Author Response to Anonymous Reviewer #1

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- 3 We would like to thank Reviewer #1 for the careful reading of the manuscript, and dedication to
- 4 evaluating this study. We also appreciate Reviewer #1's insightful comments and valuable suggestions
- 5 which are very helpful for further improving the quality of our manuscript. Please find below our
- 6 point-by-point responses to the comments and a highlight to the changes made to the manuscript.
- 7 Changes to the revised manuscript are marked in blue.

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General Comment:

- 10 I think that having this kind of inventory for VOC emissions from a large region is beneficial for
- improving air quality models and better constraining other biogenic emissions and earth system
- models (e.g. CESM, MEGAN). But it appears to me that this work is not sufficient to be much more
- than qualitative estimates. I know how difficult this work is, and with limited time and resources, it's
- impractical to get accurate emission rate data from a large number of tree species in a single season.
- And what they have done, is a reasonable attempt to at least break down a large variety of species into
- different classes. Their statistical techniques are interesting and are useful for establishing these
- 17 ranges.
- 18 **Response:** We appreciate your insightful comments and valuable suggestions on our study. We will
- 19 respond to your comments in detail point by point below, and make corresponding changes to the
- 20 manuscript for further improving its quality. Changes to the revised manuscript are marked in blue.

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Comments regarding the accuracy of the emission estimates:

- 23 1. The authors discuss the volume of the bag and talk about a "semi static" enclosure. They give some
- 24 dimensions of the bag of 160 cm \times 90 cm in the shape of a rectangle and give a volume of 400 L.
- 25 When I do the calculation using either a cylinder or a rectangular box, I get a volume closer to 1000 L.
- But if I do take the 400 L volume and their flow rate of 6 L/min into the bag, the residence time is
- 27 400/10 = 40 min. For the system to be in steady state, there needs to be $\sim 3-4$ turnovers of air, which
- would take at least two hours. The way they get around this (non-steady state nature of the system) is
- 29 to determine how much residual air is in the bag by measuring acetylene before and after zero air is
- 30 added. A typical background level of acetylene is ~500-1000 ppt, so I presume the initial
- 31 concentration in the bag was approximately this amount. Then the bag was filled with zero air for 6
- 32 minutes using 10 L/min and then another 2 L/min for 3 minutes. The total volume displaced would

- 33 then be 60*6 + 2*3 = 66 L, which is 16.5% of the total 400 L volume. So the concentration of
- 34 acetylene after this step would be $\sim (100-16.5)*C0$ or $\sim 84\%$ of the original 500-1000 ppt. What is the
- uncertainty in a measurement of 1000 ppt. vs. 835 ppt? I think the authors really need to describe the
- 36 precision of their measurements and give some results of this step, as it's important if they are going
- 37 to use equation 2 instead of waiting for a steady state condition. Uncertainty in this number could
- 38 result in large uncertainties in emission rates.

39 **Response: Accepted.**

- We got around the non-steady state nature of the enclosure system as you said through determining
- 41 the residual air in the bag by measuring acetylene before and after zero air, according to Equation (2)
- 42 in the manuscript. The accuracy of acetylene measurement is the key to the accurate calculation of
- emission rates. We think we have minimized the uncertainty of acetylene measurement.
- In this study, the quantification of acetylene was conducted by FID through the external standard
- 45 method. The calibration of acetylene was performed with five concentrations ranging from 0.4 to 8
- 46 ppbv. The correlation coefficient (R²) for its calibration curve was 0.9992. A gas standard was
- 47 measured at 1 ppbv repeatedly for ten times to determine the precision and detection limit. The
- 48 precision of the system for acetylene was 1.11% and its detection limit was 0.025 ppbv, which
- 49 indicated a high accuracy of acetylene measurement by GC-MS/FID system.
- The ratio of acetylene concentrations of background and emission sample for each experiment ranged
- from 1.12 to 1.64, resulting from differences in actual inflation of air zero and available volume of the
- 52 chamber. The measured acetylene concentrations of background and emission samples mostly ranged
- from 1.09 to 3.05 ppbv and 0.88 to 2.48 ppbv, respectively. The deviations between analytical results
- of the calibration at 1 ppbv and theoretical concentrations ranged from -7.2% to -1.5%. When the
- calibration measured at 2 ppbv, the uncertainty of quantification was 3.9%. Therefore, the uncertainty
- of acetylene measurement was considered to be < 7.2%.
- 57 We described the precision of acetylene measurements in the revised manuscript. Line 179,
- 58 "Especially, correlation coefficient of calibration curve for acetylene was 0.9992." is added. Line
- 59 179-181, "A calibration was measured at 1 ppbv repeatedly for ten times to determine precision and
- detection limits." is added. Line 181-182, "The precision of the system for VOCs ranged from 0.5% to
- 61 4%." was modified to "The precision of the system for isoprene, α -pinene, β -pinene, and acetylene
- were 2.30%, 3.65%, 3.37%, and 1.11%, respectively; the precision for other VOC species ranged
- 63 from 0.5% to 4%.".
- 64 We also added the uncertainty of acetylene measurements in the revised manuscript. Line 448-453,
- 65 "Firstly, according to Equation (2), the accuracy of acetylene is the key to the accurate calculation of

