We would like to thank the reviewers for the critical comments which helped to improve the paper a lot. Indeed, some of the important references were missing and we have added few more to support our statements.

First, we would like to address the comment made by all reviewers about the absence of continental non-fossil source and corresponding revisions to be implemented in the revised version of the manuscript.

The use of 13C as a tracer warrants a more in-depth discussion, since it is well-known that isotope fractionation effects occur during atmospheric processing. This can be due to differences between 12C and 13C in e.g. reaction rate constants, and condensation and volatilization processes in the atmosphere or on the sampling filter. This should be discussed in further details in the introduction. It is very surprising that the marine samples contain 0% continental non-fossil carbon, given that most (continental) studies find more than 50% non-fossil carbon in non-urban areas (as recently reviewed by Hodzic et al., ACP, 2010). This finding is so un-expected that it should be supported by other measurements or at least back-trajectories (using for example the freely available HYSPLIT model), before the manuscript can be accepted. This was actually also suggested by one of the reviewers of the PNAS submitted version of the manuscript ("Simple markers such as elemental tracers and meteorological conditions are given no quantitative representation in this work.") The 14C results should be compared with previous measurements.

Along with the other reviewers I am puzzled by the absence of non-fossil continental sources in the marine aerosol. But moreover also by the relatively large contribution of fossil sources to the polluted aerosol compared to the non-fossil continental sources. In various continental locations a much bigger contribution of non-fossil sources has been measured. Is there any hypothesis why this would change with transport over the ocean?

I am also very surprised by the results of "0% continental non-fossil carbon", probably more explanations and comparison with other relevant studies should be added.

We agree with the reviewers that the result is puzzling. However, air mass back trajectories together with elemental tracers (radon), presented below, agree with the dual isotope method very well. We suggest that the stormy N.E. Atlantic conditions facilitate an efficient washout of anthropogenic material from marine boundary layer and replenish it with biogenic material during the intercontinental transport.

Regarding the contribution of non-fossil sources in polluted air masses we observe about 60% of non-fossil carbon which is in agreement with the study of Hodzic et al. (2010). However, that same 60% is split between marine and continental non-fossil sources, reducing the contribution of the latter. Indeed, in many continental locations marine source contribution would be negligible, but that is not the case in the island of Ireland which is surrounded by biologically active waters in the North Sea, the English Channel and the Irish Sea.
We can hypothesize that the relatively large contribution of fossil sources in continental air masses can be due to number of reasons. Firstly, the United Kingdom

(UK) and Ireland have very little forested areas (under 10% in Ireland and just over 10% in the United Kingdom compared to an average of 46% in continental Europe) with even lower percentage of true forest ecosystems. It is well established that forests contribute significantly to SOA formation. Second, intensive shipping in the English Channel and the Irish Sea can contribute additional fossil carbon to the one originating from traffic. The latter can be supported by the fact that in the same air masses passing over the sea we observe significant contribution of marine sources, presumably from English Channel and the Irish Sea. With the continental air masses passing over relatively bare land (mainly grasslands) and intensive shipping areas can possibly explain the observed pattern. It is worth noting, however, that Heal et al. (2011) also observed large percentage of fossil fuel source in British Midlands up to and above of 50%.

The other reason for relatively large contribution of fossil fuel source in polluted air masses can potentially be due to low solubility of particles containing larger percentage of fossil fuel carbon (assuming larger contribution of elemental carbon and significant degree of external mixture). The particles containing continental non-fossil carbon would be more soluble and conceivably derived from secondary processes facilitating more efficient washout from the boundary layer.

This study is a continuation of experimental work on sources and chemical composition of marine aerosols being performed at Mace Head over the last decade, but this time using a combination of dual carbon isotope analysis. The concept of clean marine air masses being sampled at Mace Head has been discussed in several papers (Cavalli et al., 2004; Yoon et al., 2007; Rinaldi et al., 2010) showing typical air mass back-trajectories and discussing various elemental tracers (Cavalli et al., 2004 in particular) and, consequently, referenced in this study. This time in addition to back-trajectories (Figure 1, to be added to the paper and similar to the ones presented in Cavalli et al., 2004), demonstrating that clean marine air masses (sampled in this study) had no contact with land for at least 5 days before advecting at Mace Head and spending the last three-to-four days in the boundary layer, we show that our classified marine aerosol also exhibited extremely low radon concentrations (222Rn ~100mBq/m3 and 220Rn ~1mBq/m3) (Table 1). The latter fact is a quantitative proof of truly marine air masses with no contact with land for about a week (e.g. Biraud et al., 2000). Considering 222Rn concentration in polluted (continental) air masses of the order of several thousands of mBq/m3, values presented for marine samples are generally less then 10% of the continental ones. Allocating some of the 222Rn measured in marine air masses to continental shelf emissions (upfront of Mace Head) leaves room for extremely low contribution of North American continental aerosol in clean marine air masses sampled at Mace Head.

