Final Authors' response:

We thank the two anonymous referees for their thoughtful comments. We have carefully considered all the points as discussed below and have substantially revised the manuscript to improve structure and clarity.

Response to anonymous referee #3

Reviewer's comment:

The article presents calculations of yield losses due to ozone in India – both in terms of biomass and monetary value. The work is quite comprehensive and combines available literature data with new damage functions obtained from open-top chambers for various crops. Based on the new functions, which indicate a relative high sensitivity to ozone, the calculated losses are higher by a factor of more than two than previously estimated.

Authors' response:

We thank anonymous reviewer for this compliment and the comprehensive review. In particular we would like to thank the reviewer for pointing us towards several more recent studies which we now include in the revised manuscript as detailed below.

Reviewer's comment:

A deficit of the paper is that it uses various cumulative indices to be related with the damage, which all are calculated from concentrations but not from uptake. This is not state of the art (Danielsson et al., 2013, Yamaguchi et al., 2014), despite AOT40 being still in use for such exercises (Feng et al., 2015).

Authors' response:

We appreciate the reviewer's comment, that it is highly desirable to switch from an exposure based relationship to a stomatal flux based uptake-damage relationship which is based on crop models and mechanistic understanding. We have considered the possibility of including the stomatal flux based method for the present study, but found that it raised a number of serious issues and practical constraints when it comes to studying the exposure-yield relationships for South Asian cultivars. These are listed below:

- 1. The stomatal flux model DO3SE version 3.0.5 has been developed and validated mostly in European countries. In its current form the model can only handle a growing season that starts in spring and ends in summer/autumn and is unable to handle the growing season of the Indian wheat crop, which is sown between day 280-310 of one year and is harvested around day 90 of the following year. To overcome this limitation, one would have to use the measured hourly input data of ozone and meteorological parameters from the wheat growing season with fictional dates (shifted by 6 months with respect to the true dates). However, without good field observations that allow determining whether this brute force approach partially corrupts the model output, we are very hesitant to put such data into the peer reviewed literature. The developmental work required to adapt the model such that it can be used for the South Asian rabi season (wheat growing season) is beyond the scope of this paper and the model parameterization for South Asian cultivars cannot be undertaken without datasets suitable for model validation (see point 3 below).
- 2. Currently out of all the crops investigated in this study, only a parameterization for wheat is included in the model as pre-set and incorporated into the mapping manual. No parameterization for cotton, maize and rice is available as pre-set and studies adapting the stomatal flux model parameterization to other crops e.g. for rice are all recent (Yamaguchi et al., 2014). To our knowledge, no internationally agreed exposure yield relationship using flux based metrics exists for these crops.

- 3. Several recent studies have emphasised the need for a local parameterization of the stomatal flux model in particular for the Mediterranean climate, which in the European context comes closest to the climate under which wheat is grown in Northern India (Farez et al. 2012, González-Fernández 2013, Feng et al. 2015). However, the Mediterranean parameterization cannot be applied to the North West Indo Gangetic Plain, as the wheat crop in the Mediterranean is rain fed while wheat in Punjab and Haryana is irrigated. González-Fernández 2013 found that in the Mediterranean ozone fluxes were limited by soil water content limitations to the stomatal flux (g_{sto}) when O₃ concentrations were above 40 nl l⁻¹. For irrigated crops this may not be the case and fluxes may be higher. Developing and validating a local parameterization would require a dataset of colocated high time resolution observations of ozone mixing ratios, meteorological parameters, plant phenology and time resolved measurements of stomatal conductance. For South Asia, no such comprehensive dataset is available in the literature.
- 4. A major point of our paper, as recognized by the reviewer, is to highlight the fact that South Asian cultivars are more sensitive to ozone than their European and American counterparts. To make this point, we needed to use data from studies conducted on both South Asian and the other types of cultivars. Till date, there is no single experimental study reporting flux-response data using the stomatal flux based uptake model for South Asian cultivars of any of the species considered. Hence, such a comparison is only possible on the basis of AOT40 and M7 exposure-response metrics. Studies reporting ozone exposure using these two metrics have been reported for a wide range of European, American and South Asian cultivars.

We would also like to point out that, most recent global and regional modelling studies still rely on the AOT40 metrics (see e.g. Texeira et al. 2011, Avnery et al. 2011a,b, Hollaway et al. 2012, Amin et al. 2013, Ghude et al. 2014, Feng et al. 2015, Chuwah et al. 2015) for several reasons which include that exposure response relationships relying on this metric are available for a large variety of crops, internationally recognized and that the application is simple and user friendly, requires no validation for different climates and can accommodate different cropping seasons/sowing dates.

Therefore it is clear that this is not a deficit specific to the present work. However the reviewer's general suggestion is appreciated and so we have revised the description of the leaf ozone uptake based exposure indices (P 2362 line 19 onward) to be more specific about the advantages of the stomatal flux modeling approach.

The full description, however, has been shifted from the Materials and Methods section to the Introduction in response to the comment about the confusing structure of our manuscript (see below).

Moreover, we have pointed out the need to move towards ozone uptake based models for crop yield loss assessments in the "Conclusion" as an area of future research for South Asian cultivars.

Modifications in the text:

P 2362 line 19 onward the revised text now reads:

