We would like to thank the anonymous reviewer's helpful comments and suggestions which, we believe, have supported to improve the quality of the current manuscript. We have tried our best to incorporate the reviewers' comments in the manuscript. In the following responses, the reviewer' original comments are in black, authors' responses in blue and changes in the manuscript in red.

Responses to Referee #1

General comments:

This article presents a sizable set of field measurements for total suspended particulates in Bode, focusing on major inorganic ions and a wide range of markers to discuss primary and secondary emission sources. This work adopts organic molecular markers established in the published literature with tremendous effort given to chemical speciation which is commendable.

1. In addition to providing data beneficial for control strategies and relevant implications, more clearly specified scientific novelty (new scientific findings that research communities have not known yet, and need to learn) will further enhance the value of this work.

Response: Thanks for the constructive suggestion. We now added more descriptions of the scientific novelty in lines 634-633, "In addition, the current study based on the molecular level-source apportionment of OC in heavy polluted region from South Asia provides a much more specific quantification of source estimation for OC, which is different from previous studies based on the bulk carbonaceous aerosol using radiocarbon (^{14}C) measurements, PMF and CBM".

2. The major revision for the manuscript lies in the sampling artifacts and relevant impacts on the reported concentrations as well as discussion. Quartz filters are used for sample collection, which is recognized to incur positive sampling artifacts by 10–20% for OC and up to 16 % for organic tracers. The positive sampling artifacts on organics are cited based on a published study (Ding et al. 2013) instead of experimental measurements devoted for this manuscript. Authors are encouraged to examine appropriate ways (available in published literature) to assess such positive artifacts and corresponding impacts on reported data, followed by correction accordingly. At least, correction based on make-up experiments or post data analyses need to be considered. Similar correction/discussion should be given to the effects of discounted recovery rates.

Response: We understand that the main concern is about the sampling artefacts. As far as we are aware, filter sampling using a high-volume sampler is a common method to collect atmospheric particles. There are mainly two types of filters. One is fiber filter (e.g., glass, quartz), the other is porous membrane filter (e.g., Teflon). We agree with the reviewer that a positive artifact may occur during sampling due to adsorption of gaseous species on the surface of quartz fiber filters. Alternatively, a negative sampling artefact may occur during sampling due to a loss of semi-volatile organic compounds from the aerosols collected on quartz fiber filters. Both evaporation and adsorption can be affected by changing pressure or temperature.

There are some studies trying to elaborate the positive and negative artifacts using the backup filter and denuder (Genberg et al., 2011;Subramanian et al., 2004;Yttri et al., 2011b;Yttri et al., 2011a;Gelencsér et al., 2007;McDow and Huntzicker, 1990;Chow et al., 2010;Cheng et al., 2009;Turpin et al., 2000). Subramanian et al. (2004) quantified the negative artifact to be small, typically less than 10% (6.3%±6.2%) of the OC by the denuded quartz filter with a carbon-impregnated glass fiber backup filter and the positive artifact of 10–20% according to the quartz behind quartz approach, respectively for the 24 h aerosol samples from a hill in Pittsburgh, Pennsylvania. Yttri et al. (2011a) reported the mean positive sampling artifact of OC ranged from 11±2 % at the Finnish site Hyytiälä to 18%± 4% at the Birkenes site in Norway. Cheng et al. (2009) reported 10% of the OC captured by the bare quartz filter was due to the positive artifact in Beijing, China, from January to February 2009.

Similar to bulk OC, the individual organic tracers also suffer from the effect of sampling artifact. However, to our best, we did not find such detailed information in the previous literatures. Furthermore, the sampling artifacts differ from approaches, study regions and sampling period. Therefore, it is difficult for our current study to estimate the artifacts and make correction, which need a systematic and comprehensive study in the future in Kathmandu Valley and South Asia.

We reorganized the sentences denoting possible artifacts as “There may be positive and negative artifacts during the sample handling/conditioning due to the adsorption/evaporation processes of organic aerosols (Fu et al., 2010;Li et al., 2018;Boreddy et al., 2017;Oanh et al., 2016). In a comparable study, Ding et al. (2013) reported the positive artifacts of 10–20% for OC and up to 16% for organic tracers using a backup quartz filter placed behind the main quartz filter” in lines 132-136.

