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Interactive comment

# Interactive comment on "Multi-pollutants emissions from the burning of major agricultural residues in China and the related health-economic effect assessment" by Chunlin Li et al.

Chunlin Li et al.

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#1 Review comments: "Multi-pollutants emissions from the burning of major agricultural residues in China and the related health-economic effect assessment" by Li C. et al. This paper describes results from experimental investigations on EFs of multi-pollutants from crop residues open burning in China, and try to estimate the health-economic effect under different scenarios. Considering the limited EFs for crop residues open burning, new emission data for various types of biomass are always welcome addition to the literature, and should be useful to air quality communities. This paper is reasonably well written. But there are a number of revisions that should be addressed prior to publication. Reply: Thanks for your reviewing! Question 1: When

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comparing EFs in this study with literature data, I notice that only comparable data from literature is included (for example, Line 505 to 507 when comparing OC and EC EFs, Line 605 to 608 when comparing EFs of PAHs, etc). However, it is already known and acknowledged that the EFs of crop residues burning could have a wide range due to different combustion condition, properties of biomass etc...(McMeeking et al., 2009; Reid et al., 2005). Both similarity and differences in EFs should be discussed. Moreover, due to crop residues used in this study were dehydrated at 100 degrees for 24h (thus had much lower moisture content compared with elsewhere), it tends to have a much lower EFs of incomplete combustion product, such as PM and OC from chamber studies (Chen et al., 2010; Hayashi et al., 2014). However, for biomass open burning combustion intensity may be higher than those from chamber studies, and thus this would result in a lower EF. When applying EFs from this study to estimate emission inventories, I would like to suggest that the uncertainties from the effect of moisture content and the burning style should be discussed in this paper. Answer 1: Thanks so much for your suggestion. Chamber burn study has definite advantage over the field burning one, as combustion in the field will be affected by many influence factors including but not limited to meteorological condition, terrain, diffusion, air supply, contamination, fuel issue (fuel type, water content, weight), and burning intensity or fire characters etc. However, after phasing out all the influence variables, how to conduct more exercisable and comparable burning experiment in the lab, and how well the practical chamber burn study can represent field burning should be considered. In general, when combustion efficiency (CE) differences were taken into account, emission factors measured from filed will be reasonably agree with that from chamber burn (Dhammapala et al., 2007). We added more comparison with other studies, and we put the activity data and parameters for the biomass fuel (dry matter fraction, burning efficiency) into the consideration in the final uncertainty assessment for the emission inventories (Line 1071~1105). Lin 561: add in the manuscript which is consistent with the conclusion from Lee et al. (2015) and Giordano et al. (2015)." Line 564: add in the manuscript since EFs in smoke PM1.0 were seldom reported, only smoke PM2.5

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or total particulate matter emissions were collected, which were comparable with the results in this work" Line 567add in the manuscript "were in range of 3.25~15.16 and 3.04~13.20 g kg-1 for the five kinds of crop straws, a high ratio of PM1.0/PM2.5 was observed to be over 90 wt.%, which was in line with size distribution analysis of smoke particles given in Fig. S3 (SI)" Line 570: delete "8.99  $\pm$  5.55 and 7.91  $\pm$  4.67 g kg-1 for the five kinds of crop straws, and over 70 wt.% of SPM was organic components (OM and EC), with average of 73.4 wt.% in PM2.5 and 71.3 wt.% in PM1.0." Line 573: add in the manuscript "Li et al. (2007) measured the emissions from field burning of crop straws via CMB method. PM2.5 EFs for wheat and corn straw were estimated to be  $7.6\pm4.1$  and  $11.7\pm1.0$  g kg-1 (dry basis, MCE > 0.9), which were higher and presented more uncertainties than our result. As study ever found a positive relationship between particulate EFs and moisture content of agricultural residue (Hayashi et al., 2014), it was reasonable that combustion of the dehydrated crop straw produced less smoke aerosol in this work. Hayashi et al. (2014) measured particulate EFs to be 2.2 and 15.0 g kg-1 for rice and wheat straw of ~10 wt.% moisture content, while corresponded EFs increased to 9.1 and 19.5 g kg-1 when water content of straw was  $\sim$ 20 wt.%, and the linear equations between smoke EFs and straw moisture content were furtherly proposed. However, the simple linearity and its application scope should be doubted, as Hayashi et al. only considered two water content levels (10 wt.% vs 20 wt.%) and disregarded influence of combustion efficiency for the fires. PM2.5 EFs given by Dhammapala et al (2006, 2007a, b) were  $4.7\pm0.4$  g kg-1 for wheat straw and  $12.1\pm1.4$  g kg-1 for herbaceous fuel that were burnt using a chamber under flaming phase, and negative response for particulate EFs to combustion efficiency was observed. After all, smoke EFs vary with fires depend on fuel type and moisture, combustion phase, environmental conditions, and some other variables (Reid et al., 2005b)." Line 591: add in the manuscript "The carbonaceous materials (Organic matter and EC) are dominated in SPM, accounting for about 73.4 wt.% for PM2.5 and 71.3 wt.% for PM1.0 on average." Line 594: add in the manuscript "and Li et al. (2016) ever measured OM/OC ratio as ~1.3 for fresh smoke particles via volatility analysis. EFs of EC and OC from this

