We would like to thank the referees for their careful reading, review and useful questions and comments, which definitely will help to improve a revised version of the paper. We have addressed all comments below and indicate modifications planned for a revised version.

Replies to the comments of Referee #1

The paper represents important study of the PAH content in the atmosphere and can be published after some corrections.

Specific comments:

1. The first sentence of the abstract is not rather clear. It is not mentioned that measurements were made for the subset of PAHs (25 PAHs). May be there is also a need to characterize somehow what kind of PAHs were selected in the study (light, heavy, etc.) and may be to give the fraction in particulate phase as the range (not just one value – 8%).

yes, agree. For better clarity the first sentence of the abstract ' Polycyclic aromatic hydrocarbons (PAH) concentration in air of the central and eastern Mediterranean in summer 2010 was 1.45 (0.30–3.25) ngm−3, with ≈8% in the particulate phase, associated with particles< 0.25 µm.' will be re-phrased as 'Polycyclic aromatic hydrocarbons concentration in air of the central and eastern Mediterranean in summer 2010 was 1.45 (0.30–3.25) ngm−3 (sum of 25 PAHs), with 8 (1-17) % in the particulate phase., associated with particles < 0.25 µm.'

2. In the introduction (in the first statement) domestic heating also could me mentioned.

yes, agree, domestic heating will be mentioned in revised version

3. In section 2.1, the last paragraph might need reformulation as it is not rather clear that from the absence of difference between observed values on board of the ship it follows that ship-based sources were not significant.

yes, agree. For better clarity the sentence 'The PAH levels indicated by these samplers did not show significant differences, hence the ship-based contamination is negligible.' will be re-phrased as 'The PAH levels of these samples indicated that ship-based contamination was negligible.'

4. In section 2.3, in the first sentence it could be useful to note more clear what data set is meant. It is also mentioned that no data on soot or PM composition is available, but it could be said at least that this process can also affect partitioning. The underprediction mentioned later on may be connected with this selection of partitioning models.

yes; data set will be specified by '15 high-volume samples of separate gas and particulate phase concentrations'. Significance of soot for gas-particle partitioning will be emphasized: ' We refrain from testing dual adsorption and absorption models (e.g. Lohmann and Lammel, 2004) as no soot data or PM chemical composition data are available.' will be re-phrased as 'Adsorption to soot is a significant gas---particle partitioning processes for PAHs, but no soot data or PM chemical composition data are available. We, therefore, refrain from testing dual adsorption and absorption models (e.g. Lohmann and Lammel, 2004).' In the discussion of the results of the gas-particle partitioning models tested (section 3.2) the sentence ' The neglect of adsorption to soot, not covered by the gas-particle partitioning models tested, may explain a at least part of the underprediction' will be added.

5. In section 2.5 the selection of RET for the simulations of air-sea exchange can be somehow explained.

yes, agree. Short sentence 'RET is selected, because of the prevalence of one dominating source.' will be added to section 2.5.

6. In section 2.6 there is a need to give more detailed information on the generation of these retroplumes and their use for identification of potential sources.

agree. 1st paragraph of section 2.6 will start in the revised version with an introductory sentence, providing also references to respective applications i.e., 'Distributions of potential sources can be identified by inverse modelling using meteorological input data (Stohl et al., 2003; Eckhardt et al., 2007).'

7. Section 3.1: the value of total PAH concentration and its range differ from that given in the Table 1a.

The values differ because the mean values in Table 1a are referring to all 15 samples, regardless whether exceeding LOQ or not, with the ones < LOQ replaced by LOQ/2. To avoid confusion the value in the table will be mentioned in the text, too, explaining the difference.

Some comments would be useful on those PAHs that were higher than previously measured. For gaseous phase (Table 2a) these are FLT and PYR, but for particulate phase (Table 2b) there are more PAHs that were higher that previously measured. This might be connected with difference in seasons or something else. . .

