Atmos. Chem. Phys. Discuss., 14, C4521–C4524, 2014
www.atmos-chem-phys-discuss.net/14/C4521/2014/

Atmospheric Chemistry and Physics
Discussions
Open Access

Interactive comment on "The influence of physical state on shikimic acid ozonolysis: a case for in situ microspectroscopy" *by* S. S. Steimer et al.

S. S. Steimer et al.

sarah.steimer@psi.ch

Received and published: 6 July 2014

R: P. 7358, Line15: The spatial resolution should be quoted and referenced.

A: The current sentences (starting from line 13) will be replaced by: "Scanning transmission x-ray microscopy (STXM) is a microscopy technique which offers a high spatial resolution of down to less than 20 nm (Raabe et al. 2008) though it is usually more around 30 nm for environmental applications (Bluhm et al. 2006). Combining the spectroscopic technique with microscopy therefore offers the possibility to measure chemical maps at this high spatial resolution (Russell et al., 2002; Maria et al., 2004; Moffet et al., 2010, 2013) that cannot be achieved by optical methods such as Raman microscopy (Ivleva et al., 2007; Yeung et al., 2009)." Specific resolution depends on

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

instrument used and on the type of sample and energy range. For ideal cases resolutions below 10 nm have been achieved. For environmental applications at the carbon and oxygen edges, resolution is usually kept around 30 nm.

R: P. 7359, Line 20: The formula for silicon nitride should be Si_3N_4.

A: Will be changed.

R: p. 7363, Lines 15-17: When the authors refer to the peak height, how is this calculated? Is it the average over the particle, or is it the sum of the per-pixel absorbance?

A: It is the average over the particle. This will be added to the text.

R: p. 7368, Lines 1-3: The authors include a discussion of shape and include speculation that the particles are either spheres or non-spheres. How did they determine this? I would think it is possible to more quantitatively determine/estimate the thickness of the particles (pure or reacted) by assuming a density and absorption cross section.

A: Regarding shape: the attached image shows an example of a dry particle (black line) and a wet particle (71%, grey line) cross section in comparison with the projection of a circle (dark blue, dashed line) and a half circle (light blue, dotted line). From this type of analysis, general trends can be seen. However, no complete reconstruction is possible due to low contrast. This will be added to the supporting information. Particle height: we will calculate this for particles which are not oxidized using a calculated linear absorption coefficient.

R: p. 7369, Lines 24-27: The authors are not able to resolve any radial dependence to the C=C peak around 284.4 and attribute this to an extremely short reacto-diffusive length. It would be nice to see the authors suggest ways that this can be further probed to try and resolve this. Are there another set of experimental conditions or perhaps another system that could be tried to resolve a gradient in functionality? I think some discussion of this would be useful.

A: We do not think that a gradient can be observed with this particular system. However, in principle several different conditions where a gradient should be observable can be thought of. The easiest case would be where Dy is already so low that full exchange of bulk molecules does not happen on timescales of the experiments (such as in our low humidity case), but with a Dx that is still sufficiently high to guarantee reaction over an observable distance. Proteins, in which small molecules can move via percolation, might be one such system. Shiraiwa et al. (2011) have suggested such gradients to occur during nitration of proteins. Another conceivable situation would be that the ratio of the diffusion coefficient to the first order loss rate constant would be of the same order of magnitude for the two reacting species. Since for the conditions of a STXM experiment the condensed phase reactant needs to be at high concentrations to provide sufficient dynamic range, only very soluble oxidants would be feasible for such experiments, which are not simple to dose into our environmental microreactor. We will expand the final discussion in the manuscript to reflect this discussion, even though all this remains speculative.

R: Figure 6d, h: what line corresponds to what particle? It's stated in the caption, but perhaps include a legend.

A: We will do so.

R: Figure 6g: It almost appears that there is a slight ring of red around the particle in c, f, g, etc...perhaps this is an artifact due to normalizing by total carbon

A: This is correct. This “ring” is due to a mixture of alignment issues combined with the relatively low contrast of carbon K-edge measurements.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 7355, 2014.

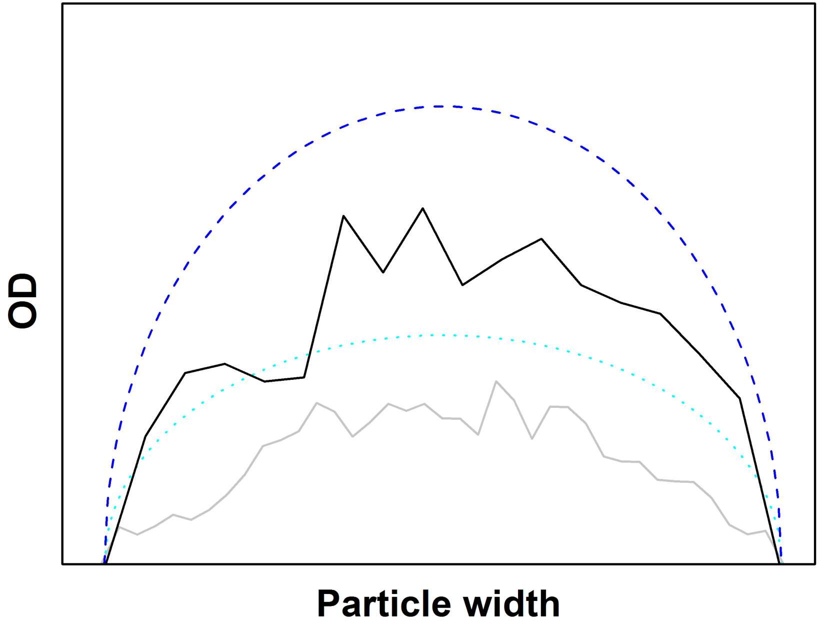

Fig. 1.