### **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<sub>sto</sub>) when O<sub>3</sub> concentrations were above 40 nl l<sup>-1</sup>. 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 AOT<sub>40</sub>-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* nmolO<sub>3</sub>, m<sup>-2</sup>, PLA, s<sup>-1</sup> with *Y* ranging from 0 to 9 nmolO<sub>3</sub>, m<sup>-2</sup>, PLA, s<sup>-1</sup> (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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### Assessment of crop yield losses in Punjab and Haryana using two years of continuous in-situ ozone measurements

### B. Sinha<sup>1</sup>, K. Singh Sangwan<sup>1,2</sup>, Y. Maurya<sup>1</sup>, V. Kumar<sup>1</sup>, C. Sarkar<sup>1</sup>, B. P. Chandra<sup>1</sup>, and V. Sinha<sup>1</sup>

<sup>1</sup>Department of Earth and Environmental Sciences, Indian Institute of Science Education and Research Mohali, Sector 81, S.A.S Nagar, Manauli PO, Punjab 140306, India <sup>2</sup>Department of Geology, Centre of Advanced Studies, University of Delhi, Delhi 110007, India

Correspondence to: B. Sinha (bsinha@iisermohali.ac.in)

#### Abstract

In this study we use a high quality dataset of in-situ ozone measurements at a suburban site called Mohali in the state of Punjab to estimate ozone related crop yield losses for wheat, rice, cotton and maize for Punjab and the neighbouring state Haryana for the years 2011–

- <sup>5</sup> 2013. We inter-compare crop yield loss estimates according to different exposure metrics such as AOT40 and M7 for the two major crop growing seasons of Kharif (June–October) and Rabi (November–April) and establish a new crop yield exposure relationship for South Asian wheat, maize and rice cultivars. These are a factor of two more sensitive to ozone induced crop yield losses compared to their European and American counterparts.
- Relative yield losses based on the AOT40 metrics ranged from 27–41% for wheat, 21– 26% for rice, 1–3% for maize and 47–58% for cotton. Crop production losses for wheat amounted to 20.8 milliont in fiscal year 2012–2013 and 10.3 milliont in fiscal year 2013– 2014 for Punjab and Haryana jointly. Crop production losses for rice totalled 5.4 milliont in fiscal year 2012–2013 and 3.2 million t year 2013–2014 for Punjab and Haryana jointly. The
- Indian National Food Security Ordinance entitles ~ 820 million of India's poor to purchase about 60 kg of rice/wheat per person annually at subsidized rates. The scheme requires 27.6 Mt of wheat and 33.6 Mt of rice per year. Mitigation of ozone related crop production losses in Punjab and Haryana alone could provide >50% of the wheat and ~10% of the rice required for the scheme.
- The total economic cost losses in Punjab and Haryana amounted to USD 6.5 billion in the fiscal year 2012–2013 and USD 3.7 billion in the fiscal year 2013–2014. This economic loss estimate represents a very conservative lower limit based on the minimum support price of the crop, which is lower than the actual production costs. The upper limit for ozone related crop yield losses in entire India currently amounts to 3.5–20% of India's GDP.
- <sup>25</sup> Mitigation of high surface ozone would require relatively little investment in comparison to economic losses incurred presently. Therefore, ozone mitigation can yield massive benefits in terms of ensuring food security and boosting the economy. Co-benefits of ozone mitiga-

tion also include a decrease in the ozone related mortality, morbidity and a reduction of the ozone induced warming in the lower troposphere.

#### 1 Introduction

India is a rapidly developing nation. Population growth, urbanization and industrial development have led to increasing emissions and have resulted in a statistically significant increase in the tropospheric ozone mixing ratios over the Indian subcontinent in the past decades (Lal et al., 2012). Tropospheric ozone mixing ratios are expected to increase further in the years to come (Giles, 2005).

Tropospheric ozone causes damage to crop at elevated levels and crop yields are extremely important to the Indian economy, as 17% of India's GDP directly depends on agri-10 culture and allied activities (RBI, 2013). However, since 54 % of the total and 72 % of the rural working population of India still relies on agriculture as their main source of income (Census, 2011), crop yields have a much larger overall effect on the economy. Rural demand for a large range of consumer products and cement depends directly on the year's crop yield. Consequently every 1 % decrease in crop yields causes a 0.36 % decrease of In-15 dia's GDP (Gadgil and Gadgil, 2006). Moreover, India has to meet the challenge of feeding 17% of the world's human population with just 2.4% of the world's geographical area and 4% of its freshwater resources (FAO, 2013). Wheat and rice are the most important food crops. In 2010 India produced 20.5 % of the world's rice and 12.4 % of the world's wheat. India is also a major producer of fibre crops (26 % of the world's fibre crops, FAO, 2013), 20 which provide raw material to the domestic textile industry. Punjab with an average cropping intensity of 190 % is considered to be the bread basket of India. It contributes 17.4 % to India's wheat and 10.9 % to India's rice production and produces 60 % of the wheat and 30 % of the rice procured and redistributed by the Department of Food and Public Distribution.

<sup>25</sup> Therefore, it is extremely important to quantify crop losses due to ozone in the North West Indo-Gangetic Plain (NW-IGP) accurately. (Agricultural Statistics, 2013).

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#### 1.1 Ozone effects on plants

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Extensive plant damage due to tropospheric ozone was first observed during the Los Angeles Smog episodes. In the early 1950's, Arie Haagen-Smit and co-workers reported that such plant damage could be reproduced in the laboratory by the reaction of organic trace gases or car exhaust with nitrogen oxides ( $NO_x$ ) in presence of sunlight (Haagen-Smit, 1952; Haagen-Smit and Fox, 1954).

The influence of ozone on vegetation is dependent on the ozone dose and plant phenotype (Pleijel et al., 1991; Heath, 2008; Iriti and Faoro, 2009). Ozone enters leaves through plant stomata during normal gas exchange in the daylight hours and impairs plant metabolism leading to yield reduction in agricultural crops (Wilkinson et al., 2012; Ainsworth et al., 2012; Leisner and Ainsworth, 2012).

In certain phenotypes, ozone exposure interferes with the hormone levels in plants and has been shown to lead to accumulation of ethylene in the leaves. The presence of ethylene in the leaves interferes with the functioning of the hormone abscisic acid (ABA), a hor-

- <sup>15</sup> mone which normally controls stomata closure and reduces water loss under drought conditions (Wilkinson et al., 2012). Consequently, such plant phenotypes when exposed to both drought and O<sub>3</sub> stress 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 <sup>BS</sup>: plants are frequently exposed to temperature or water stress 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.

In phenotypes that are unable to control their stomata opening under ozone stress,  $O_3$  enters the leaf and acts as a strong oxidant causing reactive oxygen stress (ROS) through hydrogen peroxide, superoxide, and hydroxyl radicals that alter the basic metabolic processes in plants (Heath, 2008; Iriti and Faoro, 2009; Kangasjaärvi and Kangasjaärvi, 2014). Ozone has been shown to destroy the structure and function of biological membranes lead-

- <sup>5</sup> Ozone has been shown to destroy the structure and function of biological membranes leading to electrolyte leakage causing accelerated leaf senescence and reduced photosynthesis (Calatayud et al., 2004) and can cause pollen sterility or induce flower, ovule, or grain injury and abortion (Black et al., 2000). In such phenotypes ozone causes visible leaf injury, senescence, and abscission (Kangasjaärvi et al., 2005) and can eventually reduce crop vield even if the damage occurs at early vegetative stages of crop growth by reducing the
- yield, even if the damage occurs at early vegetative stages of crop growth by reducing the amount of healthy green leaf area available for photosynthesis. Symptoms of ozone associated leaf injury have been reported for 27 agricultural crops (Mills et al., 2011a).

Certain other phenotypes respond to ozone stress by reducing their stomata aperture (Torsethaugen et al., 1999; Heath, 2008; Iriti and Faoro, 2009; Ainsworth et al., 2012).
 <sup>15</sup> While this mechanism reduces the amount of ozone taken up by the plant and hence the oxidative stress inside the leaves, it also decreases CO<sub>2</sub> uptake, leading to a reduction in photosynthesis. This affects the carbon transport to roots, reduces nutrient and water uptake and, as a result of this, limits the storage of carbohydrates in the grains. Plants of this phenotype may show little to no visible leaf damage, and often allocate significant
 <sup>20</sup> resources to induced defences following ROS, but crop yields might be very sensitive to O<sub>3</sub> stress during the grain filling stage. Picchi et al. (2010) reported that for different wheat cultivars the phenotypes with least visible leaf damage were often the ones showing maximum reduction in crop yield due to ozone.

The ozone induced physiological damage such as lower yields and inferior crop quality lead to large economic losses (Avnery et al., 2011a, b; van Dingenen et al., 2009; Wilkinson et al., 2012; Giles, 2005).

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#### 1.2 Metrics to assess the impact of ozone on crop yields

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Several large scale programs targeted at assessing the impact of ozone on crop yields have resulted in a variety of different exposure metrics. The National Crop Loss Assessment Network (NCLAN) of the USA was the first systematic and large scale study to assess the impact of  $O_3$  on crops in the world. It relied mainly on Open – Top field fumigation Chambers (OTC) (Heck et al., 1984b; Adams et al., 1989; Lesser et al., 1990) and used seasonal mean daytime exposure metrics (M7 and M12) to relate crop yield losses to ozone mixing ratios (Lefohn et al., 1988; Lee and Hogsett, 1999).

