

## Anonymous Referee #1

Comments on “Measurement report: Diurnal and temporal variations of sugar compounds in suburban aerosols from the northern vicinity of Beijing, China: An influence of biogenic and anthropogenic sources” by Verma et al., October 2020.

### General Comments:

In this manuscript the authors report observations of sugar compounds (SCs) in air at a rural site about 40 km north of Beijing from Aug 15- Oct 5, 2007. Diurnal variability is examined, and meteorological parameters are considered as explanatory variables. The SC time series were analyzed with positive matrix factorization to identify sugar aerosol types and the relative contributions of the sugars and aerosol types to organic aerosol mass are reported. Overall this is a useful contribution of measurements to a topic that is still not well-understood. The differentiation between daytime and nighttime samples is rightly recognized as important by the authors. However, the manuscript often reads like a laundry list of observations interspersed with comparisons to previous observations, and insufficient analysis to support some claims.

Response: Authors are thankful to the reviewer for his valuable comments and suggestions, which help to upgrade the quality of the manuscript. We make significant changes in the manuscript especially in section 2. Materials and methods and 3.1. - 3.2. Results and discussion. We deleted several phrases of comparisons with previous observations.

Some inferences made about the observations are provided as speculations with little explanation or supporting analysis. In fact there are a few claims made in the manuscript (noted below) that don't even seem to clearly follow from the evidence presented. The logic behind the claims needs to be clarified or the claims need to be changed or removed. A major driver of this issue is that day-night differences were also differences in temperature and humidity, and differences in air mass origin. How can these effects be disentangled to draw inferences? For example, sucrose is interpreted to be controlled by local emission related to temperature and radiation on the basis of correlation with those variables, and transport is not considered. But arabitol and mannitol correlate very closely with local RH- why in the Abstract is it claimed that these are related to transport from Beijing? Perhaps this would be clearer if a more comprehensive table of correlation coefficients were shown for the relevant quantities, for daytime, nighttime, and overall, but some more textual clarification would also help. Also, would be really nice to have some proxy of transport there too, e.g. average magnitude of the wind in the direction between Beijing and the site.

Response: We modified section 3.1.3 in the results and discussions added new lines in the revised MS. Following the reviewer's suggestions we added new correlation table (Table 2) in the revised MS. We have added a new figure (Figure 2b) showing the day and night time wind direction in the Mangshan site. Please see the revised MS.

After deeply digging the dataset we found some interesting facts about the mannitol contribution in the Mangshan aerosol samples. We added several lines in the revised manuscript. Please see lines 358 – 372 in the revised MS.

A major limitation is the lack of any air mass trajectory analysis. The authors state that there was a typical diurnal pattern to the wind direction, with daytime winds from the south and the large cities, and nighttime winds from more rural areas to the north. It would help the reader to see how consistent this pattern was in order to evaluate some of the claims made. I suggest at least a time series plot of wind speed and wind direction, perhaps as a sub-plot to Figure 2, or a wind rose diagram. A couple of representative air mass back trajectories could be instructive as well.

Response: Following the reviewers suggestions we added new figure (Figure 2b). Figure 2 shows the wind direction and magnitude of wind directions in the Mangshan site. Please see the revised MS.

The results section should have more description of the present data set, with more of the previous observations moved to the Introduction. In particular, the description of the PMF results could be expanded, perhaps with a figure showing the time series of the PMF factors.

Response: We significantly modified the result and discussion sections by adding new phrases and relocating the discussions on previous studies. Please see lines 211–214, 242–250, 261–264, 294–297, 306–317, 328–334, 345–347, 353–363, 368–375, 378–384.

We added information about the PMF analysis in the revised MS. Please see lines 187 – 201, 389 – 407, 418 – 429, 442 – 445, 453 – 457, 460 – 462 in the revised MS.

We added Figure S-1 and S-2 as supporting information in the revised MS.

Figure S-1. The Scatter plots between observed (input data) and predicted (modeled data) concentrations show statistical parameter (coefficient of determination (r), Intercept, and Slope) with linear equation of individual sugar compounds. A blue 1:1 line is provided on this plot for reference (a perfect fit would line up exactly on this line), and the regression line is shown as a dotted red line.

Figure S-2. The time series plots between observed (input data) and predicted (modeled data) concentrations of individual sugar compounds. Blue and red lines show observed (input data) and predicted (modeled data) concentrations, respectively.

Throughout the manuscript, the authors should to be more careful in their descriptions of how the SCs get into the atmosphere. They frequently state that a process or an organism “emits” a sugar compound, which reads somewhat ambiguously (i.e. are gas phase compounds being released?). I think it’s OK to use this language, but there first needs to be a clear statement in the Introduction about how SCs get into the atmosphere, or at least the state of the science on that question. How are SCs are released into the air, as fragments of organisms, as whole fungal spores, as individual molecules, etc?

Response: We added new paragraph in the introduction section, including a clear statement on the sources of individual sugar species in the atmosphere. Please see lines 55 – 61, 67 – 75, 78 – 100.

We modified the sentences where we used “emits” in the revised MS.

