Authors' Response to Anonymous Referee #1

We acknowledge the extensive comments by this reviewer which have improved the paper. Our point-by-point responses are embedded below (red) within the referee's original comments (black).

General comments:

An effort is made to model various semivolatile and non-volatile PAHs in air on short time scales and on the regional spatial scale (North America), including gas-particle partitioning. Such studies are timely as the understanding of the atmospheric fate, long-range transport and distribution of PAHs lags behind the one of other organic substance classes and other criteria pollutants. PAHs are toxic, most of the semivolatile, some of them persistent, hence, air-surface exchange (and multicompartmental cycling) and gas-particle partitioning are key processes for the atmospheric fate and long-range transport. The research field started from box models (e.g. Halsall et al. Atmos. Environ. 2001; Yaffe et al. Risk Analysis 2001; Prevedouros et al. Chemosphere 2004), while state of the art modelling includes validated dynamic aerosol sub-model and air-surface exchange parameterisations. Results from the application of an air quality model adopted to PAHs are reported. The adoption to the substances studied is incomplete.

The model is not presented in sufficient detail.
Much of the model has been described in detail in previous work referenced in the paper (e.g., Gong et al., 2006). For the new work on the addition of PAHs, 1400 words have been devoted to describing the new model components. We respond to the referees's detailed queries about more detailed aspects of the model below.

Deficiencies which render the results more or less inconclusive are an unclear aerosol sub-model (not introduced, references to validation lacking, how many components ? how is its performance with regard to major modes' surface and major components' mass concentrations at background sites ?)
Our original description in Section 2.1 gives an overview of the base model (AURAMS) upon which AURAMS-PAH is built along with referencing the paper by Gong et al. (2006) which itself includes references for the aerosol module. Section 2.1 stated that a sectional approach (rather than modal) is used to represent the aerosol particle size distribution and that 9 chemical components are used to represent aerosol particle composition. The reader was referred to the paper by Gong et al. (2006) for further details. We have modified the last sentence of the first paragraph of Section 2.1 as follows:
"A detailed overall description of AURAMS appears in Gong et al. (2006) while a description of the aerosol sectional approach and the microphysics modules of the model can be found in Gong et al. (2003a,b). Performance evaluation and model intercomparison results for AURAMS appear in McKeen et al. (2008), Smyth et al. (2009), Makar et al. (2010), Kelly et al. (2012) and Solazzo et al. (2012) among other publications."

and, most severe, lack of air-surface exchange parameterisations (inacceptable for the treatment of semivolatile substances which are long-lived in surface water or soils, which is the case for most of the substances addressed). In the same group another model, appropriate for studies of semivolatiles (including air-surface exchange), had been

developed (and used to study of chlorinated semivolatiles; Gong et al., Atmos Chem Phys 2007), but is not used in this study.
We thank the referee for raising this important issue though we respectfully disagree with the referee's comment that neglecting air-surface exchange in PAH modelling is unacceptable. This is a question that is subject to quantitative inquiry. As we have described in the manuscript, our model results are ambiguous in demonstrating whether or not air-surface exchange is important in simulating airborne concentrations of PAHs in Canada and the U.S. We have recommended that the issue be examined in greater detail in the future.

The study noted by the reviewer (Gong et al. (2007; ACP 7:4001-4013) and its companion paper by Huang et al. (2007; ACP 7:4015-4025) describe a global transport model that was tested for three species of polychlorinated biphenyls (PCBs). There are important differences between PAHs and PCBs that bear directly on the issue of including air-surface exchange in a PAH model.

PCB sources to the atmosphere are dominated by evaporation from environmental reservoirs subject to previous deposition and disposal as well as from electrical equipment, building materials, etc. that were put into service decades ago. Since PCBs are cycling through the environment with volatilisation and deposition being dominant processes, air-surface exchange is likely very important to their successful simulation. PAHs, on the other hand, are still emitted to the atmosphere primarily through combustion (Zhang and Tao, 2009, Atmos. Environ. 43:812-819) and atmospheric PAH burdens in North America are likely not dominated by air-surface exchange.

Lammel et al. (2009, Chemosphere 76:98-106) found that volatilisation was significant for semivolatile PAHs such as fluoranthene yet our model predicted atmospheric fluoranthene concentrations very well without volatilisation. We suspect that the difference in temporal and spatial scales in these two models may be responsible for these different findings. Lammel et al.'s global model examined measured values at two locations in the Arctic whereas our regional model examined measured values at locations in North America which are predominantly urban. The extent to which air-surface exchange parametrizations influence the predicted concentrations may thus depend on the locality under study – our results suggest that these parametrizations likely have a minor effect in regions subject to ongoing PAH emissions.