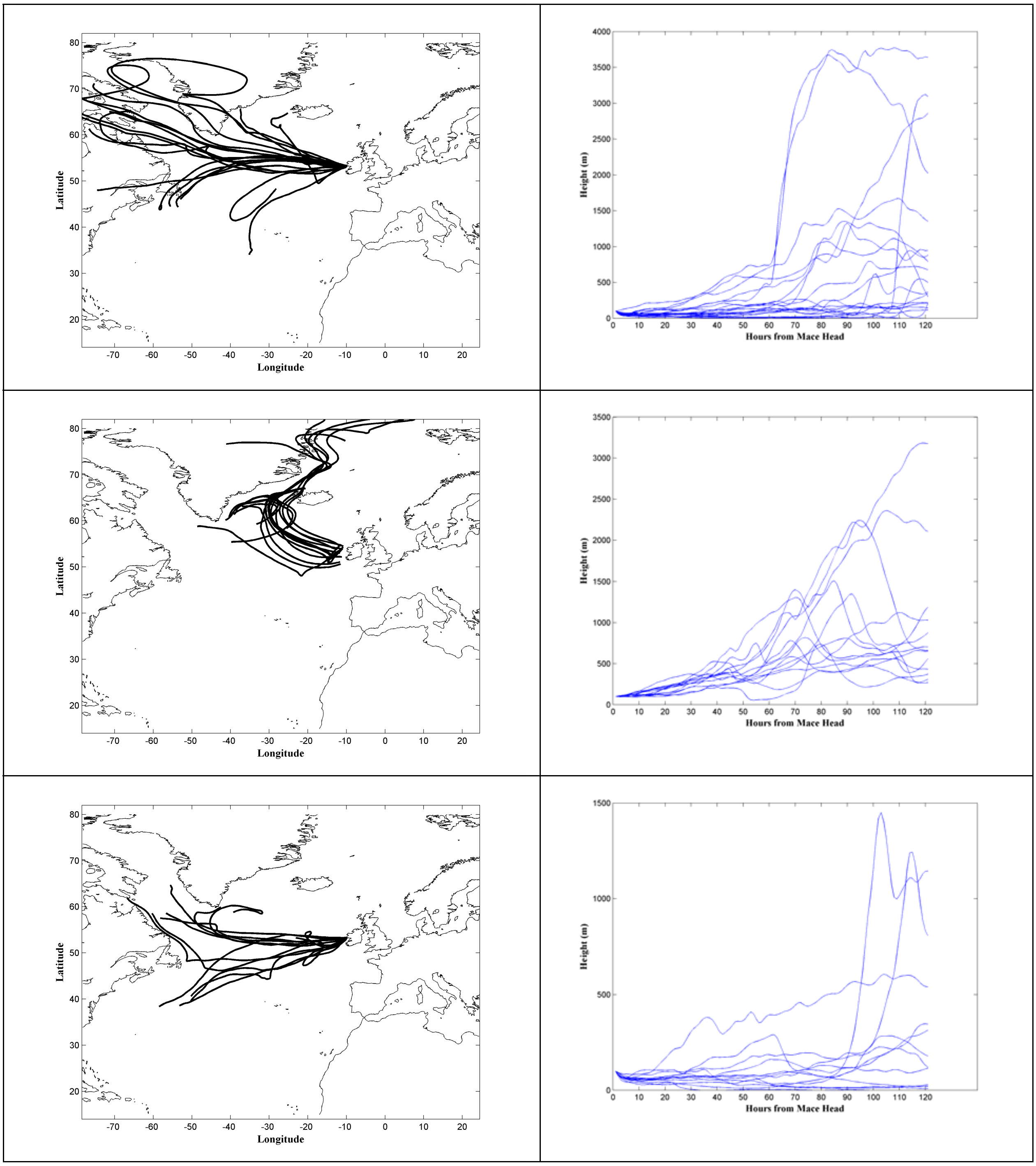

Figure 1. 120h air mass back trajectory plots (every 12hours ending at 100 AMSL) along with trajectory height along the trajectory during three marine samples presented in Figure 4 of the paper: 12-26/04/06 at the top, 12-15/06/06 in the middle, 05-12/07/06 at the bottom.

Table 1. Radon concentration (mBq m^{-3}) during clean marine sampling periods presented in Figure 3 of the paper.

Sample period	^{222}Rn	^{220}Rn
11-18/01/2006	191	0.1
29/03-05/04/2006	n.d.	n.d.
12-26/04/2006	n.d.	n.d.
12-19/06/2006	n.d.	n.d.
19-28/06/2006	145	2.53
12-15/06/2006	n.d.	n.d.
16-20/06/2006	136	0.88
05-12/07/2006	223	1.23
16-23/08/2006	166	2.36
03-08/09/2006	123	0.68
05-11/10/2006	167	0.44
15-22/11/2006	60	0.1

n.d. no data

We attempted the other line of reasoning supporting our results by performing a more detailed sensitivity analysis of the calculations leading to the uncertainty of source apportionment.

The range of marine delta 13C values found in literature is from -18‰ to -23‰ (Williams & Gordon, 1970; Bauer et al. 2002). Quite clearly, -23‰ value can not apply as clean marine samples collected during summer had heavier isotope values without even correcting for anthropogenic influence. Polluted samples could utilise isotope values of -23‰, however, we strongly believe that same marine source delta 13C value should apply for all, clean and polluted, samples. If we used -23‰ delta 13C value in marine samples, we get discrepancy between measurements and calculations in excess of 1‰, which is beyond any conceivable analytical error. It is possible to accommodate marine source delta 13C value of -18‰, but in this case continental non-fossil and fossil fuel source values would have to be even more negative than the ones used in this study in order to close the equations. However, all reviewers questioned fossil fuel source value of -29‰ as being too negative. The source apportionment was, however, quite insensitive to small variation of either continental non-fossil or fossil fuel source values as by definition 13C method can not reliably distinguish continental non-fossil and fossil fuel sources. After all, Bakwin et al (1998) presented a range of fossil fuel carbon d13C values from a global network of sites with an average value of -28.3±2.4‰ encompassing both of our values, -26‰ and -29‰.