A further issue that needs to be resolved before this is publishable is the extremely sparse description of the methods. Specific questions are raised below.

At this point this manuscript is essentially a descriptive account of measurements made at a particular location, with some interpretive claims that seem a bit ambiguous. The measurements themselves are of value, but I think for publication, it needs 1) a much more thorough method description section and 2) either a) a scale-back of the claims made, or b) additional analysis in support of the claims.

Response: We significantly modified the sections of materials and methods by including new information and new section in the revised MS. Please see lines 138 - 144, 147 – 149, 161 – 201.

Specific comments:

There are several grammatical issues of subject-verb agreement and lack of pluralization throughout the manuscript. They don't usually impede understanding, but the manuscript would benefit from a thorough grammar check.

Response: We significantly corrected grammatical issues in the manuscript. Please see the revised MS.

It would be very helpful to view the data in Figure 2 directly as a part of Figure 3.

Response: We added new figure in the (Figure 2b) in the revised MS. We also separated figure 3 in three parts (Fig 4a-c, 5a-d and 6a-c) according to the groups of sugar compounds. The new arrangement is very easy to understand and clearer than the earlier version. Please see the revised MS.

Line 66: “SCs are emitted from algae, microbes, pollen, suspended soil particle[s], and associated biota into the atmosphere” This statement reads to me a bit like sugar compounds might be released into the air as individual gas phase molecules, which I don't think is the case (?). Maybe it could be phrased “SCs are emitted as part of aerosols formed from algae...”? Same thing at line 72: I don't think mannitol and arabitol are mostly emitted as individual gases, but are a part of fungal material that gets into the atmosphere. What is physically meant here needs to be a little clearer. Are these SCs usually part of biological fragments, or is this unknown?

Response: Sentence rephrased please see lines 78 – 81 and 85 - 87 in the revised MS.

Section 2.2:

Please provide more details of the sampling apparatus and methodology. Were these samples collected with a high-volume sampler? Where was it installed specifically, and at what height? What aerosol sizes were collected? Were there any measures to avoid the sampling of gas phase components? Did the 3-hr and 9-hr samples overlap

in time? What times were the samples started? Perhaps a table of sample collection times would be helpful.

Response: We added Table S1 that contains the detailed description of the sampling procedures and collections. Other information's are added in the text. Please see section "2.1. Site description and aerosol sample collection" in the revised MS.

The twelve (n = 12) aerosol samples on 15 to 18, September and 28 September to 5 October were collected for 9-h from 9 to 18 h. While twenty six (n = 26) aerosol samples of 19 to 27 september were collected from 9 to 12, 12 to 15, 15 to 18 h.

We did not use a denuder system to remove gaseous components in high volume air sampler.

Furthermore, what were the methods for determining the WSOC and total OC? How was Ca(2+) concentration determined? Was the filter cut into sections for each analysis?

Response: Information's are added in the revised manuscript. Please see lines 185 – 186 in the revised MS.

Line 132: What is meant by C13 n-alkane? Is this an isotope standard of one n-alkane? Which one? Or do you mean C13H28, n-tridecane?

Response: C<sub>13</sub> n-alkane is n-tridecane (C<sub>13</sub>H<sub>28</sub>).

Line 157: "Hence, it is evident that increased BB activities at nighttime are associated with cool temperature (Fig. 2)." Is this saying that because it's cool at night, it makes sense that there's more BB aerosol at night? Isn't it equally likely that the different air mass origins in the day and at night are the reason?

Response: The sentence is rephrased. Please see lines 242 - 243 in the revised MS. The day-night time difference of the air masses can also influence the concentrations of BB tracers. We added related text please see lines 242 - 250 in the revised MS.

Line 209: "the meteorological conditions". Is this referring to the strong daytime winds and convective activity? It would be clearer to state that directly.

Response: Modified. Please see lines 272 - 274 in the revised MS.

Line 254: "northeasterly (99.5%)". Does this mean 99.5% of the nighttime hours the wind was northeasterly? Please clarify in the text.

Response: Yes, sentence rephrased (Please see lines 308-310 in revised MS).

Line 255: What would cause sugar emissions to decrease with lower temperature? Is there supporting literature for this?

Response: The daytime ambient temperature and solar radiations significantly affect plant activities and, subsequently, emissions of sugar enriched plant fragments. Therefore, the contribution of primary sugars at night was lower than in daytime. Miyazaki et al. (2012) reported the emissions of sugar compounds associated with

light and ambient temperature at forest site. Please see lines 311 – 317 in the revised MS.

Line 268: Trehalose paragraph. Trehalose didn't show a strong diurnal cycle, but the authors point out a correlation between trehalose and mannitol and arabitol at night, and between trehalose and Ca (2+) in the day. It would be helpful to at least report the corresponding correlation coefficients for the day and night, respectively, for comparison, and possibly to include the corresponding figures in Figure 5.

Response: We added correlation coefficient values in the text. We also added a new Table (Table 2) for correlation of sugar species and meteorological parameters in the manuscript. Please see lines 332 – 334, 336 - 337 in the revised MS.

