

Reply to Referee 1

(REFEREE)

The manuscript addresses the atmospheric deposition of organic matter in the Mediterranean, for which there is little data available. It quantifies such deposition in the small island of Lampedusa in the Central Mediterranean, in terms of carbon, nitrogen and phosphorus. It also tries to untangle possible sources of such organic matter. In this aspect the manuscript is less conclusive as there is no good relationship to aerosol origin or type of deposition. The conclusion is that the OM is mainly coming from sea spray that the different air masses pick up and transport to wind up depositing. It could be in large part but really it is just a hypothesis that needs further exploration.

(AUTHORS)

We really thank the reviewer for his/her appreciation of our work.

We totally agree that regarding the possible sources of organic matter, we just reported some hypotheses. With the available data-set in fact we were just able to make an hypothesis about the main sources of DOM in the different sampling periods. The sampling periods cover a generally wide time interval, and the deposition data are the result of integrating deposition over air masses of different origin and with different aerosol characteristics and loads. This makes very difficult to find a correlation between aerosol origin and DOC input, since the different sources are mixed in our samples.

If we would have reduced the sampling periods, we would have had less variability in the sources, but in most of them we would have not had enough DOM to do all the analysis. Two weeks was therefore the best compromise we were able to find.

We will add in the revised manuscript a sentence highlighting that in order to understand the link between aerosol origin and DOM concentration and quality further exploration is mandatory.

Also, I was surprised not to consider wind direction properties when analyzing deposited material. Lampedusa is a small island but I would not be surprised that when wind blows from directions other than due East, and especially when it blows over the island from the West, substantial OM could be picked up from the island itself.

We thank the reviewer for this comment. At first, we took into consideration the air masses trajectories, but then we realized that there are some limitations in the use of wind direction to infer the aerosol sources.

Firstly, as above reported, the sampling periods cover a generally wide time interval, and the deposition data are the result of integrating deposition over air masses of different origin and with different aerosol characteristics and loads. Thus, in some cases information on the wind might suggest what are the dominant (if existing) wind conditions during a specific period; but due to possible differences in aerosol amounts and deposition, these conditions may not be representative of the integrated samples.

Secondly, the use of wind direction and speed identify the air mass origin (which is not what the reviewer is suggesting) may be problematic, since trajectories arriving to Lampedusa may take different paths depending on the synoptic conditions. A more elaborated approach would be required (e.g., trajectory reconstruction using wind, as in Becagli et al., 2012; or modeled backward trajectories based on meteorological analyses, as in Marconi et al., 2014). However, also in this case, the relatively long duration of the sampling interval and the variability of the deposition would prevent a robust attribution of the source regions.

Wind measurements conversely, as correctly suggested by the reviewer, might potentially provide useful information on the impact of local sources. The main local source area of anthropogenic particles is in the sectors between South and South-East of the sampling site; in this sectors there are the Lampedusa town, the power plant, the airport and the port. Previous studies have shown that wind from these directions is relatively infrequent, and the impact of these sectors is estimated to be negligible (see e.g., Artuso et al., 2009, with respect to atmospheric CO₂ measurements; Calzolari et al., 2015, with respect to PM₁₀ measurements). This impact is expected to be even lower over samples integrated over are relatively large number of days.

Due to these reasons, we have preferred to associate the deposition samples' characteristics with those of PM₁₀ samples collected daily and over the same time period. This allows to use aerosol properties

measured in the same time intervals and influenced by the same sources to infer some overall conditions. Previous studies (e.g., Becagli et al., 2012, 2013, 2017; Marconi et al., 2014; Calzolari et al., 2016) have been dedicated at linking the PM10 measured composition with different aerosol sources.

These ideas will be discussed in the revised manuscript and a sentence about the main wind affecting our sampling site will be added in the materials and methods section.

A third aspect of the manuscript deals with estimating the local and Mediterranean basin-wide importance of such deposition estimates for the biogeochemical functioning of the Mediterranean. I like this part myself but I have to admit it is the least elaborated since it is based on assumptions that will be hardly met. For instance, calculations based on the extension to the whole Mediterranean of the measured OM deposition at Lampedusa. Given it is so variable and without a clear reason, I would expect variability to increase when other locations are taken into account. Also, the lability of the deposited organic matter is an unknown, so the final role of the marine biota is also unknown. But anyhow, I like these exercises. Thus, to me the main value of the manuscript is to provide a much needed data series of OM deposition measurements.

We totally agree with the reviewer and we are aware that reporting the calculations based on the extension of our results to the entire Med Sea is a risk, because it is not simple, nor appropriate, to assume that what is observed at one location is valid for the entire basin. However, we consider this as a first conceptual exercise that uses the new results from our study to give an estimate of the implications of DOM deposition for marine ecosystem, that needs to be supported by additional data.