"Recently stomatal flux-based critical levels were proposed to address concerns that the AOT₄₀-based critical levels are based on the concentration of ozone in the atmosphere whilst the ozone related damage depends on the amount of the pollutant reaching the sites of damage within the leaf (Emberson et al., 2000; Mills et al., 2011b). Models using stomatal uptake of O3 (flux; F) or its cumulative value, dose (D) have significantly improved the prediction of plant injury and have addressed the asynchronicity of maximum stomatal conductance (g_{sto}) and peak ozone in particular in plants that close their stomata when temperatures or the water vapour pressure deficit around the leaves are too high (Ainsworth et al., 2012, Fares et al. 2013, Feng et al. 2012, Danielsson et al., 2013, González-Fernández 2013, Yamaguchi et al., 2014). Stomatal flux of ozone is modelled using a multiplicative algorithm adapted from Emberson et al. (2000) that incorporates the effects of air temperature, vapour pressure deficit of the air surrounding the leaves, light, soil water potential, plant phenology and ozone concentration on the maximum stomatal conductance, i.e. the stomatal conductance under optimal conditions. The exposure yield relationships based on this algorithm

consider the accumulated stomatal flux over a specified time interval as PODY (the Phytotoxic Ozone Dose over a threshold flux of *Y* nmolO₃, m⁻², PLA, s⁻¹ with *Y* ranging from 0 to 9 nmolO₃, m⁻², PLA, s⁻¹ (Mills et al., 2011b). Studies evaluating the PODY based exposure yield relationship for a wide range of climate zones have emphasised the need for a local parameterization of the stomatal flux model (Fares et al. 2013, Feng et al. 2012, Danielsson et al., 2013, González-Fernández 2013, Yamaguchi et al., 2014) . To the best of our knowledge no parameterization for South Asian wheat and rice cultivars has been reported in the peer reviewed literature. The wheat parameterization has been developed using European cultivars (Mills et al., 2011b) and for rice the parameterization has been developed using only one Japanese rice cultivar, Koshihikari (Yamaguchi et al. 2014), which is know for its ozone resistance (Sawada and Kondo 2009). Despite the fact that the stomatal flux based model is recommended by the UNECE CLRTAP (United Nations Economic Commission for Europe Convention on Long-range Transboundary Air Pollution) for ozone risk assessment in Europe based on accumulated stomatal ozone fluxes over a threshold (UNECE 2010), exposure yield relationships have so far been internationally agreed upon only for a limited number of crops (Mills et al., 2011b)."

We have inserted a paragraph on Page 2383 line 8 and have modified the text to: "For all crops screening a large number of domestic cultivars using the new stomatal flux based exposure metrics to identify and promote those cultivars that are less susceptible to ozone damage also offers a way forward."

Reviewer's comment:

The other concern I have about this paper is that it is rather confusing because it mixes a review paper with an analysis based on newly derived functions.

Authors' response:

The newly derived functions are derived based on a literature review of the available data. We have made this clearer in the revised introduction. Moreover, we have restructured the introduction, shifted some of the text from the "Materials and Methods" section to the introduction and used the subheadings "1.1 Ozone effects on plants", "1.2 Metrics to assess the impact of ozone on crop yields" and "1.3 Present study"

to improve the clarity of the manuscript.

Modifications in the text:

Page 2359 line 12 We inserted the sub heading "1.3 Present study"

With the following text:

"In the present study, we present new ozone exposure crop yield relationship for Indian rice, wheat and maize cultivars derived through a review of the peer reviewed literature of open top chamber studies on South Asian cultivars.

We verify these new relationships using ozone monitoring data from the Atmospheric Chemistry facility in Mohali and yield data from a number of relay seeding experiments conducted in Punjab and Haryana. In these experiments crops were coincidentally exposed to different ozone levels by virtue of shifting their sowing date, but the relevant studies were not conducted to investigate the effect of ozone on yields and consequently they did not include on-site ozone monitoring or clean air control treatments.

We subsequently use a high quality dataset of in-situ ozone measurements at a regionally representative suburban site called Mohali and the newly derived exposure –yield functions to assess ozone related crop yield losses for wheat, rice, cotton and maize for Punjab and the neighbouring state Haryana for the years 2011–2013. Crop yield loss estimates calculated using two different exposure metrics, AOT40 and M7, are inter-compared for a number of sowing dates and exposure-yield functions for the two major crop growing seasons of Kharif (June–October) and Rabi (November–April)."

Reviewer's comment:

Furthermore, it is very difficult to evaluate the methodology because concentrations, indices, and response functions are described at various places and only part of what is given in the descriptions is actually used.

Authors' response:

We thank the reviewer for this feedback. To improve the clarity of the manuscript, we have removed the historical overview and retained only equations for M7 and AOT40, the two metrics used for our analysis, in the materials and methods section. We have also removed the ozone exposure according to the M12 and W126 metrics from all tables.

We have shifted part of the historical perspective and description of the flux based method to the introduction and placed it under the heading "1.2 Metrics to assess the impact of ozone exposure on crop yields"

Reviewer's comment:

In addition, the importance of when the grain is sowed is often stressed but a sensitivity analyses about different sowing dates is not provided.

Authors' response:

Sensitivity analyses using 5 different sowing/harvesting dates for both rice and wheat, 3 different sowing/harvesting dates for cotton and 2 different sowing/harvesting dates for kharif and rabi maize, had already been presented in the study. These results were in supplementary tables 1-4 and were/are discussed in the text. Since the reviewer #3 missed out on the material in the supplement, we have shifted these tables back to the main manuscript to ensure this does not happen to other readers.

Modifications in the text:

Shifted supplementary table 1-4 to the main manuscript

More specific remarks:

Introduction

Reviewer's comment:

P2357 (L20ff): The explanation about possible increased ozone damages under drought stress neglects that drought stress reduces the stomata conductance and thus the ozone uptake and damage. I guess that the somewhat strange argumentation refers to the impact of ozone to stomata regulation (Paoletti & Grulke, 2010). Differences in sensitivity to this effect could indeed cause a different ozone responses but I cannot follow the argumentation that is should occur more often in South Asia than in other regions.

Authors' response:

The yield loss mechanisms of plant phenotypes which close their stomata under stress conditions were discussed on Page 2358 (Line 13ff). We thank the anonymous referee #3 for pointing out that we did not mention that drought stress reduces ozone uptake in such plant phenotypes and have included this point.

On Page 2357 (Line 20ff) we discuss only plant phenotypes for which ozone stress interferes with stomata regulation, though we did not refer to the work of Paoletti & Grulke, (2010) on stomatal sluggishness but to the work of Mills et al. (2009) and Wilkinson & Davies (2009, 2010), referenced through a review (Wilkinson et al. 2012) which reported that for certain plant phenotypes, stomatal sensitivity to abscisic acid is compromised in O_3 -stressed plants which can result in additional drought stress (the plant hormone abscisic acid normally controls stomata closure and reduces water loss under drought conditions). We did not intend to suggest that this mechanism impacts only South Asian cultivars. We meant to suggest that losses because of such a response would be disproportionally large in South Asia for two reasons. Firstly, temperatures under drought conditions

are at the upper end of the species tolerance range (often exceed 40°C) and mid-season drought is a frequent phenomenon during monsoon season. Secondly, South Asia has a large number of rain fed landholdings with no access to irrigation.

