**Summary:**

In this scientific work, the authors have used excitation-emission matrix (EEM) fluorescence spectroscopy coupled with Fourier transform ion cyclotron resonance mass spectroscopy (FT-ICRMS) to study the chemical composition of water soluble organic carbon (WSOC) ambient aerosols in Karachi, Pakistan. In addition to that, they tried to postulate the different formation mechanisms of these organic compounds. They identified three broad components of moieties using parallel factor analysis (PARAFAC), of which two were humic-like (C1 and C2) and one was protein like (C3). They also tried to correlate these PARAFAC components to the extensive datasets of brown carbon (BrC) chromophores available in literature through the total number of carbon (C) atoms present in the molecular formula and double-bond equivalence (DBE). Except the discussion of formation mechanism, the rest of the methodology and findings of this work is quite trivial and another repetition of multiple EEM spectroscopy based ambient aerosol characterization studies.

**Strength of this work:**

The authors analysed the FT-ICR MS data and came up with the most probable formation mechanisms from different molecular signals in addition to EEM spectroscopic studies.

**Limitations of this work:**

- *Lack of discussion on seasonal variation:* As seen in Figure 1, the study site is located at a very interesting geographical location with great seasonal variation in wind direction. In pre-monsoon and monsoon, the wind flow is directed from middle-east Asia and Arabian Sea, whereas the wind trajectories arise from North Pakistan and North-West India during post-monsoon and winter. This will result in very different chemical compositions of ambient particulate matter (PM) reaching the study site. For example, in the months of May-June, the aerosol composition will be close to marine aerosol composition, but in Oct-Dec there will be molecular signals of biomass burning emissions as previous studies have pointed out extensive crop burning and biomass burning for heat generation during winter in that part of the world. Which suggests that the wintertime aerosols will probably have higher S content and less oxygenated organics because the OH radical photochemistry is limited during winter and the atmospheric transformation is driven by NOx chemistry. The authors have reported all the data in a combined way, which does not give the readers the broader picture of the regional specific atmospheric chemistry of the study site. Postulating molecular formation pathways without considering the meteorological conditions can also lead to erroneous assumptions.

- *Lack of relevant references:* In continuation to the previous point, as the authors have not discussed the geographical context of this work, they have also failed to compare their findings with previous works carried out in similar locations. Although they have mentioned a few studies carried out in the Indo-Gangetic Plane (IGP) while reporting mass absorption efficiency (MAE) and Aerosol absorption exponent (AAE), this kind of comparisons have not been made for EEM spectroscopic studies. Previous EEM spectroscopic analyses of ambient aerosol in IGP and other parts of the world have found similar PARAFAC components (two HULIS and one protein like). These references from around the globe should be mentioned and compared with the findings of this work.

- *Structure of the Results and Discussion Section*: The results and discussions section needs to be restructured. For the convenience of the readers, the discussion of "Underlying implication of PARAFAC component to BrC absorption" should be done under section 3.3 and the discussion on formation pathways should be under section 3.4. That way the flow of information will be more coherent.

- The discussion on formation pathway of S-containing compounds should be more condense and can also be moved to supplementary information (SI). The authors have mentioned that S containing compounds have almost no effect on BrC chromophores. They also reported that the two Humic-like PARAFAC components had very little S containing compounds, mostly component C3 had the highest S containing compound. Eventually it has also been shown that component C3 has the lowest overlap with the BrC region in figure 5. Therefore, in terms of climate relevant BrC chromophores, this pathway is not as important as CHO and CHON formation pathways.

- The overall grammar and clarity of the current section 3.3 (proposed to be made section 3.4 after restructuring) is unsatisfactory. Too much statistics have been used in sentences, which can instead be represented graphically. Sentences need to be written with proper grammar, for example 407-408 is unclear and needs to be rewritten, so does 413. There are many occurrences like this, so the reviewer suggests a rewriting of this whole section in a clear and concise manner. In the rewriting, the authors should also elaborate and clarify how they reached conclusions made in 520-522.

    - **But most importantly in this section**, the authors have looked at some molecular signals found through FT-ICR MS and compared those molecules with aged byproducts of certain precursors reported in previous literature. And by doing that they have tried to postulate these known reaction pathways to be present in their study samples. These are good hypotheses, but there is no concrete way of establishing these reaction pathways in the scope of this study. Therefore, it doesn't add up as novel new information. If the authors can combine other analytical study with the collected filters (if there is any remaining), for example $^{1}$H NMR spectroscopy of WSOC, in which they can quantitatively compare the spectra of the precursor and aged molecules with their sample spectra and confirm its presence, that would be a much stronger argument for the formation pathways.

**Minor corrections:**

Title: The title should be reconsidered. Instead of water soluble particulate matter, water soluble organic carbon is a preferable choice. The study location should be kept Karachi or mentioned a location in South East Asia.

**Corrections in SI:**

**Figure S2:** The title of the figure mentions February 16, but the legend in the figure shows the trajectory starting from 17 February 2016.