

Review of “Dry season aerosol iron solubility in tropical northern Australia”, by V. H. L. Winton, R. Edwards, A. R. Bowie, M. Keywood, A. G. Williams, S. Chambers, P. W. Selleck, M. Desservettaz, M. Mallet, and C. Paton-Walsh (ACP-2016-419).

General Comments

This study presents results from a measurement campaign in northern Australia (SAFIRE), and in particular focuses on the solubility of iron in aerosol sampled from air masses influenced by dust and biomass burning events, both local and distant.

The factors influencing aerosol iron solubility are still not fully understood, and this work provides interesting and valuable insights into the solubility of iron associated with biomass burning aerosol. In particular, the authors provide evidence that iron in freshly emitted combustion aerosol is not particularly soluble, which in turn suggests that atmospheric aging itself plays a significant role in observations of soluble iron in plumes sampled far from combustion sources.

Unfortunately, the paper in its current form is difficult to follow, and I find its conclusions overstated as written. Since my concerns relate to the interpretation and presentation of the data rather than the data itself, however, I do recommend publication in *Atmospheric Chemistry and Physics* after substantial revisions addressing the comments below.

In the following sections, I outline my specific scientific questions/issues regarding the manuscript, organized by manuscript section. Although I've attempted to collate my technical/presentation issues in the final section, which addresses the overall clarity of the manuscript, this wasn't always possible, since I sometimes found cosmetic and substantive issues difficult to disentangle.

Methodology

P5L11 “following the ultra-clean methodology” ... is this a reference to a specific methodology?

Section 2.3.1 From what I understand, there are a number of published methods for determining iron solubility, and a number of operational definitions of iron solubility. How does the strategy presented here fit within the literature/precedent on this topic? Specifically, in P6L1, what does “Leaches 5–etc. *were estimated*” mean? Finally, why was GP22 in particular chosen for filtration?

P7L17 Is this the correct baking temperature?

Results

P8L13 How is nss-K (*i.e.* versus total K) determined here? Is the assumption that *all* K is nss-K, since no trajectories were “mainly oceanic” (P8L1)?

P8L22 The nss-K/Fe ratio during non-fire events (1.3) is higher than that quoted for both crustal aerosol and polluted aerosol. What is the overall significance of this comparison?

P8L29 This discussion could benefit from some reference to the literature.

P9L1 Would you expect the same behaviour for samples collected under different conditions (*i.e.* during non-fire events)? And, what is the overall significance of this observation?

Section 3.4 These results could be presented more clearly—the correlations described aren't actually shown in the figures, and the figures are discussed out of order. Also, Na seems to be repeated in Figures 3 and 4. As a minor note, the section seems to be mis-titled, as it also includes discussion of elements other than iron. More importantly, I'm confused by the reference to "marine conditions" here, as earlier in the paper it was stated that no trajectories were mainly oceanic.

Discussion

P9L29 I'm wondering about the first line of Section 4.1—technically, isn't the *non-fire* PM₁₀ Fe concentration also similar to the Brazilian fire aerosol studies, given that there isn't an obvious correlation between PM₁₀ Fe and fire event presence/absence? Is this a useful comparison? Also, according to Figure 3, the maximum PM₁₀ Fe concentration actually appears to be ~0.8 ug/m³? In addition, what is the overall significance of the comparisons to other fire events if the results aren't scaled to total PM₁₀ / some proxy for proximity to the actual fires?

Section 4.3 I find this discussion confusing as written and very difficult to follow. In particular, I'm confused by this statement: "These estimates are similar to dust estimates around 0.5-2 % at relatively high Fe mass concentrations." Perhaps it might make sense to merge this section with the discussion of the fire/dust events ... why is fractional Fe solubility highest during the dust event?

P12L11 This paragraph is vague as written. "Lower enrichment factors"—lower than what? In addition, I wonder if the EF data should be presented in the results section. There is a lot of information presented here (*e.g.* detailed discussion of Pb, V, Mn sources) and in my opinion, it dilutes the overall message of the paper.

