Replay to the authors answer to the manuscript:

Estimating chemical composition of atmospheric deposition fluxes from mineral insoluble particles deposition collected in the Mediterranean region.

By Fu et al.

My reply to the authors answer is in bold character.

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In the data interpretation the authors approach is sometimes circular: they choose the main Saharan dust events deposition (demonstrated by the high load and backward trajectories analysis), besides their sampler is able to capture the only mineral fraction of the deposition and they want demonstrate that the elements they measure in these samples are marker of dust deposition (see for instance sentence on page 9 lines 31-34). This conclusion is obvious for the main crustal markers (Al, Fe, Ti, Mn).

We do not think our interpretation of data is circular. Even if the collector is limited to mineral deposition measurements, for trace metals it's not obvious that deposition is mainly due to dust but can be associated to anthropogenic particles. The use of "intense dust events" purposes to identify, even in the very constrained dusty cases, this anthropogenic influence. We use precisely the fact that it's no doubt during these events about the origin of main dust markers to identify other influence for trace metals by comparison with these markers. The conclusion mentioned in the sentence p9 lines 31-34 is only for trace metals, not for dust marker: "the results showed that the atmospheric deposition of trace metals at Le Casset, Corsica, Mallorca and Lampedusa is mainly associated to dust fluxes except Zn in Lampedusa and Cr in Corsica during intense dust event".

If the aim of the paper is find the influence of anthropic source of trace metal during Saharan dust events, the approach is correct but what is the relevance of this finding? In my opinion is more interesting to find the anthropic impact of trace metals to the total deposition. Anyway, the approach is correct, please change some sentences in order to be more clear on the aims of the paper.

Besides, the authors conclude that as these events cover the majority of deposition, the total contents of the metals arise from dust. I do not agree with such conclusion, this conclusion can be true only for the elements having only the crustal source (Al, Fe, Ti) accounting for a large amount to the total mass (Al, Fe), the fraction of metal deposited by Saharan dust for metals having also anthropic source can be quantified only by the analysis of all the samples not only those representing Saharan dust.

We agree with this comment. Our conclusions are for the total dust deposition, but it was probably not so clear. So in order to clarify this point, we added P11 lines 28-30: Chemical analyses of 107 CARAGA samples corresponding to the most intense dust deposition events observed between 2011 and 2013 in the western basin were performed to validate using CARAGA samples to estimate the origin of atmospheric deposition from dust.

The added sentence "estimate the origin of atmospheric deposition from dust", is still circular, if the deposition is from dust you already know the origin, it is dust.

Another weak point of the paper is the discussion of the elemental loss. An interesting analytical work is done to assess this loss for the metals, but the results are not well valorised. The importance of the soluble fraction of metals and especially P has to be highlighted.

The purpose of the paper is not a process study about dissolution. The experiment did in this work purpose to mimic a wet deposition then a rinsing in the CARAGA samples, we cannot highlight the result on P soluble fraction which corresponds with a very specific condition of dissolution (see detailed response in specific comments).

If the "*P soluble fraction corresponds with a very specific condition of dissolution*" and it is not representative of the real situation all the section is merely speculative.

Here below some specific (and minor) comments that I hope can help the authors to improve the discussion.
Specific and minor comments:
Page 3 lines 8-14. Sites description is very poor, the characterization of the different type of depositions needs to know more information about the aerosol source affecting the sampling sites. There is a reference to Vincent et al. 2016, but also in this paper only the geographical position of the sampling sites is reported.

We added information and references about aerosol influence on the stations in the chapter "2.1 Sampling of insoluble deposition and total mass measurements".

I'll wait for the new version to see the new section 2.1.

Page 3 line 24. I suppose that "selected samples" has to be deleted, please check this sentence and correct it.
The sentence has been corrected: Saharan origin was identified for 107 selected samples by using satellite data and air mass trajectories.

Ok

Section 3.1. The discussion in this section is reductive respect to the obtained results. I understand that the aim of this section is to demonstrate that you determine the total deposition flux and the "loss" are considered as a negative result, but, in my opinion, the quantification of metals and nutrient solubility is a very important result and deserve a deep discussion. Besides, literature data on solubility in environmental condition are scarce. In this way I strongly suggest to change the aims of this section focusing on the importance of the soluble part (and their variability in the different aerosol types) in fertilization processes. The importance of metal solubility is also claimed by the authors at page 7 lines 28-30 to explain the north –south different Al percentage in the total deposited mass than the soluble fraction seems to be not negligible as the authors state in this section.

We agree with the importance of the soluble part for studying the bioavailability of trace metals for phytoplankton. However, the methodology developed in our work was to estimate the loss of metals during the protocol of sampling and ignition. Thus, the solubility obtained in these tests is not representative of dissolution processes in rain or sea waters. Moreover, it's known that solubility of trace metals from dust is low (e.g. Desboeufs et al., 2005), it isn't a "new" finding.

