# Authors' Responses to Referees' Comments

## Anonymous Referee #2

The manuscript by Zhu and Kawamura investigates the impacts of biomass burning on TSP (total suspended particulate matter) samples collected at a distinct site (Okinawa Island). The topic is of great scientific concern because biomass burning has important and complex effects on the environment which are still far from being well understood. On the other hand, the results are meaningful for understanding the characteristics of the Asian outflows. However, I think the manuscript is not publishable in its present form and major revision is required.

Response: We thank the referee for the positive evaluation on our work. We have adopted most of the comments to improve the manuscript substantially.

Major comments

(1) The estimation of non-sea-salt water-soluble potassium (nss-K+). As mentioned by the authors themselves, there was no correlation between levoglucosan and nss-K+ during autumn, and a likely cause was the contribution of soil materials to K+. However, the authors did not make any effort to account for the influences of soil materials. Although the correlation between levoglucosan and nss-K+ was more apparent during the other seasons, contribution of soil materials to K+ could also be considerable, especially in spring and winter (when the air mass to the sampling site was mainly from the Asian continent). Therefore, the role of soil materials can not be ignored. The authors can refer to Pio et al. (Atmos. Environ., 42, 7530-7543, 2008) for this point.

In addition, the referee understands that compared to sea salt, the influences of soil materials are more difficult to account for. Another choice is to compare levoglucosan and K+ directly and use a receptor model (e.g., PMF) to quantitatively estimate the biomass burning contribution to K+. In fact, based on the comparison of levoglucosan and nss-K+, the authors just concluded that biomass burning was not necessarily the major source of K+. There is nothing new about this point (e.g., Zhang et al., Atmos. Chem. Phys., 10, 6839-6853, 2010). Therefore, this paper will be more relevant if the authors can quantitatively estimate how much K+ is associated with biomass burning.

Response: We agree with the reviewer`s comment that quantifying $K^+$ balance would be more informative. Based on the comments, we further investigated the source of $K^+$ ion using two methods: a tracer based method following Pio et al. (Atmos. Env., 2008) and the PMF model. The results show that biomass burning accounts for 23-24% of total water-soluble $K^+$.

We added the following sentences to the Experimental section: “We also investigated $K^+$ components using a tracer-based method (Pio et al., 2008) and positive matrix factorization (PMF) model (Paatero and Tapper, 1994) (Supplementary materials).” Please see lines 190-192 in the revised MS. We also modified sentences about relations of anhydrosugars with ions. Please see lines 361-369. We added new Figure 5 as below and supplementary material for PMF procedures, along with the following paragraph (see lines 370-386 in the revised MS):

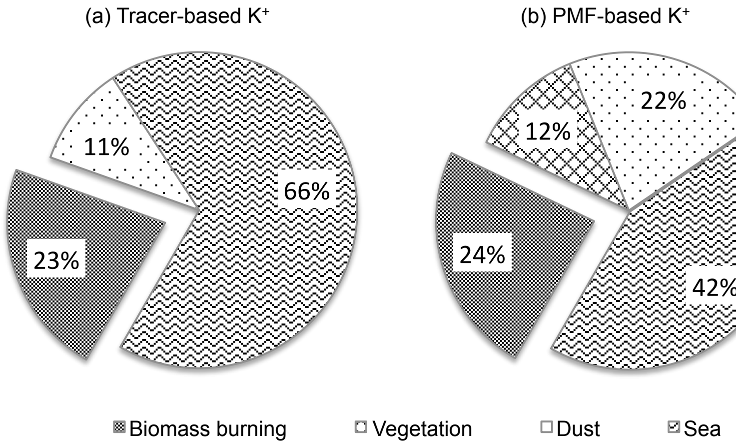

Fig. 5. Components of water-soluble $K^+$ derived by (a) tracer-based method (Pio et al., 2008) and (b) PMF analysis. Details on PMF analysis can be found in the supplementary material.

