Reply to Alexandre Albinet comments

Author's response in Blue

Line 42-43: Cite : Ravindra, K., Sokhi, R. and Van Grieken, R.: Atmospheric polycyclic aromatic hydrocarbons: Source attribution, emission factors and regulation, Atmospheric Environment, 42, 2895–2921, doi:doi: DOI: 10.1016/j.atmosenv.2007.12.010, 2008.

Reference added

Line 52: Ringuet, J., Albinet, A., Leoz-Garziandia, E., Budzinski, H. and Villenave, E.: Reactivity of polycyclic aromatic compounds (PAHs, NPAHs and OPAHs) adsorbed on natural aerosol particles exposed to atmospheric oxidants, Atmospheric Environment, 61, 15–22, doi:10.1016/j.atmosenv.2012.07.025, 2012.

Reference added

Line 53-54:

-Nalin, F., Golly, B., Besombes, J.-L., Pelletier, C., Aujay-Plouzeau, R., Verlhac, S., Dermigny, A., Fievet, A., Karoski, N., Dubois, P., Collet, S., Favez, O. and Albinet, A.: Fast oxidation processes from emission to ambient air introduction of aerosol emitted by residential log wood stoves, Atmospheric Environment, 143, 15–26, doi:10.1016/j.atmosenv.2016.08.002, 2016.

-W.F. Rogge, L.M. Hildemann, M.A. Mazurek, G.R. Cass, B.R.T. Simoneit, Sources of fine organic aerosol. 2. Noncatalyst and catalyst-equipped automobiles and heavy-duty diesel trucks Environ. Sci. Technol., 27 (1993), pp. 636-651

-B. Zielinska, J. Sagebiel, J.D. McDonald, K. Whitney, D.R. Lawson. Emission rates and comparative chemical composition from selected in-use diesel and gasoline-fueled Vehicles J. Air Waste Manag. Assoc., 54 (2004), pp. 1138-1150

-W.F. Rogge, L.M. Hildemann, M.A. Mazurek, G.R. Cass, B.R.T. Simoneit Sources of fine organic aerosol. 9. Pine, oak and synthetic log combustion in residential fireplaces Environ. Sci. Technol., 32 (1998), pp. 13-2

Two references added (Nalin et al., 2016; Rogge et al., 1993)

Line 61: Tomaz, S., Shahpoury, P., Jaffrezo, J.-L., Lammel, G., Perraudin, E., Villenave, E. and Albinet, A.: One-year study of polycyclic aromatic compounds at an urban site in Grenoble (France): Seasonal variations, gas/particle partitioning and cancer risk estimation, Science of The Total Environment, 565, 1071–1083, doi:10.1016/j.scitotenv.2016.05.137, 2016.

Reference added

Line 62:

Please explain which limitations. Sampling artifacts, etc...

Have a look there and references included:

-Albinet, A., Leoz-Garziandia, E., Budzinski, H. and ViIlenave, E.: Sampling precautions for the measurement of nitrated polycyclic aromatic hydrocarbons in ambient air, Atmospheric Environment, 41(23), 4988–4994, doi:16/j.atmosenv.2007.01.061, 2007.

-Albinet, A., Papaiconomou, N., Estager, J., Suptil, J. and Besombes, J.-L.: A new ozone denuder for aerosol sampling based on an ionic liquid coating, Analytical and Bioanalytical Chemistry, 396, 857–864, doi:10.1007/s00216-009-3243-5, 2009.

-Goriaux, M., Jourdain, B., Temime, B., Besombes, J.-L., Marchand, N., Albinet, A., Leoz-Garziandia, E. and Wortham, H.: Field Comparison of Particulate PAH Measurements Using a Low-Flow Denuder Device and Conventional Sampling Systems, Environ. Sci. Technol., 40(20), 6398–6404, doi:10.1021/es060544m, 2006.

References added and text corrected. More details added to ‘‘error evaluation’’ please see below author’s comments on Line 156.

Author’s changes in manuscript (L.62).

"However, a long sampling and averaging period creates some limitations such as sampling artefacts, notably where changing atmospheric photolysis conditions (air humidity, temperature, wind direction, ozone or other oxidant concentrations) may have a significant influence on PAHs concentrations and oxidation rates (Albinet et al., 2007b; Albinet et al., 2009, Goriaux et al., 2006, Tsapakis and Stephanou., 2003; Tsapakis and Stephanou., 2007, Ringuet et al., 2012b)."