-European researchers and policy makers focused on the critical level concept as a tool

- to identify areas where the critical ozone levels are exceeded. The accumulated exposure over a threshold of 40 nmol mol<sup>-1</sup> (AOT40) was adopted as metric during a workshop in Kuopio, Finland in 1996 and a set of critical level values based on this index has been adopted for crops, forest trees, and semi-natural vegetation (Fuhrer et al., 1997). AOT40 is the most widely used exposure plant response index. It is used by the United Nations
- Economic Commission for Europe (UNECE), the United States Environmental Protection Agency (USEPA), the World Meteorological Organization (WMO) and the World Health Organization (WHO) and is most frequently used in modelling studies targeted at assessing crop yield losses (Avnery et al., 2011a, b; Teixeira et al., 2011; Hollaway et al., 2012; Amin et al., 2013; Ghude et al., 2014; Feng et al., 2015; Chuwah et al., 2015).
- 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. Models using stomatal uptake of O<sub>3</sub> (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<sub>sto</sub>) 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; Gonzalez-Fernandez et al., 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 con-5 sider the accumulated stomatal flux over a specified time interval as  $POD_Y$  (the Phytotoxic Ozone Dose over a threshold flux of Y nmol  $O_3$ , m<sup>-2</sup>, PLA, s<sup>-1</sup> with Y ranging from 0 to 9 nmol O<sub>3</sub>, m<sup>-2</sup>, PLA, s<sup>-1</sup> (Mills et al., 2011b). Studies evaluating the POD<sub>Y</sub> based exposure yield relationship for a wide range of climate zones have emphasised the need for a local parametrization of the stomatal flux model (Fares et al., 2013; Feng et al., 2012; 10 Danielsson et al., 2013; Gonzalez-Fernandez et al., 2013; Yamaguchi et al., 2014). To the best of our knowledge no parametrization for South Asian wheat and rice 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 15 is know for its ozone resistance (Sawada and Kohno, 2009). Depite 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 internatinally agreed upon 20 only for a limited number of crops(Mills et al., 2011b).

<sup>BS:</sup>Crop yields are extremely important to the Indian economy, as 17% of India's GDP directly depends on agriculture and allied activities (RBI 2013). However, since 54% of the total and 72% of the rural working population of India still relies on agriculture as their main source of income (Census2011), crop yields have a much larger overall effect on the economy. Rural demand for a large range of consumer products and cement depends directly on the year's crop yield. Consequently every 1% decrease in crop yields causes a 0.36% decrease of India's GDP (Gadgil and Gadgil 2006). Moreover, India has to meet the challenge of feeding 17% of the world's human population with just 2.4% of the world's geographical area and 4% of its freshwater resources (FAO 2013).

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Wheat and rice are the most important food crops. In 2010 India produced 20.5% of the world's rice and 12.4% of the world's wheat. India is also a major producer of fibre crops (26% of the world's fibre crops, (FAO2013), which provide raw material to the domestic textile industry. Punjab with an average cropping intensity of 190%, contributes 17.4% to India's wheat and 10.9% to India's rice production and produces 60% of the wheat and 30% of the rice procured by the Department of Food and Public Distribution (Agricultural Statistics 2013).

#### 1.3 This study

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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).

<sup>BS:</sup> In this study we use a high quality dataset of in situ ozone measurements at a regionally rep resentative suburban site called Mohali 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 the two major crop growing seasons of Kharif (June October) and Rabi (November April).

#### 2 Materials and methods

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#### 5 2.1 Site description and analytical details

All ozone measurements were performed at the IISER Mohali atmospheric chemistry measurement facility (30.67° N–76.73° E, 310 m a.s.l., Fig. 1). The measurement site is regionally representative (Sinha et al., 2014) and located in the north-west Indo-Gangetic Plain (NW IGP). Ozone measurements from several other sites located in the IGP and the adjoining mountain regions (Fig. 1) will be discussed in detail in Sect. 3.1 to demonstrate that the measurements obtained at the facility are, indeed regionally representative.

The measurement site is located inside a residential campus of around 1.25 km<sup>2</sup> with 800–1000 residents. Local influence is expected to be significant only at low wind speeds (< 1 m s<sup>-1</sup>), which occur only rarely (Sinha et al., 2014; Pawar et al., 2015). The predominant daytime wind direction is west to northwest during winter, summer and post monsoon season and south to southeast during the monsoon season. The fetch region of air masses arriving at the site is dominated by irrigated cropland (marked in light blue in Fig. 1 in the state of Punjab, north-west of the site). During monsoon season south easterly winds bring air masses from a fetch region covering irrigated cropland in the state of Haryana, southeast of the site.

At the measurement site, inlets and meteorological measurements are co-located atop the Ambient Air Quality Station (AAQS) about 20 m above ground. A comprehensive description of the site and its representativeness for N.W. Indo Gangetic Plain can be found in Sinha et al. (2014) and a thorough description of the meteorology of the site for all seasons can be found in Pawar et al. (2015).

Ozone was measured using UV absorption photometry at a time resolution of 1 measurement every minute with an accuracy that is better than 3%, and overall uncertainty of less than 6%. Quality assurance of the large dataset was accomplished by regular calibrations using a NIST traceable ozone primary standard generator and frequent zero drift calibrations. Over the time span reported in this paper, zero drift always remained below  $\pm 0.5$  nmol mol<sup>-1</sup> between two subsequent zero drift calibrations. The drift of the calibration factor during span calibrations was usually less than  $\pm 3\%$  and always below  $\pm 8\%$  even after preventive maintenance. A detailed description of the ozone measurements and the supporting meteorological measurements can be found in Sinha et al. (2014).

#### 10 2.2 Calculation of ozone exposure metrics

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<sup>BS:</sup>The potential of ozone to damage the vegetation has been known for over 50 years, but only in the early 1980s ozone related crop yield losses became a major topic of concern in the environmental science communities all over the world Führer et al. 1997).

- In 1979, US EPA recognized the importance of O<sub>3</sub> dose plant response relationships for assessing the crop yield loss. Crop yield was chosen as parameter to assess the response of agricultural crops to ozone damage (Heck et al. 1984a). In the 1980's, National Crop Loss Assessment Network (NCLAN) of the USA was the first systematic and large scale study to assess the impact of O<sub>3</sub> on crops in the world. It relied mainly on Open — Top field fumigation Chambers (OTC) (Heck et al., 1984a; Adams et al., 1989; Lesser et al., 1990) and used seasonal mean and peak concentration
- values to relate crop yield losses to ozone mixing ratios (Lefohn et al. 1988). Subsequently data use was restricted to daytime data due to the fact that leaf stomata are open and gas exchange is maximized in daylight hours (Lee et al. 1999). 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<sup>-1</sup> (AOT40).

The Mx metric is defined as the mean daytime 7 (M7) and 12 h (M12) surface ozone concentrations during the daylight hours 09:00–15:59 and 08:00–19:59 LT respectively in

(1)

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the crop growing season (Hollaway et al., 2012).

M7 = 
$$\frac{1}{n} \sum_{i=1}^{n} [O_3]_i$$
 for 09:00–15:59 LT

 $BS: M12 = \frac{1}{n} \sum_{i=1}^{n} [O_3]_i$ -for 08:00-19:59 LT-

- European researchers and policy makers focused on the critical level concept as a tool to identify 5 areas where the critical ozone levels are exceeded. The accumulated exposure over a threshold of 4 nmol mol<sup>-1</sup> (AOT40) was adopted as metric during a workshop in Kuopio, Finland in 1996 and a set of critical level values based on this index has been adopted for crops, forest trees, and semi-natural vegetation (Führer et al. 1997). The
- AOT40 is defined as the sum of differences between the hourly ozone concentra-10 tions and 40 nmol mol<sup>-1</sup> during the crop growing season (Fuhrer et al., 1997) for  $[O_3]$  >  $40 \text{ nmol mol}^{-1}$ .

AOT40 = 
$$\sum_{i=1}^{n} ([O_3]_i - 40)$$
 for  $[O_3] > 40 \text{ nmol mol}^{-1}$  (2)

<sup>BS:</sup>AOT40 is the most widely used exposure plant response index and is used by the United Nations 15 Economic Commission for Europe (UNECE), the United States Environmental Protection Agency (USEPA), the World Meteorological Organization (WMO) and the World Health Organization (WHO) (Hollaway et al.2012).

The W126 metric was proposed and adopted in United States by United States Environmental Protection Agency (USEPA) to assess potential vegetation damage from ozone exposure. The 20 W126 metric is defined as the sum of hourly ozone concentrations (weighted by a sigmoidal function) during daylight hours (07:00-18:59 or 08:00-19:59 depending upon location of site) during the crop growing season (Eq. 1). The W126 due to its sigmoidal weighting function gives more weight to higher ozone mixing ratios and is less sensitive to ozone mixing rations between 40 and 50 nmol mol<sup>-1</sup> (Tong et al., 2009). 25

based critical level 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. Stomatal flux of ozone is modelled using a multiplicative algorithm adapted from (Emberson et al. 2000)-

 $-g_{sto} = g_{max} \times [\min(f_{phen}, f_{O_3})] \times f_{light} \times \max[f_{min}, (f_{temp} \times f_{VDP} \times f_{SWP})]$ 

that incorporates the effects of air temperature (f<sub>temp</sub>), vapour pressure deficit of the air surround-ing the leaves (f<sub>VDP</sub>), light (f<sub>tight</sub>), soil water potential (f<sub>SWP</sub>), plant phenology (f<sub>phen</sub>) and ozone concentration (f<sub>03</sub>) on the maximum stomatal conductance (g<sub>max</sub>, mmol O<sub>3</sub>, m<sup>-2</sup>, PLA, s<sup>-1</sup>), 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 POD<sub>Y</sub> (the Phy totoxic Ozone Dose over a threshold flux of Y mmol O<sub>3</sub>, m<sup>-2</sup>, PLA, s<sup>-1</sup> with Y ranging from 0 to 9mmol O<sub>3</sub>, m<sup>-2</sup>, PLA, s<sup>-1</sup> (Mills et al. 2011b).

Out of these parameters <sup>BS:</sup>While M7 <sup>BS:</sup> and M12 gives equal importance to all measurements and accounts for the yield losses due to ozone concentrations of less than 40 nmol mol<sup>-1</sup> while AOT40 <sup>BS:</sup> and W126-gives a higher weight to high ozone mixing ratios (Tuovinen, 2000). Hence, the former<sup>BS:</sup> two are the preferred metrics for will perform better

- tios (Tuovinen, 2000). Hence, the former<sup>55</sup> two are the preferred metrics for <u>will perform better</u> while evaluating plant damage and yield losses at low ozone concentration while the latter will capture the effect of events with very high O<sub>3</sub> mixing ratios on plant physiology and yields better (Hollaway et al., 2012).<sup>BS:</sup> The  $POD_Y$  based exposure yield relationship considers the stromata uptake specifically and have been evaluated using data from a wide range of climate
- 20 zones across Europe, but exposure yield relationships have so far been agreed upon only for a limited number of crops (Mills et al., 2011b).