- emission rates. In our study, the measured acetylene concentrations of background and emission samples mostly ranged from 1.09 to 3.05 ppbv and 0.88 to 2.48 ppbv, respectively. We concluded that the uncertainties of quantification of acetylene at its concentration range were < 7%, through
- 69 conducting repeated measurement of the calibration at 1 and 2 ppbv separately." is added.
- 70 Line 200-201 in the revised manuscript, "In our study, C_b/C_s ratios for the conducted experiments
- 71 ranged from 1.12 to 1.64, resulting from differences in actual inflation of air zero and available
- volume of the chamber." is added.

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- 74 2. The samples were taken to be generally in mid-day sun at ~30℃. And then standard algorithms
- were used to "normalize" the emissions to a set of standard conditions (e.g. 30°C). As far as I can tell,
- there is just one sample per tree or vegetation species, and some of the samples were taken very late
- 77 or early in the growing season. I think there needs to be more discussion on how seasons, ambient
- 78 conditions, and number of samples per species influences uncertainty.

Response: Accepted.

- 80 In our study, due to the limitation of experiment equipment and field conditions, each plant species
- 81 were measured with one or two replicates of samples for BVOC emissions. This might result in large
- 82 errors from sampling and measurement and could neither be representative to different individuals of
- 83 the same plant species. BVOC emissions from different individuals of the same plant species may
- 84 differ much owing to varied ambient conditions, such as temperature, radiation, precipitation, soil
- 85 moisture, and so on, which caused the uncertainty to our study. In addition, with limited time and

resources, it is difficult to perform field measurements to obtain emission rates from a large number of

- 87 tree species in different regions in a single season. In our study, some experiments were made in May
- 88 and October, early or late growing season. Different sampling time or season can result in different
- 89 ambient conditions which cause large differences in emissions. Additionally, the growth of plants in
- 90 different seasons usually varies, which causes radically different leaf emissions. Therefore, in future
- 91 study, measurements of emissions from different individuals of the same plant species in different
- 92 regions and seasons are urgently needed to obtain representative emission rates for different cases in
- 93 China.
- 94 A discussion on the uncertainty caused by sampling season, ambient conditions, and number of
- 95 samples for one plant species was added in the revised manuscript. Line 427-436, "In our study, only
- one or two replicates of samples for BVOC emissions from each plant species were collected. This
- 97 might introduce accident errors from sampling and measurement. The results could neither be
- 98 representative to different individuals of the same plant species because BVOC emissions from

different individuals of the same plant species may differ much owing to varied ambient conditions, such as temperature, radiation, precipitation, soil moisture, and so on. Due to limited time and resources, some experiments were made in May and October, early or late growing season. Different sampling time or season could result in different ambient conditions which cause large differences in emissions. Additionally, the growth of plants in different seasons usually varies, which cause radically different leaf emissions." is added. Line 511-513, "Besides, it is necessary to conduct measurements in different regions to obtain the representative emission rates of plants in the whole China." is modified to "Besides, it is necessary to conduct measurements from different individuals of the same plant species in different regions and seasons to obtain the representative emission rates of plants in the whole China."

