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12, C9431–C9435, 2012

Interactive Comment

Interactive comment on "Chemical characterization and stable carbon isotopic composition of particulate polycyclic aromatic hydrocarbons issued from combustion of 10 Mediterranean woods" by A. Guillon et al.

A. Guillon et al.

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Dear Referee 2,

Please find below the answers of your questions and comments to be considered for the submission of our revised manuscript to Atmospheric Chemistry and Physics:

Journal: ACP Title: Chemical characterization and stable carbon isotopic composition of particulate Polycyclic Aromatic Hydrocarbons issued from combustion of 10 Mediterranean woods. Author(s): A. GUILLON, K. LE MENACH, P.-M. FLAUD, N.



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MARCHAND, H. BUDZINSKI, E. VILLENAVE MS No.: acp-2012-409 (Research Article)

We first noted that you think that measurements of concentrations and delta13C values of polycyclic aromatic hydrocarbons in wood burning aerosols are difficult to do (especially for delta13C) and that we achieve good reproducibility. You also report that the use of stable isotopes in aerosol source apportionment is a promising research field, where data on aerosol sources are urgently needed. We have noted that you think that this data set is an important contribution to the literature. However, you also made some remarks about some conclusions that were not justified or needed to be rewritten for a better clarity. Particularly you focused on the weak level in English and on the Experimental Procedure section that needed to be reorganized. You will be happy that we have taken into account all of your remarks (see below), proposing in the new version of the manuscript a completely rewritten section, more synthetic and clear. Note also that the full manuscript has now been corrected by a native English, as requested.

Thanking you for your help in improving our article,

Best wishes,

Prof. Eric VILLENAVE

Answers to Referee 2.

1) OK. We have as proposed reorganized the section according to the recommendations of the referee. This section is now improved and intelligible (= with less jargon) for non-experts in the analysis of particulate-PAHs.

2) Right. If chemical reactions usually result in isotopic 13C fractionation when occurring in the gas phase, there was up to now no available data concerning isotopic variations during atmospheric heterogeneous processes (which are the most efficient in the troposphere in the case of PAHs). The only study has been recently performed

ACPD

12, C9431–C9435, 2012

Interactive Comment

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by our group (Guillon et al.; 2012a), showing that no significant fractionation occurred during reactions of particulate-PAHs with O3, OH and NOx or under solar irradiance, of atmospheric interest. Such result is important as it allows to consider that the methodology developed in our work permit to use isotopic measurements for source tracking of particles in the atmosphere, as potential isotopic variations induced by heterogeneous chemical reactions are much lower than uncertainties on GC/C/IRMS measurements and also considering the larger range of variations of delta 13C of particulate-PAHs from different sources. This is now better explained in the new version of the introduction section of the manuscript.

3) Sorry. It is a simple mistake (a bad "copy and paste"). 1 cm2 is the right value. It has now been corrected in the text.

4) OK. For quantification measurements, standard reference materials (previously deposited on glass fiber filter), punches of 0.64 cm2 of sampled filters and punches of 16.62 cm2 of blank filters were extracted in 10 mL cells. For isotopic measurements, remaining parts of each filter were extracted in 34 mL cells. This is now better clarified in the new version of the manuscript in the 2.4.1 section.

5) Note that this remark was already reported by referee 1. The methodologies developed in this work were validated on model particles: PAH bounded silica particles were extracted by PFE; extracts were purified using HPLC fractionation and injected in GC/C/IRMS. Isotopic compositions of PAHs, measured all along the procedure, were found to remain constant within uncertainties. Therefore, no correction was needed on isotopic composition measurements. This is now added as quoted above in the new version of the manuscript section (now 2.4.2).

6) There is little importance about the change in the different values of recovery yields of internal standards as they are deliberately introduced in the procedure to evaluate all the possible effects of the full analytical protocol (extraction and purification steps) on the PAHs of interest.

ACPD

12, C9431-C9435, 2012

Interactive Comment



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Interactive Discussion



7a) OK. All the proposed changes have been done. Note that it was also suggested by the referee 1.

7b) As presented in the introduction, due the fairly high heterogeneous reactivity of PAHs in the atmosphere (depending on both the nature of the PAHs and that of the particle surfaces), involving variations of concentrations and therefore diagnostic ratios, it is sometimes risky to apportion particulate-PAH sources by only applying such an approach.

7c) The new manuscript has been corrected by a native English.

8) Right, we of course agree with that remark and have just deleted the last sentence of the paragraph that could be misinterpreted.

9) Yes, to the first part of the question. This was also a remark from referee 1. When performing an error propagation analysis, errors are of course enlarged, accounting for global both statistical and systematic uncertainties. Considering the reproducibility of standard isotopic measurements of 0.5‰ the differentiation among investigated wood species remains significant while it is confirmed that isotopic composition of PAHs is independent of the burning type, as expected.

No, for the second part of the question. First, no woods have been measured only once; 2 ïAş errors are clearly presented in the Table 7. All woods have been at least characterized 3 times (as stated in the text (section 3.2)). The numbers are corresponding to the type of combustion and not to the number of repeated burnings.

Other (minor) comments:

Abstract, line 18: The sentence has been improved for a better clarity.

Page 20635, line 20: It is not field blanks but filter blanks as stated in the section 2.3.

Page 20636, line 22: OK.

Page 20643, line 16: The sentence has been removed as suggested by referee 1.

12, C9431–C9435, 2012

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Page 20644, line 3: OK.

Page 20644, line 5: OK. The expression has been removed.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 20631, 2012.

ACPD

12, C9431–C9435, 2012

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