Line 67: please cite also: Srivastava, D., Favez, O., Bonnaire, N., Lucarelli, F., Haeffelin, M., Perraudin, E., Gros, V., Villenave, E. and Albinet, A.: Speciation of organic fractions does matter for aerosol source apportionment. Part 2: Intensive short-term campaign in the Paris area (France), Science of The Total Environment, 634, 267–278, doi:10.1016/j.scitotenv.2018.03.296, 2018.
Reference added

Line 67-69: and other ones such as:
-Reisen, F. and Arey, J.: Atmospheric reactions influence seasonal PAH and nitro-PAH concentrations in the Los Angeles basin, Environ. Sci. Technol., 39(1), 64–73, doi:10.1021/es035454l, 2004.
-Albinet, A., Leoz-Garziandia, E., Budzinski, H. and ViIlenave, E.: Polycyclic aromatic hydrocarbons (PAHs), nitrated PAHs and oxygenated PAHs in ambient air of the Marseilles area (South of France): Concentrations and sources, Science of The Total Environment, 384(1-3), 280–292, doi:10.1016/j.scitotenv.2007.04.028, 2007.
-Albinet, A., Leoz-Garziandia, E., Budzinski, H., Villenave, E. and Jaffrezo, J.-L.: Nitrated and oxygenated derivatives of polycyclic aromatic hydrocarbons in the ambient air of two French alpine valleys: Part 1: Concentrations, sources and gas/particle partitioning, Atmospheric Environment, 42(1), 43–54, doi:10.1016/j.atmosenv.2007.10.009, 2008.
-Srivastava, D., Favez, O., Bonnaire, N., Lucarelli, F., Haeffelin, M., Perraudin, E., Gros, V., Villenave, E. and Albinet, A.: Speciation of organic fractions does matter for aerosol source apportionment. Part 2: Intensive short-term campaign in the Paris area (France), Science of The Total Environment, 634, 267–278, doi:10.1016/j.scitotenv.2018.03.296, 2018.
Three references added (Albinet et al., 2008; Reisen and Arey., 2004; Srivastava et al., 2018.) text modified.
Author’s changes in manuscript (L.67).
A few studies have used twice daily (12 h) sampling (Albinet et al., 2008; Zhang et al., 2018; Farren et al., 2015; Ringuet et al., 2012b), obtaining limited information on variability in concentrations during the daytime and night-time (Tsapakis and Stephanou., 2007). Shorter time periods for sampling (3 h and 4 h) are still very limited (Reisen and Arey., 2004; Srivastava et al., 2018).

Line 99: clean I guess
and that's not only the purpose, see:
-Albinet, A., Leoz-Garziandia, E., Budzinski, H. and ViIlenave, E.: Simultaneous analysis of oxygenated and nitrated polycyclic aromatic hydrocarbons on standard reference material 1649a (urban dust) and on natural ambient air samples by gas chromatography-mass spectrometry with negative ion chemical ionisation, Journal of Chromatography A, 1121(1), 106–113, doi:16/j.chroma.2006.04.043, 2006.
-Albinet, A., Nalin, F., Tomaz, S., Beaumont, J. and Lestremau, F.: A simple QuEChERS-like extraction approach for molecular chemical characterization of organic aerosols: application to nitrated and oxygenated PAH derivatives (NPAH and OPAH) quantified by GC–NICIMS, Anal Bioanal Chem, 406(13), 3131–3148, doi:10.1007/s00216-014-7760-5, 2014.
Author’s changes in manuscript (L.99).

All samples and blanks were purified on a SPE silica normal phase cartridge (1g/6ml; SIGMA ALDRICH) to reduce the impacts of interfering compounds in the matrix and to help maintain a clean GC injection inlet liner.

