

## Comments on Sources, solubility and acid processing of aerosol iron and Phosphorous over the South China Sea: ... Hsu et al

This is one of the most difficult manuscripts I have ever had to review. On the one hand, the manuscript presents interesting data on a potentially important subject. On the other hand the way the argument is expressed is very difficult to follow. The problem is a mixture of use of English which is OK but could certainly be better, the logical arguments used which are difficult to follow and the attempt to write this in a limited length without the results and discussion being explicitly separated. So difficult that I could simply reject the manuscript. I don't feel that it is my job to go through the text line by line suggesting how to rewrite it, so I will finish with a compromise of identifying some of the most difficult problems in reading it and evaluating it and offer to rereview a revised version. I cannot remember whether ACP has a strict length limitation. If it has, then maybe this study would be better in a journal which allowed longer text so that Fe and P can be more explicitly separated.

Some of the general problems:

Throughout the text the processes regarding Fe and P are put adjacent to one another. Actually there are real differences between them which certainly should be recognised in the introduction, where they should summarise what is known about sources, solubility and acid processing of Fe separate for those of P. There are many texts which deal with Fe in the atmosphere. There are far fewer which deal with P and the one which deals most explicitly with the sources, speciation and acid processing of P in mineral aerosols (Nenes et al., 2011) is barely referenced at all in this manuscript.

The most difficult section to follow is section 3.1. I have more specific comments on this below. The section 3.3 (which is mislabelled since it deal with fractional P solubility as well Fe) is rather easier to follow.

Specific comments:

Introduction: The authors need to separate Fe from P. Fe in aerosols is important because it essentially controls which areas of the global ocean are Fe limited. There is a lot of Fe in mineral particles (~few %) and there is very little iron in the ocean (low nM). The authors state that over one third of the worlds oceans are iron limited. I am not sure I agree it is that much but I agree it is important. By contrast the REST of the worlds oceans are essentially N & P colimited. The difference is that atmospheric P is a smaller component of the actual atmospheric aerosols (~ a few tenths of %) and the amount of P getting to the ocean from other sources is relatively much higher. Likewise there are real differences in the reactivity of Fe and P minerals to acid processes. This needs to be properly set up in the introduction and where there are relevant results, described in the results section.

P21436 line 5 remove is

Line 6 remove is widespread and replace with can be important

Page 21437 line 3 replace dominance with importance

Line 13 remove very

Line 22 remove certain

Line 27 start a new paragraph starting with The South China Sea

Use of abbreviations: I found the abbreviation EADPO and SEABB to be very confusing. I could not remember what they meant and got lost later in the text. Such long abbreviations are almost never used in US scientific journals. I would suggest that SEABB could be just BB (biomass burning) and EADPO could be DPO (dust and pollution outflows).

“The present study covers a few challenging topics in terms of distinctive dissolution characteristics of airborne Fe and P associated with varying sources and their relative significance in supplying bioavailable Fe and P to the SCS” This is an example of the problems I have with the English in this text. What is a few? Challenging to whom? What is a topic? What is a distinctive dissolution characteristic compared with a non-distinctive dissolution characteristic?

2 Materials and methods:

This is a manuscript using detailed geochemical measurements of aerosol particles to draw major conclusions about their source and behavior and yet no details of the methods of sampling, sample treatment, or analysis are given. There is no information about precision accuracy or limits of detection. In the case of extraction by Milli-Q we are told this was an acceptable procedure and essentially asked to believe the results of Hsu et al., 2013a) without any evidence to support it. This is despite knowing that the dissolution of Fe is very pH sensitive and different particles can have dramatically different pH values when added to Milli-Q water.

Results and Discussion:

In effect the first section is a results section. That is fine. However the authors have not explicitly identified what the data is actually saying about a particular important process that they discuss later. This partly because they are mixing Fe and P but mainly because they have not identified clearly enough how a particular set of results says unequivocally something about an important process.

The key initial figure is Figure 5. Yet it is almost impossible to actually identify individual elements in that figure. In particular even with a color version, I could not trace Al T vs PT which are two crucial parameters. I was trying to do a relay check on the raw data and could not. The authors should separate this figure into two figures per cruise so that the symbols don't overlap in the way they do at present, probably the total elements separate from the water soluble results.

Page 21439 line 24

The term significantly should only be used when a test of significance has been carried out

An example of text which I cannot follow: P21441 line 4:

“Further comparison showed that the June samples also had Fe concentrations nearly similar to those in the February-March samples. However %Fe was significantly distinguishable, with considerably higher solubility higher solubility in the February-March samples (11%...) than in the June samples (2.5%, excluding three background samples collected on the earlier days of the cruise) and even the ECS (7.7%)”

What is nearly similar compared with similar? What is significantly distinguishable? How do the authors justify removing the three samples from the earlier days of the cruise? Are they removed from all data sets? If not why only here? What does even the ECS mean?

Throughout this text I have problems with units. I always prefer molar units because it is easy to compare geochemical processes but I recognise others like weight units. However then molecules such as SO<sub>4</sub> and NO<sub>3</sub> require a definition of what is being shown. Is it SO<sub>4</sub> or SO<sub>4</sub>-S, NO<sub>3</sub> or NO<sub>3</sub>-N.

Section 3.2

This needs to be rewritten so it clearer what is being used to characterise the different sources and once characterised what are their particular chemical species for elements which are not characteristic of source.

Section 3.3

The title is not what the section is about. It is about fractional Fe and P solubility. The authors now use molar units.

P21446 line 24

The authors mention that the aerosols have not had sufficient time to react. What information do they have of time?

P21447 line 14:

How can you identify a diurnal trend with only such limited samples. Remove these comments

Line 20 to line 25 is another sentence in which the combination of English and scientific logic are mixed up in way which makes it impossible to understand what is actually being written.