Agree, although the basis for comparison of this and previous studies is limited, as emphasized. The discussion of the results will be expanded: 'The variability of advection or advection in combination with different routes taken by a vessel being influenced differently by coastal or ship emission plumes may explain this difference.' will be changed to:

'In the ISS some PAHs are found somewhat higher than previously measured i.e., FLT and PYR (in the gas-phase) and BAP and PER (in the particulate phase). Due to a sampling artefact BAP and other particulate phase PAHs could be underestimated by up to 25% (aforementioned, section 2.1). The seasonality of emissions and the variability of advection or advection in combination with different cruise routes being influenced differently by coastal or ship emission plumes can have a large influence and may explain these differences. On the other hand, the duration of temporal averaging atmospheric concentrations was similar across the various studies.'

8. In section 3.2 again effect of sorption on OC and EC could be mentioned, not just the absence of data.

yes; a respective sentence will be added in the revised version i.e., 'The neglect of adsorption to soot, not covered by the gas-particle partitioning models tested, may explain at least part of the underprediction.'

9. In section 3.3, the units of (> 50 ngm3 d?1) seems to be wrong.

yes, thanks, should be ng m^{-2} d^{-1}

10. In section 4, conclusions, it should be added that for most of measured PAHs the levels were lower than previously measured,

This conclusion is anyway drawn (first sentence of the section reads 'PAH pollution of the atmospheric Mediterranean environment was below previous observations at the beginning of the decade (2001-02; Tsapakis and Stephanou, 2005a; Tsapakis et al., 2006),

also considering possible losses during sampling. This might reflect emission reductions.')
again effect of sorption on OC and EC could be mentioned. Potential sources of pollution of marine atmosphere could also be mentioned as it was evaluated through the use of FLEXPART model.
We refrain from drawing a conclusion on gas-particle partitioning because of the lack of data needed to test all relevant models.
A conclusion section should include conclusions, nothing else. The results on long-range transport paths (to the ship's position, using the inverse model FLEXPART) are mentioned in the results section and are the subject of an on-going study, to be published soon.
11. Table 3a provides observed and predicted fractions of particulate phase, but the units mentioned are ng m-3 that seems to be not correct.
Yes, thanks, unit will be removed.

Replies to the comments of Referee #2
This paper on the air-sea exchange of PAHs in the Mediterranean is based on a unique dataset of air and water samples and gives new insights into the cycling of PAHs between different environmental compartments. While the idea of the paper is compelling and in general suitable for publication in ACP the paper suffers from a variety of shortcomings. Thus, I can only support publication of this work after major revisions.
First of all, the paper would strongly benefit from a revision by a native speaker. Some sentences are obscure and it is not clear what exactly the authors want to say. A few examples:
p5967 l20,21 'Each one' [...] 'as far as possible identical manipulations on site'
will be re-phrased for better clarity: "Field blank values, b, were gained from GFFs and PUFs manipulated in the field, as far as possible identical to the samples, except without switching the high-volume sampler on."
p5972 l16 'corresponds with no or beging hit of such areas'
will be re-phrased for better clarity: "…, vice versa, low concentrations corresponded with no apparent passage of such areas"
p5972 l16 'This perception' should probably be 'This finding'
yes, agree
p5973 l17,18 '...PAH was almost exclusively quantified in particle size fraction <0.25μm' should probably mean 'the majority of the particulate PAHs were found in the size fraction <0.25 μm'?
yes, agree. Will be modified to "most of the particulate PAH mass was found in the size fraction <0.25μm."
p5974 l3 'FLT and PYR are indicated to be close to phase equilibrium' should probably mean 'were found to be'
yes, agree
p5974 l24 'observable' should be 'observed'
yes, agree
Secondly, besides the title '... aerosols and air-sea exchange ...' there is only little information on the actual findings about PAHs in/on aerosols. Also the air-sea exchange was only investigated for 3 PAHs.

