Referee 3

I found that the authors of this paper designed a thoughtful online field instrument to measure Oxidative Potential (OP) of aerosols. They discussed the chemistry principles and physical characteristics underlying their instrument design. They demonstrated methodologies' functionality with calibration and case studies. At the end, it's revealing to see their method obtained a good correlation of PM2.5 data to support its technological significance. The work is reported in good alignment with the journal's theme. For these reasons I would recommend its publication.

To inspire potential improvement, I have a few questions on a major aspect. The online and off-line OP measurements both had a reaction time of 20 min. Although it's discussed the selectivity of 20min being its optimality toward DHA's stability and relevance to mimicking conditions in lung cells. Can the author discuss how different reaction time will impact the measurement outcome more clearly?

This comment addresses a very important point. Changing the reaction time will favour the sensitivity towards different components. A longer reaction time will shift the sensitivity towards metals that react catalytically and other slowly reacting components. A shorter reaction time will shift the sensitivity towards radicals and other short-lived components. Therefore, by varying the reaction time we could shift the instrument sensitivity for single compounds or classes of compounds. But to compare measurements it is essential to keep the reaction time constant. We identified 20 min as a good compromise between these opposing effects, but we are aware that this is largely an operationally defined number.

Also, Fig 3 suggests the online measurement can capture data within one min; I wonder how does this 1-min reaction be captured by a 20-min residence time?

Maybe there is a misunderstanding. There are three important time constants in the instrument:

(1) The reaction time between OP-active particle components and AA, which is 20 min (line 214). The AA solution is brought into contact with the particles *inside* the PILS, i.e. within seconds after the particles entered the OOPAAI and therefore also very reactive components are quantified, as described in detail in the method section above.
(2) The reaction time of DHA and OPDA, which is 2 min (line 222).
(3) The time resolution with which the instrument can resolve changes in OP content of particles pumped through the instrument. This time constant is about 5 minutes as illustrated in Section 2.5 and Figure 4A, where instantaneous changes of SOA are introduced into the instrument.

The time resolution (5 min) is much shorter than the reaction time of AA with OP-components (20min) because AA is mixed with the aerosol *inside* the PILS, i.e., within seconds after the particles enter the instrument. The 5min time resolution is caused by diffusional broadening during the 20 + 2 min reaction time and transport of the aerosol extract from the PILS to the detection cell (points (2) and (3) above).
In Figure 3, time on the x-axis refers to the time between aerosol generation and sampling. The OOPAAI captures the particles into the AA solution in < 1 minute. Thus, highly reactive species are almost immediately reacted with AA to produce DHA, which significantly more stable, and is subsequently quantified after the 20 min reaction time. The filter sample results shown in Figure 3 were analysed over time to demonstrate the decay of these reactive species. Comparison with the OOPAAI measurements normalised for mass, show that about two third of OP decays on filters, even when analysed a few minutes after collection.

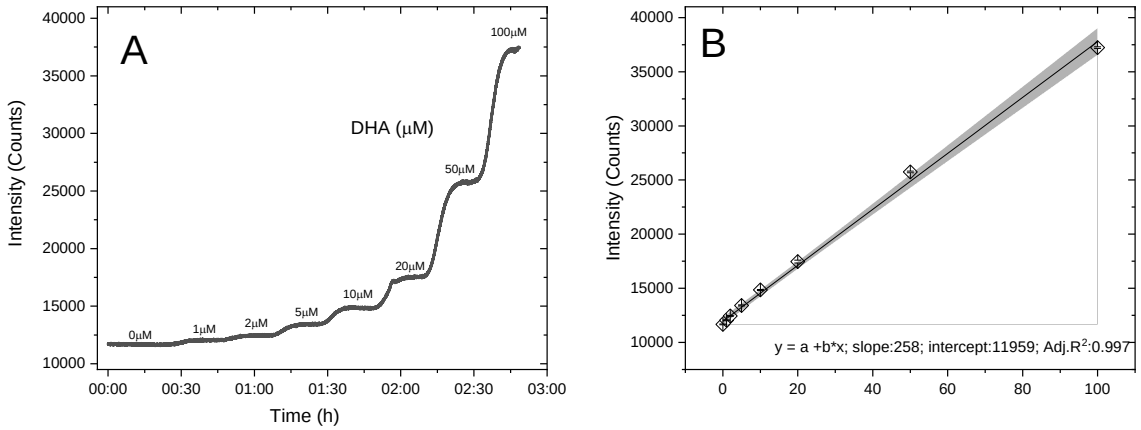
To clarify this, we added a sentence starting at line 274: "In the online instrument, the AA solution is brought into contact with the particles inside the PILS, i.e. within seconds after the particles entered the OOPAAI and therefore, also very reactive components are quantified, as described in detail in the method section above."