#### 2.3 Missing data

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For any long term dataset gaps in the data are inevitable due to preventive maintenance, calibrations and technical problems that arise from time to time. The total number and percentage of missing hourly average ambient data for each month from October 2011 to November 2013 are listed in Table 1. For calculating AOT40 and <sup>BS:W126, M7 BS:and M12</sup> continuous and complete daytime data is required, since any missing value can potentially lead to an underestimation of the real ozone exposure. Hence missing values need to be

filled in. For short data gaps of  $\leq$  3 h arising due to zero drift calibration or span calibrations we interpolated the measurements before and after the gap for filling in the missing values. Most gaps in the time series are due to calibrations. For longer data gaps we calculated the average diel ozone profile for the respective month and for each missing hour filled in the monthly average ozone value of the respective hour. In most months less than 5% of the total hours were filled in. Only during the monsoon season the requirement to occasionally purge the system with dry zero air leads to longer data gaps and up to 21% of the hourly averages had to be filled using the method described above.

#### 2.4 Cropping seasons and major crops in Punjab and Haryana

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- Rabi (winter season) and kharif (summer monsoon) are the two main crop-growing seasons in northern India <sup>BS:</sup>, however, in some districts crops are also planted in summer zaid/zayad season (April June). In Punjab, kharif crops include rice, cotton, maize, sugarcane and vegetables (Sharma and Sood, 2003). During rabi season wheat is grown in almost entire Punjab (> 90 % of the area). <sup>BS:</sup>Minor rabi crops are potato, rabi maize, sugarcane, rabi pulses
- 15 and oilseeds (Sharma and Sood). Punjab has an average cropping intensity of about 190%. This means each piece of agricultural land is sown 1.9 times in one year on an average. In recent times there is a tendency to increase the cropping intensity further, in particular in the vicinity of urban centres. In between kharif and rabi season farmers plant potato (sowing: September/October; harvest: November/December) and during zaid/zayad season (April June) farmers plant fodder crops
- 20 (sorghum), pulses (moong dal) or seasonal fruits and vegetables (musk melon, water melon, gourds and cucumber).

In Haryana kharif crops include rice, cotton, sugarcane and in most of the unirrigated areas of Haryana pearlmillet and sorghum (Panigrahy et al., 2010). Mayor Rabi crops in Haryana include wheat, gram, sugarcane and mustard (Panigrahy et al., 2010). <sup>BS:</sup>Zayad season crops include moong and vegetables (Saroj et al., 2014).

The most popular crop rotation systems in Punjab include rice-wheat >70% and cotton-wheat  $\sim 20\%$  as well as maize-wheat <sup>BS:</sup>based crop rotation systems. <sup>BS:</sup>Sorghum-wheat rotation is popular in the Shivalik mountains. In Haryana rice-wheat  $\sim 40\%$  and cotton-

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wheat  $\sim 20\%^{BS}$ ; rice mustard and rice gram rotation is popular in the north but in the dryer parts of Haryana pearlmillet-mustard and pearlmillet-wheat rotations are preferred (Panigrahy et al., 2010). Maize is currently not very popular but heavily promoted as an alternative to rice when a deficient monsoon is anticipated.

- The present study investigates crop yield losses for wheat and maize (Rabi) and rice, maize and cotton (Kharif). In the Supplement S1 we discuss the growth stages during which these crops are potentially sensitive to ozone related yield losses, as well as the time periods during which the plants reach those growth stages in the northern Indo Gangetic plain. To summarize briefly, different rice cultivars take between 90 to 140 days to reach har-
- vest maturity after the 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 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.

#### 2.5 Ozone dose exposure yield relationships

strategy to minimize ozone exposure and maximise crop yields.

 $\frac{\text{We derive specific exposure-yield relationships for Indian wheat and rice cultivars using a two pronged approach.}$ 

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

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 5 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 10 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 15 the West (Table 2) to provide a lower limit and our new the India specific functions to provide

-We use both the old (Mills et al., 2007) the 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 the two.

an upper limit to the possible loss.

<sup>BS:</sup>Till date, only a limited number of field experiments to establish ozone related crop yield losses have been carried out in South Asia. Despite the fact that open top chamber studies in particular those conducted in Pakistani Punjab (Wahid et al., 1995a, b; Maggs et al., 1995; Maggs and Ashmore, 1998; Wahid, 2006; Wahid et al., 2011) and India (Rai et al., 2007, 2010; Rai and Agrawal, 2008; Singh et al., 2009; Singh and Agrawal, 2010; Sarkar and Agrawal, 2010, 2012) suggest that

25 2008; Singh et al., 2009; Singh and Agrawal, 2010; Sarkar and Agrawal, 2010, 2012) suggest that Asian cultivars are more sensitive to ozone related crop yield losses, no ozone exposure dose response relationship specific to the Indian subcontinent has been established in the literature so far.

In this study we use exposure dose-response relationships established in several studies in the 30 West (Table 2) to provide a lower limit to the estimated crop yield losses, but also derive indepen-

dent exposure dose-response relationships for South Asian cultivars. We obtain an yield exposure relationship by relating the crop yields obtained during several regional relay seeding trials to the ozone exposure of the cropand comparing the results with the relative yields (RY) for OTC studies reported in the peer reviewed literature. This provides an upper limit to the possible loss and helps to

 establish whether optimizing the sowing date can be a suitable strategy to minimize ozone exposure and maximise crop yields.

#### 2.6 Yield loss and economic loss calculations

Table 2 summarises the ozone exposure dose–response relationships for relative yield loss (RYL) for wheat, rice, maize and cotton based on the AOT40, <sup>BS:</sup>W126, and M7 <sup>BS:</sup>, and M12 collected from the peer reviewed literature.

All the ozone exposure dose–response relationships previously reported in the literature are based on field studies conducted in the USA or in Europe. Relative yield loss is defined as the crop yield reduction from the theoretical yield that would have resulted without  $O_3$ -induced damages (Avnery et al., 2011a) calculated using the Eqs. (3) and (4)

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$$\mathsf{RYL}_i = 1 - \mathsf{RY}_i$$
 (3)

$$\mathsf{CPL}_i = \frac{\mathsf{RYL}_i}{1 - \mathsf{RYL}_i} \times \mathsf{CP}_i \tag{4}$$

wherein RY<sub>i</sub> stands for relative yield in the year i, CPL<sub>i</sub> stands for crop production loss in the year i and CP<sub>i</sub> stands for the crop production of the same year. The crop production
per fiscal year was taken from the database of the Directorate of Economics and Statistics, Department of Agriculture and Cooperation 2013.

Economic cost loss (ECL) for any crop is defined as the amount of loss in terms of money due to  $O_3$ -induced damages for particular financial year. The minimum ECL is calculated for different crops based on corresponding Minimum Support Prices (MSP) of the same fiscal year using the equation:

$$\mathsf{ECL}_i = \mathsf{CPL}_i \times \mathsf{MSP}_i$$

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The MSP are recommended by Commission for Agriculture Costs and Prices (Directorate of Economics and Statistics, 2013) and are announced by the Government of India at the beginning of each season for each year. These prices are defined as the fixed price at which government purchases crops from the farmers. All our crops of interest come under MSP valuation process. It should be noted, however, that the MSP is typically approximately 50 % less than the market value of the crop and often lower than the production costs. The upler limit for the ECL is calculated using the relationship between CPL due to deficient monsoon rains and the Indian GDP established by Gadgil and Gadgil (2006) using the equation.

Let  $ECL_i$  [%GDP] = RYL<sub>i</sub> [%] × 0.36

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#### 3 Results and discussions

#### 3.1 Ozone seasonal cycle and monthly ozone exposure indices

Figure 2 shows the seasonal box and whisker plot of the daytime (08:00-19:59 LT) 1 h average ozone mixing ratios for the period from October 2011 to January 2014. The highest ozone levels are observed in summer season in April, May and June with median ozone mixing ratios of 60–80 nmol mol<sup>-1</sup> and peak ozone mixing ratios of approximately 130 nmol mol<sup>-1</sup>. This is expected, as favourable conditions such as high temperature, low humidity and high solar radiation favour the photochemical production of O<sub>3</sub> regionally.

After summer, the next highest ozone levels are observed during post monsoon season (October and November) with median ozone mixing ratios of 50–60 nmol mol<sup>-1</sup>. The post monsoon season is characterized by lower levels of solar radiation (range of daytime maxima  $\sim 480-720 \text{ W m}^{-2}$ ) compared to summer season (range of daytime maxima  $\sim 600-920 \text{ W m}^{-2}$ ), but the occurrence of large scale agricultural burning emissions of ozone precursors and a lower boundary layer still results in comparably high ozone levels.

<sup>25</sup> The lowest median daytime ozone mixing ratios of approximately 30 nmol mol<sup>-1</sup> are observed in August, during peak monsoon season, when cloudiness and wet scavenging of ozone precursors limits the photochemical ozone production and during peak winter (DeDiscussion Paper

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cember and January). During winter reduction in the solar radiation, low temperatures and fog result in less photochemical production of  $O_3$ .

Table 3 shows the monthly <sup>BS:</sup> values of ozone exposure indices (increment in AOT40<sup>BS:</sup>, W126, and the monthly M7 <sup>BS:</sup> and M12) for the period October 2011 to January 2014. The

- yearly maximum and minimum monthly value for all indices correspond to the same months, May and August respectively in both years. All indices show maxima during summer (May and June) and post monsoon (October and November) and minima during monsoon (July to September) and winter (December to Febuary), however, the difference between the cumulative metrics (AOT40<sup>BS</sup> and W126), that give higher weight to high values and low or
- no weight to low values and the average based metrics (M7<sup>BS:</sup> and M12) comes out very clearly. For AOT40 <sup>BS:</sup> and W126- the amplitude between peaks (~ 14000 nmol mol<sup>-1</sup>h) and minima (~ 500 nmol mol<sup>-1</sup>h) is very high. The annual peak values are 30<sup>BS:</sup> and 50-times higher for AOT40<sup>BS:</sup> and W126 respectively</sup> compared to the annual minima. For M7<sup>BS:</sup> and M12 peaks are only 2–3 times higher compared to the minima.
- Few studies have so far reported ozone exposure indices over the IGP, however, a number of studies have reported average diel profiles for each month of the year (Jain et al., 2005; Kumar et al., 2010; Sharma et al., 2013) or a time series of average daytime ozone for their site (Maggs et al., 1995; Wahid, 2006; Wahid et al., 2011; Singla et al., 2011).