PAHs in aerosols: The gas-particle partitioning of semivolatile PAHs is presented in Sections 3.2 and S2.1 (and Tables 2, S2). We refrained from reporting trivial findings (e.g., on gas-particle partitioning of more volatile and non-volatile PAHs, which was expectedly close to 0 and close to 1, respectively), not needed to be communicated 25 years after publication of key literature in this topic.

PAHs air-sea exchange: Only for 4 PAHs surface seawater concentrations >LOQ were found (Table 1b). This will be stressed for better clarity in the text of section 3.1 and caption of Table 1. We prefer to refrain from speculating about air-sea exchange of PAHs which could not be quantified in surface seawater.

detailed comments:

ABSTRACT: In the Abstract (p5964 l1-6) Why are only the findings of particles <0.25µm discussed? What is the total percentage of particulate PAHs, how is the size distribution? It is later stated (p5966 l3) that particles were collected with 2.5, 1.0, 0.5, and 0.25µm filters. But the size distribution is never discussed in the paper.

Will be re-phrased for better specification. The PAH mass size distributions will be presented (new Table S4) and discussed including more literature (following suggestions of ref. #3).

Further, with the strong focus on air-sea exchange at least a rough estimate about the total annual flux should be made.

Agree. A rough estimate of total annual flux of particulate phase PAHs can be made for the season (but not annually) and will be included in the conclusion section and abstract of a revised version:

The residence time in the troposphere is longest for particles around 0.2 µm of size, with ≈0.01 cm s^{-1} being a characteristic corresponding dry deposition velocity (Franklin et al., 2000), which translates into a residence time of ≈120 days in the MBL (depth of 1000 m; see Table S3) and deposition fluxes $F_{dep} = c \times v$ = 0.03-0.13 µg m^{-2} $year^{-1}$, for the individual PAHs associated with the particulate phase (c = 0.01-0.04 ng m^{-3}; Table 2b), such as BAP, and 0.80 and 0.35 µg m^{-2} $year^{-1}$, respectively, for the total flux of particulate phase PAHs in the ISS and SEM in summer, respectively. The flux will be higher in winter because of the seasonality of the emissions.

Ref.: Franklin et al.: Quantitative determination of persistence in air. In: Criteria for persistence and long-range transport of chemicals in the environment (Klečka et al., eds.), Pensacola, USA (SETAC Press), pp. 7-62, 2000.

p5964 l10-12: 'It is concluded that future negative emission trends or interannual variability of regional sources may trigger the sea to become a secondary PAH source' How large could this source be compared to the primary sources in the Mediterranean?

Thank you for this question. This should indeed be quantified and added to the paper (conclusions). For retene it is found that one third was returned to the atmosphere during the study period: While the flux of secondary retene emissions (from surface seawaters) in the study area was 1.0 µg m^{-2} $year^{-1}$ (mean of 6 years,_UEPS), the primary sources amounted to 3.1 µg m^{-2} $year^{-1}$ (mean of 6 years). This result will be included into the abstract of the revised version ('One third of primary retene sources to the sea region in the period 2005-2010 returned to the atmosphere as secondary emissions from surface seawaters.')

METHODOLOGY: p5968 l3: Abbreviation LOQ should be explained the first time it is used.

yes, abbreviation LOQ introduced at first time mentioned

Section 2.4 seems a bit too short. It should be made clear how kw and ka were calculated. Also it should be discussed whether the air-sea exchange is dominated by ka or kw. For most species one of the two mass transfer coefficients clearly dominates the exchange process. Also the difference between 2 and 4 ring PAHs should be discussed here (e.g. what values were used to describe the diffusivity of the different species, was a temperature dependency taken into account). In general, readers of the paper should be able to reproduce the calculations done by the authors. Giving the formula and the reference to Schwarzenbach et al. is not enough here.