Modifications in the text:

On Page 2357 Line 24 "Consequently, such plant phenotypes when exposed to both drought and O_3 will continue to lose water despite the potential for dehydration. Ozone related crop yield losses in such phenotypes may be enhanced in rain fed regions where kharif cops are frequently exposed to mid-season drought during monsoon season. On the other hand, the yield of rice cultivars that show a healthy response to drought stress (i.e. close their stomata aperture rather than having a sluggish response) could substantially benefit from the system of rice intensification (SRI) cultivation practise (Turmel et al. 2011) in areas with high ozone mixing ratios. Paddy fields under SRI cultivation are irrigated only when rice plots dry too much and the crop starts withering. A healthy response of rice plants to soil drying would reduce the ozone uptake and could explain the higher yields frequently observed for SRI plots during field trials as well as the spatial variability of the yield difference between SRI plots and control treatments.

Reviewer's comment:

P2358: The overview about ozone damages seems more or less comprehensive but more recent reviews are available as references (Ainsworth et al., 2012, Kangasjärvi & Kangasjärvi, 2014, Leisner & Ainsworth, 2012). Particularly the role of induced defences, which could be the cause of yield declines without visible injuries could be mentioned (Heath, 2008, Iriti & Faoro, 2009). <u>Turmel</u>

<u>Authors' response:</u> We thank reviewer #1 for pointing us towards these interesting reviews and have revised the overview about the ozone damages to include these more recent studies as detailed below.

Modifications in the text:

Page2357 L16 Pleijel et al. 1991, Heath 2008, Iriti and Faoro 2009

Page 2357 L19 Wilkinson et al. 2012, Ainsworth et al. 2012, Leisner and Ainsworth 2012

Page 2358 L4 Heat 2008, Iriti and Faoro 2009, Kangasjärvi and Kangasjärvi 2014

Page 2358 L14Torsethaugen et al., 1999, Heat 2008, Iriti and Faoro 2009, Ainsworth et al., 2012,

Page 2358 L19 "Plants of this phenotype may show little to no visible leaf damage, and often allocate significant resources to induced defences following ROS..."

Reviewer's comment:

Materials and Methods

Here, five metrics and a historical overview about ozone damage related indices is presented although only two indices are used for further analysis. Moreover, the flux based calculation may be complemented by more recent formulations (Danielsson et al., 2013). Overall, this seems to be unnecessary comprehensive.

<u>Authors' response:</u> We have removed the historical overview and now discuss only M7 and AOT40 in Materials and methods section, as pointed out earlier in this response.

Modifications in the text:

Shifted P2361 lines 2-15 to the introduction. Replaced this text with

"We use two metrics to investigate the ozone exposure for crops in Punjab and Haryana derive south Asia specific exposure yield relationships for wheat and rice. The mean daytime surface ozone (M7) and accumulated exposure over a threshold of 40 nmol mol-1 (AOT40)."

Retained lines 16-20

Shifted P2361 line 20 to P2362 line 2

Retained P2362 line 2-5 "AOT40 is defined....

Shifted P2362 line 6 to P2363 line 9 to the introduction and revised the text (see above)

Revised P2363 line 9-14 "Out of these two parameters, M7 gives equal importance [...] while AOT40 gives [...]. Hence the former will perform better while evaluating plant damage ...

Shifted and revised P2363 line 15 to 18

Reviewer's comment:

as is also the description of cropping seasons and crops where not only the crops used in the investigation but many others are also described. However, a simple percentage of coverage and thus a reason for choosing these particular crops is not given.

<u>Authors' response:</u> We have removed text about those crops not covered in this study as specified below

Modifications in the text: f (means one line after; ff means several lines after....)

P2364 L10f however in some districts...

P2364 L13ff Minor rabi crops are potato, rabi maize, sugarcane, rabi pulses and oilseeds (Sharma and Sood, 2003)[...] or seasonal fruits and vegetables (musk melon, water melon, gourds and cucumber).

P2364 L24f Zayad season crops include moong and vegetables (Saroj et al., 2014).

P2364 L26 replaced "maize based" by "maize-wheat" and inserted % values "rice-wheat (>70%)" cotton-wheat (~20%)

P2364 L26f deleted "Sorghum-wheat rotation is popular in the Shivalik mountains."

P2364 L29 Inserted: rice-wheat (~40%) and cotton-wheat (~20%) deleted: rice-mustard and rice-gram rotation is popular in the north

P2365 L2 inserted "Maize is currently not very popular but heavily promoted as an alternative to rice when a deficient monsoon is anticipated."

Reviewer's comment:

The description of the ozone dose exposure relationships is much too short and irritating. It is not clear which calculations are done with new OTC derived functions and which are not. This is partly done in the results sections (e.g. page 2371, parts of chapters 3.2.1 - 3.2.4) where it doesn't belong. It is also not quite clear from which periods the data for the newly derived functions are obtained and of different periods are used which then might need weighting with phenological preconditions. It would be a great help if all this information could be concentrated and re-written.

<u>Authors' response:</u> We thank the reviewer for pointing this out. We have shifted the relevant text from the results on page 2371 to this section and have modified it to be more specific. Since the relationship is derived based on OTC with ozone fumigation and clean air controls it should not require weighting with phenological preconditions.

Modifications in the text:

The revised text now reads:

"We derive specific exposure-yield relationships for Indian wheat and rice cultivars using a two pronged approach.

Firstly, we use our ozone measurements conducted at a suburban site in Punjab and a number of field studies conducted in the region that reported variations in the sowing date of crops (Chahal et al., 2007; Jalota et al., 2008, 2009; Mahajan et al., 2009; Brar et al., 2012; Buttar et al., 2013; Ram et al., 2013) which lead to coincidental change in ozone exposure and one study that reported collocated yield and ozone measurements (Agrawal et al., 2003) to derive an empirical exposure-yield relationship for rice and wheat. The empirical field data supports the need to revise the exposure-yield relationship for Indian cultivars and demonstrates, that for rice optimizing the sowing date can be a suitable strategy to minimize ozone exposure and maximise crop yields.