- 3. In their evaluation and conclusion section, the authors discuss other uncertainties, and the recommendations of Niinemets et al. (2011) and readily admit that their measurements do not adhere to these guidelines. They give examples of some emission rates of 838, 707 and 2542 ugC/gdw/hr. Seeing that these are orders of magnitude greater than other reported rates seems like it should have made the authors more skeptical of their other results. The other thing that seems to be lacking in this paper is biomass distribution. It's not practical to define the whole country, but I think it's necessary to give the reader some context as to how common these different plants/trees are and how much of the total leaf area can be explained (at least in certain regions) by this listing.
- Response: Accepted.
- (1) In our study, we concluded that *B. papyrifera* had the highest isoprene emission rate of 838.62 μ g C gdw⁻¹ h⁻¹ and *L. formosana* had the highest α and β -pinene emission rates of 707.12 and 2542.13 μ g C gdw⁻¹ h⁻¹, respectively. There might be uncertainty in the data other than those described in other items of Responses to Comments. Firstly, the concentrations of emission samples of plants with the highest emissions were usually high enough to cause detector saturation to the GC-MS/FID system. For accurate quantification, the emission samples had to be diluted with nitrogen for suitable times. This would inevitably introduce errors to the quantification of concentration. Additionally, only one or two replicates of samples for BVOC emissions from these plant species were collected in our study, which introduced large uncertainties to the measured results from sampling and measurement. This uncertainty has been explained in detail in **Response to Comment 2**. Anyway, it should be concluded that *B. papyrifera* had the strongest isoprene emission potential and *L. formosana* had considerable α and β -pinene emission rates. For the uncertainties of BVOCs emission measurements from other plant species, we have made a discussion in **Reponses to Comment 1, 2, and 4 regarding the accuracy of**

the emission estimates and in the revised manuscript accordingly.

In our study, due to the small number of samples of plant species with such high emission rates, the representative emission categories could not be worked out by statistics. In the future study, to obtain more accurate emission data and more comprehensive and detailed emission categories, the quantitative measurements must be enhanced, as suggested by Niinemets et al. (2011). The detector saturation might be prevented through dynamic enclosure technique. The number of samples for one plant species should be increased. Measurements of plants within different emission categories are expected to be conducted.

We explained the uncertainty of the very high emission rates in the revised manuscript. Line 436-447, "There was another source of uncertainty of the very high emission rates measured in this study, such as 838.62 μg C gdw⁻¹ h⁻¹ for isoprene, 707.12 and 2542.13 μg C gdw⁻¹ h⁻¹ for α- and β-pinene, respectively. The concentrations of emission samples of plants with the highest emissions were usually high enough to cause detector saturation to the GC-MS/FID system, which would be overcome by dynamic enclosure technique during sampling. Errors were introduced inevitably to their quantification owing to the dilutions before analysis. However, conclusion could still be drawn from the results that the plants had high isoprene and pinene emission potential. Additionally, because of the only one or two samples of plant species with such high emission rates, their representative emission categories could not be worked out by statistics. In future study, to obtain more accurate emission data and more comprehensive and detailed emission categories, the quantitative measurements must be enhanced with increasing number of samples for one plant species and for plants within different emission categories." is added.

Reference:

- Niinemets, Ü., Kuhn, U., Harley, P. C., Staudt, M., Arneth, A., Cescatti, A., Ciccioli, P., Copolovici, L.,
 Geron, C., Guenther, A., Kesselmeier, J., Lerdau, M. T., Monson, R. K., and Peñuelas, J.:
- 157 Estimations of isoprenoid emission capacity from enclosure studies: measurements, data
- processing, quality and standardized measurement protocols, Biogeosciences, 8, 2209-2246,
- doi:10.5194/bg-8-2209-2011, 2011.
- 160 (2) In Table 2, we added the proportion of leaf biomass of each tree species to the total leaf biomass in
- Beijing, Hubei, and Sichuan provinces, respectively. The leaf biomass of dominant species in forests
- was estimated based on statistics of timber volumes at the provincial level obtained from Forest
- Resource Statistics of China with biomass-apportion models (Li et al., 2013; Li and Xie, 2014). The
- biomass of shrubs and non-dominant tree species was not listed here due to a lack of available data.

- From Table 2, 69% of measured forest trees were the dominant species locally. The leaf biomass of
- measured tree species in Beijing, Wuhan, and Chengdu comprised 87%, 35%, and 3% of the total leaf
- biomass in Beijing, Hubei, and Sichuan provinces, respectively. They totally contributed 37% to the
- national total leaf biomass of dominant tree species.
- The revised Table 2 is listed below. We also added the notes to the proportion of leaf biomass as
- follows: "b the proportion of leaf biomass of each plant species to the total leaf biomass in Beijing,
- Hubei, and Sichuan provinces, respectively; "-" for shrubs and vine means no data, "-" for trees
- means that they were not dominant species.".
- Line 212-218 of the revised manuscript, "Table 2 also shows the proportion of leaf biomass of each
- tree species to the total leaf biomass in Beijing, Hubei, and Sichuan provinces, respectively. The leaf
- biomass was estimated based on statistics of timber volumes with biomass-apportion models (Li et al.,
- 176 2013; Li and Xie, 2014). 69% of measured forest trees were the dominant species locally according to
- 177 Forest Resource Statistics of China. The leaf biomass of measured tree species in Beijing, Wuhan, and
- 178 Chengdu comprised 87%, 35%, and 3% of the total leaf biomass in Beijing, Hubei, and Sichuan
- provinces, respectively. They totally contributed 37% to the national total leaf biomass of dominant
- tree species." is added.
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- 182 References:
- 183 Li, L.Y., Chen, Y., and Xie, S. D.: Spatio-temporal variation of biogenic volatile organic compounds
- emissions in China, Environ. Pollut., 182, 157-168, doi: 10.1016/j.envpol.2013.06.042, 2013.
- 185 Li, L.Y. and Xie, S. D.: Historical variations of biogenic volatile organic compound emission
- 186 inventories in China, 1981-2003, Atmos. Environ., 95, 185-196, doi:
- 187 10.1016/j.atmosenv.2014.06.033, 2014.