In addition to this, I am slightly concerned with the linear fitting on some data sets. In figure 4, it appears to me that the linearity of the intensity-DHA calibration starts deviating after 20 uM, maybe even more at a higher concentration range. Can the author justify the linearity suitable range of the calibration maybe by adding more data points?

We extended the DHA calibration curve beyond the range of 100 µM up to 200 µM the calibration curve slightly starts to deviate from a linear fit. From 1-100uM, however, a linear fit still fits the measured data very well with a R^2=99, which we think is a good fit for the measurements shown here. Therefore, we like to keep the calibration curve up to 100 µM in Figure 4.
To address this effect, we added a sentence to the text starting from line 317: "At DHA concentrations larger than 100 µM the calibration curve starts to flatten and such DHA concentrations are therefore considered beyond the linear calibration range of the instrument (data not shown)."

What will be the statistical confidence of the slope and what magnitude of error can be caused? How to handle measurements larger than 100 µM DHA when working with ambient samples?

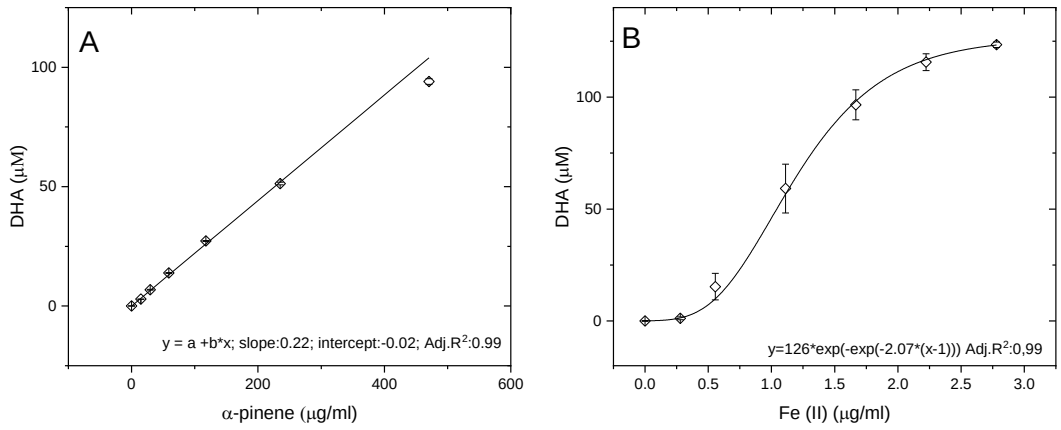
Confidence intervals are now added to Figure 4. In ambient measurements DHA concentrations were always much lower than 100 µM and thus should not cause a significant uncertainty. If this should indeed occur, the air pumped through the instrument could be diluted.



Another is Figure 5B, where it appears not very appropriate to fit the data by a line anymore- can the authors discuss the implication of the seemingly S-shaped trend?

We added sigmoidal fit to the calibration data in Figure 5B and changed the sentence starting from line 336 to:
"In Figure 5B, the OOPAAI response for iron (II) sulfate solutions is given with an adjusted R^2 of 0.99 for a sigmoidal fit."
Furthermore, we added an additional sentence starting from line 337:

“The linear range of the response for Fe(II) is between about 0.5 – 2 µg/ml, above about 2 µg/ml the calibration curve flattens.”



With these, I recommend some revision of the work to be formally published.