Line 272: Why would nighttime low RH and temp cause microorganisms to emit more trehalose? Please cite a reference. Again, the use of "emit" here can be confusing. Is it the release of spores that prefers these conditions?

Response: Several studies have reported that the meteorological conditions, i.e., high RH and low temperature, are favorable for the microbes and fungi to discharge spores. The high RH and low temperature was recorded at night time therefore the microbes emits spores at night. We added information in the revised manuscript. Please see lines 328-339 in the revised MS. References are also added in the text and references section.

Line 310: Aren't the Mt. Tai measurements higher than Mangshan, not lower?

Response: We deleted this comparison. Please see in the revised MS.

Lines 315-319: I don't understand the reasoning here. How does RH relate to transport from megacities as an explanation for fungal aerosol?

Response: Rephrased. Please see lines 368 – 375, 378-385 in the revised MS.

Line 350: Separate the Factor 3 and Factor 4 descriptions into separate paragraphs.

Response: Factor 3 and Factor 4 are described into separate paragraphs (Please see line 440 – 457 in revised MS).

Line 352: "The PMF results are very well supported by the fact that anhydrosugars are associated with BB in the Mangshan site." Is this referring to results from a previous study? Please cite it.

Response: We modified the phases. Please see lines 448-449 in the revised MS.

Line 410: "Our results also denote that secondary production of OC and WSOC from BB-derived organic precursors was crucial during nighttime at the Mangshan site." What evidence shows this? And do you mean that organic compounds went through chemical changes to form aerosol OC and WSOC, or simply that organics produced during biomass burning were incorporated into aerosol after the burning? In either case, I don't see how we know that.

Response: We deleted the sentence.

Fig. 2.

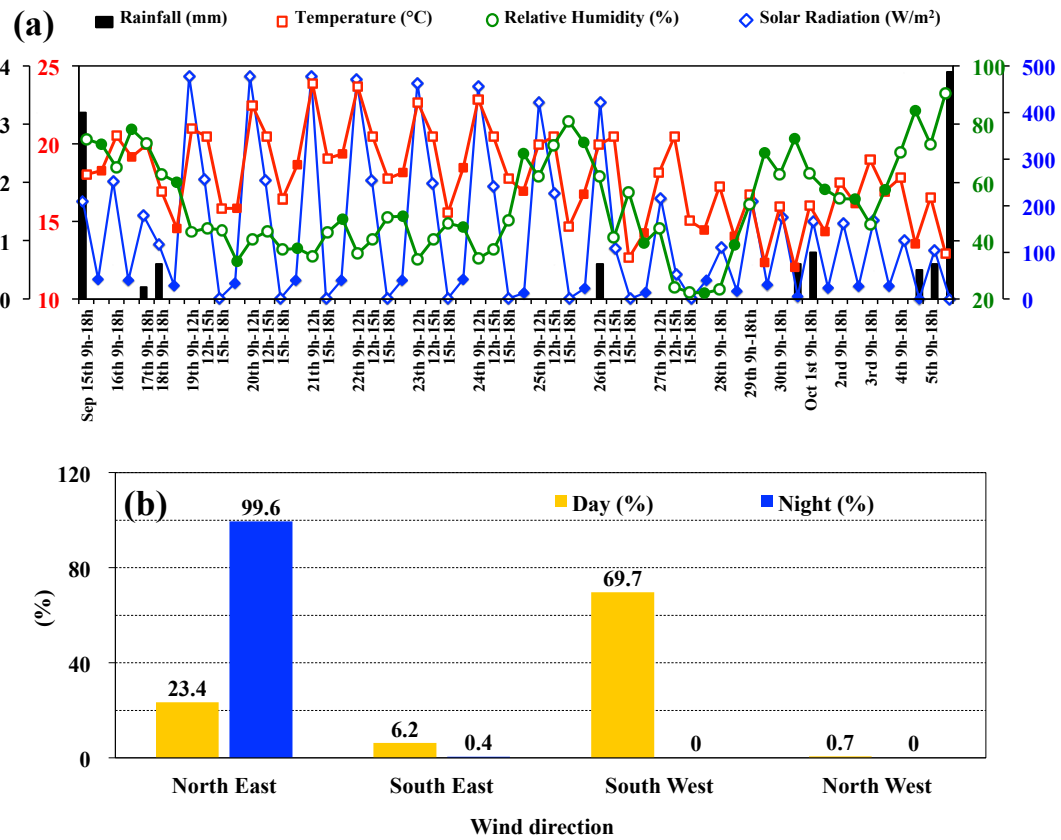


Fig. 4.