Table 2. Plant species sampled at each location, sampling time, and measured emission rates.

No.	Plant species	Vegetation type ^a	Proportion of leaf biomass ^b (%)	Sampling time	Dry biomass of enclosed leaves (gdw)	Normalized leaf-level emssion rate ^c (μgC gdw ⁻¹ h ⁻¹)			
						isoprene	α-pinene	β-pinene	other VOCs ^d
Peking University, Beijing (116°18.30′E, 39°59.82′N)		l							
1	Populus tomentosa Carr.	dbt	17.1	19 Aug. 2014	50.85	140.99	-	-	0.29
2	Platanus orientalis	dbt	0.0	25 Aug. 2014	42.30	41.67	-	-	0.74
3	Populus tomentosa Carr.	dbt	17.1	8 Sep. 2014	57.92	67.96	0.02	0.04	6.58
4	Sophora japonica	dbt	2.0	2 Oct. 2014	54.7	29.16	0.02	0.05	4.83
5	Phyllostachys aureosulcata f. spectabilis	ebt	-	2 Oct. 2014	118.6	187.73	0.00	0.01	3.21
6	Fraxinus chinensis Roxb.	dbt	0.5	5 Oct. 2014	28.7	0.18	0.01	0.08	8.71
7	Sabina chinensis (L.) Ant.	ent	5.5	5 Oct. 2014	136.8	0.00	3.87	0.45	1.45
8	Pinus bungeana	ent	-	5 Oct. 2014	106.2	NA	2.70	4.68	2.92
Jiufer	ng Mountain, Beijing (116°5.02′E, 40°3.27′N)								
9	Aceracede	dbt	-	21 Sep. 2014	26.5	NA	NA	0.06	4.62
10	Quercus wutaishanica	dbt	35.4	21 Sep. 2014	64.3	67.16	NA	NA	2.17
11	Pinus tabulaeformis Carr.	ent	14.9	21 Sep. 2014	95.5	0.07	17.78	188.50	3.49
12	Quercus variabilis	dbt	3.5	21 Sep. 2014	106	0.03	0.07	0.46	1.66
Yunn	neng Mountain, Beijing (116°42.33′E, 40°33.92′	N)							
13	Quercus variabilis	dbt	3.5	7 Oct. 2014	60.9	NA	0.02	0.17	5.45
14	Pinus tabulaformis	ent	14.9	7 Oct. 2014	89.3	0.03	2.94	11.18	1.06
15	Sophora japonica	dbt	2.0	7 Oct. 2014	52.6	142.66	0.29	1.83	3.28
16	Sabina chinensis (L.) Ant.	ent	5.5	7 Oct. 2014	162.1	0.02	1.71	0.23	0.73
17	Populus simonii	dbt	17.1	7 Oct. 2014	18.3	46.93	0.12	0.31	10.33
18	Crataegus pinnatifida	dbt	-	27 July 2016	52.10	0.13	0.10	-	1.06
19	Weigela florida (Bunge) A. DC.	dbs	-	27 July 2016	23.53	0.02	0.29	-	6.12
20	Quercus aliena Bl.	dbt	35.4	27 July 2016	77.82	40.79	0.16	-	2.22
21	Corylus heterophylla Fisch	dbs	-	27 July 2016	22.14	1.72	0.29	-	13.59
22	Betula platyphylla Suk.	dbt	4.5	27 July 2016	20.41	NA	20.42	-	9.10
23	P. thomsoni	dv	-	27 July 2016	8.86	NA	NA	-	6.65