“To further account for the fraction of BB-derived $K^+$ (bb-$K^+$), $K^+$ balance was investigated using a tracer-based method and PMF model (Supplementary materials). Pio et al. (2008) reported that bb-$K^+$ could be calculated by subtracting ss-$K^+$ and dust-related $K^+$ (dust-$K^+$) from total $K^+$, using a typical $K^+/Ca^{2+}$ ratio of 0.12 for soil dust in Europe. In East Asia, $K^+/Ca^{2+}$

ratios during dust storm periods are reported as 0.012 in Zhenbeitai, northern China (Arimoto et al., 2004), 0.07 in Xi'an, northwestern China (Shen et al., 2009), and 0.12 in Tongliao, northeastern China (Shen et al., 2007). We estimated the $K^+$ component using an average $K^+/Ca^{2+}$ ratio of 0.07 for Asian dust, following the method of Pio et al. (2007, 2008). We found that bb-$K^+$ accounted for 23% of total $K^+$. This result is similar to that (24%) obtained by PMF model (Fig. 5). Interestingly, relation between levoglucosan and tracer-based bb-$K^+$ was not strong, although positive relation was found between levoglucosan and nss-$K^+$. This phenomenon, as well as the seasonal difference of the relations between levoglucosan and nss-$K^+$, might be caused by the following reasons: (1) during open field burning, a large amount of dust-$K^+$ is mixed with bb-$K^+$; (2) dust-$K^+$ and bb-$K^+$ are mixed during atmospheric transport. Being consistent with previous studies, these results suggest that a combined analysis of $K^+$ and other tracers is recommend to characterize BB activities (Zhang et al., 2010)."

(2) The discussion on the stability of levoglucosan. The authors discussed the possible degradation of levoglucosan based on (i) the gradient of anhydrosugars from Okinawa (the sampling site of this study) to Chichijima (1400 km east of Okinawa), and (ii) the gradient of levoglucosan to OC ratio between Changdao, Okinawa and Chichijima. However, results from these locations were measured in different years. It is ridiculous to use this kind of results to discuss the possible degradation of levoglucosan!

Response: Following the comments, we compared the levels of anhydrosugars and levoglucosan/OC ratio among Changdao, Okinawa and Chichijima and reorganized the discussion in the revised MS on the longitudinal differences of the above parameters due to several possible reasons such as deposition, degradation and local OC sources in the island. Although the results were measured in different years, the comparison is still inferable for the atmospheric processes. Similar approach has been conducted by Hu et al. (Scientific Report, 2013), who compared levoglucosan levels in the literature from the Arctic to the Antarctica.

We rephrased the headline of section 3.5 from "L/OC and L/EC ratios for possible degradation of levoglucosan" to "L/OC and L/EC ratios". Please see line 450 in the revised MS. We also added the following sentences in lines 459-464 of the revised MS:

"Here, we discuss the longitudinal gradient of L/OC in Changdao, Okinawa and Chichijima in the context of the Asian outflow. It is worth noting that, in comparison to the other two sites (2001–03), the data of Okinawa aerosols were obtained in different years (2009–12) when the tropospheric oxidizing capability was likely enhanced (Itahashi et al., 2014). However, such comparison is still helpful for understanding atmospheric processes in terms of deposition and degradation of levoglucosan."

(3) Contribution of biomass burning to OC, EC and TSP. There are numerous studies investigating the levoglucosan to OC ratios in biomass burning source emissions, in addition to the two references mentioned by the authors. It has been well documented that the levoglucosan to OC ratios in biomass burning source emissions can vary by a factor of up to 10 among different types of biomass fuels, which is also strongly affected by the combustion conditions. With respect to this study, both the source areas (e.g., China, Mongolia, and Russia) and the source types (e.g., wood combustion vs. burning of crop residuals; open burning vs. domestic burning) of the biomass burning aerosol are highly variable, indicating that it is very difficult to get a representative levoglucosan to OC ratio for the biomass burning source emissions. The authors should try to use a receptor model (e.g., PMF) to estimate the contribution of biomass burning.