To the best of the author's knowledge only one paper reports that a not-well quantified fraction of atmospheric DOM can be recalcitrant. Due to the lack of information, we decided to discuss implications taking into consideration both the possibilities: DOM is labile and DOM is recalcitrant. In the revised manuscript we will better stress the need for further investigations about the biological lability of DOM coming from the atmosphere

In order to better stress that these calculations are a conceptual exercise, we can add the following sentences in the revised manuscript:

"A conceptual exercise can be made in order to give an estimate of the implications of DOM deposition for marine ecosystem."

"Even if we are aware that these assumptions are hardly met, in particular the estimate of DOC input to the whole Med Sea, based on the data collected in Lampedusa, we think that these calculations can give an idea of the relevant role that atmosphere input of DOC can have in sustaining the bacterial productivity in the surface layer, in particular when the water column is strongly stratified."

The methods are standard within the field and thus assure quality control. Maybe I am not clear whether monthly data were calculated and how or whether just sample data was provided always? or in what cases? That is, how were data treated when more than 1 sample per month was available? How was the data split when covering periods from two consecutive months?, etc.

We apologize for the inaccuracy. We did not calculate monthly data, we reported the sample data. The width of the bars in the figures 2, 3, 4, 6 and 7 refers to the duration of the sampling period (Table 1). This aspect will be clarified in the revised manuscript.

As a general rule, samples were collected every ~15 days, or immediately after strong rain or dust storm events. However, due to logistic problems the sampling period was longer than 20 days for 9 depositions (Table 1).

The DOC, DON and DOP fluxes, reported in the text and in the figures 2, 3, 4, 6 and 7 were calculated using the following formula:

$$X_{\text{Flux}} = X \cdot V / A \cdot d$$

where X is the concentration of DOC, DON or DOP measured in the sample and expressed in μM ; V is the volume of rain collected by the sampler (expressed in L) or the volume of Milli-Q water used to wash the funnel walls in case of dry deposition (250 ml); A is the area of the funnel (0.1018 m^2), and d refers to the number of days of the sampling period. The DOC, DON and DOP fluxes are reported in the figures considering the flux corresponding to each sampling period. A paragraph with this explanation can be added in the Materials and methods section of the revised manuscript in order to clarify these calculations.

I understand that sample data is clearly reported in Fig. 5, but how were the rest treated is a bit mysterious, especially since bars have unequal width within and between figures.

The bars in the figure 5 corresponded to the C:N:P molar ratios (see Table 3), so they referred to a number, not a flux, and this is the reason why the width of the bars is always the same in figure 5, in contrast to fig. 2, 3, 4, 6 and 7, where the width of the bars is different since it refers to the length of sampling periods. However, thanks to the reviewer comment, we realized that making all these figures with the same format is misunderstanding. We will therefore redo the figure 5 without bar but using a symbol and we will clarify in the text how the figures are made.

In line 150 it is also important to know the flow rate of the low-volume sampler. Also, I guess that because of physical flow rate constraints a 1 µm filter could not be used. That would have been much more desirable since there tend to be organic rich particles at the very fine particle ranges, and they would have been missed, not a minor issue in this paper on OM. I would like the authors to comment on the choice of a 2 µm filter to collect particles.

The filters used in this study are those usually used for aerosol sampling, they have a nominal porosity of 2 µm, but they are certified for 99% efficiency for particles having 0.3 µm diameter.

The sampling flow is maintained constant at 2.3 m³/h in order to maintain constant the sampling heads cut-off (10µm) as reported in the European rule UNI EN12341.

In order to clarify these concepts for a broad number of readers the text can be changed as follows:

“PM10 (particulate matter with aerodynamic equivalent diameter lower than 10 µm) is routinely sampled on a daily basis at the island of Lampedusa (Becagli et al., 2013; Marconi et al., 2014; Calzolari et al., 2015) by using a low-volume dual-channel sequential sampler (HYDRA FAI Instruments) equipped with two PM10 sampling heads, operating at constant flow of 2.3 m³/h in accord with the European rules for aerosol monitoring (UNI EN12341). Aerosol is collected on 47 mm diameter Teflon filters (PALL Gelman) having 2 µm nominal porosity but certified to have 99% retention efficiency for 0.3 µm diameter particles. The PM10 mass was determined by weighting the Teflon filters before and after sampling with an analytical balance in controlled conditions of temperature (20±1 °C) and relative humidity (50±5%).”

The manuscript is well structured and balanced. The title is informative of the contents. The language is proficient. Figures should be uniformed or clarified in aspects such as the x-axis but are otherwise well done. Other than that, I have no major concerns publishing the manuscript pretty much as it is.

We really thank the reviewer for his/her appreciation of our manuscript. As above reported, we will rework the figures in order to uniform them and to eliminate any misunderstanding.