We also add the description about results of OC and molecular tracers in the field blank filters in lines 145-146 with “The concentrations of OC and EC from field blank filters were 0.59±0.13 μg m^{-3} and 0.00 μg m^{-3}, respectively. The OC data reported here were blank corrected” and lines 169-170 of “Field blank filters were analyzed by the procedure used by the samples above, but no target compounds were detected.”

The reviewer also suggested us to consider the effects by the discounted recovery rates. Regarding this point, Stone et al. (2012) developed an empirical approach to estimate the error from surrogate quantification (EQ) based on homologous series of atmospherically relevant compounds and applied that to the study in another rural site in Kathmandu. According the method, now we also add the “estimation of measurement uncertainty” to our MS in Section 2.4 (Line 180-197) as “Since there is no commercial standard available for most SOA tracers (except for cis-pinonic acid and pinic acid), the use of surrogate standards for quantification introduces additional error to the measurements. Error in analyte measurement (EA) is propagated from the standard deviation of the field blank (EFB), error in spike recovery (ER) and the error from surrogate quantification (EQ):

$$EA = \sqrt{EFB^2+ER^2+EQ^2}$$

EFB was 0 in this study due to SOA tracers that were not detected in the field blanks. The spike recoveries of surrogate standards were used to estimate the ER of tracers, ranging from 9.2% (erythritol) to 26.1% (cis-pinonic acid). According to Stone et al. (2012), there is an empirical approach to estimate EQ based on homologous series of atmospherically relevant compounds. The relative error introduced by each carbon atom (En) was estimated to be 15 %, each oxygenated functional group (Ef) to be 10% and alkenes (Ed) to be 60%. Therefore, the EQ are calculated as:

$$EQ=En\Delta n+ Ef\Delta f+ Ed\Delta d$$

where Δn, Δf and Δd are the difference of carbon atom number, oxygen-containing functional group and alkene functionality between a surrogate and an analyte, respectively.

The estimated uncertainties in tracer measurement is presented in Table S2. The EQ ranged from 15% (2-methyltetrols) to 120% (β-caryophyllenic acid) in this study. Propagated with the error in recovery, EA were estimated in the range of 17.6% to 122.4%."

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

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

2. Quartz filters are also well known to adsorb semi-volatile inorganics (e.g. nitrate, chloride, and ammonium), another type of major positive sampling artifact. How such effects affect the various correlations and discussion involving inorganic ions mentioned in the manuscript deserve to be examined to revise the discussion accordingly.

Response: We agree with the reviewer. Both the positive and negative artifacts may occur during sampling aerosols on the quartz filters. Single filter-based sampling and filter pack systems without any denuders or without backup filters are still widely used and the extent of the sampling artifacts of volatile species in these sampling systems is not well understood. Wei et al. (2015) reported the loss of NH_4^+, NO_3^-, and Cl^- accounting for particulate matter, which ranged from 1.85% to 41.44% with a typical value of about 10%. Liu et al. (2014) showed that during 24 h sampling with denuder sampler at National Chiao-Tung University campus, Taiwan, the positive artifact of NH_4^+ and Cl^- was not important for aerosol mass concentration, and existed in NO_3^- species only, which was 5.0%±6.5% of actual NO_3^- concentration. Timonen et al. (2014) reported a positive artifact of 1.3% ± 1.8% for ammonium and 42% ± 33% for nitrate of the PM_1 samples with back-up filters from an urban, background area near Helsinki city.

During our sampling, we used the single filter-based sampling and filter pack systems without any denuders or backup filters. Therefore, we cannot quantify the positive sampling artifacts of nitrate, chloride, and ammonium. If we used some correction ratios adopted from the previous studies, it will systematically modify the concentration data for those compounds; however, it will not affect the correlations among different compounds. Therefore, we intend to keep the current dataset without correction for the sampling artifact. Definitely, in the future study, we will choose more suitable sampler to reveal the effects of such sampling artifacts.

In addition, the concentrations of major ions reported in the MS have already been blank corrected. We add the description about results of major ions in lines 141-142 of "They denoted less than 5% of the real sample concentrations in the field blank filters (Tripathee et al., 2017)".

Specific comments:

1. Line118-120: Why would comparing only BC and O_3 between Bode and Paknajol be sufficient to conclude that Bode is a representative site for Kathmandu Valley?