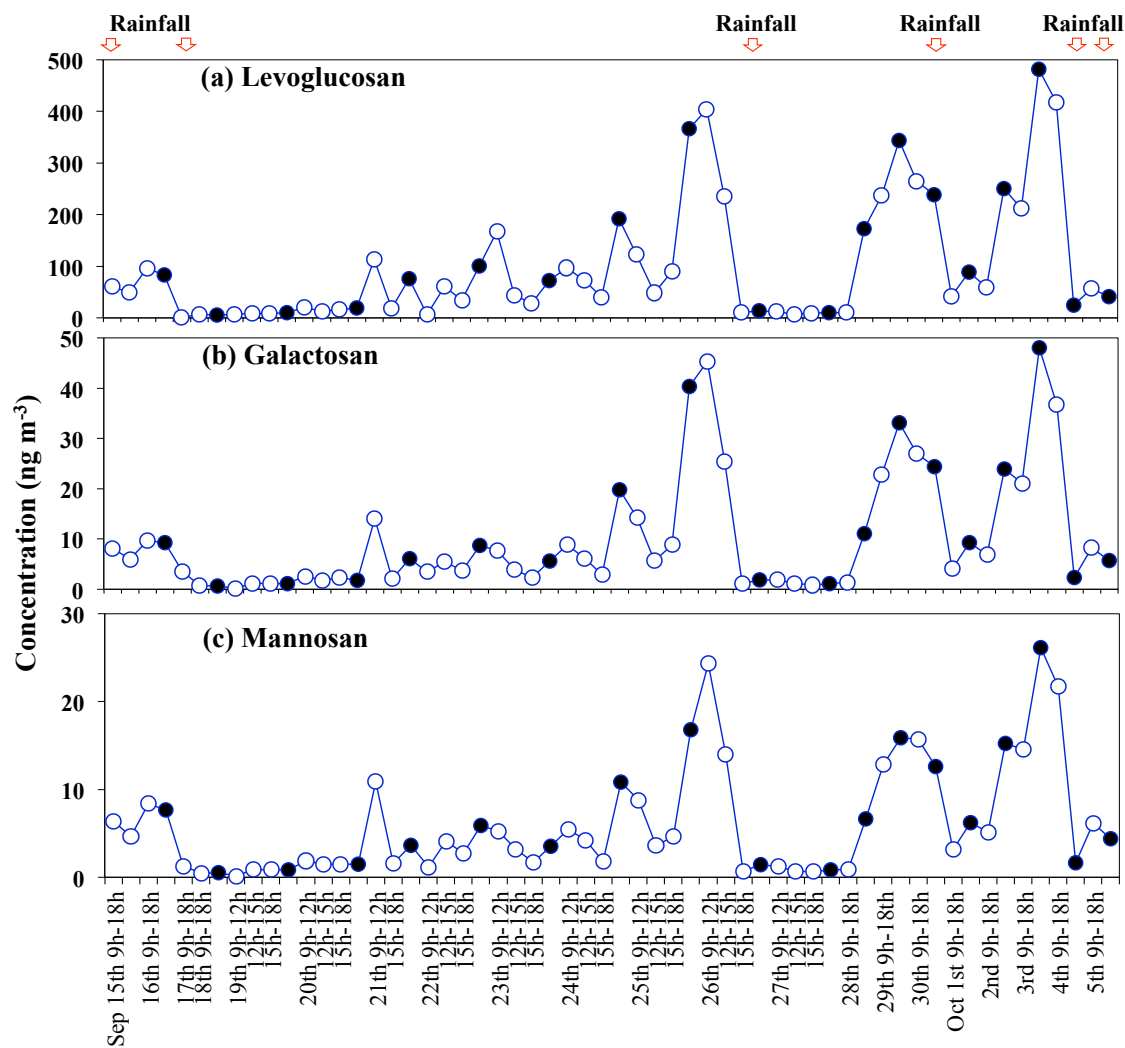
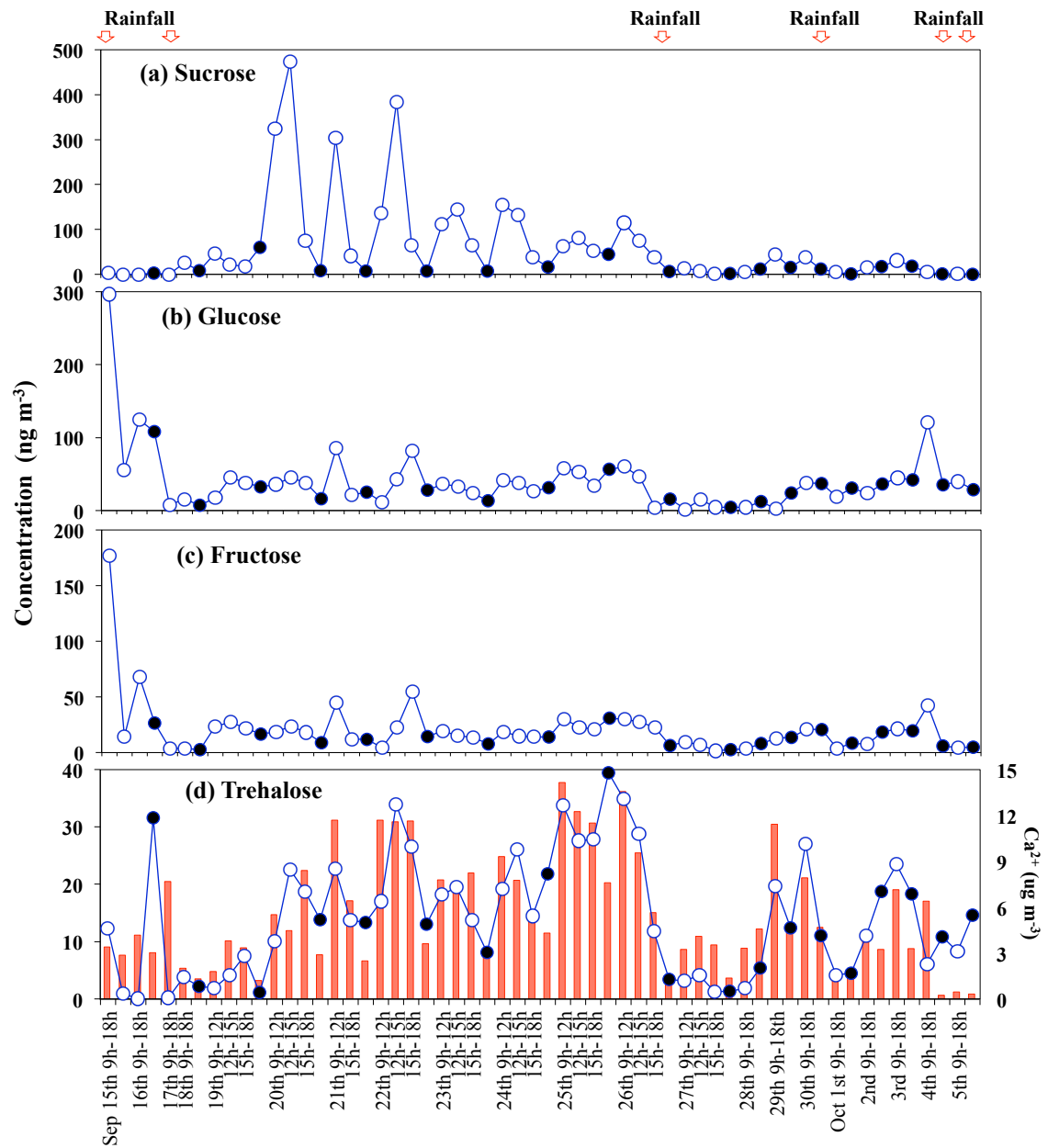


