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Molecular characterization of organic aerosols in the Kathmandu Valley, Nepal: insights into primary and secondary sources

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Improvements for future studies

There are additional improvements for future studies to be addressed.

Firstly, in our current study, we identified the organic tracers of biomass burning, plant-debris, fungal spores, biogenic secondary organic aerosols (BSOA) and aromatics-derived secondary organic aerosols. In order to explain the other sources of organic aerosols in the Kathmandu Valley, much more tracers need to be identified or if some new organic tracers special for the aerosols in the Kathmandu Valley, even better. **Secondly**, the conversion factors of tracers to organic carbon from local emissions and formation (e.g., hardwood combustion, BSOA formation, etc.) are critical for more precise source apportionments and therefore, it is valuable to increase the investigation of emission characteristics not only in the suburban region but also rural or urban regions. It would reduce the uncertainties of the source appointment using the tracer-based methods. **Thirdly**, comprehensive methods (e.g., carbon isotope and modeling) need to be integrated for the source apportionment of organic aerosols in the Kathmandu Valley. For example, constraints on primary and secondary particulate carbon sources using chemical tracers and ^{14}C methods have recently been used in many regions (e.g., California, East China, northern Italy, etc.) (Zhang et al., 2015;Liu et al., 2016;Bonvalot et al., 2016;Salma et al., 2017;Sheesley et al., 2017;Gilardoni et al., 2011). It will provide strong constraints on the relative contributions of the major sources of carbonaceous aerosols (both organic and black carbon), discriminating anthropogenic from natural, and primary from secondary aerosols. **Fourthly**, the results of the present study demonstrate that the biomass burning plays a significant role in atmospheric aerosols in the Kathmandu Valley, therefore, the influences of biomass burning on the formation of secondary organic aerosols could be further studied, especially during the heavily polluted dry season, with additional simultaneous measurements of precursors (e.g., NOx and O_3), $PM_{2.5}$ and so on at the same time. **In addition**, knowledge of various chemical species in size-segregated aerosols is important for understanding the physical and chemical atmospheric processes that affect aerosol properties, especially during haze episodes. However, such studies are limited in the Kathmandu Valley, South Asia. Therefore, the size-resolved chemical investigations are especially needed

48 in the future. **Finally**, it is necessary to conduct a systematic and comprehensive study in Kathmandu
49 Valley and South Asia to assess the potential positive and negative artifacts and corresponding impacts
50 on the organic tracers.

Table S1 Recoveries and MDLs of the target compounds

Compounds	Addition (ppb) (n=6)	Recovery (%)	MDLs (ng m^{-3})
Levoglucosan	100	81.6 ± 10.4	0.08
Mannosan	100	80.7 ± 11.7	0.07
Galactosan	100	76.3 ± 9.43	0.07
p-Hydroxybenzoic acid	100	80.8 ± 9.78	0.07
Vanillic acid	100	79.4 ± 11.5	0.11
Syringic acid	100	78.6 ± 14.2	0.11
Dehydroabietic acid	100	85.4 ± 13.4	0.11
Glucose	100	86.2 ± 13.2	0.05
Fructose	100	86.5 ± 9.57	0.06
Trehalose	100	80.9 ± 10.4	0.13
Sucrose	100	82.2 ± 12.6	0.08
Xylose	100	81.7 ± 10.1	0.09
Mannitol	100	80.6 ± 7.25	0.12
Arabitol	100	85.8 ± 8.57	0.06
Sorbitol	100	77.5 ± 11.1	0.12
Erythtitol	100	90.8 ± 3.80	0.07
Diethyl	100	87.7 ± 10.0	0.12
Di-n-butyl	100	89.5 ± 11.5	0.12
Bis-(2-ethylhexy)	100	89.9 ± 5.36	0.12
Malic acid	100	76.4 ± 5.44	0.13
Pinic acid	100	76.1 ± 7.85	0.10
cis-Pinonic acid	100	73.9 ± 5.00	0.08
Azelaic acid	100	87.2 ± 6.77	0.11
Methyl-β-D-xylanopyranoside	100	90.1 ± 13.2	
D_3-malic acid	100	70.5 ± 14.6	