Table 4 shows the M7 or average daytime ozone calculated from the data in those studies. The seasonality and monthly average daytime ozone levels are similar for all urban and suburban sites in the IGP and the adjoining mountain valleys. However, sites located further to the East report lower M7 values during May and June, due to the higher frequency of summer rain, lower temperatures and earlier onset of the monsoon in the eastern part of the IGP. The only site further to the west for which ozone measurement have been reported

is located close to the centre of the summertime "heat low" (Das, 1962) over the NW IGP and reports summertime and monsoon season M7 that are higher than those observed at our site and a strong anticorrelation of the observed ozone during monsoon season with the intensity of the monsoon rainfall.

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Given the fact that the most reliable crop yield–exposure indices are based on AOT40 and not M7 values, there is urgent need to relate the available observations to AOT40 values. Debaje (2014) did so using a linear relationship. When applied to our data presented in Table 3 the relationship estimates reasonable AOT40 values (slope AOT40 predicted vs. AOT40 observed: 0.93,  $R^2 = 0.87$ ) but performs poorly, while reproducing peak AOT40 values. We find that at our site the actual data follows an exponential curve

 $AOT40 = 0.0201 \times M7^{3.0765}$   $R^2 = 0.94$ 

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and AOT40 values predicted using this curve match peak AOT40 observations better 10 (slope AOT40 predicted vs. AOT40 observed: 1.03,  $R^2 = 0.97$ ).

Several studies attempted to model ozone levels and exposure metrics over the IGP. Deb Roy et al. (2009) modelled AOT40 over the Indian region for the year 2003 using the model REMO-CTM. For the north-western part of the IGP close to the foothills REMO-CTM models 5000–6000 nmol mol<sup>-1</sup> h in May, 1500–2000 nmol mol<sup>-1</sup> h in July and 6000– 7000 nmol mol<sup>-1</sup> h in October. We find that the model underestimates the observed AOT40 in the north-west IGP by a factor 2 to 3 during May and July and reproduces the observa-

tions well during October. Consequently the model would be able to predict crop production losses during Rabi Season better and would underestimate crop production losses during Zayad and Kharif season.

<sup>20</sup> In a more recent study conducted using WRF-Chem, Ghude et al. (2014) predicted ozone daytime concentrations of ~ 50 nmol mol<sup>-1</sup> for Kharif season and ~ 40 nmol mol<sup>-1</sup> for Rabi season for the Chandigarh UT. However, the authors considered only the time windows 15 June to 15 September and December to Febuary for kharif and rabi season respectively. For those time windows, predicted ozone daytime concentrations agree well with the measured M12.

Mittal et al. (2007) inter-compared model predicted ozone with surface observation for the HANK model. The model could not resolve the daytime ozone peak in Delhi and, hence, will perform poorly in predicting AOT40. Comparing the reported values for Chandigarh with
our measurements we find that the model has equal difficulty in resolving the seasonality, in particular the high ozone levels in summer.

Emberson et al. (2009) compared MATCH modelled M7 values with measured surface ozone for Varanarsi and Lahore and found good agreement between model and observations for both cropping seasons. For our site, too, there is an excellent agreement between modelled and observed M7 values (model: 40–50 nmol mol<sup>-1</sup> for Rabi season and 50–70 nmol mol<sup>-1</sup> for Kharif season; observations: 40–52 nmol mol<sup>-1</sup> for Rabi season and 47–64 nmol mol<sup>-1</sup> for Kharif season).

van Dingenen et al. (2009) used a global model (TM5) to predict surface ozone over India and the model reproduces surface observations for our site equally well.

#### 3.2 Ozone exposure yield relationships

Crop yield losses and associated economic losses due to ozone are well constraint for USA and Europe (Avnery et al., 2011). <sup>BS:</sup>,In Asia, even today,O<sub>3</sub>-dose-plant-response relationships developed in the United States and Europe are used to assess the crop yield loss (Deb Roy

- et al., 2009; Ghude et al., 2014), despite the fact that several studies revealed that Asian wheat and rice cultivars are more susceptible to O<sub>3</sub> induced damage than North American their counterparts (Emberson et al., 2009; Oksanen et al., 2013). The analyses of crop production losses so far made for India are based on model derived O<sub>3</sub> mixing ratios (Deb Roy et al., 2009; Ghude et al., 2014; van Dingenden et al., 2009, Avnery et al., 2011a) and apply O<sub>3</sub>-dose-
- plant-response metrics and formulae developed in the US (Adams et al., 1889; Lesser, 1990; Heck et al., 1984b; Wang and Mauzerall, 2004) or in Europe (Mills et al., 2007). Such predictions may underestimate crop yield losses. It has already been pointed out above that for some models, the model predicted daytime O<sub>3</sub> mixing ratios or AOT40 values tend to be lower than the observed O<sub>3</sub> mixing ratios or AOT40 in particular for Zayad and Kharif
   season. Hence model predictions need to be validated and improved using in-situ ozone measurements.

 $O_3$ -dose-plant-response metrics used in the modelling studies conducted so far also underestimate crop production losses due to the fact that South Asian wheat and rice cultivars

are more sensitive to ozone. Emberson et al. (2009) reviewed a large number of Asian open top chamber (OTC) and plant chamber studies, but refrained from deriving an Asia specific dose response curves for wheat and rice due to the large spread in the observational data. Emberson et al. (2009) suggested that the spread could be due to the large variety of different cultivars studied or due to the diversity of experimental conditions. In the same year 5 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). <sup>BS:</sup> This suggests that the spread in the data is indeed caused by differences in the sensitivity of different cultivars. A follow up metabolomic analysis of selected 10 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 15 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 and Agrawal (2012) reported the expression of the Ru-BisCO and several energy metabolism related proteins were adversely affected by ozone 20 exposure in two indica cultivars Malviyadhan 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. 25

<sup>BS:</sup>Consequently 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 15 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. Secondly, we derive a relationship from a series of OTC studies conducted in India (Rai et al., 2007; Rai and Agrawal, 2008; 20

Singh et al., 2009; Rai et al., 2010; Singh and Agrawal, 2010; Sarkar and Agrawal, 2010, 2012) and compare with 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 that may show a very different sensitivity to ozone
 exposure.

### 3.2.1 Rice

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Figure 3 shows the empirical correlation of rice yields and ozone exposure indices for field studies with variations in sowing in Punjab and Haryana. <sup>BS:</sup>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 (Chahal et al, 2007; Jalota et al., 2009; Mahajan et al. 2009; Brar et al., 20120). There is a significant trend of the reported crop yields as a function of

ozone exposure indices (Fig. 3,  $R^2 = 0.58$  for M7 and  $R^2 = 0.57$  for AOT40). For rice late sowing (1st of June) and late transplantation (1st of July) leads to the lowest relative yield losses 18% while early sowing (1st April) and transplantation (1st May) doubles ozone related yield losses 35% (Table 5).

Figure 4 compares the empirical ozone exposure response curve derived from the field data presented in Fig. 3 (solid line) with RY values determined in open top chamber studies (OTC) conducted in India (squares, dash and dot line fit) and Pakistani Punjab (diamonds). <sup>BS:</sup>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 re-

sponse relationship. For studies that did not report AOT40 but did report monthly or seasonal M7, M8 or M12, AOT40 was calculated using the relationship between the respective index

and AOT40 at our site. For M7 all data points of OTC studies lie close to the line derived from the empirical relationship between crop yields and ozone exposure in Punjab. The fit for the OTC studies gives a similar slope as the linear fit of the yield data. Since OTC studies compare yield losses of plants exposed to ozone with those of plants grown under identical conditions but in clean filtered air, the ozone exposure response curve derived from OTC studies of Indian cultivars provides the most accurate estimate of the RYL. A new RYL equation for Indian rice cultivars (Table 2) is derived by fitting all relative yields for Indian cultivars from OTC studies (Fig. 4). We calculate relative yields for all 5 reference periods defined in Supplement S1 both using the old (Mills et al., 2007; Adams et al., 1989) and the

<sup>10</sup> revised RYL relationships.

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It is clear from Fig. 4 and Table 5 that the RY curve derived by Adams et al. (1989) overestimates the RY of *Oryza sativa* L. *Indica* cultivars planted in the IGP significantly and interesting to note that there seems to be a East–West gradient in the sensitivity of local cultivars to ozone exposure. Bangladeshi cultivars showed the lowest sensitivity and highest

<sup>15</sup> relative yields, though this could be owed to the fact that the study was conducted in the sheltered environment of a plant chamber. <sup>BS:</sup> and Pakistani cultivars showed the highest sensitivity to ozone exposure and the lowest relative yields.

Crop production losses calculated using the equation derived based on American studies (Adams et al., 1989) underestimates crop production losses in South Asia by approximately

- <sup>20</sup> 20–30 % Table 5. For AOT 40 both the empirical relationship between crop yields and ozone exposure and the OTC studies conducted in India lead to line fits with similar slopes, however, OTC studies show an intercept of 0.95 for AOT40 = 0 indicating that in South Asia ozone levels below 40 nmol mol<sup>-1</sup> damage local paddy cultivars. While deriving the empirical relationship from field data the RY for AOT40 = 0 was defined as 1 due to the absence of
- clean air controls. The slope <sup>BS:</sup> obtained in the current of the revised equation is steeper than the slope reported by Mills et al. (2007) and the intercept of the Indian OTC studies is also lower, hence RY and crop production losses calculated using the equation derived based on European studies underestimates crop production losses in South Asia by approximately 5–15% (Table 5). Table 5 summarises relative yields for the five reference periods (which

correspond to different sowing dates) and inter-compares RY <sup>BS</sup> obtained by our calculation calculated using the new equation with RY calculated using the old relationships. It can be noted that AOT40 shows a better degree of agreement between the exposure yield relationship of Mills et al. (2007) and the exposure yield relationship for Indian cultivars (Table 5) the difference between the two is generally  $\sim 10$  %. On the other hand, M7 shows a lower degree of agreement between the exposure yield relationship of Adams et al. (1989) and the exposure yield relationship for Indian cultivars (Table 5). The difference between the two is  $\sim 20$  %. After the revision relative yields calculated using the M7 and AOT40 metrics agree within 4 % while previously the discrepancy between the crop yield losses calculated using M7 and AOT40 metrics exceeded 10 %. Our revised ozone exposure crop yield re-

<sup>10</sup> using M/ and AO140 metrics exceeded 10%. Our revised ozone exposure crop yield relationships show significantly lower relative yields than those using the previous exposure response relationships. This can be attributed to the variety of cultivars. The Indian cultivars are more sensitive to O<sub>3</sub> exposure.