The apportionment results changed somewhat more significantly when we addressed the issue of a reference marine source value of delta 14C. Note, that the value of +50‰ was used in the paper.
Beaupre & Druffel (2009) indicated for the Pacific Ocean near California that 14C (DOC) values for surface ocean waters were stable from 1991-2004. Assuming that this is valid for the Atlantic Ocean, we can take the values from Bauer et al. without

correction so that 14C (DOC) for Mace Head should be considered as -170 (±80) ‰. However, these are the values not from the open ocean but from coastal (shallow) waters and from the slope (~100km from the coast). Taking only the slope values, which are closer to the open ocean conditions, we come to -210 (±60) ‰. This shows that open ocean might even be lower than this. 14C (POC) and 14C (DIC) were 30‰ and 60‰ respectively for the slope and thus also slightly lower than the biogenic value of the atmosphere, which was 120 ‰ in 1994. In conclusion, we should assume for our campaign -210 ‰ for DOC (slope waters from Bauer without any correction for the time between 1994 and 2006), +30 ‰ for DIC (value from Beaupre and Druffel for 2004 – differences between Pacific and Atlantic Ocean should be negligible for DIC) and 0 ‰ for POC (if DIC decreases by 30 ‰ within the 10 years, we can justify the same for POC).
A reference selected value of -50 ‰ would lead to a POC vs. DOC ratio of ~3:1 following dominant contribution of water insoluble organic carbon in sea spray samples which was attributed to POC (Facchini et al., 2008). A non-modern 14C value of the reference marine source may look unexpected, but we believe it is justified.
Consequently, using marine source 14C value of -50‰, the corresponding Figure 4 of the paper changes accordingly. Applying the same error minimization approach we can now include small contribution of continental non-fossil source in clean marine air masses. The fossil fuel source contribution is still higher than continental non-fossil due to reasons considered above: solubility and shipping emissions.