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work were consistent with most studies, average OC EFs were 4.21 and 3.58 g kg-1 in smoke PM2.5 and PM1.0, and the corresponded EC EFs were 1.09 and 1.01 g kg-1, respectively. These values fell within the ranges (0.9 $\sim$ 9.3 g kg-1 for OC and 0.2 $\sim$ 1.7 g kg-1 for EC) found in other similar sources (Dhammapala et al., 2007; Hayashi et al., 2014; Li et al., 2007; May et al., 2014)." Line 603: add in the manuscript "It was ever reported chamber burn study may overestimate EC EFs due to a misassigned OC-EC split for the heavily mass loaded filter samples (Dhammapala et al., 2007b). Moreover, carbon measurement based on TOT method with NIOSH protocol may overestimate OC fraction by sacrificing EC part compared with that of TOR (Thermal-Optical Reflectance) method with IMPROVE program (Han et al., 2016)." Line 608: delete "EFs of EC and OC from this work agree well with previous study, average EFs of OC were 4.21 and 3.58 g kg-1 in smoke PM2.5 and PM1.0, and the values for EC were 1.09 and 1.01 g kg-1" Line 637: add in the manuscript "To our knowledge, seldom study ever reported source specific EFs of char- and soot-EC for crop straw burnings. Here, particulate char- and soot-EC EFs in fine mode were estimated to be  $0.56 \sim 1.76$  and 0.05 $\sim$  0.42 g kg-1, while char- and soot-EC EFs in smoke PM1.0 were 0.51  $\sim$  1.67 and  $0.06 \sim 0.41$  g kg-1, respectively." Line 646: add in the manuscript "are also fuel types and PM size dependent.. Generally, char-EC/soot-EC is also controlled by combustion mode or even moisture content of biomass fuel, and biomass burning by smoldering at low temperatures results in high char-EC/soot-EC. Chuang et al. (2013) reported char-EC/soot-EC in smoke PM2.5 was 9.4±3.8 for biomass burning (BB), and Cao et al. (2005) proposed the ratio to be 11.6 for BB sources. These values were larger than the present study, as we estimated char-EC/soot-EC in PM2.5 to be  $7.28\pm1.98$ on average. It can be explained by different techniques for EC measurement, char-EC and soot-EC were mostly measured using TOR-IMPROVE method, while TOT-NIOSH method used in this study will overestimate PC fraction in OC-EC split, resulting in less char-EC fraction (EC1-PC) and lower char-EC/soot-EC ratio. Nonetheless, the results were still comparable for the two methods (Han et al., 2016). The char-EC/soot-EC ratio was 6.29 in PM1.0, which was smaller than that in smoke PM2.5, the result indicates