Section 4.5.1 Why is the fractional Fe solubility so high during the dust event? This is discussed in Section 4.6 to some extent, but perhaps that discussion should be moved here?

Section 4.5.2 Again, I don't disagree with anything presented here, but I do think that discussion of Cr/Mo/As is distracting—more time is spent discussing sources of these elements than in discussing the fractional solubility of Fe in this air mass, which seems skewed.

Section 4.5.3 The second paragraph discusses relationships between fractional Fe solubility and various biomass burning tracers, and states that fractional Fe solubility was low during fire events FG and HI (local), but higher during fire event E (distant). However, the fractional Fe solubility in the days prior to fire events FG and HI was also low—the fire plume doesn't appear to be the only source of low-solubility Fe. More generally, to me, it doesn't appear from the data in Figure 6 that the fractional Fe solubility during fire events is significantly different from that on the surrounding days. An explanation of this would be useful, I think.

Section 4.6 The first paragraph of this section is more suited for the introduction. In P14L17, can the conclusions for GP22 be extrapolated to the rest of the campaign, when biomass burning aerosol concentrations were low and the aerosol population was primarily dust-influenced? In P15L1, again, it seems to me that the fractional Fe solubility was generally lower over the last week of the campaign, rather than that "the fractional Fe solubility was lower when fresh EC was high". Perhaps this discussion would be better framed along the lines of "Despite the influx of aerosol from

proximal fires, the fractional Fe solubility remained low”? In P16L13–14, you state an “inverse relationship between elemental carbon and soluble Fe”, yet in P13L25, you state that there is no relationship between these measurements. This is directly contradictory.

Conclusions

Generally, I think that the conclusions are somewhat overstated and not particularly well-supported by the data, *e.g.*:

- P16L21 states that the fractional Fe solubility decreased to 3% *during* the biomass burning event, whereas the data shows that the solubility was also low prior to the event.
- The discussion of the influence of oxalate on fractional Fe solubility would make sense in the introduction, but I don’t think that this data set as presented supports this conclusion (are the oxalate concentrations high enough to support this conclusion? P15L26, etc., don’t argue this convincingly).
- P16L28 is, in my mind, overstated—the fractional Fe solubility in fire event E is not substantially elevated over background conditions (*e.g.* the first sample in Figure 6).

Manuscript clarity

I often found the manuscript difficult to follow. I provide examples here, primarily from the introduction, but suggest that the manuscript be carefully edited/reorganized to maximize flow/clarity/organization on both the paragraph- and section-level scales.

- The first paragraph of the introduction states first that iron availability limits nitrogen fixation in nutrient-poor tropical waters (P2L12) and later that nitrogen fixation is iron-limited in 75% of the world oceans (P2L22)—these thoughts could be combined.
- The second paragraph of the introduction presents examples of aerosol-induced toxic algal blooms as if it were a new topic, with no link made to P2L11.
- The third paragraph of the introduction discusses two Australian aeolian dust paths, but in insufficient detail to picture them—what do “northwest” and “southeast” mean, exactly?
- The paragraph on black carbon terminology seems misplaced (P3L21–29) — perhaps this can be included in the methods section?
- The sentence beginning on the last line of P3 is unclear as written.
- In P4L3, what is meant by “smoke”? This seems vague.
- The final paragraph of the introduction is unconvincing to me (as written—I don’t mean to imply that the study isn’t worthwhile!) ... as written, it seems as though a goal is to better understand dust inputs to the Southern Ocean, whereas the rest of the introduction (and the abstract) has focused on the tropical waters north of Australia
- Table 1 could perhaps be moved to the supplemental information (or, just the most important information from it could be included in Table 2)
- Tenses are sometimes inconsistent (*e.g.* Section 2.4 has both present and past tense).
- References to personal communications in Section 3.2 should be removed.
- A number of typos throughout, *e.g.* “particulate patter” (P8L17), “biomass burring” (P13L24)
- The use of A/B for dust events and A/B/C/D for trajectory types is very confusing, and requires a lot of flipping back and forth between text and figures.
- In P11L13, “contamination” doesn’t seem an appropriate word.