Responses to the Anonymous Referee's comments

Referee #1 Evaluations

The work by Haque et al presents a range of organic compounds in atmospheric aerosols from a year-long campaign in South Korea. The authors used well established analytical techniques for their analysis. The experimental procedure is well designed and in general the data is well presented. My detailed comments are listed below.

Response: Thanks for the careful reading and valuable suggestions to improve the scientific content of the manuscript. Following the reviewer's comment, we have carefully made corrections in the revised MS. Our responses to all comments made by the reviewer are given below. Please refer to the revised MS, in which changes are highlighted in yellow.

Comment: First of all, although the work is of relatively good quality (as can be judged from the draft); I felt it is largely of a routine nature (at least the way it is presented). I believe that such type of publications can be useful but if considered for publication at ACP they need to go under the Measurement report's category.

Response: We respectfully do not comply with the reviewer's argument but would like to highlight the two key features that make this study suitable for publication in ACP under the Research article category.

1. Here, we present a new finding on the contribution of coal combustion sources in East Asia in controlling the atmospheric levels of levoglucosan (*Lev*) apart from the traditional biomass/biofuel burning emissions. This is based on the prevailing linear relationship between the radiocarbon based nonfossil-EC and *Lev* in the year-round TSP samples collected from the Korean Climate Observatory at Gosan (KCOG) site in Jeju Island. The Gosan supersite is the best location to understand how the chemical composition of source-emissions from East Asia affects the outflow regions in winter and spring. Recent studies have highlighted the potential contribution of *Lev* from residential coal combustion in China (Yan et al., 2018), with estimated annual emission of ~2.2 Gg of *Lev* from domestic coal combustion (Wu et al., 2021). Given this background information, the prevailing significant linear relationship between *Lev* and nonfossil-EC (P-value < 0.05) over the KCOG clearly emphasizes the need for reconsideration of the previous assessments on the impact of biomass burning in East Asian outflow to the western North Pacific.

2. Another important highlight is the detailed study on the atmospheric abundances of biogenic SOA tracers along with those of other primary biological particles from the KCOG for the year 2013. This dataset is further compared with the molecular distributions and relative abundances of these organic tracers in the TSP samples collected over Gosan during 2001, a decade ago (Fu et al., 2012). This comparison allows us to better understand the regional changes in the emission sources (*e.g.*, fugitive dust, biomass burning, and fossil-fuel combustion) on a decadal basis.

These points have been added in the revised MS. Please see lines 682-697.

Major comment:

Comment: I would like to know if the authors considered atmospheric stability of the measured organic compounds especially in the light of a long-range transport or/and correlation and statistical analysis of the reported species. For example, it has been demonstrated that levoglucosan can decay in particles exposed to OH. ‘’*The extent of decay ranged from 20 to 90% and was strongly correlated to the integrated OH exposure. Increased relative humidity did not enhance or impede reaction. Relative kinetics indicate that levoglucosan has an atmospheric lifetime of 0.7–2.2 days when biomass burning particles are exposed to 1 × 106 molecules cm^{-3} of OH (typical average summertime conditions). This implies that levoglucosan reacts with OH on a timescale similar to that of transport and deposition, which has important implications for the use of levoglucosan as a tracer for biomass burning emissions in source apportionment studies’’ Hennigan et al. 2010.*

I trust the above also applies to other measured organic compounds for some of which the information on reactivity is not available. My question to the authors - how does this affect the presented results? The authors present correlation of different organic compounds and discuss their potential similar source based on these correlations and statistical analysis. Does the stability of considered organic compounds can play a role in the observed absence of correlation? The same applies to the discussion on long range transport of the observed organics. I doubt that stability of considered sugars in Figure 4 is the same for all compounds.

Response: As the reviewer rightly pointed out that levoglucosan (*Lev*) undergoes a photooxidation with OH radicals during atmospheric transport (half-life: <2.2 days) and, this would cause lower abundances of this anhydrosugar (Hennigan et al., 2010). *Lev* concentration measured over the KCOG includes contributions from both fossil and nonfossil sources in the East Asian outflow. In such a case, we found the moderately significant correlations of *Lev* with nonfossil EC (i.e., biomass burning) and fossil-EC (e.g., Miocene lignite coals) over the KCOG. It is very likely that this weakened relationship is because of the photo-degradation of *Lev* during atmospheric transport. This result would be an otherwise high atmospheric abundance of *Lev* and its pronounced linear relationships with the nonfossil- and fossil-EC, which implies a much stronger impact of both these source emissions in the East Asian continental outflow in winter and spring. This is also applicable for low concentrations and poor correlations of other organics such as primary saccharides and BSOA tracers. Considering the feasibility of photooxidation, it is likely that secondary OA undergoes much faster cycling than the primary sugar compounds. This would mean a slight underestimation of BSOA over the KCOG in the East Asian outflow and their atmospheric abundances over Gosan reflect a lower limit.

These explanations have been added in the revised MS. Please see lines 370-374, 452-453, 458-460, and 676-681.

Comment: Moreover, besides anhydrosugars, the authors state that ‘’*Cis-pinonic acid showed poor correlation with pinic acid (r = 0.35, p = 0.12) (Figure 6f), suggesting that they formed from different monoterpenes such as α-pinene, β-pinene, or δ-limonene*.’’ Such assumptions can be made only if the measurements are taken very close to their emission sources **and/or** assuming that both compounds have similar atmospheric stability or reactivity. Is it the case for all considered compounds in this study? I believe this needs to be considered when discussing correlations or long range transport.