24	Picea meyeri Rehd. et Wils.	ent	-	27 July 2016	82.41	0.05	1.13	-	1.05	
25	Pirus, i, f.	dbt	-	27 July 2016	23.23	0.23	0.03	-	1.07	
26	Juglans mandshurica Maxim	dbt	1.8	27 July 2016	49.24	0.12	15.20	-	1.89	
27	Lespedeza bicolor Turcz.	dbs	-	28 July 2016	21.15	0.29	NA	-	10.34	
28	Fraxinus chinensis Roxb.	dbt	0.5	28 July 2016	20.63	0.16	0.12	-	4.85	
29	Acer mono Maxim	dbt	-	28 July 2016	26.77	0.42	NA	-	1.15	
30	Deutzia grandiflora Bunge	dbs	-	28 July 2016	9.11	20.56	0.52	-	9.80	
Wuling Mountain, Beijing (117°25.97′E, 40°37.85′N)										
31	Populus tomentosa Carr	dbt	17.1	8 Oct. 2014	23.8	40.59	0.02	0.10	6.86	
32	Salix matsudana	dbt	1.3	8 Oct. 2014	18.4	15.45	0.02	0.07	5.31	
Beijing (39°58.99′	Gardening Research Institute, Beijing (N)	(116°28.56′E,								
33	Salix matsudana f. pendula.	dbt	1.3	19 May 2016	64.85	28.62	0.05	-	3.27	
34	Pinus tabulaeformis Carr.	ent	14.9	19 May 2016	326.64	0.00	41.02	-	0.37	
35	Cotinus coggygria Scop.	dbs	_	19 May 2016	28.01	0.02	154.26	-	2.50	
36	Aesculus chinensis Bunge	dbt	_	19 May 2016	95.45	NA	5.16	-	0.94	
37	Forsythia viridissima Lindl	dbs	_	20 May 2016	34.15	0.08	0.02	-	4.95	
38	Malus 'Sparkler'	dbt	-	20 May 2016	53.34	0.03	1.05	-	2.04	
39	Buxus megistophylla Levl.	dbs	-	20 May 2016	43.11	NA	NA	-	0.59	
40	Cerasus sp.	dbt	-	20 May 2016	51.53	0.02	0.38	-	0.92	
41	Salix matsudana Koidz.	dbt	1.3	20 May 2016	36.19	27.08	0.56	-	6.32	
42	Sabina chinensis (L.) Ant.	ent	5.5	20 May 2016	108.35	0.08	18.64	-	3.05	
43	Syringa oblata Lindl.	dbs	-	20 May 2016	32.08	0.26	3.35	-	6.12	
Yunwu M	Tountain, Wuhan (114°15.11'E, 31°12.15'	N)								
44	Pteroceltis tatarinowii Maxim.	dbt	-	23 Oct. 2014	15.9	1.87	2.65	10.90	23.68	
45	Cunninghamia	ent	4.7	23 Oct. 2014	183.7	0.03	1.11	0.18	0.92	
46	Metasequoia glyptostroboides	ent	0.3	23 Oct. 2014	47.9	0.18	46.53	6.42	3.47	
47	Platycladus orientalis	ent	1.3	23 Oct. 2014	100.7	2.20	9.32	0.95	1.66	
48	Cinnamomum bodinieri Levl.	ebt	0.0	23 Oct. 2014	32.8	1.55	1.92	4.50	3.85	
Maan Mo	ountain, Wuhan (114°26.25′E, 30°31.43′N	1)								
49	Phoebe zhennan S. Lee	ebt	-	24 Oct. 2014	36.1	0.05	0.10	0.08	4.03	
50	Cinnamomum bodinieri Levl.	ebt	0.0	24 Oct. 2014	47.8	0.19	0.54	1.32	2.24	