Response: We agree with the reviewer that levoglucosan/OC ratios varied depending on biomass burning sources and conditions. Still, the levoglucosan/OC ratio of ~0.082 in the burning source have been widely used, especially in East Asia. In this work, we believe that using levoglucosan/OC ratio of ~0.082 is informative to estimate biomass burning contributions. The estimation is also comparative to other studies using the same ratio. We also agree that PMF model simulations may generate better sources apportionment of WSOC and OC, if enough tracers are available in the model run. We will conduct such work in the future once more tracers representing different source sections are available.

We added following sentences in the revised MS: "Although L/OC ratios in the BB source emissions vary among different types of biomass fuels and burning conditions (e.g., Mochida et al., 2010), the value of ~8.2% have been widely used to estimate BB contributions to OC (Fu et al., 2014; Graham et al., 2002; Ho et al., 2014; Mkoma et al., 2013; Sang et al., 2011).” Please see lines 510-514. We also added a new sentence as follows: “A further estimation on the OC components by PMF analysis may provide more information, which would be our future work.” Please see lines 518-520.

(4) Page 25593. The observed correlation between levoglucosan and ammonium can NOT support the conclusion that ammonium is either emitted by biomass burning directly or from plant nitrogen. The authors should refer to some publications about the emission inventory of atmospheric ammonia. The authors should also keep in mind that correlation does not necessarily mean a same source (in fact this point was mentioned by the authors themselves on Page 25594).

Response: Based on the comments, we revised the discussion section by referring the related publications and adding new sentences on atmospheric $NH_4^+$ as below.

“On the other hand, positive relations were found between $NH_4^+$ and levoglucosan ($r = 0.39$, $p < 0.001$) and mannosan ($r = 0.20$, $p < 0.05$). These results imply that BB in MNA could be an important direct source of $NH_4^+$ (Andreae and Merlet et al., 2001; Akagi et al., 2011). $NH_4^+$ could also be formed in the atmosphere by the protonation of $NH_3$, which was abundantly emitted from livestock excreta, fertilizer application, agricultural soil and plants, biomass burning, and so on (Asman et al., 1998; Huang et al., 2012; Paulot et al., 2014; Sutton et al., 2008; Zhang et al., 2010). The seasonal relations between levoglucosan and $NH_4^+$ showed no correlation in summer and autumn although there is a weak positive correlation in winter and spring. This may be caused by the enrichment of $NH_3$-derived $NH_4^+$ from agricultural activities, and transformation and wet scavenging during atmospheric transport. Levoglucosan did not correlate with nss-$SO_4^{2-}$ ($r$=0.12), indicating that sulfur emission from BB may not be significant.” Please see lines 387-398.

Specific comments

(1) Page 25584, the last paragraph. When describing the amount of biomass burnt by fuel types, domestic fuel is missed.

Response: Based on the comment, we added the term of domestic burnings of biomass in the revised MS as follows.

"Forest burning resulting from both natural and man-made fires for land clearing or other purposes comprises 16%, crop residue burning in the agriculture field comprises 12%, grassland and savanna burning arising from prescribed burning, natural fires, and grassland conversion comprises 7%, and biofuel consumption of fuelwood, agricultural residues, and dried animal waste for cooking and heating comprises 64% of the total burnt biomass across the Asian continent (Streets et al., 2003)." Please see lines 98-103.

(2) Page 25585. In addition to the relative humidity, the temperature at which the quartz filters were equilibrated should also be mentioned.

Response: Taking the comment, we added temperature of ~20 °C in the revised MS. Please see line 130.

(3) Page 25585. Only two filters were kept as blank in this study. Are the blank concentrations (e.g., OC and levoglucosan) comparable between them?

Response: The OC levels in the two blank filters are comparable. Based on the comment, we added new sentence as follows: "OC levels in field blanks were consistently low (1.16 and 1.54 µg per measured unit of 1.54 $cm^2$, < 5% of OC levels in most samples), indicating negligible contamination of the samples." Please see lines 133-135.

(4) Page 25587. How about the low detection limit of mannosan and galactosan?

Response: Because we used levogucosan as surrogate standard for mannosan and galactosan as described in the text, the detection limits of mannosan and galactosan should be the same with that of levoglucosan. One sentence was added in the revised MS concerning the detection limit of mannosan and galactosan. Please see lines 180-182 in the revised MS.