We have added text to Section 2.2 to reinforce statements in Section 2.1.2 regarding the exclusion of air-surface exchange in our study. We have also added text to Section 3.1.1 to reiterate that our measurements were made at predominantly urban locations. PAH concentrations there are likely not sensitive to contributions from air-surface exchange in the manner that those from rural locations appear to be as demonstrated in global modeling studies such as Lammel et al. (2009).

The results should not be published (in the peer-reviewed literature) at the present development stage of the model, unless limited to the study of nonvolatile PAHs (such as benzo(a)pyrene).
Based on our arguments above, we respectfully suggest that our model does indeed yield valid insights for semivolatile PAHs.

Unplausible results are found with regard to the predicted particulate fraction and the predicted concentrations using different gasparticle partitioning models.

Predicted particulate fractions are indeed underestimated for lower molecular weight compounds but the predictions improve for compounds with increasing molecular weight. Furthermore, our results show that particle/gas partitioning is not a major determinant of total PAH concentration at the scale and locations examined in this study. We would welcome the opportunity to address specific comments that elucidate the referee's assessment of plausibility.

Results determined by the aerosol sub-model would only be meaningful were the sub-model be evaluated and the relevant model output (lifetime of particle size modes/bins, chemical composition and number, surface and mass concentrations) be presented and discussed (see also specific comments, below).
The performance of the AURAMS model's predictions of size- and chemically-resolved particulate matter has been assessed in previously published literature (e.g., McKeen et al., 2007; Smyth et al., 2009; Kelly et al, 2012; Solazzo et al, 2012) where its performance has been found to be similar to that of other regional, peer-reviewed models in the literature. These references have been added to the paper.

The factors influencing the modelled gas-particle partitioning should be presented and then discussed, such that conclusions can be drawn. As is, the results have preliminary character. The discussion of the results in the light of previous knowledge in the field is deficient but could be improved by basing on the results being presented as atmospheric lifetimes and/or deposited and degraded amounts relative to emitted amounts.
We are cognizant of the multiple factors affecting particle/gas partitioning and have thus stated that this aspect of our model will form the basis of a subsequent publication. Examining all of the relevant factors within this work would have yielded a manuscript that would have been, in our opinion, too lengthy and unwieldy. We have chosen instead to discuss the model's general performance in simulating measured concentrations and to suggest areas for further study. As it stands, this more limited evaluation is already close to 8000 words in length.

The evaluation of model results by observational data is obviously biased by the choice of sites: Only background sites can meaningfully be compared with model output regarding the chosen model spatial resolution. Observations at sites influenced by local sources (urban, residential) will or may reflect strong spatial and temporal gradients, beyond the model resolutions, major aerosol components not included in the aerosol sub-model, and non-equilibrium partitioning. The temporal profile of the emission data (which is not specified) would be needed for discussion and interpretation.
The choice of measurement sites reflects the scale and desired use of the model and availability of measurement data. In the context of global scales, where a global PAH budget is the main desired outcome, background sites before more meaningful. In the context of human inhalation exposure, where the estimate of population exposure is the main desired outcome, regional to urban scales are of greater interest for estimating those exposures and urban sites are more relevant than background sites. Although the global distribution and budget of PAHs have been the focus of other studies, our goal was to develop a model that is capable of simulating PAH concentrations at locations and spatial scales relevant to human inhalation exposure. In the North American context, those locations are generally urban areas where the impact of local emissions is the highest and where concentrations are also highest and sometimes exceed health-based guidelines. Moreover, only a small number of PAH measurement stations are available for North America and many of them are located in urban centres. We therefore feel that an analysis using urban to regional stations is very appropriate

although any conclusions based on the use of these measurement stations must bear in mind station representativeness and point-grid cell incommensurability.

Spatial gradients in such locations are expected and simulating them should be a goal of a regional PAH model. In our analysis, we have found that measured concentrations within urban grid squares vary by roughly a factor of two. Ours is the first study to present quantitative data to suggest that even a regional model grid is insufficiently fine for the simulation of urban-scale PAH concentrations. Nonetheless, our model reproduces measured concentrations to the correct order of magnitude.