51	Fraxinus chinensis Roxb.	dbt	-	24 Oct. 2014	39.5	0.39	0.34	0.36	1.68		
52	Pinus massoniana	ent	17.2	24 Oct. 2014	81.3	0.02	5.55	14.89	2.94		
53	Liquidambar formosana Hance	dbt	0.1	24 Oct. 2014	25.2	30.13	707.12	2542.13	90.44		
54	Quercus wutaishanica	dbt	11.8	24 Oct. 2014	99.8	76.71	NA	NA	0.88		
55	Paulownia Sieb.	dbt	0.0	24 Oct. 2014	54.2	NA	0.17	0.09	2.40		
Qingcheng Mountain, Chengdu (103 °53'E, 30 °51'N)											
56	Cinnamomum bodinieri Levl.	ebt	0.1	2 Sep. 2016	30.9	NA	NA	-	1.39		
57	Metasequoia glyptostroboides	ent	0.2	2 Sep. 2016	21.3	NA	181.26	-	4.08		
58	Phyllostachys viridis	ebt	-	2 Sep. 2016	13.4	12.30	23.47	-	5.40		
59	Phoebe zhennan S. Lee	ebt	0.2	2 Sep. 2016	21.8	0.14	33.83	-	3.28		
60	Osmanthus fragrans	dbs	-	2 Sep. 2016	61.4	0.17	3.78	-	2.43		
61	Alangium chinense (Lour.) Harms	dbs	-	2 Sep. 2016	39.9	0.16	1.07	-	1.50		
Long	Longquan Mountain, Chengdu (104 °28'E, 30 °56'N)										
62	Ligustrum lucidum	ebs	-	6 Sep. 2016	19.3	0.05	1.60	-	6.72		
63	Platanus orientalis	dbt	0.0	6 Sep. 2016	34	96.52	0.33	-	2.80		
64	Broussonetia papyrifera (Linn.) L'H ér. ex Vent.	dbt	-	6 Sep. 2016	19.1	838.62	0.45	-	4.80		
65	Rhus chinensis Mill.	dbs	-	6 Sep. 2016	45.3	52.54	NA	-	1.40		
66	Axodiaceae	ent	2.0	6 Sep. 2016	54.6	0.75	77.62	-	0.81		
67	Pittosporum tobira	ebs	-	6 Sep. 2016	20.4	NA	15.85	-	1.38		
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a dbt: deciduous broadleaf tree; ent: evergreen needle-leaf tree; ebt: evergreen broadleleaf tree; dbs: decidous broadleaf shrub; ebs: evergreen broadleleaf shrub; dv: decidous vine.

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^b the proportion of leaf biomass of each plant species to the total leaf biomass in Beijing, Hubei, and Sichuan provinces, respectively; "-" for shrubs and vine means no data, "-" for trees means that they were not dominant species.

^c normalized to the standard condition (i.e., temperature=30°C; PAR=1000 μmol photons m⁻² s⁻¹) and transferred to the leaf-level emission rate from the branch-level one; "-" means no measurement, "NA" means emission was not detected.

^d including 99 quantified VOC compounds excluding isoprene, α-pinene, β-pinene, and acetylene.

4. They discuss the emissions of several other VOC compounds that are not commonly considered in biogenic emission samples. Specifically alkanes and aromatics (e.g. xyxlenes, propane, ethane, isopropylbenzene). I am not familiar of any biogenic emissions of these compounds or biochemical pathways for form them within the leaf structure. It seems like these might be artifacts of the GC system, but without knowing more details and/or seeing results from blank samples, I can't tell. But emissions of these compounds from vegetation are not commonly reported. Since their work focuses primarily on monoterpenes and isoprene, I would suggest to just omit these compounds, as they don't seem to add any valuable information, and their presence is suspicious.

Response:

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- Isoprene, monoterpenes, and sesquiterpenes are the dominant compounds emitted from vegetation. It
- is reported that plants can also emit many other BVOC species (Guenther et al., 2006; 2012). In our
- study, we developed the statistical approach for estimating representative emission rates of isoprene
- and monoterpenes, and determined their emission rates for 192 plant species/genera in China due to
- 209 lack of observations for other BVOCs.
- 210 In China, measurements of BVOC emission rates are relatively uncommon. Isoprene and
- 211 monoterpenes have often been the only VOC species measured in China, with lack of measurements
- 212 for other VOCs. To obtain more accurate BVOC emission rates and have a comprehensive
- 213 understanding on characteristics of BVOC emissions from plants in China, we conducted field
- 214 measurements using our established semi-static enclosure system and analyzed emissions of 102
- VOCs using a custom-built online GC-MS/FID system.
- Before analyzing the background and emission samples by GC-MS/FID, the blank samples were
- 217 measured and evaluated. It could be concluded that there were hardly baseline noise and the
- 218 GC-MS/FID system was clean so that no VOC species were produced. However, the errors introduced
- by quantification of concentration and calculation of emission remained inevitably, although the
- system had a high accuracy as described in **Response to Comment 1 of Reviewer #1**. Due to the very
- slight emission of some VOC species, their emission rates might be of some uncertainties. Despite
- 222 this, our study would give us some valuable information on VOC emissions from plants in China.
- In the added uncertainty analysis in Section 5 in revised manuscript, we stated the uncertainty of other
- VOCs measurements. Line 458-460, "Additionally, the errors introduced by quantification and
- 225 calculation remained unavoidably, which caused some uncertainties to measurements of other VOCs
- emissions due to their slight emission potential." is added.
- In Section 6, we also made a further explanation on the measurements of other VOCs. Line 480-482,
- 228 "The results from measurements of other VOC species are expected to provide perspectives on

229 understanding VOC emissions from plants in China, despite of possible uncertainty." is added.