Line 126: You can say that it's a modified version of previous published methods:
-Albinet, A., Leoz-Garziandia, E., Budzinski, H. and ViIlenave, E.: Simultaneous analysis of oxygenated and nitrated polycyclic aromatic hydrocarbons on standard reference material 1649a (urban dust) and on natural ambient air samples by gas chromatography-mass spectrometry with negative ion chemical ionisation, Journal of Chromatography A, 1121(1), 106–113, doi:16/j.chroma.2006.04.043, 2006.
-Albinet, A., Nalin, F., Tomaz, S., Beaumont, J. and Lestremau, F.: A simple QuEChERS-like extraction approach for molecular chemical characterization of organic aerosols: application to nitrated and oxygenated PAH derivatives (NPAH and OPAH) quantified by GC–NICIMS, Anal Bioanal Chem, 406(13), 3131–3148, doi:10.1007/s00216-014-7760-5, 2014.
-Kawanaka Y, Sakamoto K, Wang N, Yun S-J (2007) Simple and sensitive method for determination of nitrated polycyclic aromatic hydrocarbons in diesel exhaust particles by gas chromatography-negative ion chemical ionisation tandem mass spectrometry. J Chromatogr A 1163:312–317
-Bezabeh DZ, Bamford HA, Schantz MM, Wise SA (2003) Determination of nitrated polycyclic aromatic hydrocarbons in diesel particulate-related standard reference materials by using gas chromatography/mass spectrometry with negative ion chemical ionization. Anal Bioanal Chem 375:381–388
References added and text corrected
Author’s changes in manuscript (L.126).
The method development for OPAHs and NPAHs was based on previous studies (Albinet et al., 2006; Albinet et al., 2014; Bezabeh et al., 2003; Kawanaka et al., 2007) and conducted using Negative Chemical Ionisation

Line 126: 155 eV, really?
Yes 155 ev (autotune settings), According to Agilent manual the maximum can be used in NCI is 240 ev.

Line 156-157:
Clearly I doubt about it. If you really want to consider all the possible errors, you should applied a GUM approach. https://sisu.ut.ee/measurement/9-iso-gum-modeling-approach-bottom-approach
In addition, to estimate the uncertainties, one of the main factor is linked to the extraction efficiency and you don't mention anything about that. Standard reference materials are useful for that (for both PAHs and PAH derivatives). There are many paper about that:
-Albinet, A., Leoz-Garziandia, E., Budzinski, H. and ViIlenave, E.: Simultaneous analysis of oxygenated and nitrated polycyclic aromatic hydrocarbons on standard reference material 1649a (urban dust) and on natural ambient air samples by gas chromatography-mass spectrometry with negative ion chemical ionisation, Journal of Chromatography A, 1121(1), 106–113, doi:16/j.chroma.2006.04.043, 2006.
-Albinet, A., Nalin, F., Tomaz, S., Beaumont, J. and Lestremau, F.: A simple QuEChERS-like extraction approach for molecular chemical characterization of organic aerosols: application to nitrated and oxygenated PAH derivatives (NPAH and OPAH) quantified by GC–NICIMS, Anal Bioanal Chem, 406(13), 3131–3148, doi:10.1007/s00216-014-7760-5, 2014.
-Schantz MM, McGaw E, Wise SA (2012) Pressurized liquid extraction of diesel and air particulate standard reference materials: effect of extraction temperature and pressure. Anal Chem 84:8222–8231
-Wise S, Poster D, Kucklick J, Keller J, VanderPol S, Sander L, Schantz M (2006) Standard reference materials (SRMs) for determination of organic contaminants in environmental samples. Anal Bioanal Chem 386:1153–1190

-Delgado-Saborit JM, Alam MS, Godri Pollitt KJ, Stark C, Harrison RM (2013) Analysis of atmospheric concentrations of quinones and polycyclic aromatic hydrocarbons in vapour and particulate phases. Atmos Environ 77:974–982
-Nocun M, Schantz M (2013) Determination of selected oxygenated polycyclic aromatic hydrocarbons (oxy-PAHs) in diesel and air particulate matter standard reference materials (SRMs). Anal Bioanal Chem 405:5583–5593
-Ahmed, T. M., Bergvall, C., Åberg, M. and Westerholm, R.: Determination of oxygenated and native polycyclic aromatic hydrocarbons in urban dust and diesel particulate matter standard reference materials using pressurized liquid extraction and LC–GC/MS, Anal Bioanal Chem, 1–12, doi:10.1007/s00216-014-8304-8, 2014.

