Tong et al. report systematic investigations on RS yields of ambient PM in clean and polluted environments using EPR spectroscopy. Furthermore, they found that using the surrogate mixtures can reproduce the relative fractions of different types of radicals formed by ambient PM, which would be useful for future laboratory studies. This ms is generally well written, highlighting the complicated mechanisms responsible for radical formation, but it lacks mechanistic insight into how different chemical compositions in PM generate different relative fractions of RS. I hope to see more detailed discussions about the mechanisms in the text, although it is understood that It may be beyond the scope of this study. Overall, I support this ms to be published in ACP after addressing the following points.

Major points

Can you elaborate why naphthalene and beta-pinene were used as representative SOA in Beijing and Hyytiala, respectively? Anthropogenic and biogenic representatives?

For the sample treatment procedure of HA or FA water suspensions, why was the procedure necessary? So, does it mean that a water-soluble fraction of HA or FA was extracted and only analyzed?

Relative fractions of different radicals, particularly organic and OH radicals, seem to be dependent on the origin of PM samples. What would be the most important factor determining such relative fractions?

In Figure 4, why were only transition metals shown in its absolute concentration, whereas the others were in the relative abundance (%)?

Lines 369-373: Why did naphthalene SOA have a lower radical yield than β-pinene SOA with the same mass concentration? This information should be important to generalize the findings of this study.

It seems that consistently the higher relative fraction of C-centered radicals is associated with the lower relative fraction of OH radicals. Does it suggest that OH radicals are consumed by something to generate C-centered radicals? This point deserves detailed discussions in the text.

When HA was replaced by FA, the dominant radical species becomes O2- from C-centered radical. Do you any explanation on the different radical distribution?

Lines 496-498: It is said that “the higher relative amount of detected radicals and H2O2 formed by urban PM2.5 can see as a measure of higher potential oxidative damage”, but the totally formed radical amount decreases with PM mass. The statement seems contradicting and needs clarification.

Minor points

Line 236: The -> the