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- 5. My analysis of this work is that it's a nice start, but the uncertainties are very large (and not
- sufficiently addressed) and these are not sufficient to extrapolate emissions to other times of year or
- 233 different individuals within the same plants species.

234 Response: Accepted.

- 235 (1) We added the uncertainties analysis for our emission measurements and suggestions on the future
- study in the revised manuscript, referring to your valuable comments.
- We changed the title of Section 5 to "Evaluation and uncertainties" in the revised manuscript. In this
- section, the uncertainty analysis of measurements and calculations was added as follows:
 - "The measured emission rates from collecting samples by the semi-static enclosure system were believed to be realistic ones for each vegetation species tested (Zimmerman, 1979). But there were some uncertainties regarding the emission rate measurement and calculation in this study. In our study, only one or two replicates of samples for BVOC emissions from each plant species were collected. This might introduce accident errors from sampling and measurement. The results could neither be representative to different individuals of the same plant species because BVOC emissions from different individuals of the same plant species may differ much owing to varied ambient conditions, such as temperature, radiation, precipitation, soil moisture, and so on. Due to limited time and resources, some experiments were made in May and October, early or late growing season. Different sampling time or season could result in different ambient conditions which cause large differences in emissions. Additionally, the growth of plants in different seasons usually varies, which cause radically different leaf emissions. There was another source of uncertainty of the very high emission rates measured in this study, such as 838.62 µg C gdw⁻¹ h⁻¹ for isoprene, 707.12 and 2542.13 µg C gdw⁻¹ h⁻¹ for α- and β-pinene, respectively. The concentrations of emission samples of plants with the highest emissions were usually high enough to cause detector saturation to the GC-MS/FID system, which would be overcome by dynamic enclosure technique during sampling. Errors were introduced inevitably to their quantification owing to the dilutions before analysis. However, conclusion could still be drawn from the results that the plants had high isoprene and pinene emission potential. Additionally, because of the only one or two samples of plant species with such high emission rates, their representative emission categories could not be worked out by statistics. In future study, to obtain more accurate emission data and more comprehensive and detailed emission categories, the quantitative measurements must be enhanced with increasing number of samples for one plant species and for plants within different emission categories.

Uncertainty existed in calculation and normalization of emission rates. Firstly, according to Equation (2), the accuracy of acetylene is the key to the accurate calculation of emission rates. In our study, the measured acetylene concentrations of background and emission samples mostly ranged from 1.09 to 3.05 ppbv and 0.88 to 2.48 ppbv, respectively. We concluded that the uncertainties of quantification of acetylene at its concentration range were < 7%, through conducting repeated measurement of the calibration at 1 and 2 ppbv separately. Secondly, PAR sensor was placed horizontally on the ground to monitor PAR outside the bag due to limitation of actual operation, which resulted in a difference of 5–10% for PAR between inside and outside the bag (Ortega et al., 2008; Yaman et al., 2015). This would introduce uncertainties to BVOC normalized emission rates, 0.9–3.1% for isoprene and 0.2–2.4% for other compounds.

Additionally, the errors introduced by quantification and calculation remained unavoidably, which caused some uncertainties to measurements of other VOCs emissions due to their slight emission

274 potential."

- (2) Thank you for your insightful comment. In our study, only one or two replicates of samples for BVOC emissions from each plant species were collected and some experiments were made in May and October, early or late growing season, due to limited time and resources. This introduced large uncertainties to the measured results and their representativeness to different individuals of the same plant species in different regions and seasons, because BVOC emissions from different individuals of the same plant species may differ much owing to varied ambient conditions in different time of year and regions. Additionally, the growth of plants in different seasons usually varies which cause radically different leaf emissions. In future study, measurements of emissions from different individuals of the same plant species in different regions and seasons are urgently needed to obtain representative emission rates for different cases in China.
- We have described the above uncertainties and gave suggestions for reducing the uncertainties in the revised manuscript. Please refer to **Response to Comment 2 of Reviewer #1** for details.

Other Comments:

1. The classifying of the emissions into different categories (low, medium, high, etc.) based on statistical distributions of the emissions is valid. And I think this approach is useful for model inputs where the modeler could input a certain mean (+/- range) of emissions based on the species distribution and leaf area index. I just question the accuracy of the emissions for the reasons and examples cited above.