The estimated error (L.157) is due to available information on laboratory test performance. The word ‘‘uncertainty’’ (L.157) was replaced by ‘‘error’’.
Based on our type of data we chose the ‘‘top-down’’ approach where the bias determination can be based on recovery efficiency. Using the step-by-step approach (bottom up) will increase the uncertainty on individual compound and mostly used for ISO accreditation. It is shown in the text that the average recovery efficiencies have ranged from 85% to 96% (L.102). The %RSD average for deuterium labelled compounds was about 3.6% (L.149). In this study we spiked 10 filters covering the different sampling time (3h, 9h, 15h). We agree that SRM are widely used for a better assessment of the analytical method and their use is probably required for publishing in analytical technique Journals. SRM do not provide certified values for most of the derivatives compounds we have used.

Author’s changes in manuscript (L.156).
Therefore, the overall estimated error, combining the precision and the systematic effects, is less than 20%.
Another source of error can be attributed to sampling artefacts and this has been discussed in previous studies (Schauer, C. et al., 2003, Goriaux, M. et al., 2006, Tsapakis and Stephanou, 2003). The absence of an ozone denuder to trap the gas phase oxidants may lead to an underestimation of the true values of PAHs due to chemical decomposition. Therefore, data from long sampling times and under high ozone ambient concentrations may be biased by sampling artefacts by more than 100 % (Schauer et al., 2003, Goriaux et al., 2006). However, at low ozone levels, negative artefacts were considered not significant (Tsapakis and Stephanou 2003), whilst, at medium ozone levels (30-50 ppb), PAHs values were underestimated by 30 % (Schauer et al., 2003). In addition, heterogeneous reactions during particles sampling may occur on the monolayer surface coverage only with limited diffusion of oxidants to the bulk particles (Keyte et al., 2013 and references therein). Previous studies reported that the formation of NPAHs during high-volume sampling is not significant and calculated to be < 3 % (Arey et al., 1988) and < 0.1 % (Dimashki et al., 2000).
Considering the predominant role of ozone levels (below 30 ppb in this study, mean: 8.4 ppb), sampling time and temperature on the magnitude of PAHs concentrations, the estimation of the negative sampling artefacts on our data range between 10 and 20 %, with the highest error estimation attributable to longer sampling time (15h).

Line 159: lab or field blanks? How many blanks?
Line 160: if they are detected they are not <LOD
Line 161: how much?
Text corrected
Author’s changes in manuscript (L.159,160,161).
To determine any sources of contamination during sample preparation and the analytical procedure, the solvent (acetonitrile) and field blanks (n=2) were analysed following the same procedure as for the samples (Extraction, SPE, Evaporation). Most target compounds were found to be below LOD (S/N=3) or orders of magnitude (up to 10^3- 10^4) lower than was found in the samples. A small number of compounds found in field blanks (1,8-Naphthalic anhydride, Benzo[a]fluorenone, 1-Nitronaphthalene, 9-Nitroanthracene) have a higher contribution (4-30 %) to very few filters (2 to 5 samples) collected

over a 3 h time period, if this was co-incident with low particulate loading conditions. The contribution to each compound from field blanks has been corrected in the final data.

Line 218: All this part should be moderated or removed.
There are clearly evidences that the validity of this kind of ratios is limited
-Dvorská, A., Lammel, G. and Klánová, J.: Use of diagnostic ratios for studying source apportionment and reactivity of ambient polycyclic aromatic hydrocarbons over Central Europe, Atmospheric Environment, 45(2), 420–427, doi:10.1016/j.atmosenv.2010.09.063, 2011.
-Dvorská, A., Komprdová, K., Lammel, G., Klánová, J. and Plachá, H.: Polycyclic aromatic hydrocarbons in background air in central Europe – Seasonal levels and limitations for source apportionment, Atmospheric Environment, 46, 147–154, doi:10.1016/j.atmosenv.2011.10.007, 2012.
-Katsoyiannis, A., Sweetman, A. J. and Jones, K. C.: PAH molecular diagnostic ratios applied to atmospheric sources: a critical evaluation using two decades of source inventory and air concentration data from the UK, Environ. Sci. Technol., 45(20), 8897–8906, doi:10.1021/es202277u, 2011.
-Katsoyiannis, A. and Breivik, K.: Model-based evaluation of the use of polycyclic aromatic hydrocarbons molecular diagnostic ratios as a source identification tool, Environmental Pollution, 184, 488–494, doi:10.1016/j.envpol.2013.09.028, 2014.
Limitations for source apportionment using Diagnostic Ratios (DR) increase when the receptor site is far from evident emission sources, e.g. long-time sampling (24h) or by using volatile and reactive compounds such as the ratio ANT/(ANT+PHE) which indicate petrogenic sources. This ratio has not been used in this study and the sampling time scale was short (3h during daytime) which reduce limitations. In addition, the site location (Fig S1) was at urban area and surrounded by busy traffic road, residential buildings, underground, restaurants and thermal power stations.
A recent study (Feng et al., 2019) has used PMF models and DR and showed consistent results from both methods.
Accordingly, we consider the obtained results from diagnostic ratios are representative and may be important for future studies and method comparison.
Reference:
Feng, B., Li, L., Xu, H., Wang, T., Wu, R., Chen, J., Zhang, Y., Liu, S., Ho, S.S.H., Cao, J., Huang, W.: $PM_{2.5}$-bound polycyclic aromatic hydrocarbons (PAHs) in Beijing: Seasonal variations, sources, and risk assessment, Journal. of Environmental. Sciences., 77, 11-19, doi:10.1016/j.jes.2017.12.025, 2019.