Response: We comply with the reviewer's view. Likely, the extent of loss of BSOA by the photooxidation during transport would also cause such poor correlations, influencing their abundances over KCOG. We have added a few sentences in the revised MS. Please see lines 476-478 and 499-501.

Minor comment:

Comment: It is hard to read Figure 2 (especially Figure 2a). Please either split the concentration axis so that all minor compounds can be clearly seen in the plot or move it into the SI and increase the size of the figure.

Response: Following the reviewer's suggestion, we have added an additional figure in the supplement for the visualization of minor compounds in the temporal plot. Please see Figure S2.

Reference:

Hennigan, C.J., Sullivan, A. P., Collett, J. L., Robinson, A. L.: Levoglucosan stability in biomass burning particles exposed to hydroxyl radicals, Geophys. Res. Lett., 37, https://doi.org/10.1029/2010GL043088, 2010.

Response: Added in the reference list of the main text.

Reviewer #2 Evaluations

The authors present the results of the chemical composition of organic aerosol in South Korea with a focus on primary organic compounds and biogenic secondary organic aerosol tracers. Seasonal variations and sources of studied compounds were analysed. The paper is suitable for publication in the journal Atmospheric Chemistry and Physics, however, several comments reported below should be addressed before acceptance for publication. Minor revisions of the paper are requested.

Response: We thank the reviewer for the positive assessment of this work. We have carefully revised the manuscript following the reviewer's comments and suggestions. Our responses to all comments made by the reviewer are given below. Please refer to the revised MS, in which changes are highlighted in yellow.

Comments:

Comment: Lines 277-281: Glucose and other saccharides may be partly formed also during biomass burning, which explains their contribution during the winter period.

Response: Following the reviewer's comment, we have added the following sentence in the revised MS as: "Since glucose, fructose, and sucrose showed moderately significant correlations (R^2 = 0.44-0.48, p<0.01) with levoglucosan in winter, it is somewhat possible that BB source emission could also influence the concentrations of these saccharides in this season (Haque et al., 2019; Fu et al., 2008)." Please see lines 279-283.

It can be noted that levoglucosan showed a poor/negative correlation with other saccharides except xylose (lines 404-405) and inositol in winter (discussed in the main text). Please see lines 302-305 in the revised MS.

Comment: Line 283-294: Sources of inositol and other polyols are only perfunctorily characterized focusing on prevailing sources. Sources of inositol are entirely missing.

Response: Following the reviewer's suggestion, more discussion on sources of polyols along with inositol has been added in the revised MS as: "The major sources of arabitol and mannitol are airborne fungal spores (Bauer et al., 2008), accompanying detritus from mature leaves (Pashynska et al., 2002). Heald and Spracklen (2009) reported that mannitol and arabitol are considerably associated with terrestrial biosphere activity. Inositol is largely derived from the developing leaves in summer (Pashynska et al., 2002) and BB in winter (Fu et al., 2010b). Zhu et al. (2015b) found similar seasonal behavior of inositol with those of other sugar alcohols with the predominance in summer, associated with microbial activities in local forests from Okinawa. Inositol showed a moderately significant correlation with levoglucosan (R^2 = 0.33, p< 0.01) in winter; however, there were no positive linear relationships between levoglucosan and other sugar alcohols, implying a partial emission of inositol from the BB during winter in Gosan aerosols." Please see lines 296-305. More information regarding the sources of polyols are already interpreted in section 3.4 (lines 409-419).

Comment: Line 306-307: Unlike other studies, the concentration of mannosan in this study is surprisingly lower than those of galactosan, do you have an explanation of this fact?

Response: Following the reviewer's comment, we have added the following sentences in the revised MS as: "Galactosan is more abundant in crop-residue burning emissions than mannosan (Engling et al., 2009; Sheesley et al., 2003). It is very much likely that the impact of crop-residue burning emissions in East Asia over Gosan is more prominent in winter/spring. Such high abundances of galactosan over mannosan were found in the North China Plain (Fu et al., 2008) and in the Indo-Gangetic Plain outflow sampled over the Bay of Bengal (Bikkina et al., 2019)." Please see lines 320-325.

Comment: Line 400-404: Ratios levoglucosan/mannosan and levoglucosan/(mannosan + galactosan) also allow to distinguish biomass burning and lignite combustion as the source of these anhydrosugars. In addition, the empirical equation using levoglucosan and mannosan data allows you to calculate the contribution of softwood and hardwood to the total amount of combusted wood.

Response: Based on the reviewer's comment, we have added the results of these diagnostic ratios in the revised MS as "Ratios of *Lev*/*Man* and *Lev*/(*Man* + *Gal*) can be useful to distinguish BB and coal combustion contributions. The average ratios of *Lev*/*Man* (15.1 ± 6.76) and *Lev*/(*Man* + *Gal*) (4.27 ± 1.23) in Gosan aerosols are much closer to those from wood burning and coal combustion sources emissions, respectively (Yan et al., 2018). It reveals that *Lev* could originate from both biomass and coal burning source emissions, which is consistent with the linear relationship between *Lev*-C and fossil-/nonfossil carbon fraction (section 3.7). Please see lines 422-427 in the main text. Unfortunately, we are not familiar with the empirical equation to calculate the contribution of softwood and hardwood.

Comment: Line 426: Add references to levoglucosan degradation.

Response: Added. Please see line 453 in the revised MS.

Comment: Line 536: Add missing information into Table 3, such as analysed PM fraction and studied season.

Response: Added. Please see Table 3 in the revised MS.

Comment: Line 562: Another key factor is a higher concentration of ozone and other oxidation agents in summer.

Response: Added. Please see lines 591-592 in the revised MS.