**Anonymous Referee #3**

This MS presents an interesting assessment of anhydrosugars in ambient air in Okinawa, aiming to identify their origin and the impact of biomass burning emmissions on air quality as a function of back-trajectories and ratios between the sugars. The paper is well written and interesting for the general scientific community. I have only one major concern, which is the discussion on the degradability of levoglucosan over time. Almost all of the interpretations in the MS depend on the assumption that levoglucosan is mostly stable in the aerosol phase, such as source region analysis (with back-trajectories) and ratios between anhydrosugars. However, there is no discussion (or very little reference) to whether levoglucosan is stable or not in the atmosphere, and under which conditions. I therefore would suggest publication after this issue is duscussed in detail.

Response: We thank the referee for the positive evaluation of our work. We have newly considered a possible degradation of levoglucosan and its implications to the current work. Specifically, we added one paragraph in the Introduction section on current understandings of the degradation of levoglucosan, and discussed the issue in another paragraph in the Results section. Please see lines 65-82 and 323-337, respectively.

Some specific comments:

- line 1, page 25582: "compositions" should be "composition".

Response: Corrected. See line 7 in the revised MS.

- page 25583, line 28: this is the first reference to the issue of the stability of levglucosan. The discussion is too short, and should be improved considering the implications for the analyses presented in the following sections.

Response: Based on the comments, we added a new paragraph describing the current progresses on studies of levoglucosan degradation as follows.

"Recent studies, however, demonstrated that degradation of levoglucosan occurred on the exposure to high levels of hydroxyl radicals (OH). Kinetic studies on the reactivity of levoglucosan with OH, $NO_3$, and $SO_4^-$ radicals in aqueous phase and the model simulations indicated that levoglucosan can be oxidized by OH radicals in daytime by the rates of 7.2 ng $m^{-3}$ $h^{-1}$ in summer and 4.7 ng $m^{-3}$ $h^{-1}$ in winter for a polluted continental plume (Hoffmann et al., 2010). Based on the chamber experiment on levoglucosan oxidation by OH, Hennigan et al. (2010) reported that levoglucosan has an atmospheric lifetime of 0.7–2.2 days when BB particles are exposed to $1.0 \times 10^{6}$ molecules $cm^{-3}$ of OH. A recent study by flow reactor indicated that pure levoglucosan is degrading by OH with rate of $(9.17 \pm 1.16) \times 10^{-12}$ $cm^{3}$ $molecules^{-1}$ $s^{-1}$ at 25 °C and 40% relative humidity (RH), and the average atmospheric lifetime is 1.2–3.9 days (Lai et al., 2014). The effect of temperature on levoglucosan degradation had been reported consistently positive (Bai et al., 2013; Lai et al., 2014), while the effect of RH is divergent (Hennigan et al., 2010; Lai et al., 2014; Slade et al., 2014). Nevertheless, the study by Bai et al. (2013) indicated that levoglucosan reacts with OH at a rate of $2.21 \times 10^{-13}$ $cm^{3}$ $molecule^{-1}$ $s^{-1}$ at 25 °C, resulting in an atmospheric lifetime of 26 days under the OH level of $2.0 \times 10^{6}$ molecule $cm^{-3}$. Although these results are divergent, levoglucosan is subjected to degradation in BB aerosols during atmospheric transport." Please see lines 65-82 in the revised MS.

- page 25584, line 24: "grassland and savanna burning", just for information, are these natural or man-made fires?

Response: These burnings were not separated for natural and man-made sources. For the clarity, we rephrased the sentences including open-field and domestic burnings to "Forest burning resulting from both natural and man-made fires for land clearing or other purposes comprises 16%, crop residue burning in the agriculture field comprises 12%, grassland and savanna burning arising from prescribed burning, natural fires, and grassland conversion comprises 7%, and biofuel consumption of fuelwood, agricultural residues, and dried animal waste for cooking and heating comprises 64% of the total burnt biomass across the Asian continent (Streets et al., 2003)." Please see lines 98-103 in the revised MS.