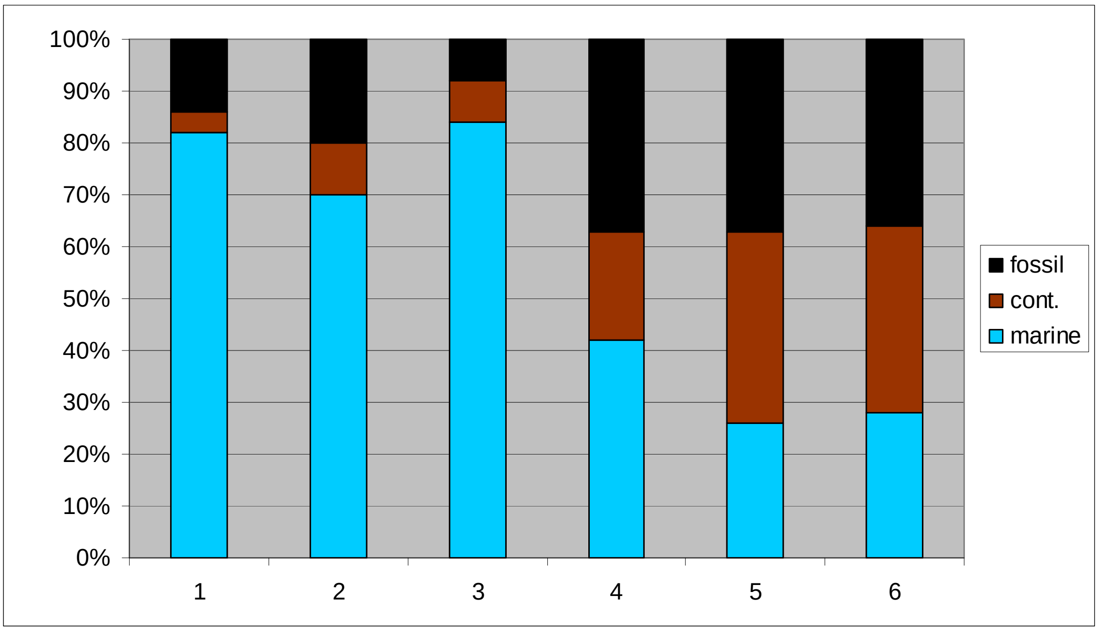

Figure 4. Contribution of three principal sources when changing marine source 14C value to -50‰ instead of +50‰ as in the paper.

Other specific comments are addressed individually.

Reviewer 1

The introduction lacks a thorough introduction to carbon-isotopes and their use in atmospheric studies. Such a section is now hidden in the experimental section (page 2754) and I suggest to move relevant parts to the introduction (leaving experimental details in the experimental section). Also state typical delta 13C values in terrestrial and marine environments.
Introduction will be expanded as requested by other reviewers. Authors would like to thank the reviewers for pointing out valuable references.

Specific comments

The title: The wording "dual carbon isotope analysis" will not be clear to most readers, and I suggest to be more specific and just use 13C and 14C instead. Is it necessary to state both aerosol and particles in the title?
Title will be amended: Quantification of the carbonaceous matter origin in submicron marine aerosol by 13C and 14C isotope analysis

Affiliations: Please state all affiliations at the same level of detail - affiliations 3, 5, 6, and 7 are too short.
Affiliations will be corrected as requested.

Page 2751 line 22-23: " Organic matter has been observed, to different degrees, in marine aerosol particles for many decades" - be more specific and avoid the use of vague statements such as "to different degrees" and "for many decades".
Appropriate references will be added, starting with D.C.Blanchard.

P2752 L7: Define MSA before using.
It was defined a few lines above as methane-sulfonic acid. Abbreviation “MSA” will added next to it.

P2752 L15: unclear statement: inconclusive due to varying pollution degree – please clarify.
No sampling criteria were applied in the cited studies (continuous sampling), therefore, samples were affected by polluted air to a different degree. This clarification will be added to the text.