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that SPM comprises a considerable amount of char-EC and char particle has a larger size than soot, in consistent with the conclusion that soot particles are mainly tens of nanometers in size and cluster together into loose aggregates of hundred nanometers, while char particles were reported to be larger with diameter in the range of  $1\sim100~\mu\text{m}$ " Line 680: add in the manuscript "Oxalic acid is the dominated dicarboxylic acids measured in the ambient environment and biomass burning aerosol (Falkovich et al., 2005; Kundu et al., 2010), and oxalic acid EF was measured to be  $2.2 \sim 4.8$  and  $1.6 \sim 3.6$ mg kg-1 for smoke PM2.5 and PM1.0 in present work." Line 752: add in the manuscript "Statistical analysis showed WSA/NH4+ was 0.16  $\pm$  0.03 and 0.18  $\pm$  0.06 in smoke PM1.0 and PM2.5, respectively, which were almost one order of magnitude larger than that in the ambient aerosol (Liu and Bei, 2016; Tao et al., 2016). Tao et al. (2016) ever measured the ratio as a function of particle size during NPF days in Shanghai, and a noticeable enrichment of aminiums for ultrafine particles (<56 nm) was observed with WSA/NH4+ over 0.2, highlighting the competitive role for amines to ammonia in particle nucleation and initial growth of the nuclei, the ratio was then decreased with the increasing particle size, and the final increasing trend was found after  $\sim$  1.0  $\mu$ m, and average WSA/NH4+ for ambient bulk PM1.0 and PM2.5 were 3.2% and 3.5%, respectively." Line 752: add in the manuscript"Hays et al. (2005) estimated total EFs of 16 PAHs to be 3.3 mg Kg-1 in wheat straw burning PM2.5. Korenaga et al. (2001) measured PAHs EFs from rice straw burning to be 1.9 mg Kg-1 in particulate phase. while the value from Jenkins et al. (1996) was 16 mg Kg-1. Dhammapala et al. (2007b) found negative linear response for biomass burning source PAHs emissions to burning efficiency, and under flaming combustion, particulate total 16 PAHs EFs were 2  $\sim$  4 mg Kg-1. Zhang et al. (2011) simulated burning of rice, corn, and wheat straws, the corresponded PAHs EFs were measured as 1.6, 0.9, and 0.7 mg Kg-1 in fine smoke particles, respectively. Great uncertainties for PAHs EFs were evident that relied on burning phase, fuel types, moisture content, and also measurement techniques." Line 806: add in the manuscript "EFs for the sum phenols were  $9.7 \sim 41.5$  and 7.7 and 23.5

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mg Kg-1 for smoke PM2.5 and PM1.0, respectively. Dhammapala et al. (2007a) esti-

mated particulate methoxyphenols emissions to be 35  $\pm$  24 mg Kg-1 for wheat straw burning, while Hays et al. (2005) measured the same compounds to be 6.8 mg Kg-1. Carbonaceous materials like PAHs and Phenols or aromatic and phenolic deviates are the main chromophores in the atmosphere, and the considerable fractions of PAHs and Phenols justify biomass burning as a significant source of brown carbon (Laskin et al., 2015), study has proved  $\sim$  50% of the light absorption in the solvent-extractable fraction of smoke aerosol can be attributed to these strong BrC chromophores (Lin et al., 2016). " Line 866: add in the manuscript"in line with result from domestic burning of wood and field investigation of crop straw burning (Li et al., 2007; Zhang et al., 2012)" Line 990: add in the manuscript "Qin and Xie (2011, 2012) developed national carbonaceous aerosol emission inventories from biomass open burning for multi-years with dynamic burning activity, they believed BC and OC emissions followed an exponential growth from 14.03 and 57.37 Gg in 1990 to 116.58 and 476.77 Gg in 2009. Cao et al. (2006, 2011) calculated smoke aerosol emissions from biomass burning in China for 2000 and 2007 using the same activity data from BAU-I scenarios, national OC and EC emissions were reported to be 425.9 and 103.0 Gg in 2000, however, no evident changes were found for the emissions in 2007, which were assessed to be 433.0 and 104.0 Gg. Huang et al. (2012b) estimated crop burning in the fields with unified EFs and burning rate ( $\sim$ 6.6 %) for all kinds of crops across China in 2006, the estimated annual agricultural fire emissions were about 270, 100, and 30 Gg for PM2.5, OC, and BC, respectively. In present work, agricultural fire PM2.5 emissions in 2012 were allocated into six zones, average contribution in percentage for each zone was compared: NPC (23.1 %) > NC (21.6 %) > PRD (18.4 %) > CC (18.2 %) > WC (9.8 %) > YRD (8.8 %). Furtherly, contribution for summertime emissions was: NPC (35.5 %) > CC (28.8 %) > PRD (21.1 %) > YRD (9.1 %)> WC (5.4 %) > NC (0.1 %), and for autumn harvest emissions: NC (27.8 %) > NPC (19.6 %) > PRD (17.6 %) > CC (15.1 %) > WC (11.1 %) > YRD (8.8 %)" Line 1011: add in the manuscript"It was obviously that the North Plain experienced extensive crop fire emissions during the whole harvest periods, where PM2.5, PM1.0, OC, and BC emissions in 2012 were 233.6, 209.8, 102.3, and 29.4 Gg