Moreover, for the nominal horizontal grid spacing used in this study (42 km independent of map scaling factor), the Nyquist wavelength will be 84 km and hence the nominal resolvable spatial feature will be 4 x 42 km = 168 km (e.g., Grasso, Bull. Amer. Meteor. Soc., 81, 579-580, 2000). As shown in Galarneau et al. (2007), there are pronounced regional spatial gradients in the distribution of North American PAH emissions. These emissions gradients will contribute an important fraction of the spatial variability of measured concentrations and the model grid resolution employed in this study will be capable of representing such regional spatial gradients on the order of 150 km or greater.

No known major aerosol components have been excluded from our dynamic aerosol sub-model. Major inorganic ions (sulphate, nitrate, ammonium), carbonaceous components (elemental carbon, primary and secondary organic matter), crustal material, sea salt and particle-bound water have all been included.

The impacts of assuming equilibrium partitioning are difficult to assess due to the time scales of the currently available measurement data. The available measurements are for 24 h averages whereas the time scale of mass transfer has been shown to be minutes to hours. This is a factor that we discuss in our upcoming companion paper about partitioning.

The emissions fields used in the model are discussed in the work referenced in the submitted manuscript (Galarneau et al., 2007). The temporal factors assumed for the suite of different PAH-emitting processes are the best available at the current time. The temporal profile of the emissions varies from source to source and the examination of this factor would lead to a study in its own right. The following text has been added to Section 2.2 to clarify this point.
“As discussed in Galarneau et al. (2007), hourly PAH emissions fields were estimated with an emissions processing system using source-specific temporal profiles. The temporal profile library included 3020 month-of-year, 64 day-of-week, and 2672 hour-of-day temporal profiles for Canada and 1500, 49, and 680 analogous temporal profiles for the US. The overall temporal profile thus varies from grid cell to grid cell due to the different mixtures of source types found in each one.”

Specific comments and minor corrections:
Introduction, p 18419: It should be mentioned that benzo(a)pyrene is a criteria pollutant in many countries, regulated under a regional UN convention (Arhus protocol to the CLRTAP Convention) and discussed in the context of a global one (UNEP Stockholm or POPs Convention). As to the study region itself: please complement by information on respective policy in Mexico

We have added text to the first paragraph of the Introduction to elaborate the international importance of PAHs.

Although the northern portion of Mexico is included in our study region, no estimates of Mexican PAH emissions were available for this study (see Galarneau et al., 2007). As a result, we have not discussed this country in any detail in our Introduction.

l 27: also box model applications (Yaffe et al. Risk Analysis 2001; Prevedouros et al. Chemosphere 2004) should be cited; should read 'Shatalov et al.'; this group's modelling, however, has been advanced and Gusev et al. 2011 (Persistent Organic Pollutants in the Environment. EMEP Status Report No. 3/2011, Meteorological Synthesizing Centre – East, Moscow) would be more appropriate to be quoted; Sehili and Lammel's modelling was global; other models in use should be quoted, too: Zhang et al. Atmos. Environ. 2011, 45, 2820-7.
Thank you. We have added citations to our Introduction as suggested. Prevedouros et al. (2004) and Zhang et al. (2011) were included in our original manuscript.

p 18420 ll 19-21: Be more quantitative. Semivolatiles are compounds with partial pressures in the range 1e-6 – 1e-2 Pa at 298K. Rephrase avoiding 'use'. ll 24-26: Not true as several partitioning models have been tested by Aulinger et al. 2007 and Lammel et al 2009.
We prefer definitions of semivolatile that are operational, especially given the wide range of particle/gas partitioning that is found at different ambient temperatures encountered over an annual cycle in temperate regions.

We are the only study to have examined PAH particle/gas partitioning in a paired modelled vs. measured data comparison framework. We have clarified this point in the manuscript.

p 18421, l 8: be more concise, avoid 'usually', introduce and reference aerosol submodel used
The aerosol submodel used is described in Gong et al. (2006) and in more detail in Gong et al. (2003a,b). Section 2.1 of the paper has been modified to add this information.

p 18422, l 24-24: unclear which kinetics: first order or second order ? remove 'equilibrium' as misleading here
Kinetics are first order as specified on line 24. We have removed the term "equilibrium" as requested.

p 18423, ll 11-15: trivial, drop
We prefer to leave this text as is. Many workers in the field are more familiar with SOA partitioning and we wanted to specify that this different approach taken with PAHs is applicable to other compounds as well.

p 18424, ll 1-2: 0.172 J/m2 was suggested by Junge, 1977, as default for all condensable vapours, but 1.71 J/m_2 bei Pankow, 1987 (using other units), for PAHs, which should be preferably used. Here a justification is needed for the selection of Junge's value for the 'constant' and a discussion of the influence of choice of this parameter.