P2752 L16-18: The sentence seems out of context.
Introduction will be extended as suggested by all reviewers to bring the sentences better into context.

P2752 L23: How can D<1.5 micrometer aerosols be submicron?
Many previous studies made at the same site or region (Cavalli et al. 2004; Yoon et al., 2007; Ceburnis et al., 2006) demonstrated that separation between submicron and supermicron aerosol occurs at around 1-1.5um, i.e. strictly submicron aerosol is below 0.5um and strictly supermicron – above 2um. It was attempted to make a distinction

from fine particles which is usually assumed to be D<2.5um. It will be changed to "fine particles (D_{50}<1.5um)".

P2753 L11: I do not agree with the postulate that the marine samples were likely the cleanest possible to obtain in the Northern Hemisphere. There are more remote areas in e.g. Greenland and Canada. Please rephrase.
Indeed, remote areas like Greenland or Northern Canada are cleaner overall, however, only a subset of clean marine air masses was sampled at Mace Head using strict sampling criteria. In this respect, clean marine samples collected at Mace Head can be among the cleanest in the Northern Hemisphere. It will be rephrased accordingly.

P2754 L4: What do you mean by "the samples were treated to carbon isotope analysis"? The sentence should be rephrased.
The sentence will be rephrased to:
"Carbon-13 and carbon-14 isotope analysis was performed to quantify the biogenic marine carbon ..."

P2754 L5: Remove "fuel" in "continental non-fossil fuel carbon". There are many other sources to non-fossil carbon, so state that those mentioned are some of the primary sources.
Will be removed.

P2754: The discussion of isotopic fractionation (of primarily 13C?) should be more indepth as stated above. Why should condensation favor the lighter isotope? Why do the authors assume that a "dominant primary source of marine organic aerosol" should not be affected by isotope fractionation? Could isotope fractionation occur during evaporation of compounds from primary aerosols or aerosol phase reactions or as a sampling artefact? The discussion should reflect the associated uncertainties.

Physicochemical properties of isotopes arise from quantum mechanical effects with lighter isotopes possessing higher vibration energy levels and, therefore, weaker intermolecular bonds (Hoefs, 2009). Also lighter isotope species have lower vapour pressures facilitating faster phase transitions. Those are the reasons behind condensation favouring lighter isotope. However, most of the marine organic aerosol is primary in origin according to the latest research (Russell et al., 2010; Rinaldi et al., 2010). If primary organic matter is preserved and only gaining mass through oxidation (addition of oxygen) then carbon isotope ratio should remain unchanged. Condensation of lighter organic species would drive isotope ratio to more negative values while evaporation – to more positive. Rinaldi et al. (2010) suggested that there is likely a limited amount of truly secondary organic carbon in marine aerosol with the majority being either primary or processed primary material, hence, little organic mass contributed via condensation processes. Fractionation during evaporation in marine aerosol would only be possible if primary sea spray organics would be losing mass during oxidation process producing small volatile organic molecules when, for example, braking unsaturated double bonds. Evaporation of entirely primary organic compound will not change isotope ratio and is probably hardly possible due to chemical species present (long chain hydrocarbons, typical of phospholipids (Facchini et al., 2008). In summary, due to origin and nature of marine organic aerosol matter isotopic fractionation is expected to be small and within analytical uncertainty of

~0.2‰. Moreover, there is a lack of specific targeted studies at isotopic fractionation in aerosol phase.

P2754 L17-19: Please find a more suitable reference for this general statement.
We will include Currie, L. A.: The Remarkable Metrological History of Radiocarbon Dating [II], J. Res. Natl. Inst. Stand. Technol., 109, 185-217, 2004.

P2754 L27: Should start with "Measurements of TC concentrations..." Has TC been defined?
It will be defined as total carbon.

P2755 L4-5: Which analysis protocol was applied?
OC/TC ratios of selected samples for 14C analysis were determined with a commercial thermo-optical transmission instrument (Sunset Laboratory, Tigard, OR, USA) using the EUSAAR-2 protocol.

P2755 L15: The sentence is unclear. I suggest rephrasing into something like this: blanks were measured and were also estimated from regression analysis.

The sentence will be clarified as suggested.

P2756 L16-20 and 21-26: These lines should be rewritten using better English grammar and clarity.
Sections will be revised.