#### 3.2.2 Wheat

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- <sup>15</sup> Figure 5 shows the empirical correlation of wheat yields and ozone exposure indices for field studies with variations in sowing in Punjab and Haryana. <sup>BS:</sup>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) re-
- <sup>20</sup> ported 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. There is a significant decrease in yield as a function of increasing ozone exposure (Fig. 5) for both ozone exposure indices ( $R^2 = 0.55$  of M7 and  $R^2 = 0.7$  for AOT40). Based on the values
- <sup>25</sup> ozone exposure indices ( $R^2 = 0.55$  of M7 and  $R^2 = 0.7$  for AO140). Based on the values of slopes and y-intercept, we determined our own values of the relative yield relationship (solid line, Fig. 6) by calculating the relative yield at the observed M7 compared to the yield that would be obtained for background concentrations of 25 nmol mol<sup>-1</sup> ozone. For AOT 40

the relative yield is determined with respect to the yield that would have been obtained for AOT40 = 0).

Figure 6 compares the empirical ozone exposure response curve derived from field data (solid line) with RYL relationships reported in the literature (Mills et al., 2007; Heck et al., 1984b; Lesser et al., 1990; Adams et al., 1989) and with open top chamber studies (OTC) conducted in India (squares, dash and dot line) and Pakistani Punjab (diamonds). Circles show plant chamber studies on Bangladeshi wheat cultivars conducted in Japan. For studies that did not report AOT40 but did report monthly or seasonal averaged M7 or M12, AOT40 was estimated. For M7 most data points of OTC studies with Indian cultivars lie close to the line

- derived from the empirical relationship between crop yields and ozone exposure in Punjab. However, the exposure response relationship for wheat can only be appropriately described by fitting a Weibull function. Since OTC studies compare yield losses of plants exposed to ozone with those of plants grown under identical conditions but in clean filtered air, the ozone exposure response curve derived from OTC studies of Indian cultivars provides the
- <sup>15</sup> most accurate estimate of the RYL. A new RYL equation for Indian wheat cultivars (Table 2) is derived by fitting all relative yields for Indian cultivars form OTC studies (Fig. 6). We calculate relative yields for all 5 reference periods defined in Supplement S1 both using the old (Mills et al., 2007; Adams et al., 1989) and the revised RYL relationships. It is clear from Fig. 6 that the RY curves for winter wheat derived by Lesser et al. (1990) and Heck et al.
   20 (1984b) overestimates the RY of most *Triticum aestivum* L. *cultivars* planted in the IGP.
- For *Triticum aestivum* L. there is no significant trend between cultivars planted in the fort countries. Crop production losses calculated using the M7 index and the equation derived based on American studies (Lesser et al., 1990; Heck et al., 1984b) underestimates crop production losses in South Asia by approximately 10 and 20% for the equation of Heck
   et al. (1984b) and Lesser et al. (1990) respectively (Table 6).

For AOT 40 both the empirical relationship between crop yields and ozone exposure and the OTC studies conducted in India lead to line fits with similar slopes and intercepts. The slope obtained in the current study is steeper than the slope reported by Mills et al. (2007), although a limited number of cultivars planted in the IGP show an exposure RY relationship

similar to that reported by Mills et al. (2007). Cultivars with lower sensitivity to ozone include Bijoy (Akhtar et al., 2010a), Ingilab-91, Punjab-96 and Pasban-90 (Wahid, 2006), HUW234, PBW343 and Sonalika (Singh et al., 2009; Sarkar and Agrawal, 2010). For HUW468 the sensitivity obtained by Singh et al. (2009) and Singh and Agrawal (2010) differ. However, for most cultivars crop production losses calculated using the equation derived based on Eu-5 ropean studies underestimates crop production losses in South Asia. Table 6 summarises relative yield that are obtained by our calculation. For AOT40 the exposure yield relationship of Mills et al. (2007) and the exposure yield relationship for Indian cultivars (Table 6) differ by  $\sim 10-15$  %. For M7 the exposure yield relationship of Lesser et al. (1990) overestimates the yields by  $\sim 20$  % and the exposure yield relationship of Heck et al. (1984b) by  $\sim 10$  % 10 (Table 6). After the revision relative yields calculated using the M7 and AOT40 metrics still show  $\sim 15$  % discrepancy. The quality of the fit for M7 is better than the fit for AOT40. however, given the very steep slope of the M7 curve at > 35 nmol mol<sup>-1</sup> it is credible that cultivars with such a sensitivity to ozone would respond very strongly to even a few days with extremely high ozone and such behaviour will only be captured by the AOT40 index. 15 Daytime peaks with  $\sim$  70–100 nmol mol<sup>-1</sup> are observed in March and April (Fig. 2) during the grain filling stage of the plants and the M7 for the full growth period does not capture such extreme events. AOT40 is the better indicator to accurately reflect exposure when the variance of the amplitude of daytime peak ozone is high. Picchi et al. (2010) reported high sensitivity of wheat cultivars to ozone exposure during the grain filling stage and our obser-20 vations agree well with their finding. Therefore, for South Asian wheat cultivars the revised exposure-response curve using AOT40 will provide the best estimate of the crop production losses. Our revised ozone exposure crop yield relationships show significantly lower relative yields than those obtained by previously used exposure response relationships (-15%)for AOT40). This can be attributed to the variety of cultivars. Most Indian cultivars are more 25 sensitive to high  $O_3$  concentration, though few individual cultivars show higher resistance.

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# 3.2.3 Cotton

Cotton yield data for this region has only been reported in two studies (Jalota et al., 2008; Buttar et al., 2013) and OTC studies on cotton in India have not been conducted till date. Buttar et al. (2013) reported yields for different number of pickings (period 2 and 3) and hence his observations cannot be used to investigate the crop response to ozone. 5 Exposure-yield relationships acquired abroad indicate that cotton is potentially extremely sensitive to ozone induced damage. The yield data from India shows very high variability and no significant influence of ozone on yields, when the results are averaged over 2 years (Jalota et al., 2008). However, there is a significant intra and inter-annual variability of yields as a function of rainfall reported from the site on which the crop was grown (Jalota et al., 10 2008). Since the crop was irrigated sufficiently, this yield dependence on rain should not be related to drought stress. Ozone levels in Punjab during monsoon season are strongly influence by wet scavenging of precursors and cloudiness, hence rain spells can be taken as a proxy for times of low photochemical ozone production. The lowest yields were observed for Period 1 sowings in 2004 that were affected by a prolonged dry spell from 60 15 days after sowing to 100 days after sowing. This corresponds to the period of maximum square production and peak bloom in a cotton plant. In 2005 the same Period 1 sowings received regular rain (every 5-7 days) in the same time period (total 400 mm between 60 to 100 days after sowing) and showed the highest yields (2.4 times the yield of the previous year on average). The Period 2 sowings in 2005 received rain from 40 to 80 days after 20 sowing but were subjected to a dry spell during the second half of the square production and peak bloom period. Observed yields were 1.9 times higher compared to the plants that were subjected to a dry spell during the entire period. Period 2 sowings in 2004 received a short ( $\sim$  7 day) rain spell around 80 days after sowings during the peak square production period and showed yields that were 1.4 times the dry spell yields. Considering the average 25 difference between dry spell and rain spell M7 of approximately 10-20 nmol mol<sup>-1</sup> the observations described above seem to suggest a strong sensitivity of the plant to ozone levels during square production and peak bloom (60-100 days after sowing) but it is difficult to

separate the effect of yield losses due to adverse meteorological conditions from that due to ozone exposure. In the absence of dedicated OTC fumigation studies conducted in India that separate the two effects we use the relationship of Mills et al. (2007) and Heck et al. (1984b) to calculate relative yields (Table 7).

<sup>5</sup> For cotton there are extreme differences of 30–60 % between the relative yields calculated using AOT40 (Mills et al., 2007) and M7 (Heck et al., 1984b). Ozone fumigation studies on Indian cultivars are urgently required to constrain relative yields and crop production losses due to ozone more accurately.

# 3.2.4 Maize

- <sup>10</sup> Maize is planted both as Rabi and Kharif crop, however, cultivation occurs only on a limited area, <u>but maize is heavily promoted as an alternative to rice when a deficient monsoon is</u> <u>anticipated</u>. We could not find any study reporting crop yields for maize planted in Punjab or Haryana in the peer reviewed literature. A recent <sup>BS:</sup>Neither could be identify any 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) add[] and find that in the real world both
- <sup>20</sup> the differences between the revised and old relationship and the overall losses are minor." <sup>BS:</sup> In the absence of suitable data we were unable to derive a ozone exposure RY relationship for Indian maize cultivars and use the relationship of (Mills et al. 2007) and (Heck et al. 1984b) to calculate relative yields (Table 8).

# 3.3 Yield loss and economic loss in Punjab and Haryana

<sup>25</sup> Table 9 summarises the relative yield loss calculated according to different exposure indices. In general crop production losses calculated using the M7 index exposure response

relationships based on American studies conducted in the 1970's and 1980's (Heck et al., 1984b; Adams et al., 1989; Lesser et al., 1990) tend to underestimate the actual yield losses of Indian cultivars, as the M7 index fails to capture the effect of extreme events on plant physiology and yields (Tuovinen, 2000; Hollaway et al., 2012). The old AOT40 exposure-response-relationship by Mills et al. (2007) does not capture the sensitivity of 5 most South Asian cultivars. Only Bangladeshi rice cultivars and a few select wheat cultivars follow this relationship while most Indian wheat and rice cultivars are far more sensitive to elevated ozone levels. We propose a revised relationship (Table 2, Figs. 4 and 6) based on a literature review of OTC studies conducted on Indian cultivars and demonstrate that this relationship adequately describes the empirical relationship between crop yield and AOT40 in field trials that were not aimed at studying the effect of ozone on crops. The revised equation (Table 2) predicts that RYL for Indian cultivars are 1.5–2 times the RYL predicted based

on the equation by Mills et al. (2007).