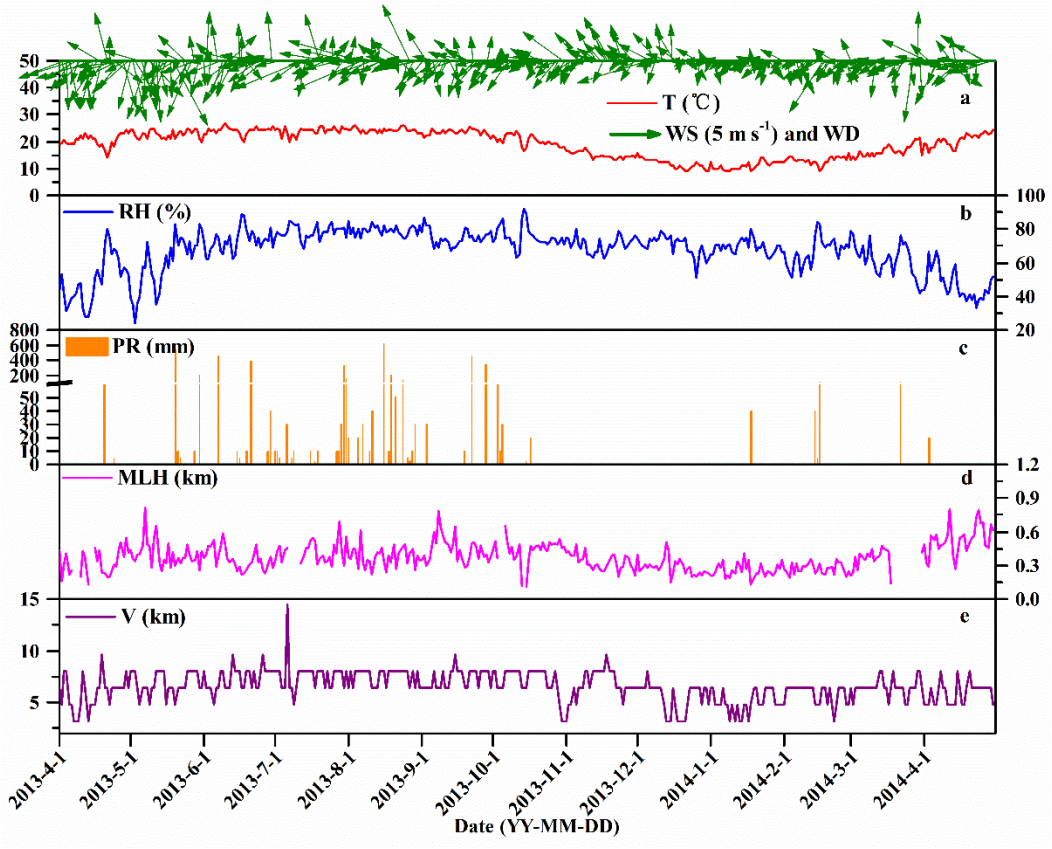
53 Table S2 Estimation of measurement uncertainty