Response:

Thank you for your valuable comment.

As you concern, the base of the approach in our study is the accuracy of emission rates database used. When we selected the original emission rate observations from China and abroad, strict evaluation and screening on the data quality were made, as suggested by Niinemets et al. (2011). For the foreign researches with large quantity of emission measurements, the emission rate observations mostly from dynamic open systems were summarized. In China, the emission rate measurements were mainly derived from simple static systems and also our semi-static system, primarily due to a lack of other available measurements.

When extrapolating the original measurements to standard ones, to minimize the errors, we only included measurements conducted during the day on temperature and light conditions approximate to the standard conditions. Our field measurements were also conducted during the daytime on the temperature and light conditions of 26-33 °C and 600-1300 µmol photons m⁻¹ s⁻¹, respectively.

We have evaluated our established statistical method on the quality of original emission rate observations in the manuscript. Line 408-418 in Section 5, "Certainly, the foundation of our method was the evaluation and screening based on the quality of the emission rate observations and the use of reliable extrapolations, as suggested by Niinemets et al. (2011). Firstly, they recommended that only two quality classes (quantitative measurements and semi-quantitative measurements) associated with dynamic systems could be used to construct BVOC emission inventories. Meanwhile, non-quantitative measurements (i.e., those conducted using static enclosure systems or possibly semi-static and some dynamic systems) should not be used in BVOC modeling. In our study, the summarized reported emission rate measurements from abroad were mainly derived from dynamic open systems. While those in China were measured using simple static systems, primarily due to a lack of other measurements, which should only be used in emission rate estimates when there are no other available observations for a region." The errors from extrapolation have also been explained in Section 2.2.

We should realize that the accuracy of local emission rate observations in China should be questioned because most of them were performed with simple static systems, which were non-quantitative measurements according to Niinemets et al. (2011), although the measured results could still be used in emission rate estimates when there were no other available quantitative measurements and semi-quantitative observations for China. Our study did provide a comprehensive understanding of BVOC emissions from plants in China and explored a scientific method for the accurate estimation of species-specific representative BVOC emission rates for plants based on reliable original emission rate observations. In future study, more local reliable quantitative measurements are needed for

- 328 estimating accurate representative emission rates for China.
- 329 Line 463-465 in the revised manuscript, "In future study, more local reliable quantitative
- measurements are needed for estimating accurate representative emission rates for China." is added.

- 332 Reference:
- Niinemets, Ü., Kuhn, U., Harley, P. C., Staudt, M., Arneth, A., Cescatti, A., Ciccioli, P., Copolovici, L.,
- Geron, C., Guenther, A., Kesselmeier, J., Lerdau, M. T., Monson, R. K., and Peñuelas, J.:
- Estimations of isoprenoid emission capacity from enclosure studies: measurements, data
- processing, quality and standardized measurement protocols, Biogeosciences, 8, 2209-2246,
- 337 doi:10.5194/bg-8-2209-2011, 2011.

338

- 2. Figure 1 looks suspiciously like Figure 1 in Ortega et al. (Chemosphere, 72, p. 365, 2008).
- 340 **Response: Accepted.**
- We drew Figure 1 in the manuscript referring to the drawing of Figure 1 in Ortega et al. (2008). In the
- revised manuscript, we added the reference. "(referring to the drawing of Figure 1 in Ortega et al.
- 343 (2008))" is added in the title of Figure 1.

344

- 3. Tables 1 and 9 are almost redundant.
- 346 **Response: Accepted.**
- Thank you for your suggestion. We deleted Table 1 in the revised manuscript.
- Line 90 in the revised manuscript, "Table 1 shows the different isoprene emission categories used in
- several studies and the resulting emission rates for some plants with high emission potentials." is
- deleted. Line 87-88, "... resulting in different emission rates for specific plants." is revised to "...
- resulting in different emission rates for specific plants (Table 8).".
- 352 The numbers of tables in the text and table captions are reordered.

- 4. The authors frequently refer to "pinene", which seems useless to me. I think they should distinguish
- between this and general Monoterpenes, or specify if they mean alpha pinene, beta pinene, or the sum
- of the two.

357 **Response: Accepted.**

- 358 Thank you for your comment and we are sorry for the confused expression.
- 359 The "pinene" in the manuscript is the sum of α -pinene and β -pinene. We specified it in the revised
- 360 manuscript.
- Line 18-19, "... while needle-leaf trees emitted more pinene." is changed to "... while needle-leaf trees
- 362 emitted more pinene (sum of α -pinene and β -pinene).".