Transfer coefficients were calculated for one 3 ring and two 4 ring PAHs (stated in section 3.3), $k_w << k_a$ throughout (will be specified section 3.3). Derivation of k_w and k_a now explained in detail, including the references to the input data sources: ’$k_a = (0.2U_{10} + 0.3)*(D_{i,air}/D_{H2O,air})^{0.61}*36$, $k_w = (0.45U_{10}^{1.64})*(Sc_i/Sc_{CO2})^{-0.5}*0.01$. U_{10} is the wind speed at 10 meter height above sea level (m s^{-1}), $D_{i,air}$ and $D_{H2O,air}$ are the temperature dependent diffusivities of substance i and H_2O in air, and Sc_i and Sc_{CO2} are the Schmidt numbers for substance i and CO_2 (see Bidleman and McConnell, 1995; Zhong et al. 2012; and references therein). U_{10}, T_a, T_w and air pressure are taken from the ship based measurements.’

Section 2.5: Is river runoff considered as a source for PAHs? Is it an important source?

Many PAHs are enriched in coastal waters as opposed to the open sea, most likely as river runoff is a significant source. As retene is rarely monitored this is not obvious for this substance. In an urban estuary in the USA, the linkage with river runoff was found less obvious for retene than for other PAHs (Lohmann et al., Env Sci Technol 2011, 45, 2655-2662). The neglect of the riverine retene source in our modelling may cause an underestimate of the emission flux, Faw, and might explain part of the discrepancy (underestimate when compared to observed flux). This will be added to the discussion text (section 3.6): The sentence ' The sensitivity to input uncertainties (SM S1.2) may explain part of the underestimate, but not up to one order of magnitude.' will be extended: 'Neglected retene sources to seawater, such as riverine input may explain part of the discrepancy.'

Section 2.6 is much too short (2 sentences) to justify an additional section.

This part of the methodology is different from others. Its results are included in the text of section 3.1 (1st paragraph).

RESULTS: Section 3.1: It should be discussed whether earlier crusies are comparable. Different seasons, different wind conditions, different cruise routes can potentially have a large influence on the observed concentrations.

This discussion is expanded as suggested by reviewer #1. ('The seasonality of emissions and the variability of advection or advection in combination with different cruise routes being influenced differently by coastal or ship emission plumes can have a large influence and may explain these differences. On the other hand, the duration of temporal averaging atmospheric concentrations was similar across the various studies.')

Table 2 should state precisely when the different cruises were taking place.

Months identified in the revised version

The reference (1) in Table 2b is not used.

Ref. (1) is used in Table 2a

Section 3.2: As stated before there should be more information about the size distribution of the PAHs from the 2.5, 1.0, 0.5, 0.25μm filters.

The PAH mass size distributions will be presented, listed for the size range <0.25μm (below, new Table S4) and discussed including an earlier study of size segregated PAH measurements in the eastern Mediteranean (Kavouras and Stephanou, JGR 2002).

Table S4. Concentrations of 4-6 ring PAHs found in particulate phase size fraction corresponding to <0.25 μm a.e.d. (ng m^{-3}) as time-weighted mean (min-max). n_{LOQ} = number of samples > LOQ (out of 3). PAHs with concentrations <LOQ in all size fractions not listed. LOQ = 0.001 ng m^{-3} except for FLT (0.011) and PYR (0.020).

	n_{LOQ}	mean (min-max)
FLT	3	0.046 (0.035–0.054)
PYR	3	0.047 (0.040–0.051)
RET	3	0.048 (0.046–0.050)
CPP	1	0.007 (<0.001–0.022)
CHR	1	0.002 (<0.001–0.005)
BJF	1	0.002 (<0.001–0.007)
BEP	1	0.003 (<0.001–0.008)

CONCLUSIONS: The Conclusions seem to be very short. It would be beneficial to give an estimate about the relation of total annual RET deposition to net revolatilisation. Also there should be more information about the differences between the observed PAHs and on the observed particulate PAHs.