Fig. 5.







**Table 2.** Statistical summary of correlations among the chemical species and meteorological variables in aerosol samples collected at a forest site in northern Japan.

<b>Linear regression</b>	<b>Correlation coefficient</b>	<b>p value</b>	<b>Significance of correlation at P value &lt; 0.05</b>
<b>Overall (n = 58)</b>			
Levoglucon vs. Galactosan	0.98	< 0.05	Significant
Levoglucon vs. Mannosan	0.97	< 0.05	Significant
Mannosan vs. Galactosan	0.98	< 0.05	Significant
Sucrose vs. Temperature	0.52	< 0.05	Significant
Sucrose vs. Solar radiation	0.55	< 0.05	Significant
Arabitol vs. Mannitol	0.81	< 0.05	Significant
Arabitol vs. RH	0.69	< 0.05	Significant
Mannitol vs. RH	0.57	< 0.05	Significant
Glucose vs. Fructose	0.94	< 0.05	Significant
Trehalose vs. Arabitol	0.58	< 0.05	Significant
Trehalose vs. Mannitol	0.58	< 0.05	Significant
Trehalose vs. Ca <sup>2+</sup>	0.70	< 0.05	Significant
<b>Daytime (n = 38)</b>			
Sucrose vs. Ca <sup>2+</sup>	0.32	> 0.05	Not significant
Glucose vs. Ca <sup>2+</sup>	0.02	> 0.05	Not significant
Trehalose vs. Arabitol	0.49	< 0.05	Significant
Trehalose vs. Mannitol	0.51	< 0.05	Significant
Trehalose vs. Ca <sup>2+</sup>	0.81	< 0.05	Significant
Fructose vs. Mannitol	0.79	< 0.05	Significant
Levoglucon vs. OC	0.45	< 0.05	Significant
Levoglucon vs. WSOC	0.40	< 0.05	Significant
<b>Nighttime (n = 20)</b>			
Sucrose vs. Ca <sup>2+</sup>	0.37	> 0.05	Not significant
Glucose vs. Ca <sup>2+</sup>	0.27	> 0.05	Not significant
Trehalose vs. Arabitol	0.76	< 0.05	Significant
Trehalose vs. Mannitol	0.85	< 0.05	Significant
Trehalose vs. Ca <sup>2+</sup>	0.61	< 0.05	Significant
Fructose vs. Mannitol	0.86	< 0.05	Significant
Levoglucon vs. OC	0.81	< 0.05	Significant
Levoglucon vs. WSOC	0.70	< 0.05	Significant

## Supporting Information

Table S-1 Sampling informations.