- page 25587, line 22: if major WSIIons were determined using IC, why wasn't K+ determined? Why was an indirect measure should be "are probably associated"through Na+ used?

Response: Following the comments, we added/revised sentences as follow: "Based on IC-determined $K^{+}$, $SO_4^{2-}$ and $Na^{+}$, we estimated the fractions of $K^{+}$ ion derived from sea salt (ss) and none-sea-salt (nss-) sources following the method of George et al. (2008). The results of nss-$K^+$, $NH_4^+$ and nss-$SO_4^{2-}$ were discussed in this study." Please see lines 187-190 in the revised MS.

- page 25589, line 19: "although" should be "where", given that the statement "dense fire spots were detected" is not a consequence of the prior statement.

Response: Corrected. Please see line 240.

- page 22590, line 11: "galactosan did not show such a trend", please provide an explanation for this. Given that they originate from the same source, wouldn't a similar trend be expected?

Response: Based on the comment, we added the following sentences in the revised MS: "This is likely associated with changes in the relative content of cellulose and hemicellulose in varying BB fuel types along with changes in seasons and source regions. The second possible reason may be the different degradation rate of galactosan with those of levoglucosan and mannosan." Please see lines 269-273.

- same page, line 16: "the primary cause... levoglucosan and mannosan...", what about galactosan? What are the sources/processes affecting galactosan?

Response: We modified the statement to "The primary cause of the seasonality of three anhydrosugars in Okinawa aerosols is the shift of air mass source regions." Please see lines 276-277.

- page 22591, line 11: "... largely contributes" should be "may have largely contributed", given that at this point this is only a hypothesis, there is no evidence to support this statement.

Response: Corrected. Please see lines 297-298.

- same page, line 19: same here, "are associated"

Response: Corrected to "may have been associated" as suggested. Please see lines 305-306.

- section 323: a discussion of the degradation of levoglucosan should be included somewhere in the results section, to assess whether it may be transported over such long distances.

Response: Based on the comment, we added a new paragraph about the degradation of levoglucosan in the revised MS as follows.

“Clear diurnal cycle of OH was observed at Okinawa showing a daytime maximum of around 4 × $10^6$ radicals $cm^{-3}$ in summer (Kanaya et al., 2001). In light of literature reports on levoglucosan degradation by OH (Hennigan et al., 2010; Lai et al., 2014), the average atmospheric lifetime of levoglucosan could be roughly estimated as 1 day in summer and 3 days in winter. The corresponding transport distances of levoglucosan in summer and winter are estimated by weighted arithmetic means of trajectory clusters as represented in July and January, respectively. In July, air masses traveled 564 km for 1 day before arriving at Cape Hedo, where these regions were mostly ocean with no BB sources. Therefore, the observed low levels of anhydrosugars in summer represent a regional background level, incorporating atmospheric deposition and degradation. Similarly, Hu et al. (2013) reported that levoglucosan was detected in marine aerosols from the Arctic in boreal summer to the Antarctic in austral summer at ng $m^{-3}$ levels. In comparison, air masses traveled 2480 km for 3 days before arriving at Cape Hedo in January, a distance in which MNA was broadly passed through. It can be inferred that degradation plays a minor role in winter during strengthened continental outflow under low temperature and high wind speed conditions.” Please see section 3.2.3 and lines 323-337.

- same page, line 28: "background level", same here, if levoglucosan degrades over time during transport, what can be the regional background concentration?

Response: We appreciate for the referee’s comments. We modified the headline of 3.2.3 to “Anhydrosugar levels in summer” in the revised MS. Please see line 310. We also added some discussion on levoglucosan degradation. Please see lines 323-337.

- page 25592, line 8: interesting. Can the authors estimate approximately the max distance which levoglucosan may travel in their region in summer before degradation? Basically, I think the readers would be interested to know is whether levoglucosan emited in Mongolia or Russia may be detected in Okinawa in winter.