P2757 L17: How do you define continental sources? As all non-fossil sources, whether natural or man-made? If so, then it would be better to state this more clearly.
Correct, continental sources were assumed to be all non-fossil sources whether natural or man-made. It will be stated accordingly in the text.

P2759 L7-10: It is not clear how these other studies support the interpretation.
Those studies support less negative ratios found in marine aerosols and not the sentence preceding it. This will be revised accordingly.

P2760 L25: It is a very strong statement that the NH marine boundary layer can be exceptionally clean and largely devoid of anthropogenic material. Such a statement should be documented by measurements of e.g. elemental tracers. See also general comments.
This statement is supported by air mass back-trajectories and radon concentrations in aerosol (Rn220 and Rn222) as explained above.

P2761: The discussion should reflect that the conclusions are drawn on a limited study of only 6 samples. Are these results in line with previous investigations at the site?
A statement of validity of the results based on limited number of samples will be added. These results are the first quantitative estimates of the magnitude of marine versus anthropogenic sources, however, they are very well in line with other measurements performed at the site (O’Dowd et al., 2004; Cavalli et al., 2004; Yoon et al., 2007). A corresponding sentence will be added to the conclusions.

P2761 L25: Should be changed to "Dual carbon isotope.." and "marine and polluted aerosol samples"
It will be revised accordingly.

P2761 L27-: Should be rephrased to reflect that the conclusions are drawn from a study of 6 samples.
Stated as above.

P2762 L7-: The sentence is not clear and should be more descriptive. What are the 30%? Table 2. The notation delta 14C should be described in the text or footnotes. Are the uncertainties due to counting statistics? What is the overall uncertainty on the results?
Marine sources can contribute around 30% to the amount of total carbon in polluted air masses sampled at Mace Head which has not been quantified so far. The sentence will be rephrased.
P 2757 L9-10 says: "Δ14C values of the filter samples were calculated according to Stuiver and Polach (1977) and corrected for a filter blank." In principle, this citation is standard within the 14C community. Referring to the comment of the referee, however, we will include the following equation into the revised version of the manuscript.

$\Delta^{14}C = [R_{\text{sample (norm)}}/R_{\text{standard (corr)}} - 1] \times 1000$,
where the ^{13}C isotopic fractionation in the sample, irrespective of environment, is taken into account by normalizing to $\delta^{13}C = -25‰$ and the standard is corrected for its decay since the reference year 1950.

The 14C results in Table 2 are accompanied by total uncertainties (around 3-5%) including contributions from the 14C measurements (mainly from counting statistics) and the correction of the field blank.

Reviewer 2

1) The literature on 13C measurements for aerosol source apportionment is not very well reviewed and many important citations are missing. For example, the term dual isotopic characterization and some first applications were already presented by Currie et al., 2000 (references therein). This should obviously be cited. Other examples follow later in the review, but the authors should conduct a thorough literature study and use it for introduction and discussion of results.

We admit the lack of a thorough literature study (especially regarding 14C) and thank the reviewer for pointing at some of the important papers. The introduction will be expanded accordingly.

2) The methods still need to be discussed in more detail before they are sufficiently clear and can be evaluated.

a. Pg 2755: How was TC converted to CO2 for 13C analysis?

The filters were analysed with the elemental analyzer FlashEA 1112 connected to the stable isotope ratio mass spectrometer ThermoFinnigan Delta Plus Advantage. The 1/8th part of the filter was placed into the tin capsule and combusted in the oxidation furnace at the temperature of 1020C and with the oxygen excess. Later this gas was transferred into the reduction furnace (650C). The water from the sample in the helium flow was removed using the magnesium perchlorate trap. Then the gas mixture was separated in the column PoraPlot Q (50C). Separated gas was delivered to the mass spectrometer ionization cell through the gas distribution device ConFlow III. Before the analysis of a series of samples the calibration CO2 gas was delivered to the mass spectrometer until the isotopic ratio uncertainty was better than 0.15%.
We will include a shorter version of the above description as it is already published in Garbaras et al. (2008).

b. Pg 2755, line 15ff: Please describe exactly how field blanks were treated. What was used for C_blank in eq 2?