Tracers	Tracer formula	Surrogates	Surrogate formula	EQ (%)	[a] ER (%)	EA (%)
cis-Pinonic acid	$C_{10}H_{16}O_3$	*cis*-Pinonic acid			26.1	
Pinic acid	$C_9H_{14}O_4$	Pinic acid			23.9	
3-Methyl-1,2,3-butantricarboxylic acid	$C_8H_{12}O_6$	*cis*-Pinonic acid	$C_{10}H_{16}O_3$	60	26.1	65.4
3-Hydroxyglutaric acid	$C_5H_8O_5$	*cis*-Pinonic acid	$C_{10}H_{16}O_3$	95	26.1	98.5
3-Hydroxy-4,4-dimethylglutaric acid	$C_7H_{12}O_5$	*cis*-Pinonic acid	$C_{10}H_{16}O_3$	65	26.1	70.0
cis-2-Methyl-1,3,4-trihydroxy-1-butene	$C_5H_{10}O_3$	Erythritol	$C_4H_{10}O_4$	85	9.2	85.5
3-Methyl-2,3,4-trihydroxy-1-butene	$C_5H_{10}O_3$	Erythritol	$C_4H_{10}O_4$	85	9.2	85.5
trans-2-Methyl-1,3,4-trihydroxy-1-butene	$C_5H_{10}O_3$	Erythritol	$C_4H_{10}O_4$	85	9.2	85.5
2-Methylglyceric acid	$C_4H_8O_4$	Erythritol	$C_4H_{10}O_4$	20	9.2	22.0
2-Methylthreitol	$C_5H_{12}O_4$	Erythritol	$C_4H_{10}O_4$	15	9.2	17.6
2-Methylerythritol	$C_5H_{12}O_4$	Erythritol	$C_4H_{10}O_4$	15	9.2	17.6
β-Caryophyllenic acid	$C_{13}H_{20}O_4$	Pinic acid	$C_9H_{14}O_4$	120	23.9	122.4
2,3-Dihydroxy-4-oxopentanoic acid	$C_5H_8O_5$	Azelaic acid	$C_9H_{16}O_4$	90	12.8	90.9

54 [a] ER is the difference between 100% and mean recovery of each surrogate standard.

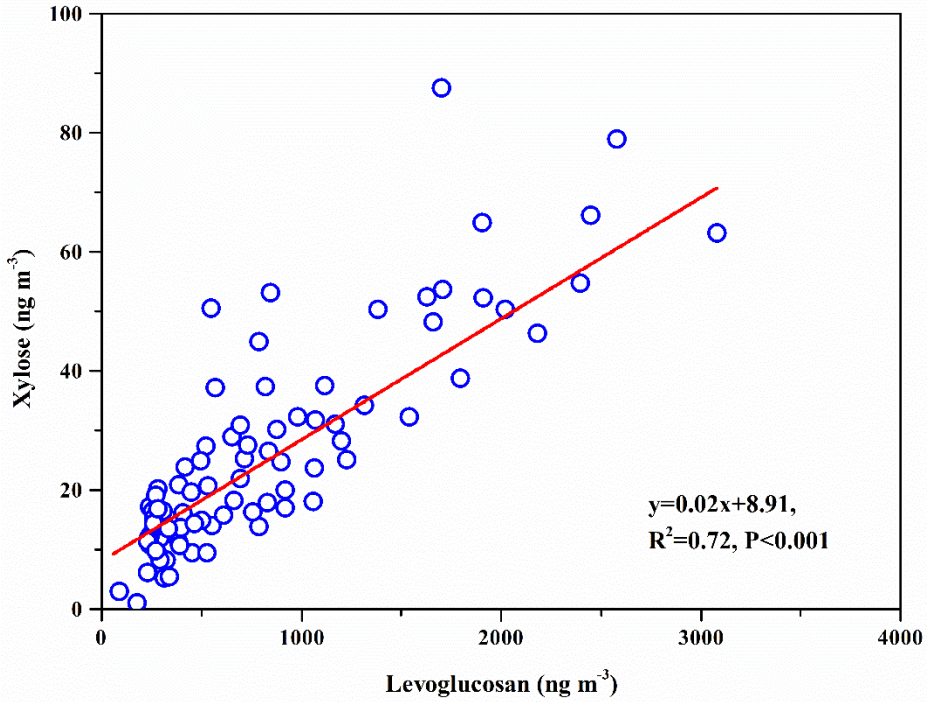
Table S3 Uncertainties using different ratios from other studies for biomass burning estimation

