This paper investigated the molecular compositions of urban organic aerosols by ultra-high resolution FTICR MS. The results give an insight into the sources of organic aerosols with different SOA loadings in urban Tianjin, China on a molecular level. The data of this kind is informative and valuable and related discussions would inspire some new aspects for further OA studies. While some statements and conclusions in this study need to be described more clearly. After addressing the following issues, it is suitable to be published on ACP.

Comments:

1 My major concern is the sample selection and classification. Discussion and results in this work are too much dependent on the sample selection and classification. It is possible to draw a opposite conclusion with different samples or other classification way. A simple statement of “according to biogenic SOA tracers....” in section 2.1 is not very sufficient.

For example, the high SOA-loading sample is not the one with highest SOA tracer concentration, especially for the nighttime one (Figure 1). The daytime high SOA-loading sample also accompanied with highest levoglucosan concentration, how to evaluate the variation of relative contribution from biomass burning and secondary formation? Is it better to make the classification based on the tracer contribution among total organic aerosol concentration? What’s more, the biogenic SOA concentrations in the night moderate SOA-loading sample are comparable or slightly higher than the high SOA-loading one. I may suggest a more detailed statement on the sample selection.

2 In the section of Materials and Methods, some important information directly related to the discussion needs to be added.
The discussion on compound categorization throughout the main text, which support some key points in the work. Only a very short statement (lines 24 in page 4) on compound categorization referring another two papers may not a good way.

Ion suppression in the ionization source of FTICR MS could influence the ionization efficiency a lot. How much would this effect the further discussion in this work?

Sampling strategy needs to be briefly introduced in this section. "Described in a previous study" (line 29 in page 3) may be not a clear statement.

3 Some conclusion in section 3.3 and 3.4 may need more evidence.

In section 3.3, authors draw a conclusion that biomass burning as the major source of CHON compounds. Why the sample with highest levoglucosan concentration (high SOA loading day sample) show very low CHON contribution (figure 2a), while low-levo sample (low SOA loading day sample) with high CHON contribution (figure 2e).

In section 3.4, authors suggest CHOS from biogenic VOC oxidation (line 27 page 8), while biogenic VOC emissions should be very low in winter.

The statements in lines 18 page 8 and lines 10-13 page 9 need more evidence. Could the author show other evidence or data to support this conclusion?

**Minor comments**

Figure 1: I think the data in Figure 1 is from a published paper (Fan et al., 2020). It should be better to show Figure 1 in supplementary.

Page 1

line 20-23: this sentence needs to be re-organized. How to arrive the conclusion of "sensitive to ...chromophores"?
Line 25: Why photochemistry processes produce compounds with low oxygen content under low SOA loading condition? SOA should generally be more oxidized.

lines 30-31: this sentence is a little confusing. Suggest to be clearer.

Page 5

line 5: change “of” to “for”

lines 12-16: These sentences are unclear.

Page 6

Lines 4-5: Do you mean the compound number or contribution with a difference of one order of magnitude? Pls be clear.

Lines 6-7: Based on figure 2, the contribution of CHO compounds increased from moderate SOA loading sample to high SOA loading sample.