Response: Actually, the descriptions from lines 103-122 are all about the explanation of Bode as a representative site for the Kathmandu Valley. More specifically, the BC and O_3 between Bode and Paknajol (in lines 118-120) are chosen as example to illustrate this claim.

2. Line 252: Incense burning can also emit levoglucosan. Would such emissions be significant at the study site?

Response: Yes, most incenses are made of wood powder (https://en.wikipedia.org), which can emit levoglucosan when they are burnt. There are religious activities in the Kathmandu Valley, so Bode may be influenced by the incense burning. However, we don't know how large its effect is. Now we add a sentence in lines 308-309 as "We must point out that the incense burning in Kathmandu Valley may also influence the levoglucosan concentration".

3. Line 450-451: Reference is needed for "During the fires, substantial amounts of aerosols and VOCs including isoprene and monoterpenes would generate, ...". Similar description also appears at other locations.

Response: Missing citation has been included in the reference. Please see lines 405 and 476.

4. Line 452-453: An R^2 value of 0.32 does not indicate good linear correlation between levoglucosan and 3-HGA, even though the p-value is less than 0.001. There is also inconsistent use of "r" vs. "R^2" throughout the work. The use of statistical mean deserves more careful consideration and application.

Response: Suggestion taken. All r were changed into R^2. Please see lines 257-259 and lines 320-322. Though the linear correlation coefficient is not very good, it indicates to some degree that monoterpene tracers may be influenced by biomass burning.

5. Line 489-490: Typically, atmospheric samples show greatly fluctuated concentrations. Prior to calculating and using the mean values for various comparison, a distribution of concentrations can be examined to evaluate whether a median or mean should be used to convene corresponding discussion. Use of Lev/OC ratio among major biomass types deserves re-consideration. This use assumes that atmospheric degradation pattern of levoglucosan and overall OC at any time remains the same. This assumption is questionable, especially under varied temperature, relative humidity, locations, types and abundance of major biomass burnt, dominant burning conditions, varied transport, etc. It is also worth noting that oxidation intermediates of levoglucosan (and other organics) remain part of overall OC, which adds additional questions on the validity of adopting the ratio. The ratio at a given time point is a net result of multiple atmospheric processes on overall OC and levoglucosan therein.

Response: Now we add the median concentrations in Table 1.

We totally agree with the referee that levoglucosan/OC (Lev/OC) ratios varied depending on biomass burning sources and conditions and degradation. The degradation of levoglucosan is affected by radicals (OH), temperature, and relative humidity (Hoffmann et al., 2010;Bai et al., 2013;Lai et al., 2014;Slade and Knopf, 2014). However, given the complicated biomass burning sources, conditions and degradation mechanism, it is not applicable to estimate the uncertainty for the moment.

Still, the Lev/OC ratio of ~8.2% in the burning source have been widely used (Graham et al., 2002;Fu et al., 2014;Ho et al., 2014;Sang et al., 2011;Zhu et al., 2016;Mkoma et al., 2013), especially in Asia. Although the ratios in the BB source emissions vary among different types of biomass fuels and burning conditions (Mochida et al., 2010). In this work, the Kathmandu valley is considered as a source region of organic aerosols, therefore, we believe that using Lev/OC ratio of 8.14% is reliable to estimate biomass burning contributions. The estimation can also be compared to other studies using the same ratio. Now we estimate the uncertainties using different ratios from other studies in Table S3.

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

We added the sentence of “although the ratios in the BB source emissions vary among different types of biomass fuels and burning conditions (Mochida et al., 2010)” in lines 508-509 and “The mean value of Lev/OC value of biomass burning from main biomass types was 10.1%. In this study, we choose the mostly used values of 8.14% for biomass burning estimation (Graham et al., 2002;Fu et al., 2014;Ho et al., 2014;Sang et al., 2011;Zhu et al., 2016;Mkoma et al., 2013). In addition, we also calculated the uncertainties of using different ratios (see Table S3), the diagnostic ratios among molecular tracers and OC (e.g., Lev/OC) from direct emissions are critical for more precise results. It’s meaningful to understand the emission characteristics for individual OC emission categories, as well as in different locations, especially in South Asia.” in lines 518-524. We reorganized the precaution as the second reviewer suggested in lines 501-503, which is “It should be noted here that tracer methods can provide a reasonable estimation, but uncertainties are introduced considering the site differences and the lack of representative source profiles for the given study location. The contribution evaluated from each source to OC in the current study is still inferable".

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