Response: Based on the comments, we estimated the approximate travel distances in winter and summer. In winter, “air masses traveled 2480 km for 3 days before arriving at Cape Hedo”, a distance in which MNA was broadly passed through.” Thus, degradation may play a minor role in winter. These points are added in the revised MS. Please see lines 323-337.

- same page, line 18: what is the distance between Okinawa and Philipines? If the authors state in line 1 of this page that levoglucosan in Okinawa in summer originates from background aerosols from Chichijima, but also that levoglucosa, decomposes from Philipines to Okinawa, isn’t this contradictory? Again, it would be useful to estimate the max distance which levoglucosan may travel in summer before it decomposes or it is removed by wet/dry deposition.

Response: The distance between Okinawa and Philippines is ~2000km. We agree that not only deposition, but also degradation may contribute to the undetectable influence of BB from the Philippines. Following the comment, we modified the sentence to “Anhydrosugars emitted from the Philippine region are likely decomposed and/or scavenged from the atmosphere by dry/wet deposition in summer before arriving at Okinawa, given the long travel distance of ~2000 km, more frequency of precipitation and short lifetime of levoglucosan (roughly 1 day, corresponding to 663 km in the trajectory cluster). ” Please see lines 346-350.

- section 324: the data presented in this section are not results from this study, I’d suggest to move them to the introduction.

Response: The first half of the section are from literature. We attempted to evaluate the BB types affecting Okinawa aerosols. Thus we consider this part being more suitable as discussions than introduction. However, following the comments, we moved this discussion point to section 3.1 because it best fits in the context. Please see lines 243-257.

- same page, line 23: "patterns", the authors detected fires in MNA in winter on page 25589, lines 10-14. Do their results coincide with the literature patterns?

Response: The fire pattern in MNA obtained by satellite is consistent with the literature. This point is briefly mentioned in the revised MS as follows: “Monthly total fire counts in MNA indicated consistent pattern with a major peak in May (total counts 45020) and secondary peak in October (total counts 17838) (Fig. S1).” Please see lines 246-248. We also added Figure S1 in the revised MS as follows.

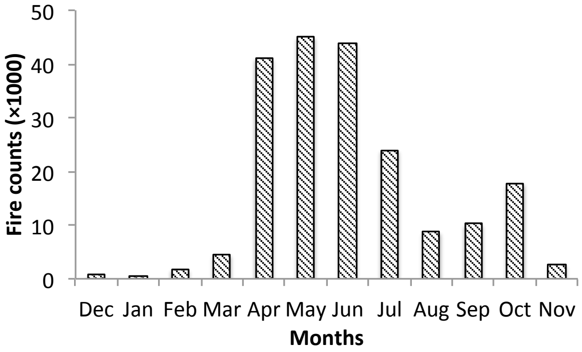

Figure S1. Monthly fire counts in Middle to North Asia (MNA, 30–60 °N, 80–130 °E) during December 2009–November 2011.

- page 25593, line 15: please provide sone interpretation of this, even if only hypothesizing.

Response: Following the comment, we added one sentence in the revised MS : "Galactosan did not co-vary with other anhydrosugars, probably due to different BB types that vary with time and source region and/or different degradation rate." Please see lines 356-358.

- same page, line 22: how does NOx evolve to NH4+? Please provide the pathway.

Response: Thanks for the comment. We re-reviewed literatures about sources and transformations of atmospheric $NH_4^+$ and then revised the sentence to "$NH_4^+$ could be formed by the protonation of $NH_3$." Please see line 390 in the revised MS.

- same page, last line: why does the correlation (r) increase from winter to summer? I would have expected higher values in winter, when the source is strongest. How do the authors explain this trend, or the difference between seasons?

Response: Following the comment, we examined the sources of $K^+$ using a tracer-based method and PMF model and added the following sentences in the revised MS. Please see lines 370-386.