In the revised version retene deposition will be compared with revolatilisation (detailed above).

The results on PAH level trends are partly inconclusive (discussion in section 3.1, expanded as suggested by reviewer #1). We prefer to refrain from drawing conclusion from inconclusive results.

Replies to the comments of Referee #3

General comments The current manuscript presents PAHs concentrations in the air and the seawater that were obtained during a 15-day cruise (27 Aug-12 Sept 2010) of the research vessel Urania across central and southeastern Mediterranean. Although the g-p partitioning of PAHs is presented, the authors focus mainly at the diffusive air-sea exchange fluxes of the three PAHs measured in sea water. The manuscript contains interesting original data concerning the occurrence of PAHs in the open sea atmosphere. Nevertheless, it suffers from various scientific and technical deficiencies (several obscurities, inconsistencies, insufficient comparison with literature data, etc), most of which are listed below. In my opinion, the manuscript needs revision before it can be accepted for publication in ACP.

Specific comments:

Title The tile of the manuscript is not consistent with the content, I would suggest the following (Gas-particle partitioning and air-sea exchange of polycyclic aromatic hydrocarbons in the Mediterranean)

agree, title changed into 'Air-sea exchange and gas-particle partitioning of polycyclic aromatic hydrocarbons in the Mediterranean'

Methods - Section 2.1: It should be clarified whether results presented in the manuscript have been corrected for possible losses during sampling due to oxidation by ozone.

This is anyway clarified in the text of section 2.1 ('Based on such sampling artefact quantification studies (Tsapakis and Stephanou, 2003; Galarneau et al., 2006) and ozone levels (Table 1a) and sampling times (Table S1) we expect that total PAHs are underestimated by up to 50% in the gas-phase and by up to 25% in the particulate phase.'). Implications now included in the discussion (section 3.1).

- Section 2.2: The section title about organic trace substances is confusing, please, change to the following "PAHs analyses and quality assurance". Also, change "organic" with "PAHs" in the first sentence of this section.

agree, will be modified accordingly

- Section 2.2: It is stated that no QFF field blank was taken for impactor sampling, but instead the mean of values of the QFF substrates of the 2 uppermost impactor stages (in total 5) was taken". Hence, more information about the concentrations of PAHs in the 1-2.5 µm and the 2.5-10 µm particle fractions needs to be provided.

agree, will be specified in section 2.2 ('No QFF field blank was taken for impactor sampling. As no PAHs were detected in the stages corresponding to 2.5-10 µm (all PAHs < limit of detection in all such samples), instead the mean of values of the QFF substrates of the 2 uppermost impactor stages (in total 5) was taken.')

- Section 2.4: It is unclear which concentrations were used in the air-sea diffusive mass exchange calculations. The FR equation requires the dissolved aqueous concentration Cw. However, as stated in Section 2.2,"Water samples (2-2.5 L) were extracted immediately after their collection using solid phase extraction on C18 Empore discs", which means that total PAHs concentrations (dissolved and adsorbed on suspended particles) were determined (those presented in Table 1b).

Indeed total PAH concentrations were determined and dissolved concentrations used for mass exchange calculations: To this end dissolved aqueous concentrations were derived from the bulk seawater concentrations using Cw = Cbulk /(1 + kPOC*CPOC + kDOC * [DOC]), with POC and DOC from Pujo-Pay et al 2011, and kPOC and kDOC from Lüers & ten Hulscher 1996, Ma et al 2012, Karickhoff et al 1981, and Rowe et al 2009. This sentence now added to the section text.

- Section 2.4: Please correct the concentration units for Cw.

unit is correct, ng m^{-3}.

- Section 2.6: The analysis of the long-range advection of air needs further clarifications, especially the statement “Hereby, 50000 particles per hour were released and followed backwards in time for 5 days”.

will be clarified by re-phrasing (virtual particles)

Results and discussion

- Section 3.1: Statement about the long-range transport (illustrated in Fig. S5) is not very clear.

