

This manuscript presents a comprehensive study on PM<sub>2.5</sub> water-soluble metals regarding their temporal and spatial variations, as long as the source apportionments of these metals. A new method of quantifying water-soluble metals in filter extractions was introduced based on a commercially available metal monitor. Source apportionment of important water-soluble metals and WSOC are performed by using PMF method. Overall, this manuscript is very well written, fits perfectly for the scope of ACP journal, and more importantly provides significant insight on important redox-active water-soluble metals in PM<sub>2.5</sub>. Findings described in this manuscript are very informative in both ambient aerosol studies as well as aerosol related health studies. In my view, this manuscript is acceptable for publication after the authors address my minor comments as below.

### **Specific comments:**

1. Line 90: The author's statement regarding total and water-soluble metals is too strong and needs to be rephrased. The exact role of metal solubility on introducing health effects is not clear yet and apparently still being studied. There have been at least quite a few studies showing significant evidences on the role of insoluble/total metals leading to different health outcomes. Although it is widely accepted that water-soluble metals are more likely bioavailable and therefore having a more obvious effect in generating oxidative stress, making such as statement is likely to be too subjective at this stage.
2. Line 188: The description about the system purging is not clear and more clarifications are needed. Is the DI (with 2% nitric acid) also coming from the same sampling streamline out of the auto sampler? Based on the description I assume that the DI with acid is also aerosolized and collected by filter on Xact? If so, was this collection spot skipped?
3. Line 380-397: In Figure 7(b), it seems that the brake/tire ware factor is also showing higher contributions in winter time comparing to summer, although the authors mentioned in previous sections (line 320) that higher loadings of Cu, Fe and etc. were observed in summer and were attributed to drier conditions, which enhance the re-suspension of road dust. These two observations seem to be inconsistent and more explanations are needed on this point.
4. Line 410, 435-446: The authors discussed the possible reasons why a large fraction of Fe is associated with Secondary sources, but not for Cu, which is also a major species shown in secondary sources. Additional discussions on this point are needed as well. It is not intuitively obvious to me why Cu is found in the "Secondary source" factor given the known origins of this metal