		Lev/OC ratios					
		8.14%	8.27%	7.94%	14.0%	12.0%	10.1%
Pre-monsoon	Average	28.5	28.0	29.2	16.6	19.3	23.0
	Stdev	10.3	10.1	10.5	5.96	6.96	8.29
	Median	28.0	27.5	28.7	16.3	19.0	22.6
Monsoon	Average	17.7	17.4	18.2	10.3	12.0	14.3
	Stdev	5.11	5.03	5.24	2.97	3.47	4.13
	Median	17.2	16.9	17.6	9.99	11.7	13.9
Post-monsoon	Average	36.3	35.8	37.3	21.1	24.7	29.4
	Stdev	10.4	10.3	10.7	6.07	7.08	8.44
	Median	32.3	31.8	33.2	18.8	21.9	26.1
Winter	Average	27.9	27.5	28.6	16.2	18.9	22.6
	Stdev	8.63	8.50	8.85	5.02	5.86	6.98
	Median	24.9	24.5	25.5	14.5	16.9	20.1
Annual	Average	24.9	24.6	25.6	14.5	16.9	20.2
	Stdev	10.4	10.3	10.7	6.07	7.08	8.44
	Median	22.4	22.1	23.0	13.0	15.2	18.1



57

58 **Fig. S1**. Temporal variations of (a) wind speed (WS), wind direction (WD), temperature (T), (b) relative
59 humidity (RH), (c) precipitation (PR), and (e) visibility (V) monitored at Tribhuvan International Airport
60 and (d) mixing layer height (MLH) from Vaisala ceilometer at Bode site, Kathmandu Valley from April
61 2013 to April 2014.



62

63 **Fig. S2.** Correlations between levoglucosan and xylose during the sampling period (April 2013 to April

64 2014).