We selected Junge's original value of c because Pankow's 1987 analysis was based on only two measurement datasets (Yamasaki et al., 1982; Bidleman et al. 1986) and a single estimated value of 4E-10 moles of sorption sites per cm3 of aerosol. The value of c has little effect at the extreme ranges of vapour pressure for these PAHs though particulate fraction is sensitive to the value of c for mid-volatility species. Our simulations of particulate fraction did not show a pattern consistent with the value of c being a causal factor in poor simulation of particulate fraction. Instead, our simulations showed the worst model performance for the lowest molecular weight compounds which are insensitive to the value of c. Nevertheless, we agree that our choice for this factor should be noted in the manuscript and we have added text to section 2.1.4 to this effect.

p 18425, l 2: K_aw with temperature dependence (according to Table S1.1), should be noted here
We have noted that K_{AW} values are temperature-dependent in the revised manuscript.

p 18425, ll 10-13: trivial, drop
We prefer to leave this sample calculation in the manuscript. We believe that this calculation might be difficult to follow for a reader who is less familiar with this subject and that giving a short example might be helpful to them.

p 18426, section 2.2: the emission's temporal pattern, spatial resolution, and major limitations (which emissions were neglected) should be explained
The reader is referred to Galarneau et al. (2007) in Section 2.2 for a thorough discussion of the emission inventory and accompanying emissions processing, including spatial and temporal allocation. As noted above, a few sentences have been added to Section 2.2 to describe the temporal allocation step.

l 21: better 'observational data'
We have changed this in our revised manuscript as requested.

section 2.3: number of sites, site type and sampling protocol (e.g. 24 h, were oxidant denuders used or not?) should be mentioned here
Considerable information about the measurement networks and observational data is provided in the Supplement to this paper. Oxidant denuders were not used at any sites as they were not standard in any North American monitoring programs at that time. Nor are they now. This has been made explicit in Section 3.1.1 of the revised manuscript.

p 18427 l 16: 'particulate' rather than 'particle'
We have made this change in the revised manuscript as requested.

p 18428 ll 15-16: be quantitative here: What was the temporal emission pattern like ? Should be discussed in the light of the emissions' temporal resolution
We apologize for the lack of clarity. We meant to state that location-specific temporal profile of emissions, which are developed using multiple disaggregation factors applied to annual emissions figures for different source types to arrive at month-of-year, day-of-week, and time-of-day emissions, likely yields less variation in emissions than is seen in reality. Text has been added to Section 2.2 of the revised manuscript to describe the temporal allocation of emissions.

ll 17-19: agreement between partitioning models is very surprising and not plausible regarding that other modelling studies using a model appropriate for semivolatiles with

a dynamic aerosol sub-model found very significant differences (e.g. Lammel et al 2009)
We have checked our results and confirmed that our two parametrizations yielded similar partitioning results. We suspect that this is due to the values of the constants used and the underestimation of particle mass by the model. This topic is explored in our partitioning manuscript that is currently in preparation.

ll 17-24: very deficient discussion: How do the differences look like for the entire domain, and at the subset of background sites ? The model resolution (42km) is such that polluted sites cannot be compared with model output, as the gradients can never be reproduced. The comparison between predicted and observed values in this manuscript is strongly biased by the inclusion of a number of urban or industrial site, it seems (cf. p 18430 ll 9-10; which is nowhere told, though). Try for background stations only and discuss these results.
See above for a discussion on the relative importance of background versus urban sites for model evaluation and the impact of Nyquist wavelength on the resolution of spatial gradients. Major urban areas in North America have spatial scales that are not much smaller than our grid spacing. For instance, the city of Toronto is covered by 2 grid squares in the model and the three measurement sites in that city differ in their PAH concentrations by less than a factor of 2. Though we agree because of our analysis that a finer resolution might improve urban results (indeed, that is one of our conclusions), we do not agree that our study is invalid in such areas and would argue that the results demonstrate why comparisons in urban locations are necessary for estimating the impact of PAHs on human health. As seen in Figure 4, the model performance does not differ much between sites. No consistent pattern is seen in performance for urban/industrial vs. rural sites.