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A recent modeling studies for the year 2005 predicted RYL of 1 and 1.2% for Punjab and Haryana respectively for wheat and 8.1% for Punjab for rice (Ghude et al., 2014). These 15 relative yield losses are a factor of 15-30 lower compared to the RYL calculated using the same equation (Mills et al., 2007) but employing in-situ measurements for calculating AOT40 for wheat and a factor of 1.5 to 1.8 lower for rice (Table 9 Column RYLAOT40, Mills et al., 2007).

Debaje (2014) estimated the crop production loss of winter wheat based on a review 20 of measured ozone mixing ratios published in the peer reviewed literature for the years 2000–2007. The calculated relative yield losses both based on the M7 exposure response relationship for winter wheat proposed by Lesser et al. (1990) of 10.8% and for the AOT40 based exposure response relationship by Mills et al. (2007) of 29.8 % RYL for Punjab and

Haryana agree well with crop yield losses calculated by applying the same equations, to 25 our in-situ observations (Table 7) for the years 2011–2014 (Table 9 Column RYLAGT40, Mills et al., 2007). This indicates that the underestimation of RYL by Ghude et al. (2014) is due to an underestimation of the AOT40 values during the wheat growing season in the north west IGP caused by the fact that the Ghude and co-workers only considered December

to February as the ozone sensitive growth periods and excluded the months of March and April, which show the highest AOT40 values in the growing season of wheat. However, in the NW-IGP the grain filling stage of the crop is only reached in March and wheat has been shown to be extremely sensitive to high ozone during the grain filling stage (Picchi et al., 2010). Avnery et al. (2011a) used the Mozart-2 model to predict national average RYL of 25–30 % for wheat using the AOT40 based equation, which agrees well with our

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- observations. van Dingenen et al. (2009) using TM5 model predicted RYL ranging from 20-30% for wheat, 10-15% for rice and 1-3% for maize for the year 2000, which agrees well with the observations.
- Table 10 shows the crop production, crop production loss and MSP for the fiscal year 2012–2013 and 2013–2014. Data on crop production was obtained from the following sources: Directorate of Economics and Statistics (2013); Agricultural Statistics (2013), Procurement data was obtained from the Food Corporation of India (2013). For the fiscal year 2013–2014 data for Punjab are based on estimates while final data for Haryana was obtained from Department of Agriculture Haryana (2014). The table also presents economic
- cost losses calculated for wheat, rice, maize and cotton using the old (Mills et al., 2007) and revised exposure-yield relationship. The losses are present for both Haryana and Punjab separately and cumulatively.

The highest crop production loss is seen for wheat: 20.8 million t in fiscal year 2012–2013 and 10.3 million t in fiscal year 2013–2014 for Punjab and Haryana jointly. Ghude et al. (2014) predicted crop production losses of 0.25 million t only for the year 2005 for both states. The discrepancy is mostly due to the fact that this study assumed the ozone sensitive growth period of wheat lasts only from December to February and, hence, did not capture the effect of the high AOT40 during the grain filling stage of the crop in March (factor ~ 15–30) and partially due to the revision of the exposure response relationship (Table 2; factor ~ 2). Debaje (2014) estimated crop production losses of 10.9 million t year<sup>-1</sup> on average for both states combined. The estimate falls within the same order of magnitude as our estimate. Avnery et al. (2011a) estimated CPL of 26 million t for entire India but did not resolve losses for individual states. Economic cost losses amount to INR 243.67 billion

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Rice shows crop production losses of 5.4 million t in fiscal year 2012–2013 and 3.2 million t in fiscal year 2013–2014 for Punjab and Haryana jointly. Ghude et al. (2014)
<sup>5</sup> predicted crop production losses of 0.85 million t only for the year 2005 for both states. The discrepancy is caused both by an underestimation of the AOT40 due to the fact that the author considered a shorer ozone sensitive growth period (factor 1.5–1.8) and the revision of the exposure yield relationship (Table 2) to account for the sensitivity of Indian rice cultivars (factor 1.9). Economic losses amount to INR 67.42 billions and INR 42.34 billions for the fiscal year 2012–2013 and 2013–2014 respectively. At an exchange rate of 60 INR/\$ this amounts to USD 1.1 and 0.7 billion respectively.

The Indian National Food Security Ordinance entitles  $\sim 820$  million of India's poor to purchase about 60 kg of rice/wheat per person annually at subsidized rates. The scheme requires 27.6 Mt of wheat and 33.6 Mt of rice per year. Cutting down ozone related crop production losses in Punjab and Haryana alone could provide > 50% of the wheat and 10% of the rice required for the scheme.

For cotton and maize economic losses amount to INR 79.15 billion and 47.50 billion (USD 1.3 and 0.8 billion) for cotton and INR 0.18 billion and INR 0.24 billion (USD 3 and 4 million) for maize in the fiscal year 2012–2013 and 2013–2014 respectively.

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The total economic losses for the agricultural sector in Punjab and Haryana amount to INR 390.89 billion (USD 6.5 billion) in the fiscal year 2012–2013 and INR 223.34 billion (USD 3.7 billion) in the fiscal year 2013–2014. The loss estimates presented above underestimate the real economic losses due to ozone on several accounts.

Firstly, the crop is valued only at the MSP for common grade crops. The MSP is often even lower than the actual production cost and the economic value of the crop is typically much higher. This is particularly true for high quality rice varieties like Basmati.

Secondly, we do not account for the losses in the food processing sector and other allied industries. The value gain from MSP to final end consumer product ranges from a factor of 2 to 20 for food crops to a factor of > 100 for cotton.

Thirdly, this calculation does not consider the relationship between the rural demand for consumer products and rural income. 78% of the rural population depends on agriculture as primary source of income. Hence, rural income is affected strongly by crop yields.

Previous studies investigating the relationship between monsoon rainfall, food grain production and the Nations GDP for the years 1951–2003 (Gadgil and Gadgil, 2006) found that one percent decrease in food grain production due to deficient monsoon lead to a 0.36 % decrease in Indias GDP. Ozone related crop production losses are likely subject to the same multiplication factor. With relative yields losses currently ranging from 10 to 58 % for the different crops (Avnery et al., 2011a; van Dingenen et al., 2009), the real economic burden
of current ozone levels in terms of the Indias GDP is likely to fall into the range from 3.6 to 20 % (Eq. 8).

### 4 Conclusions

Using a high quality dataset of in-situ ozone measurements in the NW-IGP and yield data from the two neighbouring states of Punjab and Haryana we derived a new crop yield ozone exposure relationship for Indian rice and wheat cultivars. Indian cultivars are a factor of 2–3 more sensitive to ozone compared to their European and South East Asian counterparts. Relative yield losses based on the AOT40 metrics ranged from 30–42 % for wheat, 22–26 % for rice, 9–11 % for maize and 47–58 % for cotton.

Crop production losses for wheat amounted to 20.8 million t in fiscal year 2012–2013 and 10.3 million t in fiscal year 2013–2014 for Punjab and Haryana jointly. Crop production losses for rice totaled 5.4 million t in fiscal year 2012–2013 and 3.2 million t year 2013–2014 for Punjab and Haryana jointly. Cutting these ozone related crop production losses alone could provide 50 % of the wheat and 10 % of the rice required to provide 60 kg of subsidized wheat/rice to  $\sim$  820 million of India's economically weaker sections of society.

The lower limit for economic cost losses in Punjab and Haryana amounted to USD 6.5 billion in the fiscal year 2012–2013 and USD 3.7 billion in the fiscal year 2013–2014. The upper limit for the ozone related economic losses incurred at current ozone

levels for entire India amount to 3.5–20% of India's GDP. The wealth gained by mitigating tropospheric ozone and decreasing ozone related economic losses would be distributed among a large group of beneficiaries, as 54% of the India's population and 79% of India's rural population still rely on agriculture as their principle source of income. Co-benefits of ozone mitigation include a decrease in the ozone related mortality and morbidity, a reduction of healthcare related costs and the number of workdays lost and a reduction of the ozone induced warming in the lower troposphere.

At current tropospheric ozone levels optimizing the sowing date of rice towards sowing at the start of June and transplantation in the first week of July can increase crop yields substantially by reducing ozone exposure of the crop. Reaching out to farmers for promot-10 ing this change in cropping practise will yield co-benefits in terms of increasing the water productivity of the crop and preserving precious ground water. It will also increase the profit margin, as farmers often run tubewells on diesel, whenever grid power supply is not available.

- For wheat, too, timely sowing is crucial to minimize ozone exposure during the grain 15 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 <sup>BS:</sup> or machinery to rapidly clear rice residues from the fields are urgently required to facilitate timely sowings. Proving a "Happy Seeder"
- machine to every village in Punjab would cost ~0.04 billon USD. The Happy Seeder sows 20 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
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technology include a higher carbon sequestration in the soil and a higher water productivity of the crop.

<sup>BS:</sup>However, 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.

<sup>BS:</sup> Mitigating the increasing tropospheric ozone levels in India remains a challenging task for policy makers and the regulatory authorities. Enforcing existing legislation aimed at reducing the emission of ozone precursors remains a challenge even in metropolitan cities. Most area sources of ozone precursors, which include domestic cooking and heating, emissions from cottage industries, waste

burning and crop residue burning are either not within the purview or not within the reach of the regulatory authorities. Low cost indigenous solutions which are attractive alternatives to the existing technologies are urgently required to curb precursor emissions and should be a major research focus. Developing suitable solutions requires interdisciplinary efforts, as technical feasibility, costs and social acceptability of the proposed solutions needs to be assessed in order to ensure widespread

15 implementation.