Sampler:High Volume Air Sampler (Kimoto-AS810A)						
Sample ID	Start Time	Finish Time	Center time	Total Time (Hours)	Total Air V (m <sup>3</sup> )	Filter Total (cm <sup>2</sup> )
CHN-131	2007/9/15 9:08	2007/9/15 17:57	15/09/07 13:32	8:49	533.25	405.3
CHN-132	15/09/07 18:01	16/09/07 8:25	16/09/07 1:13	14:24	873.09	405.3
CHN-133	16/09/07 8:29	16/09/07 17:53	16/09/07 13:11	9:24	563.03	405.3
CHN-134	16/09/07 17:57	17/09/07 8:56	17/09/07 1:26	14:59	907.48	405.3
CHN-135	17/09/07 9:00	2007/9/17 17:38	17/09/07 13:19	8:38	515.44	405.3
CHN-136	18/09/07 9:58	18/09/07 18:03	18/09/07 14:00	8:05	490.5	405.3
CHN-137	2007/9/18 18:06	19/09/07 8:54	19/09/07 1:30	14:48	905.69	405.3
CHN-139	19/09/07 9:04	19/09/07 12:00	19/09/07 10:32	2:56	177.5	405.3
CHN-140	19/09/07 12:04	19/09/07 15:01	19/09/07 13:32	2:57	180.14	405.3
CHN-141	19/09/07 15:05	19/09/07 18:01	19/09/07 16:33	2:56	179.44	405.3
CHN-142	19/09/07 18:04	20/09/07 9:01	20/09/07 1:32	14:57	920.18	405.3
CHN-143	20/09/07 9:05	20/09/07 12:02	20/09/07 10:33	2:57	172.93	405.3
CHN-144	2007/9/20 12:06	2007/9/20 15:01	20/09/07 13:33	2:55	177.25	405.3
CHN-145	20/09/07 15:04	20/09/07 17:59	20/09/07 16:31	2:55	178.6	405.3
CHN-146	20/09/07 18:02	21/09/07 8:57	21/09/07 1:29	14:55	919.91	405.3
CHN-147	21/09/07 9:00	21/09/07 12:02	21/09/07 10:31	3:02	180.96	405.3
CHN-149	21/09/07 15:06	21/09/07 17:59	21/09/07 16:32	2:53	177.52	405.3
CHN-150	21/09/07 18:02	22/09/07 9:00	22/09/07 1:31	14:58	915.67	405.3
CHN-151	22/09/07 9:02	22/09/07 11:59	22/09/07 10:30	2:57	178.6	405.3
CHN-152	22/09/07 12:02	22/09/07 14:59	22/09/07 13:30	2:57	177.73	405.3
CHN-153	22/09/07 15:02	22/09/07 17:58	22/09/07 16:30	2:56	180.68	405.3
CHN-154	22/09/07 18:01	23/09/07 8:56	23/09/07 1:28	14:55	911.27	405.3
CHN-155	23/09/07 9:00	23/09/07 11:57	23/09/07 10:28	2:57	177.82	405.3
CHN-156	2007/9/23 12:00	23/09/07 14:59	23/09/07 13:29	2:59	179.3	405.3
CHN-157	23/09/07 15:02	23/09/07 17:59	23/09/07 16:30	2:57	180.75	405.3
CHN-158	23/09/07 18:03	24/09/07 8:55	24/09/07 1:29	14:52	918.89	405.3
CHN-159	24/09/07 8:59	24/09/07 11:56	24/09/07 10:27	2:57	178.16	405.3
CHN-160	24/09/07 11:59	24/09/07 14:57	24/09/07 13:28	2:58	180.62	405.3
CHN-161	2007/9/24 15:00	24/09/07 17:56	24/09/07 16:28	2:56	179.84	405.3
CHN-163	24/09/07 18:11	25/09/07 9:01	25/09/07 1:36	14:50	899.43	405.3
CHN-164	25/09/07 9:04	25/09/07 12:07	25/09/07 10:35	3:03	184.92	405.3
CHN-165	25/09/07 12:09	25/09/07 14:58	25/09/07 13:33	2:49	169.63	405.3
CHN-166	25/09/07 15:01	25/09/07 17:58	25/09/07 16:29	2:57	180.45	405.3
CHN-167	25/09/07 18:01	26/09/07 9:01	26/09/07 1:31	15:00	912.34	405.3
CHN-168	2007/9/26 9:04	26/09/07 11:59	26/09/07 10:31	2:55	176.33	405.3
CHN-169	26/09/07 12:02	26/09/07 14:58	26/09/07 13:30	2:56	176.46	405.3
CHN-170	26/09/07 15:01	26/09/07 17:58	26/09/07 16:29	2:57	181.09	405.3
CHN-171	26/09/07 18:01	27/09/07 8:56	27/09/07 1:28	14:55	916.56	405.3
CHN-172	27/09/07 8:59	27/09/07 12:00	27/09/07 10:29	3:01	183.05	405.3
CHN-173	27/09/07 12:03	27/09/07 15:01	27/09/07 13:32	2:58	180.88	405.3
CHN-174	27/09/07 15:04	27/09/07 17:59	27/09/07 16:31	2:55	179.04	405.3
CHN-175	27/09/07 18:02	28/09/07 9:15	28/09/07 1:38	15:13	936.45	405.3
CHN-176	28/09/07 9:19	28/09/07 17:57	28/09/07 13:38	8:38	519.1	405.3
CHN-178	28/09/07 18:10	29/09/07 8:59	29/09/07 1:34	14:49	907.18	405.3
CHN-179	29/09/07 9:01	29/09/07 17:59	29/09/07 13:30	8:58	539.66	405.3
CHN-180	29/09/07 18:01	30/09/07 8:57	30/09/07 1:29	14:56	922.34	405.3
CHN-181	30/09/07 9:00	30/09/07 17:57	30/09/07 13:28	8:57	531.89	405.3
CHN-182	30/09/07 18:00	01/10/07 8:55	01/10/07 1:27	14:55	896.33	405.3
CHN-183	01/10/07 8:58	01/10/07 17:54	01/10/07 13:26	8:56	538.04	405.3
CHN-184	01/10/07 17:57	02/10/07 8:56	02/10/07 1:26	14:59	926.33	405.3
CHN-185	02/10/07 8:59	02/10/07 17:56	02/10/07 13:27	8:57	531.43	405.3
CHN-186	02/10/07 17:59	03/10/07 8:59	03/10/07 1:29	15:00	913.14	405.3
CHN-187	03/10/07 9:01	03/10/07 17:52	03/10/07 13:26	8:51	523.19	405.3
CHN-188	03/10/07 17:54	04/10/07 8:57	04/10/07 1:25	15:03	919.84	405.3
CHN-189	04/10/07 9:00	04/10/07 17:56	04/10/07 13:28	8:56	535.79	405.3
CHN-190	04/10/07 18:00	05/10/07 8:59	05/10/07 1:29	14:59	918.46	405.3
CHN-191	05/10/07 9:04	05/10/07 17:56	05/10/07 13:30	8:52	535.52	405.3
CHN-192	05/10/07 17:59	06/10/07 8:53	06/10/07 1:26	14:54	906.91	405.3
CHN-138	19/09/07 9:00	blank	19/09/07 9:00			405.3
CHN-148	21/09/07 13:52	blank	21/09/07 13:52			405.3
CHN-162	24/09/07 17:58	blank	24/09/07 17:58			405.3
CHN-177	28/09/07 18:00	blank	28/09/07 18:00			405.3