Agree; will be re-phrased for better clarity, as ‘corresponded with air masses having resided over large urban areas, and, vice versa, low concentrations corresponded with air masses without apparent passage of such areas’

- Section 3.1: The west-east gradient of PAHs concentrations could be evidenced if the sum concentration of PAHs (_PAHs) for ISS and SEM were included in Tables 2a and 2b.

Sum concentration will be included and referred to in the text

- Table 1a: Total concentrations (sum of gaseous and particulate phase) of PAHs provided in the Table are not consistent with the values provided in Table 2a and 2b for the separate phases (For instance, the total concentration of PHE is 0.581 ng/m3, whereas the gaseous and particulate phase concentrations are 0.35 and 0.51 ng/m3). Please, check and correct wherever needed.

The data are consistent. The apparent inconsistency is the consequence of time-weighted averaging of different data subsets, 15 samples in Table 1a, but each 5 samples in Table 2 for ISS and SEM data subsets.

- Table 2a,b: Some of the data provided in these Tables indicate an unexpected g/p partitioning for certain PAHs, for instance, for PHE and FLT greater distribution in the particle than in the gaseous phase is shown. This strange g/p partitioning should be explained.

Thanks for pointing to the presentation of these data, which was imperfect. No data suggest unplausible gas-particle partitioning, but the way they were presented (Table 2b) was misleading: mean values <LOQ were erroneously not flagged as such (but only the range shown in brackets showed values for min and max eventually flagged for <LOQ). Will be corrected unambiguously. Also the number of valid digits will be harmonized.

- Table 1b: Only 3 PAHs (PYR, FLT, RET) were quantifiable in sea water? Some comparison with literature data for open sea levels of PAHS would be useful.

Yes, will be compared with earlier data, which suggests similar levels: These observed seawater contamination levels are comparable to levels found in the region 2 and 1 decades ago (Lipiatou et al., 1997; Tsapakis et al., 2003).

– Section 3.2: The g/p partioning of PAHs should be more thoroughly investigated. The authors are claiming that the effect of temperature was not possible to be examined because of only little temperature variation during the cruise (21-27C). I encourage them to examine the logKp-logPLo relationship and evaluate the slope and intercept values.

Such type of examination was inspiring the research research 2 decades ago (Pankow & Bidleman Atmos Environ 1992; Pankow et al. Environ Sci Technol 1994), but is inherently inconclusive, at least as long as supporting parameters are not measured (but fixed values adopted assuming standard or characteristic conditions) and time resolution is not improved. We have been applying this method and will continue to do so for the interpretation of field data, assuring measured (rather than default) aerosol parameters, high time resolution, and an extended chemical characterization of the particulate phase (Lammel et al Atmos Environ 2010, J Environ Mon 2011, and unpublished in urban environments). Neither of these was possible on board RV Urania (our data set).

The examination’s limitations apart from lack of supporting parameters are as follows: If the gas-phase concentrations were controlled by local relaxation to (liquid–vapour) phase equilibrium, then the relationship between these and ambient temperature should be described by the Clausius–Clapeyron equation, physically equivalent to plots of the form