Secondly, we derive India specific exposure yield relationships by plotting relative yields (RY) and ozone exposure for all OTC studies on Indian cultivars reported in the peer reviewed literature and

fitting the data to obtain an exposure yield relationship. (Rai et al., 2007; Rai and Agrawal, 2008; Singh et al., 2009; Rai et al., 2010; Singh and Agrawal, 2010; Sarkar and Agrawal, 2010, 2012) For maize only one OTC study on two Indian cultivars has been conducted and we use the fit of this data to obtain an exposure yield relationship (Singh et al., 2014). We compare these exposure-yield relationships for rice and wheat with RY observed for cultivars commonly grown in Pakistan and Bangladesh (Wahid et al., 1995b; Maggs et al., 1995; Maggs and Ashmore, 1998; Wahid, 2006; Akhtar et al., 2010a,b; Wahid et al., 2011) to investigate to which extent the results can be extrapolated to entire South Asia. We refrain from including cultivars popular in South East Asia into our study, as they have been reported to show a very different sensitivity to ozone exposure (Sawada and Kohno, 2009). We provide an upper and lower limit for RY and crop yield losses for a set of 5 different sowing dates for rice and wheat, 3 for cotton and 2 for rabi and kharif maize both using exposure dose–response relationships established in several studies in the West (Table 2) to provide a lower limit and our new India specific functions to provide an upper limit to the possible loss. We use both the old (Mills et al., 2007) AOT40 based exposure yield function, as well as our revised AOT 40 based relationship to calculate crop production losses and economic cost losses and contrast

Reviewer's comment:

the two."

Results and Discussion

P2377, L1ff: I agree that rainfall can will reduce ozone related precursors but it would obviously be correlated with low radiation also. So the ozone forming potential would be low and the stomata would be less open, reducing uptake and relative yield loss. Can this be confirmed from the data?

<u>Authors' response:</u> The reviewer is possibly correct in pointing out that radiation plays a larger or equal role compared to the wet scavenging of precursors in reducing the ozone mixing ratios.

We are not aware of any observational evidence from South Asia reporting stomata opening/conductance with sufficient time resolution to investigate whether stomata would be less open during rainy/cloudy conditions. It is clear that the ozone is lower (on average by about 20 ppbv) during rain spells and under heavy cloud cover and if stomata closure reduces uptake further this would only enhance the effect. But then, since plants cannot keep the stomata closed perpetually this would also mean that stomata would preferably open during dry spells when the ozone is much higher. If that is true it would make AOT40 (which is usually high during sunny days) a much better proxy for stomatal flux compared to M7 for the kharif season. Unfortunately, all this discussion is speculative. We are not aware of any experimental data that would allow verification.

Reviewer's comment:

It is also a bit frustrating to read and think about the possible mechanistic relationships and then learn that no new exposure relationships exist for cotton and maize. In my opinion, the article should focus on wheat and rice (as implied in the title). The other crops may however complement the analysis in order to judge the relative importance of the new findings.

Authors' response: For maize we have included a revised relationship based on a recent study by Singh et al. 2014, which also indicates that South Asian cultivars are a factor 2 more sensitive, into the final revised manuscript. When it comes to cotton we are equally frustrated. India grows 25% of the world's cotton and the relative yield losses are potentially very high (almost 50%) even with the old Mills et al. 2007 relationship. Yet there is no data to verify or derive a revised relationship. We, therefore, prefer to retain the discussion of cotton. Removing it would send the wrong signal and would imply losses are not worth discussing, when in fact they are higher than those for rice.

Modifications in the text:

Adding the new relationship for maize has resulted in the following changes:

Table 2 (additional equation), Table 5 (AOT40 based yields in the "this study column") and Table 6 (crop production losses and economic cost losses) as well as an additional column in the table with the results for maize, which has been shifted for the supplement back into the manuscript.

While calculating this revised relationship, we found a mistake in the excel spread sheath. Accidentally the RY for maize had been calculated with the equation for rice. We have corrected this and now RY are higher and RYL are lower. We have checked all spread sheaths and now the correct equations have been used everywhere.

The Abstract has been modified to: "... and established a new crop yield exposure relationship for South Asian wheat, rice and maize cultivars..."

Section 3.2.4 was revised as follows:" Maize is planted both as Rabi and Kharif crop, however, cultivation occurs only on a~limited area, but maize is heavily promoted as an alternative to rice when a deficient monsoon is anticipated. We could not find any study reporting crop yields for maize planted in Punjab or Haryana in the peer reviewed literature. A recent study investigating ozone related crop yield losses for Indian maize cultivars (Singh et al., 2014), found Indian maize cultivars are twice as sensitive to ozone compared to their American and European counterparts. However, maize is one order of magnitude less sensitive to ozone compared to rice and wheat and is, therefore, a suitable alternative for drought years. We use all three ozone exposure RY

relationships (Heck et al., 1984b; Mills et al., 2007; Singh et al., 2014) to calculate relative yields (Table~8) and find that in the real world both the differences between the revised and old relationship and the overall losses are minor."

Reviewer's comment:

What I feel is missing is an analysis about the relative sensitivity of the results to 1)

weather conditions in different years and the determination of ozone concentrations for

the region and seasons, and 2) the exposure – damage functions used. To which degree can damage be avoided if sowing dates are adapted?

Is it necessary to include a seasonal dynamic sensitivity to judge this and in which way would a cumulative uptake calculation be beneficial to the analysis?

Authors' response: Response to 1) Currently there is too little temporal overlap between the yield data and our ozone and meteorological dataset to attempt a detailed analysis investigating the influence of weather conditions in different years. With only 2 kharif and rabi seasons worth of data for which the yields have been finalized and reported in the statistical yearbook, we do not have a sufficiently large dataset for a comprehensive sensitivity analysis. This can be a topic of a future study. Response to 2) This data, including a cumulative exposure calculation for different sowing dates has been presented in supplementary table 1-4. Since reviewer #3 has shown substantial interest in this information and could not find it in the supplement, we have shifted these tables back into the main text and have added to the discussion the following statement.