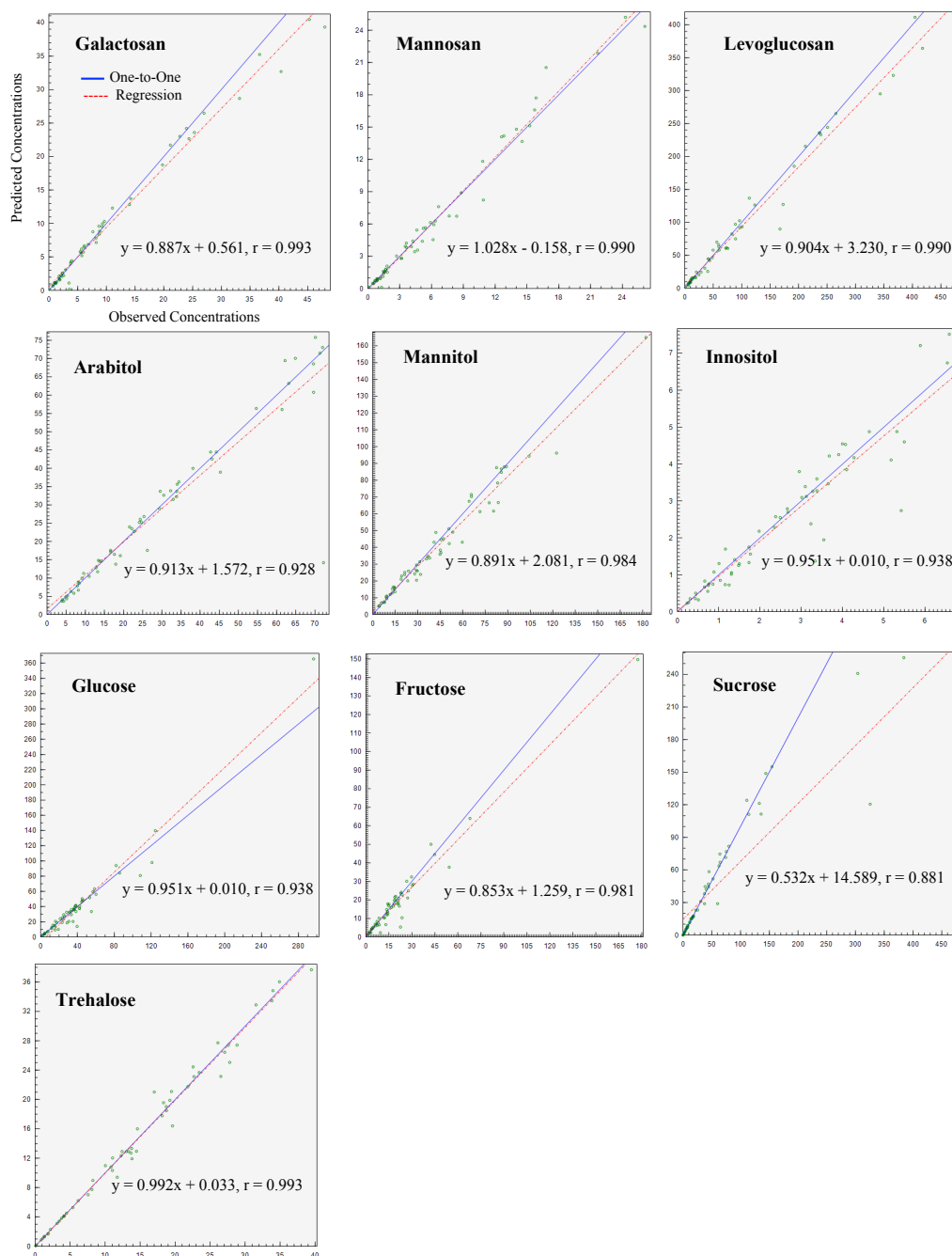


Figure S-1. The Scatter plots between observed (input data) and predicted (modeled data) concentrations show statistical parameter (coefficient of determination (r), Intercept, and Slope) with linear equation of individual sugar compounds. (A blue 1:1 line is provided on this plot for reference (a perfect fit would line up exactly on this line), and the regression line is shown as a dotted red line).