"To further account for the fraction of BB-derived $K^+$ (bb-$K^+$), $K^+$ balance was investigated using a tracer-based method and PMF model (Supplementary materials). Pio et al. (2008) reported that bb-$K^+$ could be calculated by subtracting ss-$K^+$ and dust-related $K^+$ (dust-$K^+$) from total $K^+$, using a typical $K^+/Ca^{2+}$ ratio of 0.12 for soil dust in Europe. In East Asia, $K^+/Ca^{2+}$ ratios during dust storm periods are reported as 0.012 in Zhenbeitai, northern China (Arimoto et al., 2004), 0.07 in Xi'an, northwestern China (Shen et al., 2009), and 0.12 in Tongliao, northeastern China (Shen et al., 2007). We estimated the $K^+$ component using an average $K^+/Ca^{2+}$ ratio of 0.07 for Asian dust, following the method of Pio et al. (2007, 2008). We found that bb-$K^+$ accounted for 23% of total $K^+$. This result is similar to that (24%) obtained by PMF model (Fig. 5). Interestingly, relation between levoglucosan and tracer-based bb-$K^+$ was not strong, although positive relation was found between levoglucosan and nss-$K^+$. This phenomenon, as well as the seasonal difference of the relations between levoglucosan and nss-$K^+$, might be caused by the following reasons: (1) during open field burning, large amount of dust-$K^+$ is enriched in and mixed with bb-$K^+$; (2) dust-$K^+$ and bb-$K^+$ are mixed during atmospheric transport. Being consistent with previous studies, these results suggest that a combined analysis of $K^+$ and other tracers is recommend to characterize BB activities (Zhang et al., 2010)."

- page 25594, line 10: "significanlty" hould be "partly", given that the r values are very low.

Response: Corrected. Please see lines 402.

- same page, line 11: after "such relations" please add something like "and fossil fuel is most probably a more significant source of EC i terms of mass in this region".

Response: Based on the comment, we added the following sentences in the revised MS as follows: "Observation of black carbon (BC) by aethalometer at Rishiri Island, northern Japan indicated outflow of high level BC from fossil fuel combustion from the Asian continent (Zhu et al., 2015). Fossil fuel combustion in the Asian continent is probably a more significant source of EC in Okinawa aerosols compared to BB sources. Seasonally, levoglucosan correlates significantly with EC in summer, autumn and winter. No correlation in spring may be associated with higher EC source from fossil fuel combustion in the upwind Asian continent." Please see lines 403-409.

- section 3.4: again, these ratios may be altered if levoglucosan or mannosan degrade with transport along such distances. Please discuss this. The ratios are probably not stable over time.

Response: Based on the comment, we added few sentences in the revised MS as follows: "It is worthy noting that the possible different degradation rates between levoglucosan and mannosan would contribute to L/M variations. With respect to this point, the low L/M ratios in May–June may imply that levoglucosan is degrading faster than mannosan during atmospheric transport." Please see lines 423-426.

- page 25597, line 25, "significantly affects" should be "may significantly affect". Also, regarding this statement, please claryfy over what period of time may BB burning affect air quality in Okinawa? Days? Weeks?. At the end of the sentence "air quality in Okinawa", please add "mostly during specific episodes", given that on an annual basis the contribution is rather low, 2.9% of OC.

Response: We appreciate the comments and suggestions. We revised the sentence as: "These results indicate that BB, especially from MNA in winter, may significantly affects the regional air quality in Okinawa in winter, especially during Asian outflow episodes." Please see lines 520-522.

- conclusions: please add a comment on the degradation of levoglucosan and its implications regarding the results presented here.

Response: Following the comments, we added one sentence in the revised MS as follows: "The degradation of anhydrosugars during atmospheric transport may contribute to the summer low levels, while in winter the observed levels of anhydrosugars are less influenced by degradation." Please see lines 528-530.

**Anonymous Referee #4**

This manuscript presents ambient measurement results of anhydrosugars in aerosol particles collected at a remote background site (northern tip of Okinawa island) in Japan over a period of more than two years. Seasonal patterns and various diagnostic ratios, along with air mass history and fire count data, are used to explain qualitatively as well as semi-quantitatively the source influence from different types of biomass burning in various upwind regions of the Asian continent.