Modification in the text

Shifted supplementary table 1-4 back into the main paper and changed the references to these tables. Moreover, we added the following text to the discussion P2372 L5: "For rice late sowing (1^{st} of June) and late transplantation (1^{st} of July) leads to the lowest relative yield losses (18%) while early sowing (1^{st} April) and transplantation (1^{st} May) doubles ozone related yield losses (35%)."

Reviewer's comment:

Conclusion

P2383, L10ff: The polical demands seem to be quite unrelated to the research presented here. Despite they might generally be valid I don't think they should be voiced here.

Authors' response:

Removed P2383, L10ff

Reviewer's comment:

Others

Despite an overall understandable stile, there are some problems with spelling and grammar as well as referring to the correct equation number (p.2366), full description of equation variables and other abbreviations (IGP). The text should also be checked for repetitions (e.g. p2370) and caption descriptions which belong beneath the figures (e.g. p.2371, 2374) that give some room for shortenings.

Authors' response:

We have corrected the equation number and the abbreviations and removed the repetition on . p2370 and have shifted part of the text to the figure caption as detailed below:

Shifted to figure caption Page 2371 Line 28-Page 2372 Line 3: "Ozone exposure for rice sowed on different sowing dates has been calculated using our data Table~5 Yield data for rice has been taken from the peer reviewed literature (<u>Chahal</u> et al, 2007; <u>Jalota</u> et al., 2009; <u>Mahajan</u> et al. 2009; <u>Brar</u> et al., 20120)."

Removed text that was already present in the figure caption Page 2372 Line 9-12 "Large diamonds indicate studies on Basmati, all other studies were conducted on paddy. Circles show plant chamber studies on Bangladeshi rice cultivars conducted in Japan and the dashed line delineates the European (AOT40, (Mills et al., 2007) and American (M7, (Adams et al. 1989) dose response relationship."

Shifted to the figure caption Page 2374 Line 3-12 "Ozone exposure for wheat sowed on different sowing dates has been calculated using our data (Table~6). Yield data for wheat have been taken from the peer reviewed literature (Agrawal et al., 2003; Chahal et al., 2007; Jalota et al., 2008; Coventry et al., 2011; Buttar et al., 2013; Ram et al., 2013). Agrawal et al. (2003) reported co-located measurements of ozone exposure and yields for a~number of urban locations that included residential areas and kerb site locations, where NO titration leads to low wintertime ozone levels. Other studies reported yields corresponding to different sowing date. The yield data has been positioned in conformation to the emergence dates (Period 1 to 5) defined in Supplement S1."

Removed text that was already present in the figure caption page 2374L23, "Circles show plant chamber studies on Bangladeshi wheat cultivars conducted in Japan"

Reviewer's comment:

Figures and Tables

I a bit irritated by seeing cumulative exposure indices per month. I thought that the cumulative index always refers to the period of a plants (leaves) exposure to ozone. If any, the index should be steadily increasing until harvest. Could you thus please explain what the relevance or meaning of the values presented in Table 2?

Authors' response:

We have given the cumulative index month wise, to provide data that can be of use to a variety of Authors'. We now call it "Monthly values of M7 and increment in AOT40 in the respective month" in the figure caption and have modified the text to "Table 3 shows the monthly increment in AOT40 and the monthly M7..." to avoid confusion. The purpose of giving the information in this format is twofold.

The region is notorious for its diversity; it is not uncommon to see that on one field the farmer is still burning the crop residue of the previous crop, while on the neighbouring field the flag leaves of the wheat crop sown more than a month ago are already several cm tall. Similarly, some farmers sow early and try to transplant their rice in May or early June in the hope of squeezing another crop in between rice and wheat while others will sow in June and transplant early in July. Month wise data will allow the interested user to sum up himself/herself, for the relevant growth period of their crop and can be useful for agricultural scientists in the region.

Moreover, this data can also be used for model validation. The winter growing season for example includes both persistent winter fog in December and January as well as heat waves with temperatures in the upper 30s later during the grain filling stage. Month wise indices allow a more detailed evaluation of model performance. Models could predict the right cumulative exposure for the whole growing season for the wrong reasons (e.g. if both the extreme fog episodes and the heat waves in March are not well captured).

Response to anonymous referee #1:

Reviewer comment:

The paper covers an important and interesting topic: Assessment of crop yield losses in Punjab and Haryana using two years of in-situ measurements. The study calculates the impact of present-day reductions of crop yield due to the background ozone from the measurements at Mohali and then extrapolates these fields to states of Punjab and Haryana. The most interesting part of the paper is new crop yield exposure relationship for South Asian wheat and rice cultivars which Authors' tired to develop based on scattered literature from south Asian specific studies. The manuscript is easy to read and the results are important. This paper is definitely a first step in achieving the objectives the Authors' have set up to achieve. My overall recommendation is acceptance after careful revision of the text and queries as under:

Authors' response:

We thank the anonymous reviewer #1 for the support to publish this paper and for his review. Addressing the comments will greatly improve the clarity of the manuscript. Detailed below is our response to the queries raised by the reviewer and a list of the specific changes made in the text.

Reviewer comment:

Specific comments

I have some reservations about the Authors' finding that new crop yield exposure relationship are a factor of two more sensitive to ozone induced crop losses compared to European and American Indices, and Authors' have not specified likely explanation for the dissimilarity. Is it because only few OTC (inconsistent) experiments are available over this region and lack of consistent OTC experimental and robust data set could be the prime reason (compared to European and American counterpart)?

Authors' response:

We agree that too few studies on South Asian cultivars are available - but this does not mean the studies available are of poor quality. Some of the studies have included metabolites and have elucidated the damage mechanism for individual cultivars. So far, different South Asian cultivars have been investigated by different author teams and hence at this stage there is no scope for revealing inconsistencies of the datasets. More detailed studies are clearly required.