The CO2 signal in millivolts was used in Eq.2 assuming a constant contribution from filter field blank. A proper equation will be added in the text:

$$\delta^{13}TC_{\mathrm{sample}} \times CO_{2(\mathrm{sample})} = \delta^{13}TC_{\mathrm{blank}} \times CO_{2(\mathrm{blank})} + \delta^{13}TC_{\mathrm{aerosol}} \times (CO_{2(\mathrm{sample})} - CO_{2(\mathrm{blank})})$$

c. Pg 2756, line 10ff: This is confusing. What was the motivation for calculating marine 13C this way? I assume this does not refer to the blank correction, so please give the exact equation that was used. Was only BC subtracted? What about other fossil sources? Moreover, there are several other studies that give significantly higher delta 13C values for BC closer to -27 ‰ (Huang et al., 2006; Ho et al., 2006) or even higher for biomass burning BC. There are more examples in the literature, which I encourage to authors to look up.

The equation for calculating marine 13C was the following:

$$\delta^{13}TC_{\mathrm{sample}} \times CO_{2(\mathrm{sample})} = \delta^{13}TC_{\mathrm{marine}} \times (CO_{2(\mathrm{sample})} - CO_{2(anthr.)}) + \delta^{13}TC_{\mathrm{anthr.}} \times CO_{2(\mathrm{anthr.})}$$

where $CO_{2(\mathrm{anthropogenic})}$ was calculated from Figure 2 regression equation.

BC measurements (by aethalometer AE16 and multi-angle absorption photometer MAAP) are routinely performed at Mace Head station and show low but discernable BC levels even in clean air masses. 14C analysis is suggesting the same. We attempted to calculate marine delta 13C ratio assuming certain amount of carbon being anthropogenic. The concentration of BC in clean marine air masses is always below 50ng/m3 (in selectively sampled clean marine air masses as outlined in the Methods section), but some of this carbon must be organic carbon (absorption measurements can not distinguish between black carbon and other absorbing OC species). We agree with the comment that there are other studies suggesting somewhat different anthropogenic delta 13C values. Marine delta 13C will be presented with the uncertainty level encompassing the range of anthropogenic delta 13C values found in different studies.

d. Equation 5, more explanation is needed here: Which data sets were used to minimize this equation? The same subset of data that is used later for source apportionment? How would this method perform if the variation in the data was not governed by 3 main sources, but maybe 4 or 5 sources would be important?

Indeed, the same subset of data was used to minimise this equation (i.e. subset of 6 samples for which both 13C and 14C data were available). We can't comment on how this method would perform if 4 or 5 sources would be apportioned. We can speculate that with more potential sources more uncertainty would be expected, especially with less well constrained equations for those additional sources.

3) Contrary to the authors assertion there is evidence of isotopic modification carbonaceous aerosol by photochemical processing (e.g. Wang and Kawamura, 2006 and some more recent studies). The more convincing argument to neglect it, would be the absence of a seasonal variation in polluted 13C values.

We agree that the topic of isotopic fractionation has been started to be addressed. The aforementioned study suggests isotopic fractionation based on different isotopic ratios of di-carboxylic acids (DCAs). However, one could obtain different delta 13C ratios of different DCAs if precursors of various DCAs have different sources, e.g. some produced by secondary processes, some by heterogeneous ageing of primary material. In summary, while acknowledging the issue of isotopic fractionation it is not clear of how to obtain a quantitative estimate of it. We have not attempted to separate secondary and primary marine sources and consider marine aerosol in bulk. We have little confidence in addressing the effect of process dependent isotopic fractionation until more robust quantitative estimates of primary versus secondary organic aerosol in marine environment become available. The issue of isotopic fractionation will be expanded in the next version of the manuscript.

4) The estimate of delta 13C of 29‰ for fossil carbon is somewhat in contradiction to most of the literature values that I am aware of, which are in fact closer to -27‰ to 26‰. See e.g. Ho et al., 2006 (roadside); Hueng et al., (2006) (tunnel); Widory et al., 2004, Currie et al., 2000. The values estimated here should be compared with a larger number of literature values, (not only the ones cited in this review).

