Dear Editor,

Please find enclosed the manuscript for submission as a research article to Atmospheric Chemistry and Physics journal entitled "Quantification of the carbonaceous matter origin in submicron marine aerosol particles by dual carbon isotope analysis".

This manuscript was originally submitted to Proceedings of the National Academy of Sciences journal, but was rejected, because only one reviewer recommended it for publishing in PNAS while the other did not. The reviewers were mainly concerned with clarification of the many points and the method itself which are now addressed in the expanded manuscript due to non-limited space in ACP.

On behalf of all authors I would like to include and comment on the main issues raised by the PNAS reviewers which we believe now adequately addressed in the manuscript.

1st Reviewer's comments:

As described by the authors and as employed in the cited literature, marine versus terrestrial (fossil fuel + vegetative) sources can be differentiated based on only δ13C for which many more samples were characterized. Δ14C was used primarily to differentiate fossil fuel versus modern vegetative sources the latter of which were unimportant in clean marine air. I encourage the authors to differentiate and interpret the marine source based on the larger number of samples that were characterized for δ13C. These results would increase the robustness of the analysis substantially.

Discussion on marine source separation based on δ13C data alone is added. However, such a separation is unconstrained and therefore has significant uncertainty due to the lack of established typical source ratios.

The analysis is based almost exclusively on relative percentages rather than absolute values. However, in most cases TC in polluted samples is greater than that associated with clean samples. Focusing on relative differences masks relevant aspects of variability.

TC concentrations are given in Table 1 and we can see that polluted sample TC concentrations are not always very different from marine ones as it depends on the degree of anthropogenic perturbation. The more important result, however, is the presence of significant amount of marine biogenic organic matter even in polluted air masses.

Based on reference 6, it appears that the indicated thresholds for particle number concentrations and black carbon correspond to averages values for multiple measurements over each period of active sampling. Under all flow regimes at Mace Head (including those associate with openocean trajectories arriving from the marine section), local ship traffic results in occasional shortterm spikes in CN that are many times greater than 700 cm-3. The text should clarify this point.

It has been clarified that automatic clean sector control system is actively controlling CN and wind direction parameters, therefore, short-term spikes are excluded from the samples as well.

The statement that the carbon isotopic composition of aerosols is stable during transport should be supported with a citation. To my knowledge, this issue has not been resolved unequivocally. Isotopic theory suggests that chemical processing and associated changes in partitioning between the condensed and vapor phases can cause isotopic fractionation [e.g., Hoefs, 1987, Stable Isotope Geochemistry]. For example, for a given compound, the material depleted in the heavy isotope condenses preferentially. Deviation from the implicit assumption that the carbon isotopic composition is stable during transport and chemical evolution could bias the inferred source attribution. A caveat to this effect should be added to the text.

We added a section on this topic highlighting a possibility of isotopic fractionation, however, as most of the studies to date are mostly supporting primary sources to dominate biogenic marine organic matter production, isotopic fractionation is probably of limited importance to source attribution.

The regression technique used to estimate the line and coefficients in Fig. 1 should be specified. It appears that the authors may have used standard linear regression but, because both sets of variables are subject to significant measurement error, this approach does not yield reliable results. The reduced major axis procedure (explained in most standard statistics books) should be used to regress such data. Same comment for Fig. 2.

The reduced major axis procedure was followed to produce the essentially same result proving the robustness of the regression method in obtaining the blank value.

Manuscript, Figure 1 and caption. As presented, this information is potentially confusing. For example, in line 1, the terms "marine" and "polluted" are ambiguous. The subset of data corresponding to relatively clean conditions includes significant contributions from pollution sources and the subset of data corresponding to "polluted" conditions includes significant contributions from "marine" and terrestrial vegetative sources. Explicit terminology should be used to differentiate the filtered data subsets from the corresponding sources for OC associated with each.

The ambiguity between "clean marine" and "polluted" terms is now highlighted in the Experimental Methods section, but quantified backing of the terms was an inevitable scope of the study. The results demonstrate that clean marine samples are indeed very clean with 80% of organic material being marine biogenic, hence, named appropriately. Polluted samples have 40% contribution from fossil fuels and certain percentage from continental non-fossil but still anthropogenic sources (biomass/biofuel burning) which justifies the term "polluted".

Since multiple samples were involved, it is not clear how exactly the equations were solved to yield the indicated values or by what criteria convergence between the solution and measurements was defined.

Error minimisation approach is now expressed with the formula and the convergence is explained in more detail.

Finally, the suggestion that measurements by Chesselet et al. at Eniwetok Atoll are significantly impacted by local or regional anthropogenic sources is incorrect. Relative to available evidence based on clean sector sampling at Mace Head [e.g., Savoie et al., 2002, JGR], back trajectories and measurements of anthropogenic tracers published in several companion papers from that campaign suggest that the samples collected on Eniwetok were substantially less impacted by anthropogenic sources than those at Mace Head.

We disagree with this comment.
First, Chesselet et al. showed that based on isotopic ratios (-26 to -27‰) submicron particles at Eniwetok contained largely continental particulate organic carbon with only supermicron ones being of marine origin.
Second, Savoie et al. (2002) did not use clean sector sampling system in that particular study but rather collected daily samples regardless of sector. It was only in the post-processing of data when they applied anthropogenic tracers (NO3, Sb) to obtain unperturbed nssSO4 signal. Our main argument is not that the North Atlantic is cleaner overall than the Western Tropical Pacific, but rather than the clean marine sector in the Eastern North Atlantic (i.e. in prevailing westerlies) is now cleaner than the Eniwetok in the late seventies (based on data) or even today due to the proximity to Southeast Asia.

2nd Reviewer's comments:

The manucript may be suitable for a more specialized journal if the discussion is clarified, the calibrations are provided, the inversion is clarified and the associated uncertainty is discussed.

It was indeed very economical description of the method due to limited space requirements in PNAS. It was significantly expanded this time.

As it stands, the work has failed to make the fundamental case for the uniqueness of the apportionment to three sources based on two measurements that it has not shown to be linearly independent. Simple markers such as elemental tracers and meteorological conditions are given no quantitative representation in this work. Furthermore, the results are based entirely on a coastal site, which is in no way reflective of open ocean conditions, despite the generality of the title and discussion.

We admit a couple of misprints in the submitted version which gave the reviewer an impression that the unique apportionment has failed. Using independent ^{13}C and ^{14}C measurements the three principal sources are indeed linearly independent. Elemental tracer (BC) is quantified as $<50 ng/m^3$ level for samples to qualify for clean marine and backward air mass trajectories were used retrospectively to confirm that there was no contact with land for the last 4-5 days. As for the coastal vs open ocean conditions there is a recent publication by Rinaldi et al. (2009) which discusses the very details of such comparison with the main conclusion that measurements performed at Mace Head actually are representative of open ocean conditions.

Using Carbon isotopes to determine the contribution of marine, continental, and fossil fuel sources is really interesting. This can be used in future studies if the technique is properly explained and quantified. It would be interesting to discuss the fossil-fuel influence more explicitly. Make sure to only include what is necessary in the paper and supplementary materials. A lot of overlap is not necessary and can be removed to make room for more discussion on the implications of the findings.

It was unfortunate that the reviewer rejected the paper while admitting it being really interesting. We believe that the current version of the manuscript is significantly improved.