Reviewer comment:

Or, Asian crops itself are highly sensitive to ozone than European and American crops?

Authors' response:

We have not commented in detail on the difference between European, American and South Asian cultivars as no comparative study of these cultivars has been conducted under identical conditions. Therefore, only speculations are possible at this stage.

However, we pointed out on page 2371 line 7-10 " ... Sawada and Kohno (2009) compared 20 different rice cultivars under identical conditions in a plant chamber and showed that most Oryza sativa L. Japonica cultivars were resistant to ozone damage (11 out of 12) while most Oryza sativa L. Indica cultivars showed significant yield losses (5 out of 8)."

Changes in the manuscript:

We replaced the text "This suggests that the spread in the data is indeed caused by differences in the sensitivity of different cultivars." page 2371 line10 with a longer statement that is more comprehensive to stress clearly that the differences are most likely related to the differential response of cultivars to ozone and that more data is required:

"A follow up metabolomic analysis of selected cultivars by the same authors' Sawada et al. 2012 showed that the only japonica cultivar with high yield losses, Kirara 397, down-regulated proteins associated with photosynthetic electron transport as a response to ROS induced by ozone. One of the indica cultivars with high yield losses, Takanari, showed no noteworthy changes in the metabolic pathway of photosynthesis resulting from ozone exposure but its yields were equally sensitive to ozone and most down-regulated proteins were associated with protein destination and storage and unknown functions. In one of the japonica cultivar, which did not suffer yield losses, Koshihikari, ozone stress up-regulated the expression of certain proteins in the Calvin cycle of the energy metabolism. Sarkar & Agrawal 2012 reported the expression of the RuBisCO and several energy metabolism related proteins were adversely affected by ozone exposure in two indica cultivars

Malviya dhan 36 and Shivani. These results seem to indicate that the responses to ozone are indeed cultivar specific. More studies are required to understand the damage mechanisms in different cultivars at a fundamental level and identify high yielding cultivars, that are resistant to ozone stress, which can be promoted by the relevant government agencies in affected areas."

Reviewer comment:

Or, crop exposure period for ozone to derive crop specific E-R function is different in SA, European and American (see below comments)?

AOT40 exposure requires accumulation of ozone concentrations over 90 days of crop growing period in order to assess the crop loss. Mills exposure functions are based on consistent 3 months (except for tomato which based on 3.5 months) growing period for wheat, rice, cotton and maize from various literatures.

Authors' response:

All studies used in this work to derive the ozone exposure relationship, expose the crop from emergence to maturity for wheat, and from transplantation to maturity for rice. Mills exposure functions are based on crops that were exposed 3 months to ozone for wheat and from emergence/transplantation to maturity for rice, cotton and maize. The paper explicitly states that for crops other than wheat and tomato, Mills et al. 2007 used only studies that satisfied the condition as follows: "*Experiments were conducted in the open field using a field release system or in open-top chambers. The crop should have been planted directly in the soil and should have been exposed to ozone from emergence to harvest. Only data from well-watered experiments were included in the analysis.*" Mills et al. 2007, p 2632 Therefore, the concern raised here and below regarding applying the Mills exposure -yield curve to the AOT40 accumulated over the full growth period is only valid for wheat not for rice, maize & cotton.

The 3 month period considered for wheat has historical reasons. Most of the early studies for wheat looked only at shorter time spans of ~3 months prior to harvest. This has been caused by the fact that "... *in most experiments, fumigations with ozone began several weeks after emergence.*" Adams et al. 1989 p 962. For wheat, Mills et al. 2007 relies on the compilation of older experiments by Fuhrer et al. 1997 and the 3 month limitation is again imposed by the fact that "...*duration of exposure varied between experiments, with an upper limit of about 90 days.*" Fuhrer et al. 1997 p95.

The fact that many early studies on wheat did not fumigate throughout, should not be used to imply that no damage occurs in the initial growth stages, though some select studies have shown, that wheat is more sensitive to ozone levels during anthesis & grain filling (Amundsen *et al.*, 1987, Pleijel *et al.*, 1996, Picchi et al. 2010). Hence our approach takes into account these relevant aspects.

Reviewer's comment:

This study derives empirical exposure-yield relationship based on various OTC studied conduced in India and Pakistan for wheat and rice (section 2.5 (last para), 3.2, 3.2.1 and 3.2.2). Here, author failed to mention what time-frame (exposure days, number of days from emergence to maturity) studies in India and Pakistan considered for the yield loss due to ozone (for wheat and rice)? Is it 3 months period? If not, whether the growing period is consistent in all these regional studies? This is important because if the exposure period differs within the various studies for the same crops (eg. wheat) then obviously crop exposed for longer duration (eg 120 days) will show higher yield loss compared to the same crop exposed for shorter duration (eg 90 days), and therefore derived empirical exposure yield relationship based on different exposure periods will be unrealistic. Author should cite (probably in table) the growing period/exposure period considered in OTC studies in India and Pakistan for different crops

Authors' response:

All studies presented in this paper exposed crops from the date of transplantation till harvest for rice. For wheat exposure, this was from emergence till harvest in all cases. We have added a sentence clarifying this in the relevant figure captions.

The number of days the crop takes from emergence to maturity varies from cultivar to cultivar. It also varies from year to year for multi-year field studies of the same cultivar; as the speed at which the cultivars reach maturity in the fields depends on meteorological conditions which vary from year to year. Listing this information for such a large number of different multi-year studies several of which included multiple different cultivars will make the paper lengthy. It would also imply that each

cultivar should be labelled differently in figure 4 & 6 which would obscure the clarity of the figure. Since there is no evidence supporting systematic differences between e.g. rice cultivars that reach maturity rapidly (90 day) and those that take longer (120 or 140) we believe that it is better if the interested reader refers to the original papers for these details. All the references have been provided in the figures and in the text. The fact that the ozone sensitivity is not systematically correlated with the time the respective cultivars take to reach harvest maturity can be most clearly seen from two studies that included a large number of rice cultivars Akhtar et al. (2010) and Sawada et al (2009).