p 18429 l 9: Is it applicable yes or no ? Losses occur unless oxidant denuders are used, which, nowadays, is the case in many networks.
Yes, we believe that this comment is applicable. The North American networks did not employ oxidant denuders for PAH monitoring during the period considered. As noted above, a comment has been added to Section 3.1.1 to make this explicit.

ll 13-15: Very severe deficiency of the model, renders is unsuitable to simulate fate of semivolatiles. Re-volatilisation is expectedly very significant for the cycling of e.g. fluoranthene and anthracene. Air-surface exchange needs to be included in the processes covered by the model. Why was no appropriate model chosen (e.g. Gong et al Atmos Chem Phys 2007) ?
Please see our earlier comments about revolatilisation and the differences between PCBs and PAHs. Our results present ambiguous evidence that suggests that revolatilisation MAY be needed in the model. It is not unequivocal.

We have added text to Section 2.2 of our revised manuscript to make it clear that volatilization has not been considered in this study. We have also added text to this Section (3.1.1) to clarify that the model spatial scale and the observational data locations used in the model's evaluation can lead to perceived differences in the relative importance of atmospheric fate processes such as air-surface exchange of PAHs, noting that many of our observation locations are in urban areas and noting also that volatilization may play a greater role in non-urban locations.

ll 24-25: conclusion is not justified as the results are biased by the selection of sites

We have added text to clarify the role of model spatial scale and observational data locations as described previously.

p 18430 ll 9-20: The whole paragraph is not conclusive without a site and emissions' temporal patterns description (which both is not there). Obviously are sites included which are strongly influenced by local sources (see above). Otherwise such large ranges would not occur.

The sites are indeed strongly influenced by local sources. This is one of the reasons for using a regional rather than a global model for this work. We have clarified the statement by adding text to note again the context for our work: the importance of providing model estimates of PAHs in regions where human exposure is high which are usually close to emissions sources.

p 18431 l 3: number of sites should be introduced in section 2.3. Here a number is given which differs from the one in section 4. Why?

A number of sites, specifically from the California Air Resources Board (CARB) network, included measurements only for benzo[a]pyrene. This is described in the Supplement. A short explanation has been added for clarification.

p 18432 ll 3-10: should be discussed in the light of type of site and model spatial and emission' temporal resolutions

This paragraph was intended to describe the sites in question and notes whether they are rural or urban, and for the former, details the presence of a major point source emitter. We would welcome the opportunity to add further detail not included here or in subsequent lines 15-24 if directed by the referee.

ll 20-22: not true, horizontal concentration gradients within urban areas can be expected even for 24h samples as a consequence of relative position of sites to major sources in the area (subject to wind direction)

The stated example is of a single major point source, a large smelter located in an otherwise wilderness area.

ll 27-29: the effective sink process for ozone by reaction with NOx should be mentioned/ discussed, too, as the list probably contains sites which are directly influenced by road traffic emissions (?). Then, the characteristic time for ozone formation is not relevant for its temporal variation.

Yes, some of the urban sites are affected by road traffic. Ozone levels close to major roadways will be affected by NO_x titration, whereas those immediately outside the urban regions may have enhanced O_3 variation. This spatial variation supports the use of resolutions higher than global for simulating PAHs in human inhalation exposure regions. We have mentioned the spatial variation of NO_x impacts on ozone in the revised manuscript in Section 3.1.3.

p 18433 ll 3-13: quite trivial and, in particular, known a priori. Hence, move to introduction or drop section 3.2: should be discussed in the light of predicted atmospheric lifetimes (nowhere given)

We disagree with the referee's statement. This result is not known a priori in that we are the first to support the statement with quantitative results.

As mentioned above, our paper doesn't not have a global PAH budget or long-range transport as its primary focus. Rather, we focus on simulating concentrations in areas where these pollutants are relevant to human health, e.g., cities.

p 18434 ll 5-15: Performance of partitioning models should be discussed in the light of the aerosol sub-models performance. Was it evaluated ? What is the range of organic carbon fractions and aerosol surface predicted by the model for various type of sites?
As noted above, the performance of the AURAMS model's predictions of size- and chemically-resolved particulate matter have been assessed in previously published literature (e.g., McKeen e tal., 2008; Smyth et al., 2009; Kelly et al., 2012; Solazzo et al., 2012) where its performance has been found to be similar to other regional, peer-reviewed models in the literature.