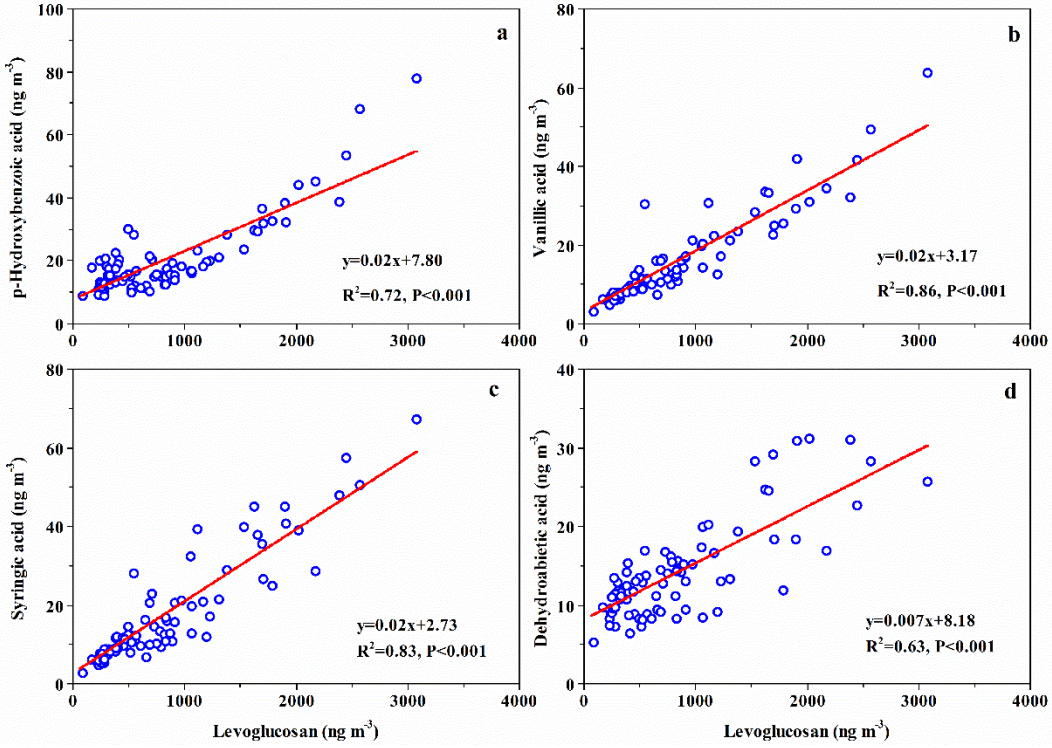
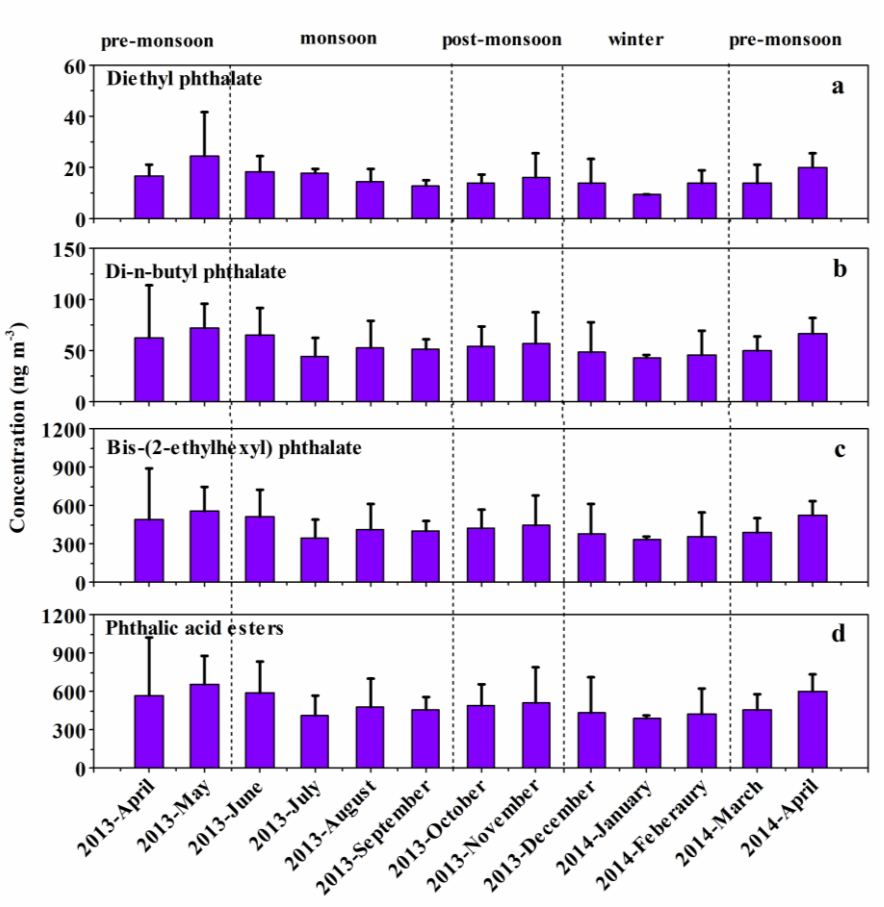