$\log K_P = A/T + B$. $\log K_p$ of PAHs based on differentiating sampling of the gaseous and particulate fractions was found to be correlated with 1/T (or $\log p_{L0}$) in a number of studies, but these tests were largely inconclusive for several reasons: Correlation with 1/T is in accordance with both adsorptive and absorptive mechanisms of gas-particle partitioning (Pankow et al. Environ Sci Technol 1994). Adsorption had been claimed to lead to a slope of -1, inconclusively, however, as various possible reasons for non-ideality exist which usually cannot be addressed (such as non-equilibrium, temperature change during sampling, and absorption into OM; Pankow & Bidleman Atmos Environ 1992; Goss & Schwarzenbach Environ Sci Technol 1998). This would in particular apply to our data set, as OM was not quantified and long sampling intervals, >12h, with significant temperature variation during sampling prevailed. Non-equilibrium can be caused by kinetic constraints (introduction of fresh particles or clean air) or by the presence of nonexchangeable compounds in the particles. None of these hypotheses explanations could be rejected based on our data set. Furthermore, standard or characteristic aerosol parameters had to be assumed (rather than determined). A large data scatter is anyway expected, as attributed to lack of representativeness of the temperature measurement for the volatilisation process (which may have occurred several days back, in particular if local substance levels are influenced by long-range transport; Hoff et al.: Non-linearities in the slope of Clausius-Clapeyron plots for semivolatile organic compounds. Environ. Sci. Technol. 1998, 32, 1793–1798), again, certainly also in our data set.

The explanation given in the text ('little temperature variation') will be replaced by one sentence identifying the key limitations of the method with regard to its application to our data set.

- The paragraph concerning the size distribution of PAHs needs substantial improvement, otherwise remove. Results are presented in one single phrase "In size-segregated samples PAH was almost exclusively quantified in particle size fractions <0.25 μm a.e.d." The particle mass size distribution is not reported. The authors are claiming that there are no available data in literature for the size distribution of PAHs in the open sea atmosphere; however comparison could be made with some background coastal sites in Mediterranean, such as Finokalia on the Crete Island, for which there are published PAH size distribution data.

Re-phrased for better specification in the abstract (following suggestion of ref. #1). More results from impactor samples presented (see above, following ref. #2)

- Table 3 provides calculated theta and logKp values for 4 PAHs only, whereas values for PAHs that were quantified in both phases (such as for FLN, PHE, ANT, FLT, PYR) are missing. Missing data shall be provided.

We limited the substances studied for testing gas-particle partitioning models to those showing obvious semivolatility i.e., concentrations in both atmospheric phases, C_g and C_p, >LOQ in at least half of the samples (n = 15), and with input data available. This led to the rejection of FLT and PYR, BBN (C_p <LOQ), CPP, BBF, BJF (C_g <LOQ), and BGF (no p0_L available). These criteria will be explained in the revised version.

- Section 3.3: The FR values reported for FLT, PYR and RET appear to follow the same order with the water solubility of the specific PAHs, FLT (265 μg/L) > PYR (135 μg/L) > RET (0 μg/L). Consequently, I am concerned that, if FRs were calculated from the total (soluble+adsorbed) concentrations in sea water, the FR of RET has been possibly underestimated. The FR equation requires the dissolved aqueous concentration Cw to be known. However, as described in Section 2.2, sea water samples were extracted immediately after their collection without filtration, which means that total PAHs concentrations (dissolved and adsorbed) were determined. The authors need to address this issue.

Indeed total PAH concentrations were determined and dissolved concentrations used for FR and mass exchange calculations (detailed above)

- Figure S2: Should I suppose that the units of the PM10 concentrations presented in Fig. S2 are µg/m3 and not ng/m3. Moreover, PM10 concentrations need to be clarified regarding the way they were obtained. If they were obtained from the low volume impactor sampling, they could not be more than 3 (according to sampling description in Section 2.1.). If they were obtained from the high volume sampling, they should be reported as TSP because the high volume sampler was not equipped with a PM10 inlet. Please, make the appropriate corrections and clarifications.

Yes, µg/m^3, now corrected in Fig. S2.

Conclusions: The conclusion that “PAH pollution of the atmospheric Mediterranean environment was below previous observations at the beginning of the decade (2001-02; Tsapakis and Stephanou, 2005; Tsapakis et al., 2006)” is not true, since some PAHs such as FLT, PHE and ANT in the particle phase were found in the current study at much higher concentrations. Therefore, it would be better if comparisons were made on the sum concentration of PAHs (_PAHs).

The discussion of these results has now been expanded to the individual PAH level (as suggested by reviewers #1 and #2)