p 18434, l 23: be more concise, replace 'volatility' by the quantity addressed
Since volatility is VP for J-P and KOA and KSA for D-E, we used the generic term. These quantities are directly related to each other. For example, VP and KOA are perfectly correlated for PAHs.

p 18436 ll 4-27: almost no conclusions in this text (except ll 20-22), drop or move to other section
The Conclusions section has been modified to focus more on the main results of the study vis-à-vis human exposure estimates using the model and to identify key areas for future model improvements.
"This study described the first known modelling results for atmospheric PAHs at the regional scale over North America. Predictions from the AURAMS-PAH model were compared to roughly 5,000 24-hour-average PAH measurements from 45 sites, eight of which also provided data on particle/gas partitioning which had been modelled using two different partitioning schemes.

The evaluation of the model is key to determining its potential utility as an input for estimating the impacts of PAH inhalation exposure on human health. Annual average modelled total (gas + particle) concentrations were statistically indistinguishable from measured values for fluoranthene, pyrene and benz[a]anthracene, indicating the model's potential utility for providing inputs to health impact estimation for these species. The model annual average concentrations for phenanthrene, anthracene and chrysene+triphenylene were biased low. For these species, the negative bias would have to be considered if used as inputs to human health impact estimates as the model in its present form underestimates long-term concentrations.

The utility of the model for prediction purposes may also be considered on a day-to-day basis though this is less relevant to the chronic health effects associated with carcinogenic PAHs. The model simulated total PAH concentrations to the correct order of magnitude 64-86% of the time. That level of accuracy must be considered when assessing human health impacts; annual exposure estimates are likely of more utility with the model in its current state.

The partitioning approach chosen did not have a significant impact on the model results for total concentrations though differences resulting from the choice of parametrization approached the 95% significance level for benzo[a]pyrene. At this time, neither of the two approaches used here provided a clear advantage for simulation accuracy of total concentrations.

As a first work of this nature, the analysis has suggested several avenues for further model development and improvement. Improved temporal emissions estimates for PAHs are key to improving model simulations of these species; simulated PAHs showed less temporal variability than the measurements. The reactions of particulate PAH species with atmospheric oxidants should given further consideration since the more reactive species were overestimated in the current model. Model resolution has been shown to be a key factor in improving air pollution estimates in areas with high human exposures. While the 42-km horizontal grid spacing used in this study is finer than that used in global models, it was insufficient to capture the distribution of concentrations in densely populated areas. A more detailed analysis of the factors influencing modelled particle/gas partitioning is needed to improve the distribution of PAHs between the gas and particle phases in the atmosphere given that both partitioning schemes used here showed increasing negative biases for particle-bound PAH concentrations of increasing volatility."

l 17: be more concise, avoid 'majority'
We have replaced the term "majority" with quantitative data in the revised manuscript (see Conclusion text above).

ll 23-24 a priori knowledge, drop or move to other section
This was not known a priori. It was suspected, perhaps, but never quantified before this study.

p 18436 ll 1-2: yes: preliminary character of this communication
We have called this a "first-generation" model but we believe that our results are sufficiently useful to report to the scientific community and may help give direction for ongoing research.

Table 1 caption: n should be explained.
n = number of model-measurement data pairs. We have added this as a footnote to the table.

Fig. 2 is it a mix of urban, residential/urban background and continental background stations ?
The observational data sites are predominantly urban/industrial though some regional background sites are included as well.

Fig. 5 caption: the word 'ratio' is missing (as in caption of Fig 4)
Corrected as noted.

Table S1.1: No reference is given for OH degradation rate coefficients. Is it estimated using the EPIsuite model ? Experimentally determined rate coefficients should be used whenever available. The OH degradation rate coefficients for anthracene were determined higher (190e-12) and for fluoranthene lower (11e-12; ; Brubaker and Hites, 1998) than used here.

We are grateful that the reviewer has asked this question since it has made us realize that we mistakenly used an outdated version of this table. For the final model runs described in the manuscript, the k_{OH} values for PHEN, ANTH and FLRT were taken from Brubaker and Hites (1998) at values of 2.7E-11, 1.9E-10 and 1.1E-11 cm^3 $molec^{-1}$ s^{-1}, respectively. We used the ANTH value for BaP as well in light of BaP's noted reactivity though the gas phase reactivity has little effect on the fate of this predominantly particulate PAH.