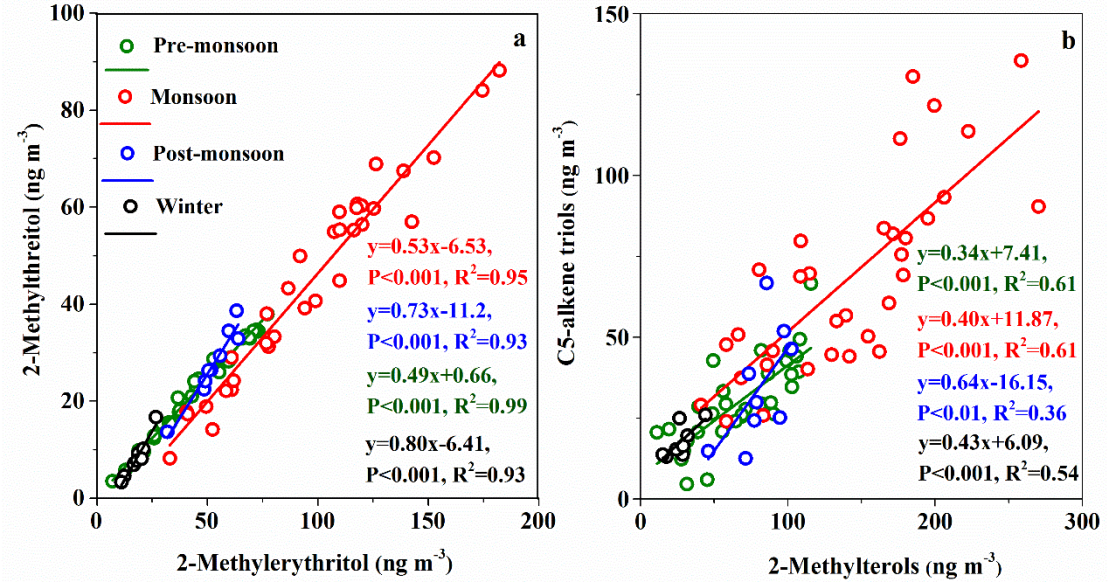
Fig. S3. Correlations between (a) levoglucosan and *p*-hydroxybenzoic acid, (b) levoglucosan and vanillic acid, (c) levoglucosan and syringic acid, (d) levoglucosan and dehydroabietic acid in Bode aerosols during the whole year (April 2013 to April 2014).



69

70 **Fig. S4.** Monthly variations of phthalic acid esters at Bode site, Kathmandu Valley during April 2013-

71 April 2014.



72

73 **Fig. S5.** Correlations between (a) 2-methylthreitol and 2-methylerythritol, (b) C5-alkene triols and 2-

74 methylterols in Bode aerosols during the sampling period (April 2013 to April 2014).

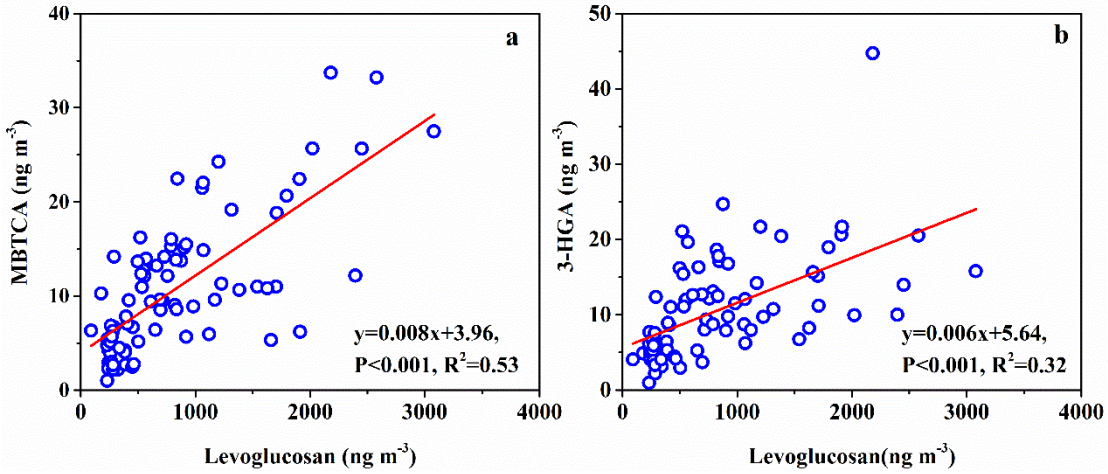


Fig. S6. Correlations between (a) MBTCA and levoglucosan, (b) 3-HGA and levoglucosan in Bode aerosols during the sampling period (April 2013 to April 2014).

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