Akhtar et al. 2010 studied four different Bangladeshi cultivars two of which had a longer (120 day) growth period and two of which had a shorter 90 day growth period. Both sets of cultivars, the one with the shorter 90 and the one with a longer 120 day growth period, included one ozone sensitive and one ozone resistant cultivar. Similarly Sawada et al. 2009 studied cultivars that took between 99 and 143 days from emergence to harvest. Two cultivars with almost identical growing periods IR 64 and IR36 (~120 days) stand at opposing ends when it comes to the ozone sensitivity of the studied indica cultivars, while suphanburi a cultivar with a ~140 day growth period shares its lower sensitivity to elevated ozone mixing ratios with IR64.

We would like to stress that the anonymous reviewer's viewpoint is incorrect in terms of implying that exposure for the full growth period will lead to unrealistic high yield losses! Exposure for the full growth period will lead to more robust estimates, while exposure-response curves based on experiments that limited fumigation to certain growth stages, can suffer from a systematic bias. It should be noted, that in the real world, the crop has no shield that protects it from ozone from emergence till 3 months prior to harvest.

If indeed the damage for wheat occurs mostly during anthesis & grain filling as suggested by Picchi et al. 2010 and Mills et al 2007, (i.e. damage is limited to the last 3 months prior to harvest), the slope of the curve in Figure 6 would become steeper for the South Asian wheat cultivars (i.e. the implication would be that the cultivars are even more sensitive). According to that hypothesis, early fumigation does not affect the crop yield and hence the observed loss would not change for a delayed onset of fumigation (anthesis & grain filling are part of the 3 month prior to harvest time window) while AOT40 would decrease (due to the fact that AOT is a cumulative index and a shorter time window necessarily leads to a lower number). It is, therefore, unlikely that the manner in which we presented the results are biased towards higher sensitivity, by considering a longer rather than shorter exposure period while deriving the exposure-yield relationship. As the data presented in figure 4&6 was acquired from crops exposed through the above ground growth stages, we considered ambient ozone for the same period in order to calculate RY and economic losses.

We would also like to emphasize that this criticism cannot be applied to crops other than wheat, as Mills et al. 2007 derived the exposure-yield relationship for those crops only based on studies that exposed the crops to ozone from emergence to harvest. Mills et al. 2007, p 2632

Changes in the manuscript:

We added the following text to clarify this

Figure caption of figure 4. "In all studies presented in this figure rice plants were exposed to elevated ozone from the date of transplantation till harvest."

Figure caption of figure 5."In all studies on South Asian cultivars wheat was exposed to elevated ozone levels from emergence to harvest, while the European and American exposure-response curves include datasets acquired on wheat crops that exposed to elevated ozone during the last 3 months prior to harvest."

Reviewer comment:

(Table 6 and sections 3.2, 3.2.1, 3.2.2, 3.3) Mills exposure functions are based on 3 months growing season, therefore while estimating crop yield losses based on Mills functions one generally consider 3 months growing period of exposure regardless of days from emergence to maturity. Here, Authors' have considered around 4-5 months period for rice and 5-5.5 months for wheat, and 6 months for cotton. Using Mills exposure functions and accumulated ozone above 40 ppb for more than 3 months will therefore provide unreal estimates.

Authors' response:

As stated in the supplementary material we have considered 4 months for rice and 4 to 4.5 months for wheat (not 4-5 months period for rice and 5-5.5 months for wheat). Mills et al. 2007, p 2632

considered only crops exposed from emergence to harvest except for wheat and tomato. Therefore, for crops other than wheat this criticism is not valid.

The results in table 6 computed according to the Mills et al. relationship for wheat changes from a RY of 0.27 to 0.26 and 0.18 to 0.21 for the years2011-12 and 2012-13 respectively, if only the last 3 months prior to harvest (February to April) are considered for calculating losses. The extremely high ozone mixing ratios observed in April during the 2 week period when the flag leaves have already turned yellow, but kernel moisture is too high for harvesting, are not of much consequence for ozone damage but result in higher AOT40, if this 2 week period is included. Compared to this, considering the earlier growth stages but removing this period when the crop can no longer be damaged by ozone from consideration results in overall lower AOT40. The harvesting date used in our study can easily be verified by obtaining Modis fire counts for Punjab region as the post harvest crop residue burning occurs right after harvest. This activity peaks in May & November every year (Kumar et al. 2015).

Changes in the manuscript:

We added the following text to Materials and Methods, section 2.4 for readers to keep a few essential details in the main paper.

"To summarize briefly, different rice cultivars take between 90 to 140 days to reach harvest maturity after the \sim 20-30 day old seedlings have been transplanted into the fields. In this study we calculate the accumulated and average ozone exposure (AOT40/M7) for a 4 month period (120 days), which is typical of cultivars popular in the NW-IGP."

"Wheat cultivars take between 4 to 4.5 months from emergence to maturity. High temperatures and water stress during the grain filling stage result in a shorter growth period. Therefore, accumulated and average ozone exposure (AOT40/M7) was calculated for a 4.5 month period for timely sowings and for a 4 month period for late sowings."

Reviewer comment:

Same apply for the exposure functions derived in this study, and therefore author should clearly state that what period of exposure used in deriving the relationship.

Authors' response:

Both exposure-yield relationship and our calculations are based on crops exposed throughout i.e. for more than just 90 days. We have clarified these in all relevant places.

Reviewer comment:

Further: how relevant is the AOT40 or M7 observed in an urban/suburban environment for crops which are likely to be produced in a more rural environment (where ozone levels can be much different)? (Table 3)

Authors' response:

Measurements at the IISER Mohali Atmospheric Chemistry station, are usually not influenced by NO sources that lead to titration of ozone (Sinha et al. 2014, Kumar et al. 2015). High wind speeds prevail during daytime and the prevalent wind direction is from the rural sector (Pawar et al. 2015); therefore, the site is regionally representative. Some of the urban stations in table 3 are likely to be affected by NO titration. In that case, the ozone mixing ratios at urban site should be considered to represent a lower limit for exposure of agricultural crops in the NW-IGP as rural sites downwind of urban centres are usually impacted by equal or higher ozone levels (Logan, 1989) and truly remote sites do not exist in the densely populated NW-IGP.