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**Table 1.** Total number (*N*) of missing hourly average ambient data (mh) and total number of hours per month (th) and percentage (%) of missing hourly average ambient data for each month and number of short ( $\leq$  3 h) and long (> 3 h) data gaps.

month	mh/th [ $N/N$ ]	missing values [%]	short gaps [N]	long gaps [N]
Oct 2011	0/670	0.2		0
Nov 2011	2/0/2	0.3	2	0
Dec 2011	2/120 A/7AA	0.5	י 2	0
Jan 2012	3/711	0.5	1	0
Eab 2012	1/606	0.4	1	0
Mar 2012	4/744	0.1	0	1
Apr 2012	45/720	6.3	2	1
May 2012	13/744	17	5	1
Jun 2012	3/720	0.4	2	0
Jul 2012	153/744	20.6	1	1
Aug 2012	57/744	7.7	2	1
Sep 2012	92/720	12.8	2	1
Oct 2012	8/744	1.1	2	1
Nov 2012	4/720	0.6	4	0
Dec 2012	33/744	4.3	2	2
Jan 2013	1/744	0.1	1	0
Feb 2013	1/672	0.1	1	0
Mar 2013	25/744	3.4	1	1
Apr 2013	5/720	0.7	2	0
May 2013	3/744	0.4	1	0
Jun 2013	108/720	15.0	1	3
Jul 2013	63/744	8.5	1	2
Aug 2013	73/744	9.8	1	1
Sep 2013	33/720	4.6	1	3
Oct 2013	42/744	5.6	1	1
Nov 2013	49/720	6.8	2	2
Dec 2013	2/672	0.3	2	0
Jan 2014	2/720	0.3	1	0
Feb 2014	4/744	0.5	2	0

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Crop	Index	Exposure-RY relationship	References
Rice	M7 AOT40 POD <sub>1</sub> 0 M7 AOT40	$\begin{aligned} RY &= e^{-(M7/202)^{2.47}} / e^{-(25/202)^{2.47}} \\ RY &= -0.0000039 \times AOT40 + 0.94 \\ RY &= 0.996 - 0.487 \times POD_10 \\ RY &= e^{-(M7/86)^{2.5}} / e^{-(25/86)^{2.5}} \\ RY &= -0.00001 \times AOT40 + 0.95 \end{aligned}$	Adams et al. (1989) Mills et al. (2007) Yamaguchi et al. (2014); ozone resistant rice this study, Indian rice cultivars this study, Indian rice cultivars
Wheat	M7 M7 AOT40 POD <sub>6</sub> M7 AOT40	$\begin{split} &RY = e^{-(M7/137)^{2.34}} / e^{-(25/137)^{2.34}} \\ &RY = e^{-(M7/114)^{1.8}} / e^{-(25/114)^{1.8}} \\ &RY = e^{-(M7/186)^{3.2}} / e^{-(25/186)^{3.2}} \\ &RY = -0.0000161 \times AOT40 + 0.99 \\ &RY = 1 - 0.038 \times POD_6 \\ &RY = e^{-(M7/62)^{4.5}} / e^{-(25/62)^{4.5}} \\ &RY = -0.000026 \times AOT40 + 1.01 \end{split}$	Lesser et al. (1990); winter wheat Heck et al. (1984b); winter wheat Adams et al. (1989); spring wheat Mills et al. (2007) Mills et al. (2011b) this study, Indian wheat cultivars this study, Indian wheat cultivars
Maize	M7 AOT40 AOT40	$\begin{aligned} RY &= e^{-(M7/158)^{3.69}} / e^{-(25/158)^{3.69}} \\ RY &= -0.0000036 \times AOT40 + 1.02 \\ RY &= -0.0000067 \times AOT40 + 1.03 \end{aligned}$	Heck et al. (1984b) Mills et al. (2007) Indian maize Singh et al. (2014)
Cotton	AOT40 M7	$\begin{split} \mathbf{RY} &= -0.000016 \times \text{AOT40} + 1.07 \\ \mathbf{RY} &= e^{-(\text{M7}/152)^{2.2}} / e^{-(25/152)^{2.2}} \end{split}$	Mills et al. (2007)Beck et al. (1984b)

**Table 2.** Exposure–relative yield (RY)relationships established in the literature and comparison with our own exposure–relative yield relationships. RY stands for Relative Yield.

Table 3. Monthly values of BS: ozone exposure indicesM7 and increment in AOT40 in the respectivemonth for the period October 2011 to January 2014.

Month	AOT40	M7
Oct 2011	7770	71
Nov 2011	6150	63
Dec 2011	2879	46
Jan 2012	1705	39
Feb 2012	2729	47
Mar 2012	5391	57
Apr 2012	7286	64
May 2012	14783	83
Jun 2012	12544	77
Jul 2012	4005	49
Aug 2012	478	32
Sep 2012	2760	46
Oct 2012	6951	63
Nov 2012	5041	57
Dec 2012	1820	42
Jan 2013	1372	32
Feb 2013	1133	37
Mar 2013	3714	51
Apr 2013	7608	64
May 2013	13381	80
Jun 2013	8123	63
Jul 2013	3014	46
Aug 2013	883	37
Sep 2013	3310	49
Oct 2013	4968	55
Nov 2013	4730	56
Dec 2013	2617	43
Jan 2014	1370	36

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**Table 4.** Comparison of the average montly ozone exposure indices observed at at suburban site in Mohali with measurements at other urban<sup>b, c, d, e, f, g, h</sup> and suburban <sup>i, j</sup> sites in the IGP and nearby remote mountain<sup>1</sup> and suburban valley<sup>k</sup> sites indicated in Fig. 1. Letters denote the following sources: <sup>a</sup>this study, <sup>b</sup>Maggs et al. (1995), <sup>c</sup>Wahid (2006), <sup>d</sup>Wahid et al. (2011), <sup>e</sup>Jain et al. (2005), <sup>f</sup>Ghude et al. (2008), <sup>g</sup>Satsangi et al. (2004), <sup>h</sup>Singla et al. (2011), <sup>i</sup>Tiwari et al. (2008), <sup>j</sup>Rai and Agrawal (2008), <sup>k</sup>Sharma et al. (2013), <sup>I</sup>Kumar et al. (2010). Except for a, f and i values in this table were calculated from the available diel profiles or time series plots.

Site	Mohaliª	Mohaliª	Lahore <sup>b</sup>	Lahore <sup>c, d</sup>	New Delhi <sup>e</sup>	New Delhi <sup>f</sup>	Agra <sup>g</sup>	Agra <sup>h</sup>	Varanarsi <sup>i, j</sup>	Kullu <sup>k</sup>	Nainital <sup>I</sup>
Years	2011-	2011-	1992-	2003-	2001	1997-	2000-	2008-	2003-	2010	2006-
	2014	2014	1993	2004;		2004	2002	2009	2005		2008
				2007							
Index	M7	M12	10:00-	08:00-	M7	11:00-	09:00-	09:00-	M12	M7	M7
			16:00	16:00		18:00	18:00	17:00			
Jan	36	32	40	66	35	32	56	28	35	46	38
Feb	42	37	48	80	57	46	11	45	41	53	42
Mar	54	48	47	92	60	50	45	52	48	70	43
Apr	64	58	52	96	62	55	19	60	53	65	61
May	82	74	-	-	50	55	19	61	56	77	63
Jun	70	66	61	95	41	41	27	46	51	62	41
Jul	48	45	43	93	51	30	16	22	34	48	27
Aug	35	31	48	84	30	24	11	12	25	-	23
Sept	48	42	55	69	45	30	25	29	29	-	27
Oct	63	51	58	60	56	40	36	42	42	58	40
Nov	59	46	33	53	53	41	53	51	41	53	43
Dec	44	38	36	57	56	34	30	34	37	53	39

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**Table 5.** Ozone exposure according to different exposure indices and relative yields for rice. Data for the five periods used to plot Figure 3 is provided in the table. Period (P) 1-3 correspond to the periods in which rice is usually grown in Punjab and Haryana and the average yield loss of these three periods is used to calculate crop production loss and economic loss for each fiscal year.

Time	AOT40	M7	RY <sub>AOT40</sub> Mills et al. (2007)	RY <sub><i>M</i>7</sub> Adams et al. (1989)	RY <sub>AOT40</sub> Indian OTC studies	RY <sub>M7</sub> Indian OTC studies
2012 P1	25641	55	0.84	0.97	0.69	0.72
2012 P2	19788	51	0.86	0.97	0.75	0.77
2012 P3	16715	49	0.87	0.98	0.78	0.79
2012 P4	35640	64	0.80	0.95	0.59	0.62
2012 P5	31853	60	0.82	0.96	0.63	0.67
Average P1-3	20715	52	0.86	0.97	0.74	0.76
2013 P1	20839	53	0.86	0.97	0.74	0.75
2013 P2	15330	49	0.88	0.98	0.80	0.79
2013 P3	12623	47	0.89	0.98	0.82	0.81
2013 P4	29259	60	0.83	0.96	0.66	0.67
2013 P5	25498	56	0.84	0.96	0.70	0.71
Average P1-3	16264	49	0.88	0.98	0.79	0.78

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**Table 6.** Ozone exposure according to different exposure indices and relative yields for wheat. Data for the five periods used to plot Figure 5 is provided in the table. Period 2 (P2) and Period 3 (P3) correspond to the periods in which wheat is usually grown in Punjab and Haryana in the rice-wheat cropping cycle, while Period 4 (P4) and 5 (P5) correspond to the cotton-wheat cropping cycle. The average yield loss of the rice-wheat cycle is used to calculate crop production loss and economic loss for each fiscal year as most of the area is cultivated in the rice-wheat cropping system.