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on average. Liu et al. (2015) developed emission inventories from agricultural fires in the North Plain based on MODIS fire radiative power, emission for PM2.5, OC, and BC in 2012 was reported to be 102.3, 37.4, and 13.0 Gg, respectively. However, EFs were also treated as unified values (e.g., Crop burning EFs for PM2.5, OC, and BC was 6.3, 2.3, and 0.8 g Kg-1) in the work of Liu et al. (2015) that was cited directly from Akagi et al. (2011) without considering fuel type dependence of EFs. Zhao et al. (2012) established comprehensive anthropogenic emission inventories for Huabei Region including the North Plain, Inner Mongolia, and Liaoning province, all crop straws were assumed to be burnt in the field, resulting in much more emissions of 446 Gg OC and 160 Gg BC in 2003. A specific temporal pattern for agricultural fire emissions was observed in the Northeast of China (Heilongjiang, Liaoning, and Jilin), where the open burning were mainly occurred in autumn harvest to produce great amount of pollutants (217.5 Gg PM2.5, 89.4 Gg OC, and 29.7 Gg EC), while emissions in the summertime can be neglected."

Question 2: China maps used in Figure 7 are incomplete, part of Xinjiang and Tibet is missing from maps in Figure 7, there should be a reason to explain this. Answer 2: Thanks for your comment. Figure 7 displays geographic distribution of pollutants which is drawn by ArcGIS software, the final graph was designed to contain the figures for all the five versions and also the average one, the map was clipped and zoomed in to show more detailed information of subgraph (the legend). Moreover, information of provincial emissions for Xinjiang, Tibet, and Heilongjiang was not lost. Question 3: Line 228, the definition of MCE (Modified Combustion Efficiency) should be given. MCE= $\Delta$ CO2/( $\Delta$ CO2+ $\Delta$ CO), where  $\Delta$ CO2 and  $\Delta$ CO are the excess molar mixing ratios of CO2 and CO, and thus cannot be monitored directly, as stated on Line 228. Answer 3: Thanks for your reminding, definition of MCE has been corrected and added in Line 229. Line 238: add in the manuscript"with CO and CO2 measuring to determine the burning phase and ensure the repeatability. MCE is defined as  $\Delta$ CO2/( $\Delta$ CO2+ $\Delta$ CO), where  $\Delta$ CO2 and  $\Delta$ CO are the excess molar mixing ratios of CO2 and CO (Reid et al., 2005b)."

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Question 4: When stating there are "significant differences" between means, the statistical tests should be conducted and the results should be also given. Otherwise, there are no significant evidence that one mean differs from the other. The statistical test should be conducted in Line 495, 519, 617, 766... Answer 4: Thanks for your comment, we have added the significance test for the corresponded statistical conclusions in the manuscript, e.g., from multivariate statistical analysis considering fuel type and size range effect on the chemical compositions for smoke PM2.5 and PM1.0, significant difference was found (P<0.05 at 95% CI) Table 1 Multivariate statistical analysis for chemical compositions of smoke PM2.5 and PM1.0 from five agricultural residues burning Species PM2.5 PM1.0 Emission factor Mass fraction Emission factor Mass fraction PM2.5 0.000 PM1.0 0.000 0.650 0.000 OC 0.000 0.275 0.000 0.170 EC 0.000 0.013 0.010 0.189 WSOA 0.004 0.040 0.003 0.049 WSA 0.000 0.011 0.000 0.015 WSI 0.001 0.000 0.000 0.000 SO42- 0.000 0.000 0.000 0.020 Cl- 0.000 0.000 0.000 0.000 NH4+ 0.000 0.000 0.000 0.000 K+ 0.000 0.000 0.000 0.000 THM 0.000 0.030 0.000 0.017 PAHs 0.001 0.008 0.001 0.037 Phenols 0.000 0.019 0.006 0.006 Note: SPSS analysis, P<0.05 means significant difference at 95% confidence interval (CI)

Question 5: Although several ways to estimate uncertainties of the emissions were mentioned in Section 3.3.4 (Line 827 to Line), it is not clear which method is used in this study. For the emission inventory in this study, a discussion of the overall inventory uncertainty is needed and this could be given by considering the uncertainties in each of the terms in the inventory (Eq 5). Answer 5: Thanks for your comment, in the previous manuscript, we only considered the uncertainties for the average emission inventory from the 5 versions using the uncertainty propagation calculation as:  $U_total = \sqrt{(\sum_i \Theta_i(U_i \times x_i) \tilde{A} \tilde{U}^2)}$  $)/(\sum\_\Theta x\_i)[1]U\_total = \sqrt{(\sum\_\Theta tU\_\Theta)}[2]Where U\_is uncertainty in percentage for variatei, x\_i is the ugricate, and equation the property of the property of$ 