Reviewer comment:

General:

Page 1, Line 27-28: Authors' have not calculated the technological and economic cost for sustainable mitigation of ozone in India. It is therefore unknown to the reader that how much investment would required for mitigating ozone. I would suggest avoiding line from the abstract 'Mitigation of high : : :: :: Incurred presently"

Authors' response:

We have added the following details in this regard:

Changes in the manuscript:

Page 2383 line 7ff :"For wheat, too, timely sowing is crucial to minimize ozone exposure during the grain filling 5 stage of the crop. New tillage practises that facilitate timely sowing such as relay seeding into cotton and zero or low tillage regimes that incorporates rice straw or machinery to rapidly clear rice residues from the fields are urgently required to facilitate timely sowings. "

has been replaced by:

"For wheat, too, timely sowing is crucial to minimize ozone exposure during the grain filling stage of the crop by advancing the harvest from April end to (March/ early April). New tillage practises that facilitate timely sowing such as relay seeding into cotton and zero or low tillage regimes that incorporates rice straw are urgently required to facilitate timely sowings. Providing a "Happy Seeder" machine to every village in Punjab would cost ~0.04 billon USD. The Happy Seeder sows through the crop residue and leaves it as mulch on the fields. Promoting this technology would not only reduce ambient ozone mixing ratios by curbing crop residue burning, which contributes significantly to ozone precursor emission in post monsoon season (Sarkar et al. 2013), it would also protect the young seedlings against ozone as the mulch acts as protective cover and reduces the dry deposition of ozone onto the leaf surface. Co-benefits of this technology include a higher carbon sequestration in the soil and a higher water productivity of the crop."

Reviewer comment:

Page 1, Line 13-14: Why wheat loss is a factor of two higher in 2012-13 compared to 13-14?

Authors' response:

Ozone levels were a factor 2 higher in 2011-12 compared to 2012-13. The winter 2012-13 had a higher than usual number western disturbances which brought rain, including some very late in the season. The associated wet scavenging of ozone precursors resulted in much lower ozone levels during the grain filling stage of the crop.

Reviewer comment:

Section 3.2, 3.2.1 and 3.2.2: Figure 3 and Figure 4: Variation in sowing dates and exposure shows the significant trend of the crop yields as a function of ozone exposure indices. Here, how can one ignore the influence of micro climate suitable for more yields based on sowing dates and year to year variation of crop yield (because crop yield of rice/wheat reported in figure 3 and 4 are for different years) Is this relationship mere a coincidence? Can Authors' verify whether the yield of rice and wheat is similar during 2007 -2013 for same sowing dates?

Authors' response:

The data presented in Figure 3 and 5 covers different years ranging from 2003-2011. The year to year variations of crop yield have already been accounted for by the fact that individual studies shown were replicated in atleast 2 years. The concerns regarding micro-climate too were addressed in the original experimental design as most studies were performed on different plots in some cases even in different districts. Moreover, studies included different cultivars and tillage practises. The variability in the form of the standard deviation, introduced by all these factors combined, is indicated by the vertical bars on each data point. Similarly the variability in ozone mixing ratios for the same period in different years are indicated as horizontal bars. Different studies were started in different years, therefore the overall period covered is 2004-2008 for rice and 2003-2011 for wheat. It is true that it is difficult to completely disentangle the effect of ozone from that of heat and water stress without a clean air control grown under identical conditions. Heat waves and ozone episodes unfortunately coincide and are likely to reinforce each other when it comes to yield losses. However, the fact that the empirical exposure response curve agrees so well with exposure response curve from OTC studies that do have a clean air control grown under identical conditions in the same field, seems to suggest that most of the yield loss is due to the ozone and not due to meteorological factors.

Reviewer comment:

Section 3.2.1: East-west gradient in sensitivity of local cultivars to ozone exposure is due to difference in exposure period considered in these various studies?

Authors' response:

No. All cultivars were exposed from transplantation to maturity but the data seems to indicate that length of growth period is not the factor controlling sensitivity. Akhtar et al. 2010 had four different Bangladeshi cultivars two of which had a 1 month longer (120 day) growth period. Both the cultivars with the shorter 90 day period from emergence to maturity and the cultivars with a longer 120 day growth period included one more sensitive and one resistant cultivar. Similarly Sawada et al. 2009 studied cultivars that took between 99 and 143 days from emergence to maturity. Two cultivars with almost identical growing periods IR 64 and IR36 (~120 days) stand at opposite ends when it comes to the ozone sensitivity of the studied indica cultivars, while suphanburi a cultivar with a ~140 day growth period shares its lower sensitivity to elevated ozone with IR64. However, it could be that

relative yields obtained during plant chamber studies, in a completely controlled and sheltered system in which temperatures remain within the optimum range throughout and water stress never occurs, are systematically higher (i.e. losses are lower) compared to RY obtained in open top chamber studies under field conditions. We have added a note of caution regarding this.

Changes in the manuscript:

"Bangladeshi cultivars showed the lowest sensitivity and highest relative yields, though this could be owed to the fact that the study was conducted in the sheltered environment of a plant chamber. Pakistani...."

Reviewer comment:

Pl. check. Table 2: I suggest to normalize these RY calculations by the RY obtained for AOT40 = 0, such that the intercept of the relative yield equals 1. Because the value of "a" in the Mills regressions and also the regression obtained in the present study is not always equal to 1 as would be expected for Table AOT40 = 0 (particularly for rice and cotton) (for rice it would mean an additional 5

Authors' response:

We have checked table 2 carefully. Equations taken from other publications are shown as reported by the respective authors. Our equation is based on the regression of the data presented in this study.

We do not agree with the anonymous reviewer that regression lines should be forced through 0 as AOT40=0 does not mean $[O_3]=0$. Forcing the regression through 0 has never been the practice of the scientific community. The "a" value of the regression line carries scientific meaning. If the intercept is less than one then ozone levels below 40 ppbv have a negative impact on the cultivar in question. An intercept > 1 suggest that the plant is only sensitive to higher levels of ozone and does not suffer much damage if ozone levels only slightly exceed the threshold of 40 ppbv.

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