I'm not asking for a big change of this section, just few sentences to highlight the importance of the soluble part, but if the Authors declare that "the solubility obtained in these tests is not representative of dissolution processes in rain" and especially during the rain episode sampled by CARAGA, the section is merely speculative and have to be deleted.

Concerning the decreasing of Al percentage, the soluble part of metals is not mentioned, only the soluble species as Ca are pointed (p8, lines 5-6): "This increase was explained by the fact that the mass

loss due to dissolution of highest soluble species during settling modifies the mass percentage of the less soluble elements'

Ok

Section 3.2.1 PCA shows that all elements (excluding one in each site) are grouped in F1 representing the crustal source. This is expected due to the choice of samples, and the exclusion of anomalous samples do not change the general result. Figure 2 caption need to be revised and please increses the size of characters in the figure plots.

We agree that such PCA result on common factor for the majority of elements was expected because of the choice of samples. However, the objective in this approach was not to show the crustal origin of all the elements whatever the samples, but precisely to identify the anomalous samples in order to discriminate the potential other sources. Actually, after removing outlier samples, the obtained fluxes for several elements were changed, e.g. Zn in Lampedusa.
Figure 2 has been corrected.

This is not a common use of PCA but could be right

Section 3.2.2 Page 7 line 20-25 and related table 4. The percentage of Al respect to total mass of deposition is sometime (Le Casset and Corsica) higher than the percentage of Al in the average upper continental crust. Is it possible a loss of carbonate during the ash procedures?
The decomposition of $CaCO_3$ is from 800 °C, but the ignition protocol is just at 550 °C, so we don't think there was a loss of carbonate during ignition. The method of LOI (loss on ignition) in 550°C is traditionally used to determine SOM (soil organic matter), and just the loss of SOM and structural water for several minerals were reported, according to Sun et al. (2009).The higher Al contents are due to the loss of the most soluble species such as Ca carbonate or sulfate as detailed in p8 between lines 4 and 19.

Ok

Page 8 lines 25-30. The source of Zn from waste incineration is true in general over Mediterranean region, but not at Lampedusa, where Zn arises from manufacturing of non-ferrous material (largely use in extreme marine environment) as correctly stated by the authors in the previous sentence.
We agree that Lampedusa Island is not a metropolitan area, but there is a Power plant close to the sampling site (added on page 8 line 28), we are waiting for more information about this power plant form other colleagues, it will be added in article while possible. But we can say that the abundance of Zn in Lampedusa samples showed anthropogenic Zn sources affecting this sampling site.

It is well known that both waste incineration and power plant do not emit only Zn, but also other metals and you do not observe other anomalous enrichment of other metals, especially if you consider samples characterises by high Saharan dust content. I'm still not convinced about the Zn source from waste incineration or power plant.

Section 3.3: This section is the most interesting of the paper but need to be rewritten. Page 10 lines 7-10 such information is not inferable from table 6. Page 10 line 21. I suppose the sentence should be "… fluxes of metals and P associated to intense dry deposition events." Page 10 lines 21-29. The sentence is not clear. I do not understand what the authors want to demonstrate.

1) 2 categories added in table 6: dust deposition and mixed deposition.

2) The sentence has been corrected: "These results show that the fluxes of metals and P associated to wet deposition predominate in western Mediterranean Sea environment".

3) Page 10 lines 21-29: two points were developed: firstly, the mass fluxes estimated in this paper are averaged weekly mass fluxes for the most intense dust events account in mass 50-84 % of the total

deposition, as studied by Vincent et al. (2016), so we could not compare with references values in literature which are typically annual. Secondly, due to the effect of dissolution during the CARAGA sampling protocol, these fluxes are underestimated at the worst case by 13 % for P and 10 % for Zn, Cu and Mn, and 5% for other trace metals.
Ok these results have to be shortly reported in the conclusion.

Conclusion and abstract are too general, some specific results have to be reported.

In abstract sentence added: 'the mass fluxes strongly depend on the distance from dust sources and the most intense events, proximity from anthropogenic sources strongly impacted the masse fluxes of Zn and Cu at Lampedusa and Frioul'

Ok
In conclusion sentences added : 'High average and strong standard deviation of Al (dust marker) mass fluxes were observed at Lampedusa and Mallorca due to several most intense dust events, and extreme Zn mass fluxes (122.63 ± 1765.23 µg m^{-2} wk^{-1}) was observed at Lampedusa because of contamination of the incinerator sources'.

Too many digit in the Zn mass fluxes.
I strongly disagree with the incinerator source for Zn, the sentence have to be changed in: "…and extreme Zn mass fluxes (123 ± 1765 µg m-2 wk-1) was observed at Lampedusa that need to be deeply investigate but likely due to the large use of non-ferrous metal manufacturing in this environment strongly impacted by sea spray."

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