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Interactive comment on “Atmospheric oxidation of isoprene and 1,3-butadiene: influence of aerosol acidity and relative humidity on secondary organic aerosol” by M. Lewandowski et al.

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We would like to commend the authors for a very interesting study. The few comments we have regard the interpretation with respect to acidity, which hopefully can be considered in a revision of the manuscript.

The interpretation of the experimental results relies heavily on the ability of $[H^+]_{air}$ to capture the in-situ pH of atmospheric particles. The methodology of Surrat et al.(2007) involves extracting filter samples in water, measuring its pH and then back-calculating the amount of H^+ in the original aerosol (assuming that H^+ is conserved). This is

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Discussion Paper



Interactive
Comment

largely equivalent to an “ion balance” method, because the H^+ measured corresponds to an aerosol sample diluted by orders of magnitude, so that the ions present are largely dissociated (this is affected by the degree of dilution used, but does not change the inherent nature of the methodology). However, two studies currently in discussion in ACPD (Guo et al., 2014; Hennigan et al., 2014) raise doubts on whether such methods can be used as a proxy for in-situ aerosol pH, as it is shown that H^+ from ion balance is not correlated with in-situ aerosol pH for a wide range of aerosol conditions. This is especially relevant for this study as chamber RH is varied; the latter affects the amount of in-situ aerosol liquid water (hence pH) in a way that is not reflected in $[\text{H}^+]_{\text{air}}$. Even if a constant RH were used as in Surrat et al.(2007), the variable chemical composition (and potentially phase state) of the particles across experiments can still have a nontrivial effect on aerosol liquid water content, hence in-situ pH.

With the above said, we are not implying that the study here is fundamentally flawed. However, it would be good to see the acidity interpretation accompanied by both $[\text{H}^+]_{\text{air}}$ and pH calculations from a thermodynamic model (e.g., AIM, ISORROPIA, MESA) applied to the chamber conditions. Guo et al.(2014) and Hennigan et al.(2014) have shown that models applied to complex SE US and Mexico City aerosol, even at relatively low RH, can provide a much better representation of aerosol pH than ion-balance methods, provided that they are applied in “forward mode”. This is what we recommend here as well to be done. In the absence of gas-phase inorganic component measurements, one can still apply thermodynamic models in a useful manner (e.g.,Guo et al.,2014).

Thank you in advance for considering our comments.

Respectfully,

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