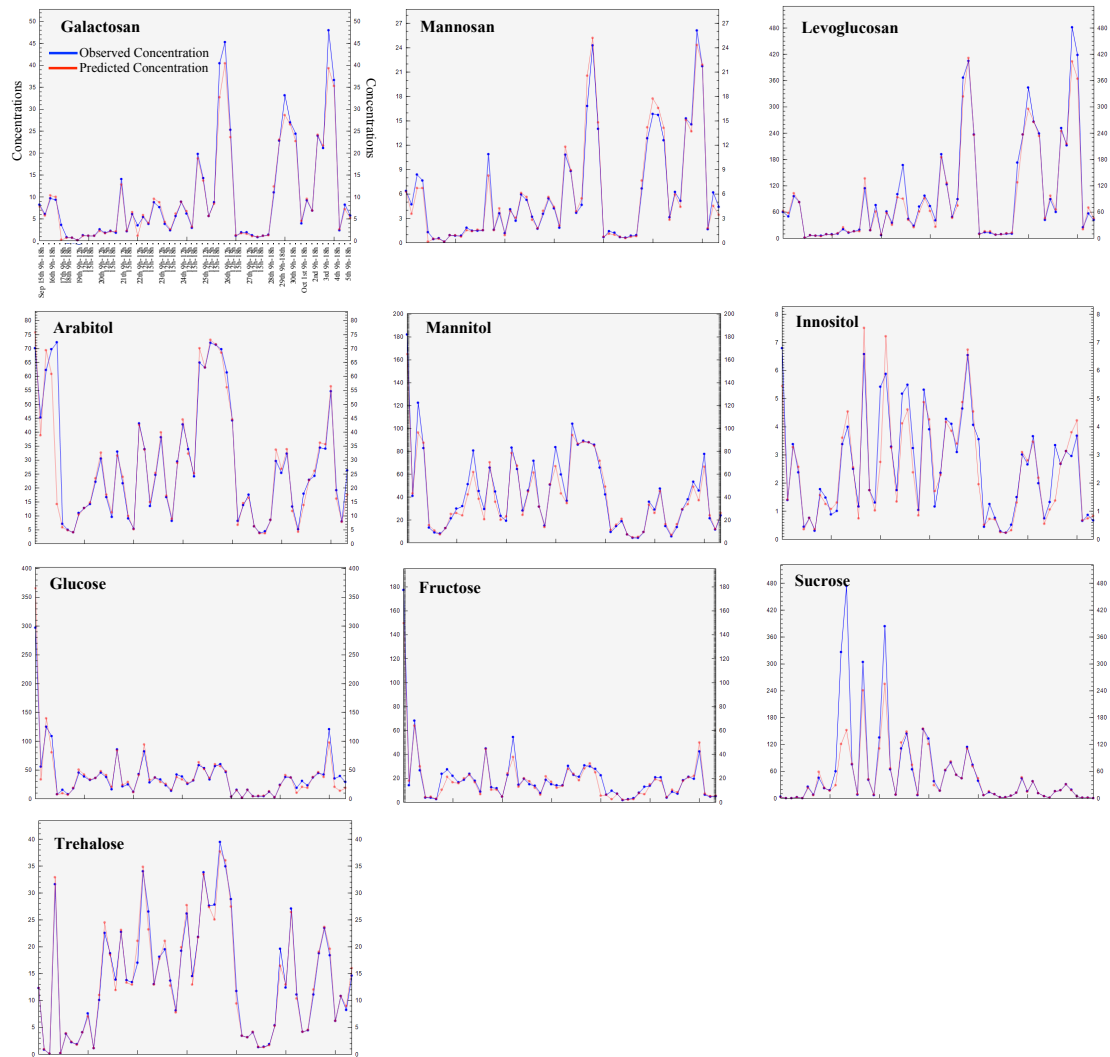


Figure S-2. The time series plots between observed (input data) and predicted (modeled data) concentrations of individual sugar compounds. (A blue line and redline shown as observed (input data) and predicted (modeled data) concentrations, respectively).

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