The estimate of delta 13C of -29‰ comes from error minimisation. While we acknowledge some contradiction with the references pointed by the reviewer, we clearly stated that our values were obtained by solving the equations. Using any other values would prevent closing the equations (using error minimisation). In fact, it should also be acknowledged that there are references presenting anthropogenic delta 13C values (e.g. that of BC) below our estimate of -29‰. For example, -29.2‰ (Rumpel et al., 2006) or -31‰ (Ulevicius et al., 2010) as referenced in the paper. Also Bakwin et al. (1998) presented a range of fossil fuel carbon d13C values from a global network of sites with an average value of -28.3±2.4‰. We agree that our delta 13C value is at the lower negative end of literature values which will be stated in the text. Having said that, we state in the paper that by varying 13C source ratios by ±1‰ and 14C by ±50‰, the corresponding uncertainty in contribution of sources comes to 3 to 5%. Therefore, the value of -29‰ should be considered as -29±1‰ with a fair degree of tolerance in it, despite finding it by error minimisation.

5) This relatively low estimate of fossil delta 13C could also be the reason for larger ma-rine contributions than in the literature, or the absence of non-fossil continental sources. At least a sensitivity study should be conducted that uses the more commonly measured isotopic value of fossil sources.

It is not correct to state that relatively low estimate of fossil delta 13C could be the reason for a larger marine contribution. It would only be true considering 13C data alone. 14C data do not allow for a larger proportion of fossil fuel contribution. It is a combination of both carbon isotopes which leaves little room for source specific isotope ratio uncertainty.

References (provided by the reviewer):

Widory, D., S. Roy, Y. L. Moullec, G. Goupil, A. Cocherie, and C. Guerrot (2004), The origin of atmospheric particles in Paris: a view through carbon and lead isotopes, Atmos. Environ., 38, 953-961.
Huang, L., et al., Atmos. Environ., 40, 2690-2705, (2006).
Wang, H. B., Kawamura, K., J. Geophys. Res., 111, D07304 doi.:10.1029/2005JD006466, (2006).
Currie, L.A., Evolution and multidisciplinary frontiers of 14C aerosol science, Radiocarbon, 42 (1), 115-126, 2000.
Ho, K.F., S.C. Lee, J.J. Cao, Y.S. Li, J.C. Chow, J.G. Watson, K. Fung, Variability of organic and elemental carbon, water soluble organic carbon, and isotopes in Hong Kong, Atmos. Chem. Phys., 6, 4569-4576, 2006.

We would like to thank the reviewer for pointing at valuable references.

Short Comment

The first look of the title makes me confusing, since it mentions submicron particles, but indeed it refers to Dp <1.5 micrometer throughout the paper, is that strange? Also, "aerosol particles" is awkward, why not just use "particles"?
"Submicron" will be substituted by "fine particles (D_{50}<1.5um)".

I also think the paper might require expansion. In the introduction, little information is given for the stable carbon isotope analysis, I think more relevant studies should be mentioned, for example,
Fisseha et al., Rapid Commun Mass Spectrom. 2006;20(15):2343-7;
Li et al,, Anal. Chem., 2010, 82 (16), pp 6764–6769;
Zhang et al., Chemosphere, 75, 2009, 453-461.
Introduction will be expanded to discuss stable carbon isotope analysis in the broader context.

Page 2752, line 5: "other postulated to marine sources". I think it is probably better to cite the references for the specific species individually, not to mix them together.
Additionally, there are two recent papers on amines:
Ge et al., Atmos. Environ., 2011, 45, 524-546 and 561-577)
The sentence will be amended in the expanded introduction section.

References (added by authors):

Ceburnis et al., J. Environ. Monit., 8, 479-487, (2006)
Rinaldi et al., Advances in Meterology, Vol. 2010, Article ID 310682
Heal et al., Atmospheric Environment, 45, 2341-2348, (2011)
Biraud et al., J. Geophys. Res., 105 (D1), 1351-1366, (2000)
Beaupre & Druffel, Deep Sea Research I, 56, 1717-1726, (2009)
Williams & Gordon, Deep Sea Research, 17, 19-27, (1970)
Bauer et al., Deep-Sea Research II, 49, 4387–4419, (2002)
Bakwin et al., Global Biogeochemical Cycles, 12(3), 555-562, (1998)
Hoefs J. Stable Isotope Geochemistry. Springer-Verlag, Berlin Heidelberg, 2009