as 15- and 20-year trends and current values, averaged for 2010-2014 (Mills *et al.*, submitted). Furthermore, grid-cell modelled values for ozone metrics are available for the EMEP grid at http://www.emep.int/mscw/mscw_data.html. A second reason could be a lack of collation of process-effects data in suitable format for process-based modelling which is currently being addressed (Emberson *et al.*, 2018), including initiatives involving the AgMIP modelling community (Agricultural Model Intercomparison and Improvement Community; <http://www.agmip.org/>). Lastly, it is possible that some earlier concentration-based studies that have

estimated relatively large effects of ozone in some regions (e.g. 17.5% to 55% yield losses estimated for India by Burney and Ramanathan, 2014; Ghude *et al.*, 2014; Avnery *et al.*, 2011a, b), may have inadvertently hindered acceptance within the agricultural community as the values may be higher than expected by agronomists. The new stomatal uptake-based method presented here, supported by field-based ozone filtration experiments from around the world, could provide the impetus for renewed interest in inclusion of ozone in crop yield predictive modelling.

In conclusion, our results indicate that the negative effects of ozone on wheat yield need to be mitigated in order to help close current yield gaps and achieve the UN SDG 2 of providing food security and eradicating hunger by 2030 (UN, 2016). Negative effects on wheat production in countries receiving ODA total 50% more than those in DC, meaning that both within-country production and imports will be impacted. Importantly, we also show that the benefits of water management practices and crop breeding strategies designed to increase yield potential by increasing carbon capture in wheat (IFPRI, 2016; Hawkesford, *et al.*, 2013; Jägermeyr, *et al.*, 2016; Mueller, *et al.*, 2012; Roche, D., 2015; Tanaka, *et al.*, 2015;) may not be as pronounced as anticipated owing to the effects of co-occurring uptake of ozone pollution. Ozone impacts on yield might be mitigated by both (i) exploiting the genetic variation in ozone resistance in wheat cultivars in breeding programmes using approaches discussed recently for rice (Frei, 2015), and (ii) developing management strategies that protect against ozone damage (Wilkinson *et al.*, 2012). Ultimately, though, the largest benefit would accrue from global-scale international efforts to reduce the emissions of ozone precursors, with co-benefits for the production of other staple food crops known to be sensitive or moderately sensitive to ozone (e.g. maize, soybean and rice (Mills *et al.*, 2007) as well as for human health, ecosystems and climate (UN SDGs 3, 15 and 13, UN (2016)).

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Data Accessibility Statement:

The Supporting Information provides the National average crop yield losses calculated using M7, AOT40 and POD3IAM, and 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. (a) – (c) The seasonal mean M7 (ppb), AOT40 (ppb h) and POD₃IAM (mmol m⁻²) and (d) - (f), the negative effects of ozone pollution on wheat yield presented as the % yield loss for M7, AOT40 and POD₃IAM. All values are the means per 1° x 1° grid cell where the wheat production exceeds 500 Mg and are averaged for 2010 - 2012. Values for (c) POD₃IAM and (f) yield loss calculated from POD₃IAM use the POD₃IAM weighted by the proportion of production in the cell that is rain-fed (using POD₃IAM calculated using soil moisture limitation) and irrigated (using POD₃IAM calculated without soil moisture limitation). Yield losses for AOT40 and POD₃IAM are relative to 0 ppm h and 0 mmol m⁻² respectively, whilst those for M7 are relative to 25 ppb.

Fig. 2: Production loss for wheat due to ozone modelled from the POD₃IAM and percentage yield losses presented in Fig. 1 and averaged for 2010-2012.

Fig. 3: Box plot of percentage yield loss by climatic zone for the (a) northern (NH) and (b) southern hemisphere (SH), calculated using POD₃IAM as indicated in Fig. 1. The box represents the interquartile range, with the top line being the third quartile (Q3), the middle line the median and the bottom line the first quartile (Q1), with the whiskers indicating the range of the data calculated as an upper limit of Q3 + 1.5 (Q3-Q1) and a lower limit of Q1 - 1.5 (Q3 - Q1), and * representing outliers.

Fig. 4: (a) Yield loss and (b) production loss (Tg) by country category, calculated using POD₃IAM (see Fig. 1) and averaged over 2010-2012. Countries have been assigned by the Organisation for Economic Co-operation and Development (OECD) Development Assistance Committee (DAC) into the following categories receiving Official Development Assistance (ODA): Least Developed Countries (LDC); Other Low Income Countries (OLIC); Lower Middle Income Countries and Territories (LMIC); and Upper Middle Income Countries and Territories (UMIC). The countries included in these categories are listed in OECD (2017), with their category added to Table S2. For comparison, effects of ozone on wheat yield are also shown for developed countries (DC), assumed here to be all other countries that are not included in the ODA recipients list.

Fig. 5: Production losses in wheat due to ozone for the seven most affected countries. Values are sums of production losses per 1° x 1° grid cell calculated using production data and irrigation weighted POD₃IAM (see legend for Fig. 1 for explanation) within each climatic zone. The mean percentage yield loss per country is provided above each bar.

Fig. 6: The effects of removing soil moisture limitation of stomatal conductance on ozone-induced yield loss by assuming that irrigation can be used as needed, presented as the added % yield loss due to ozone if irrigation is increased from current usage to 100% for each grid cell. Yield losses with current irrigation usage are presented in Fig. 1f. Note: The values presented are per 1° x 1° grid cell where the wheat production exceeds 500 Mg and are averaged for 2010 - 2012.

Fig. 7: Additional lost production due to ozone, assuming 100% irrigation, presented by country category. See Fig. 4 legend for key to county groupings.

Fig. 8. Additional wheat production losses associated with expansion of irrigation in dry climatic zones. The figure shows the percentage irrigation currently in use in each country (black filled circles) together with the additional production losses if irrigation were increased to 100% (grey bars) for (a) Cool temperate dry, (b) Warm temperate dry and (c) Tropical dry climatic zones. Values are means for 2010 – 2012, are calculated using POD₃IAM, and are shown for the eight most affected countries per climate zone.