Table 2 Uncertainties for the national smoke PM emissions in 2012 (pollutant emission in unit of Gg/yr, 95% CI in percentage) Species BAU-I BAU-II EM-I EM-II NDRC Average PM2.5 1001.1 (-52.3%, 73.5%) 835.4 (-48.7%, 68.8%) 1211.9 (-63.6%,

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from literature (BAU-I and BAU-II), and a uniform distribution with  $\pm$  30% deviation for the rest activity data (crop-to-residue ratio, dry matter fraction, and burning efficiency). Regarding the emission factors, Bond et al. (2004) assumed that most particulate EFs followed lognormal distributions with CV of  $\pm$  50% for domestic EFs, and of  $\pm$ 150% for EFs obtained from foreign studies. Here, we applied the CV of smoke EFs as we measured ones, which were chemical species and fuel type dependent. With randomly selected values within the respective probability density functions (PDFs) of EFs and activity data for each biomass type, Monte Carlo simulation was implemented for 10,000 times, and the uncertainties in national yearly multi-pollutants emissions at 95% CI were obtained for all the 5 versions. Afterwards, uncertainties for the average emission inventories were assessed using the propagation of uncertainty calculation that suggested by IPCC (1997) (method in SI), and all the emission uncertainties were presented in percentage in Table 6. Emissions for water soluble aminiums and organic acids had the vast uncertainties, due to their large deviation in EFs compared with other smoke species. Besides, emissions of BAU versions were more accurate than EM versions, because of more uncertainty addition in the burning rates conversion using economic data for EM versions. Otherwise, burning rates derived from NDRC report were assumed to have less uncertainty, resulting in the least uncertainties in smoke emission assessments. On average of all the 5 versions, mean, 2.5th percentile, and 97.5th percentile values for smoke PM2.5 emissions in 2012 were 1005.7, 758.3, and 1344.6 Gg, respectively. As to OC emissions, mean, 2.5th percentile, and 97.5th percentile values were 432.4, 327.8, and 576.4 Gg, the figure for EC was 134.2, 100.9. and 187.9 Gg. Therefore, the overall propagation of uncertainties for smoke PM2.5, OC, and EC at 95% CI was [-24.6%, 33.7%], [-24.4%, 33.5%], and [-24.2%, 33.3%], respectively. The uncertainties for OC and EC emissions were much less than the work of Qin and Xie (2011), in which emission and uncertainties were 266.7 Gg [-55.9%, 96.1%] for OC and 66. 9 Gg [-53.9%, 92.6%] for EC in 2005" Question 6: Line 215, "costume-built" should be "custom-built"; Citation formatting and styling errors should be corrected carefully. For example, Line 360, References should be cited with

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publication year. Chen et al. (2001) is cited under Cao's publication... Line 374, Qin et al. (2012) is cited, but is missing from References list. Answer 6: "custom-built" has been corrected in Line 215, citation errors have been carefully checked and modified. Line 224: "custom-built" has been corrected Line 65: "Andreae and Merlet, 2001" has been corrected Line 71:"Qin and Xie, 2012"has been corrected and added in the reference list Line 79: "Andreae and Merlet, 2001" has been corrected Line 81:"Qin and Xie, 2012"has been corrected Line 94: "Arora and Jain, 2015" has been corrected Line 123: "Qin and Xie, 2011, 2012"has been corrected Line 148: "Ostro and Chestnut, 1998" has been corrected Line 182: "Reddy and Venkataraman, 2000" has been corrected Line 226: "Zhang et al., 2008a, 2011" has been corrected Line 404: "CAREI, 2000" deleted Line 435: "Cermak and Kuntti, 2009" has been corrected Line 490: "Bell and Hipfner, 1997" has been corrected Line 522: "Aunan and Pan, 2004" has been corrected Line 625: "Arora and Jain, 2015" has been corrected Line 633: "Andreae and Gelencsér, 2006" has been corrected Line 673: "Arora and Jain, 2015" has been corrected Line 702: "Andreae and Gelencsér, 2006" has been corrected Line 711: "Qiu and Zhang, 2012" has been corrected Line 715: "Lee and Wexler, 2013"has been corrected Line 718:"Schade and Crutzen, 1995"has been corrected Line 744: "Arey and Atkinson, 2003" has been corrected Line 798: "Berndt and Boge, 2006"has been corrected Line 849: "Amdur and Chen, 1989" has been corrected

Please also note the supplement to this comment: http://www.atmos-chem-phys-discuss.net/acp-2016-651/acp-2016-651-AC1-supplement.pdf

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