Time	AOT40	M7	RY <sub>AOT40</sub> Mills et al. (2007)	${ m RY}_{M7}$ Lesser et al. (1990)	${ m RY}_{M7}$ Heck et al. (1984b)	RY <sub>AOT40</sub> Indian OTC studies	RY <sub>M7</sub> Indian OTC studies
2012 P1	15843	49	0.73	0.93	0.85	0.60	0.74
2012 P2	15807	49	0.74	0.93	0.86	0.60	0.75
2012 P3	16168	49	0.73	0.93	0.86	0.59	0.75
2012 P4	14754	49	0.75	0.93	0.85	0.63	0.74
2012 P5	17110	52	0.71	0.92	0.84	0.57	0.69
Average P2-3	15987	49	0.73	0.93	0.86	0.59	0.75
2013 Period-1	11384	42	0.81	0.96	0.91	0.71	0.88
2013 Period-2	9887	40	0.83	0.96	0.92	0.75	0.90
2013 Period-3	11375	41	0.81	0.96	0.91	0.71	0.88
2013 Period-4	10012	41	0.83	0.96	0.91	0.75	0.89
2013 Period-5	13817	46	0.77	0.94	0.88	0.65	0.81
Average P2-3	10631	41	0.82	0.96	0.91	0.73	0.89

Time	AOT40	M7	RY <sub>AOT40</sub> Mills et al. (2007)	$\begin{array}{c} RY_{M7} \\ Heck \\ et al. \\ (1984b) \end{array}$
2012 P1	47926	57	0.30	0.91
2012 P2	33728	53	0.53	0.91
2012 P3	48342	56	0.30	0.92
Average P1-2	40825	55	0.42	0.91
2013 P1	40029	55	0.43	0.92
2013 P2	27312	51	0.63	0.92
2013 P3	41046	53	0.41	0.93
Average P1-2	33670	53	0.53	0.92

**Table 7.** Ozone exposure according to different exposure indices and relative yields for cotton. Period1 (P1) and Period 2 (P2) correspond to the periods in which cotton is usually grown.

**Table 8.** Ozone exposure according to different exposure indices and relative yields for rabi and kharif maize.

Time	AOT40	Μ7	RY <sub>AOT40</sub> Mills et al. (2007)	${ m RY}_{M7}$ Heck et al. (1984b)	RY <sub>AOT40</sub> Indian OTC
2012 P1	11346	46	0.98	0.97	0.95
2012 P2	7522	43	0.99	0.99	0.98
Average	9434	45	0.99	0.99	0.97
2011/2012 P3	9824	48	0.98	0.99	0.96
2011/2012 P4	15406	56	0.96	0.98	0.93
Average	12615	52	0.97	0.99	0.95
2013 P1	9496	46	1.00	0.99	0.97
2013 P2	7209	44	0.98	0.99	0.98
Average	8353	45	0.99	0.99	0.97
2012/2013 P3	6219	40	0.99	0.99	0.99
2012/2013 P4	12455	51	0.99	0.99	0.95
Average	9337	46	0.99	0.99	0.97

**Table 9.** Relative yield losses calculated according to different ozone exposure response relationships for rice, wheat cotton and maize.

Time	RYL <sub>AOT40</sub> Mills et al. (2007)	RYL <sub>M7</sub> Adams et al. (1989)	RYL <sub>M7</sub> Heck et al. (1984b)	RYL <sub>M7</sub> Lesser et al. (1989)	RYL <sub>M7</sub> this study	RYL <sub>AOT40</sub> this study
Rabi 2011–2012 Wheat Maize	0.27 0.03		0.14 0.01	0.07	0.25	0.41 0.05
Kharif 2012 Rize Cotton Maize	0.14 0.58 0.01	0.03	0.09 0.01		0.21	0.26 0.03
Rabi 2012–2013 Wheat Maize	0.18 0.01		0.09 0.01	0.04	0.11	0.27 0.03
Kharif 2013 Rize Cotton Maize	0.12 0.47 0.01	0.02	0.08 0.01		0.19	0.21 0.03

**Table 10.** Crop production (CP) for Punjab (PB) and Haryana (HR) and MSP for the fiscal year 2012–2013 and 2013–2014. Crop production loss (CPL) and economic cost losses (ECL) are calculated for wheat, rice, maize and cotton using the old AOT40 based exposure-yield relationship (Mills et al., 2007)<sup>a</sup> and for wheat and rice also using the revised AOT40 based exposure-response relationship<sup>b</sup>. CP and CPL for rice, wheat and maize are given in tonnes (t); CP and CPL in bales (b.)

	CP	CP	CP	MSP	CPL <sup>a</sup>	CPL <sup>a</sup>	CPL <sup>a</sup>	ECL <sup>a</sup>	ECL <sup>a</sup>	ECL <sup>a</sup>	CPL⁵	CPL <sup>♭</sup>	CPL⁵	ECL <sup>b</sup>	ECL <sup>b</sup>	ECL <sup>b</sup>
	РВ	HR	Total		PB	HR	Total	PB	HR	Total	PB	HR	Total	PB	HR	Total
2012– 2013	10 <sup>6</sup> t	10 <sup>6</sup> t	10 <sup>6</sup> t	INR/kg	10 <sup>6</sup> t	10 <sup>6</sup> t	10 <sup>6</sup> t	10 <sup>6</sup> INR	10 <sup>6</sup> INR	10 <sup>6</sup> INR	10 <sup>6</sup> t	10 <sup>6</sup> t	10 <sup>6</sup> t	10 <sup>6</sup> INR	10 <sup>6</sup> INR	10 <sup>6</sup> INR
Wheat	17.28	12.69	29.97	11.7	6.39	4.69	11.09	74 777	54915	129692	12.01	8.80	20.81	140 495	103176	243 671
Rize	11.37	3.98	15.35	12.5	1.85	0.65	2.50	23 137	8099	31 235	4.00	1.40	5.39	49 936	17480	67416
Maize	0.48	0.02	0.50	11.75	0.005	0.0002	0.005	56	2	59	0.015	0.001	0.015	173	7	180
	10 <sup>6</sup> b	10 <sup>6</sup> b	10 <sup>6</sup> b	INR/b	10 <sup>6</sup> b	10 <sup>6</sup> b	10 <sup>6</sup> b	10 <sup>6</sup> INR	10 <sup>6</sup> INR	10 <sup>6</sup> INR						
Cotton	2	2.5	4.5	12737	2.8	3.5	6.2	35 1 7 9	43974	79154						
2013– 2014	10 <sup>6</sup> t	10 <sup>6</sup> t	10 <sup>6</sup> t	INR/kg	10 <sup>6</sup> t	10 <sup>6</sup> t	10 <sup>6</sup> t	10 <sup>6</sup> INR	10 <sup>6</sup> INR	10 <sup>6</sup> INR	10 <sup>6</sup> t	10 <sup>6</sup> t	10 <sup>6</sup> t	10 <sup>6</sup> INR	10 <sup>6</sup> INR	10 <sup>6</sup> INR
Wheat	16.11	11.80	27.91	12.85	3.54	2.44	6.13	45 442	33 285	78727	5.93	4.36	10.32	76 567	56 0 8 2	132 649
Rize	8.16	4.00	12.16	13.1	1.11	0.55	1.66	14 577	7142	21719	2.17	1.06	3.23	28 4 1 5	13922	42 338
Maize	0.56	0.03	0.60	13.1	0.006	0.0003	0.006	74	4	78	0.017	0.001	0.018	228	11	239
	10 <sup>6</sup> b	10 <sup>6</sup> b	10 <sup>6</sup> b	INR/b	10 <sup>6</sup> b	10 <sup>6</sup> b	10 <sup>6</sup> b	10 <sup>6</sup> INR	10 <sup>6</sup> INR	10 <sup>6</sup> INR						
Cotton	2.1	2.0	4.1	13064	1.9	1.8	3.6	24 329	23170	47 499						
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Post-flooding or irrigated croplands Artificial surfaces and associated areas (Urban areas >50%) Rainfed croplands 📕 Water bodies abore Mosaic cropland (50-70%) / vegetation Mohalie (grassland/shrubland/forest) (20-50%) Nainita Mosaic cropland (20-50%) / vegetation New Delhis (grassland/shrubland/forest) (50-70%) Closed to open (>15%) broadleaved Agra evergreen or semi-deciduous forest (>5m) Closed (>40%) broadleaved deciduous forest (>5m) Open (15-40%) broadleaved deciduous forest/woodland (>5m) Open (15-40%) needleleaved deciduous or evergreen forest (>5m) Mosaic forest or shrubland (50-70%) / grassland (20-50%) Mosaic grassland (50-70%) / forest or shrubland (20-50%) Closed to open (>15%) herbaceous vegetation (grassland, savannas or 1000 kn lichens/mosses) Bare areas Sparse (<15%) vegetation

**Figure 1.** Location of our site and surrounding sites for which ozone measurements have been reported superimposed on a land classification map (courtesy ESA GlobCover 2009 Project).



**Figure 2.** Seasonal box and whisker plot of the 1 h average daytime (08:00–19:59 LT) ozone mixing ratios. Whiskers denote the monthly minimum and maximum value, the box the upper and lower quarter value and the horizontal line within the box the median.



**Figure 3.** Empirical correlation of rice yields and ozone exposure indices for field studies with variations in sowing date. 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 (Chahal et al., 2007; Jalota et al., 2009; Mahajan et al., 2009; Brar et al., 2012). Error bars on the *x* axis show the variance in the ozone exposure metrics for the same growth period (see Supplement S1 for definition) for different years. Error bars on the *y* axis show the variance in the yield obtained. Variance is introduced by replicating the study on several test plots, in different districts, plots with different soil properties using different cultivars, replicating the study in several years or transplanting seedlings with a different age at the time of transplanting.



**Figure 4.** Comparison of the empirical exposure response relationship based on field data (solid line) with OTC studies conducted in India (squares with dash and dot fit) and Pakistan (Diamonds, not included in line fit). Large diamonds indicate studies conducted 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. In all studies presented in this figure rice plants were exposed to elevated ozone from the date of transplantation till harvest.





**Figure 5.** Empirical correlation of wheat yields and ozone exposure indices for field studies with variations in sowing date. 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. Error bars on the *x* axis show the variance in the ozone exposure metrics for the same growth period (see Supplement S1 for definition) for different years. Error bars on the *y* axis show the variance in the yield obtained. Variance is introduced by replicating the study on several test plots, in multiple years or varying growing conditions, number of irrigations and tillage practises.



**Figure 6.** Comparison of the empirical exposure response relationship based on field data (solid line) with OTC studies conducted in India (squares with line fit) and Pakistan (Diamonds, not included in line fit). Circles show plant chamber studies on Bangladeshi wheat cultivars conducted in Japan. The exposure response relationship based on American and European studies is plotted in the same graph for comparison. 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.