Responses to Jianzhong Ma (Editor)

We greatly appreciate the editor for taking time to read this manuscript and providing insight comments. Below, we have addressed your concerns and provided our point-by-point responses in blue.

Solubility refers to the extent to which a substance dissolves in a particular solvent. According to IUPAC definition, solubility is the composition of a saturated solution, expressed in terms of the proportion of a designated solute in a designated solvent (http://goldbook.iupac.org/S05740.html). The solubility of a substance fundamentally depends on the physical and chemical properties of the solute and solvent as well as on temperature, pressure and the pH of the solution (http://en.wikipedia.org/wiki/Solubility).

In this pater, Li et al. (2015) defines element solubility as the percentage of water-soluble concentrations of an element divided by its total concentration (Page 13018, Line 3-4). This definition has a conflict with the traditional concept of solubility in chemistry. The word "solubility" is used in the title of the paper, and appears frequently in the Abstract and the Introduction, but its own definition is not available until Sect. 3.4.

The definition of solubility in our study is indeed different from its traditional concept in chemistry defined by IUPAC and Wikipedia. Apart from the total content of trace elements, we also determined the concentration of their water-soluble fraction which are highly reactive in aqueous phase of aerosols and can bring great damage to human health and ecosystem due to their high mobility and bioavailability.

Solubility is usually calculated as the ratio of dissolved trace metal concentration to total concentration in work on aerosol dissolution (Deguillaume et al., 2005). The solubility here represents the capacity of an element's chemical reactivity/mobility as well as the degree of its bioavailability. In this study, the solubility concept, defined as the percentage of water-soluble concentrations of an element divided by its total concentration, was modified from sequential extraction procedures for trace metals

speciation (see the next responses for detail).

In order to avoid misunderstanding and to distinguish from traditional concept of solubility, we have shifted forward our definition of solubility into the Introduction.

The authors also use the phrase "total fraction" to describe the total concentration of trace elements in fine particles. In addition to ionic compounds (water-soluble fraction) and elementary substances of trace elements, are there any other forms of elements existing in the total fraction which were experimentally analyzed in this study? Again, "element solubility" in this study appears to have nothing to do either with the solubility of ionic compounds or the solubility of elementary substances.

The phrase "total fraction" here means the total content of water soluble and water insoluble fraction of trace elements. Tessier et al. (1979) developed a sequential extraction procedure for partitioning the speciation of particulate trace metals, including *Water soluble* + *Exchangeable, Bound to carbonates, Bound to Fe-Mn oxides, Bound to organic matter, and Residual* fraction. Fernández-Espinosa et al. (2004) divided chemical speciation of trace metals in fine particles into four metallic fractions: *Soluble and exchangeable metals; Carbonates, oxides and reducible metals; Bound to organic matter, oxidisable and sulphidic metals; and Residual metals.* As we primarily focused on the bioavailability (water solubility) and total concentration of trace elements, extraction procedure in this work concentrated on the aqueous extraction (approximating the total content of aerosol elements), regardless of the other partitioned speciation. The element solubility defined in this study was accordingly derived from the modified extraction procedure.

It is worth noting that the above partitioning method for trace elements speciation using sequential extraction procedure is based upon their likely released fraction under various simulated environmental conditions, rather than the specific chemical forms like ionic compounds or elementary substances, etc.

Reference

Li, T., Wang, Y., Li, W. J., Chen, J. M., Wang, T., and Wang, W. X.: Concentrations and solubility of trace elements in fine particles at a mountain site, southern China: regional sources and cloud processing, Atmos. Chem. Phys. Discuss., 15, 13001-13042, 10.5194/acpd-15-13001-2015, 2015.

References:

- Deguillaume, L., Leriche, M., Desboeufs, K., Mailhot, G., George, C., and Chaumerliac†, N.: Transition metals in atmospheric liquid phases: Sources, reactivity, and sensitive parameters, Chem. Rev., 105, 3388-3431, doi:10.1021/cr040649c, 2005.
- Fernández-Espinosa, A. J., Rodríguez, M. T., and Álvarez, F. F.: Source characterisation of fine urban particles by multivariate analysis of trace metals speciation, Atmos. Environ., 38, 873-886, 10.1016/j.atmosenv.2003.10.046, 2004.
- Tessier, A., Campbell, P. G. C., and Bisson, M.: Sequential extraction procedure for the speciation of particulate trace metals, Anal. Chem., 51, 844-851, 10.1021/ac50043a017, 1979.