The paper is well structured, coherent, and concise, while more detailed discussions could enhance the presented findings in some cases, such as regarding the stability of the measured biomass burning tracers. Overall, this study is valuable to the atmospheric chemistry community, as the number of such measurements at background sites is limited, especially in East Asia and in the western North Pacific Rim. Therefore, I recommend publication of this manuscript in ACP upon consideration of the specific and technical comments given below.

Response: We appreciate the referee's comments. All the comments are helpful to improve the manuscript significantly. We have newly added description about levoglucosan degradation in the Introduction section and its implications in the Results section. Please see lines 65-82 and 323-337, respectively.

Specific comments

1. The authors correctly point out (page 25591, lines 7-10) that agricultural residue burning on farm fields may not be detected by Satellite imaging due to the small scale of such fires. However, then they should be careful with the statement on page 25591, lines 1-2, saying that the observed hot spots are associate with open field burning.

Response: Following the comment, we changed "Fire spots in north and northeast China indicate the burning in the open fields" to "Fire spots in north and northeast China indicate the large scale burning in the open fields". Please see lines 287-288.

2. For the discussion of the 27 October–3 November 2009 episode (page 25595, lines 1-8) the study by Zhang et al. (2007) could be cited here again, as it included maize straw (besides two other common types of biomass burned in south China: rice and wheat straw), and the average L/M ratio was very high (55), which indicates maize straw smoke to have a rather high L/M ratio as well, since it has been shown in other studies (e.g., Engling et al., 2009, Fu et al., 2008) that the respective ratios for rice and wheat straw can be in excess of 40.

Response: Based on the comment, we added discussions about L/M ratios of maize (see lines 434-440) as : "High L/M ratios had been reported for smoke from chamber burning of rice straw (40–42) and ambient aerosols affected by burning of wheat straw (40–46) (Engling et al., 2009; Fu et al., 2008). A higher average L/M ratio of 55 was obtained for the burning of rice straw, wheat straw and maize straw by chamber experiment (Zhang et al., 2009). These results implied that maize straw smoke may have higher L/M ratio. Our results gave a relatively high L/M ratio of 38.9 even after long-range transport to Okinawa, supporting the above proposition. ”.

3. The statement that the measured Lev/OC fractions indicate degradation of Lev (page 25596, lines 2-3) is rather presumptive and not really supported by the data presented here. The authors further explain that in the following statement, but they may want to tone down the statement in this sentence.

Response: Following the comment, we focus on the comparison of Lev/OC ratios among different sites, instead of using it to evaluate degradation of levoglucosan. We then proposed several causes for the differences.

The headline of section 3.5 was changed to “L/OC and L/EC ratios”. Please see line 450. We reorganized section 3.5 and added the following sentences in lines 459-464:

“Here, we discuss the longitudinal gradient of L/OC in Changdao, Okinawa and Chichijima, in the context of the Asian outflow. It is worth noting that, in comparison to the other two sites (2001–03), the data of Okinawa aerosols were obtained in different years (2009–12) when the tropospheric oxidizing capability was likely enhanced (Itahashi et al., 2014). However, such comparison is still helpful for understanding atmospheric processes in terms of deposition and degradation of levoglucosan.”

Technical comments

1. Please, use consistent acronyms, i.e., either "Lev" or "L" for levoglucosan, and don’t say "L/M" one time and then "Lev/Man" in other places.

Response: We formatted the acronyms to use “L/M”, “L/OC” and “L/EC” throughout the MS.

2. The proper use of articles (direct vs. indirect) and singular vs. plural forms needs to be checked throughout the entire manuscript. For example, on page 25582, line 10 the indirect article "a" is not necessary, on page 25583, line 9, say "smoke" instead of "smokes", or on page 25583, line 13 write "burning" rather than "burnings".

Response: We corrected the articles and singular/plural forms being mentioned and checked throughout the manuscript.

3. What are "smack and planting practices" (page 25583, line 12)?

Response: We now rephrased to “burning and land cultivation practices” in line 45.

4. What do the authors mean with "open field cooking for entertainment" (page 25595, line 13)?

Response: We rephrased to “open field cooking for barbeque” in line 445.

## Other changes

1. We added Bhagawati Kunwar as coauthor.

2. Un-bolded fonts in Figure 2.