We thank the reviewers for the insightful comments. We have addressed the reviewers' comments point by point as indicated below and revised the manuscript accordingly. The reviewers' comments are in italics and changes made to the manuscript are in quotation marks.

Reviewer #1

General comments:

The authors described the relatively low-cost analytical method for measurement of water-soluble and total Fe and Cu in aerosols and analyzed the aerosol samples over Atlanta.

Such a high-frequency monitoring measurement is extremely useful for validation of the models and environmental assessment. I can recommend this paper for publication in Atmospheric Measurement Techniques and have minor comments to improve the paper.

Response: We thank the reviewer for the positive and constructive comments. We appreciate the suggestion of improving the language. We have also addressed the specific comments by the reviewer accordingly.

Specific comments:

1) *Title: The reader might expect measurement of various chemical forms of iron and copper mentioned in introduction. Please consider rephrasing it by total and water-soluble, etc.*

   Response: We have changed the title, as suggested, to: A Method for Liquid Spectrophotometric Measurement of Total and Water-Soluble Iron and Copper in Ambient Aerosols

2) *p.7, l.191: Please discuss feasibility of in-situ measurements of ambient aerosols to investigate the speciation of WS Fe and WS Cu.*

   Response: We thank the reviewer for this comment. We have added "In-situ measurements of Fe(II) is feasible using this colorimetric method when connected to a PILS or similar system (Oakes et al., 2010; Rastogi et al., 2009). It is not feasible to perform near-real time measurements of Fe(III) due to its long reduction reaction time (overnight). Quantifying speciation of Cu is challenging due to the tendency of Cu(I) to undergo disproportionation in aqueous solutions (Johnson et al., 2015). In-situ measurements of Cu(I) is possible with the presence of reducing agents and masking ligand to inhibit Cu(II) interference (Moffett et al., 1985), but further studies are necessary to achieve an acceptable detection limit." to p.7, l.192-197.

3) *p.10, l.289: Please specify the chemical composition for the mass fraction of 5.63%.*

   Response: The abandance of iron element in the Earth's crust is 5.63%. To clarify, we rewrote the sentence in p.10, l.297 to "As expected the total Fe concentration was much higher than total Cu since Fe is a common element in the Earth's crust (mass fraction of iron element is 5.63 %) (Tomaszewski, 2017) and ubiquitous in mineral dust, while $PM_{2.5}$ Cu in urban environments is mainly derived from vehicle brake and tire wear, and tail-pipe emissions resulting from motor-oil impurities;".

4) *Conclusion: Please discuss feasibility of high-frequency monitoring of Fe and Cu in size-resolved aerosol and rainwater samples.*

   Response: We do not attempt to make high-frequncy monitoring in the size-resolved samples since the ultrafiltration procedure takes time (30-60 min) and cannot be done automatically.

5) *Data availability: How did you estimate the solubilities when the total and WS concentrations were blank?*

   Response: We thank the reviewer for the comment. We have added the following text for clarity "Those solubilities when total or WS concentrations were blank were estimated using half of the detection limit of the total or WS concentration." to p.10, l.292-293.

6) *Data availability: Please comment on the data such as 2017/3/20, which showed higher concentration through ultrafiltration than a 0.45 µm filter.*

   Response: We thank the reviewer for catching this. In most cases, the higher concentration through ultrafiltration than a 0.45 µm filter was because those two actual concentrations of that specific sample were similar and there were uncertainties during both measurements. We also deleted the data of the samples whose concentrations through 30k ultrafiltration were significantly higher than that through 0.45 µm filter (WS Fe of 3/20/2017 and 3/22/2017) and added the following text "The data of the samples whose concentrations through 30k ultrafiltration were significantly higher than that through 0.45 µm filter was removed." to p.10, l.306-307.

7) *Data availability: Please comment on the data such as 2017/4/10 and 2017/12/18, which showed higher solubilities than 100%.*

   Response: We thank the reviewer for pointing this out. The higher solubility than 100% was probably because the actual solubility was close to 100 % and there were uncertainties during both water-soluble and total measurements. To address the reviwer's concern, we changed higher solubilities than 100% to 1 and added "Due to uncertainties of both water-soluble and total measurements, a few calculated Cu solubilities were higher than 1 and were treated as 1 when calculating the average fraction of Cu." to p.10, l.290-292.

8) *p.16, l.510: Please correct the unit.*

   Response: Revised. Thanks!

9) *Supplement, p.2, l.32: Please correct "colloidal??".*

   Response: Revised. Thanks!

10) *Supplement, p.2, l.53: Please correct "Figure S8".*

Response: Revised. Thanks!

References

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