Line 246 please cite actual references which showed that, e.g.
-I.J. Keyte, A. Albinet, R.M. Harrison.On-road traffic emissions of polycyclic aromatic hydrocarbons and their oxy- and nitro- derivative compounds measured in road tunnel environments.Sci. Total Environ., 566–567 (2016), pp. 1131-1142
-J.K. Schulte, J.R. Fox, A.P. Oron, T.V. Larson, C.D. Simpson, M. Paulsen, N. Beaudet, J.D. Kaufman, S. Magzamen. Neighborhood-scale spatial models of diesel exhaust concentration profile using 1-nitropyrene and other nitroarenes.Environ. Sci. Technol., 49 (2015), pp. 13422-13430
-B. Zielinska, J. Sagebiel, W.P. Arnott, C.F. Rogers, K.E. Kelly, D.A. Wagner, J.S. Lighty, A.F. Sarofim, G. Palmer Phase and size distribution of polycyclic aromatic hydrocarbons in diesel and gasoline vehicle emissions Environ. Sci. Technol., 38 (2004), pp. 2557-2567
-B. Zielinska, J. Sagebiel, J.D. McDonald, K. Whitney, D.R. Lawson. Emission rates and comparative chemical composition from selected in-use diesel and gasoline-fueled vehicles. J. Air Waste Manage. Assoc., 54 (2004), pp. 1138-1150.
Two references added (Keyte et al., 2016, Magzamen et al., 2015)

Line 252: Tomaz, S., Jaffrezo, J.-L., Favez, O., Perraudin, E., Villenave, E. and Albinet, A.: Sources and atmospheric chemistry of oxy- and nitro-PAHs in the ambient air of Grenoble (France), Atmospheric Environment, 161, 144–154, doi:10.1016/j.atmosenv.2017.04.042, 2017.
Reference added

Line 312-313: OK but ANT is mainly in the gas phase and you have data only about the PM phase

As it was shown in the text (L. 299-300) the dominant formation pathway of NPAHs is secondary formation in the gas phase. Heterogeneous reactions may play a role and contribute to the formation of NPAHs. 9-nitroanhtracene was reported to be one of the major products for the heterogeneous reaction of adsorbed anthracene on different types of particles (NaCl, SiO_2, MgO) in presence of NO_2 (Jinzhu et al., 2011; Wenyuan and Tong., 2014).

Zhang et al, (2013) have reported that ANT in the gas phase can be adsorbed on particles and they showed a high production of 9-NANT during the heterogeneous reaction of adsorbed ANT in presence of O_3-NO, suggesting that NO_3 radicals were formed and reacted with anthracene. In our study the positive strong correlation between ANT and 9-NANT in particle phase does not prove but it supports the heterogeneous formation pathway of 9-NANT. On the other hand, previous studies reported that the formation of NPAHs during high-volume sampling is not significant and calculated to be < 3% (Arey et al., 1988) and < 0.1 % (Dimashki et al., 2000). The mechanisms and levels of formation are still far from being fully understood, especially that the reaction of PAHs with oxidants is highly influenced by surface coverage and PAHs loading (Keyte, I.J., 2013 and references therein) i.e. particles were shown to exhibit a potential inhibiting factor on the reactivity of PAHs due to slow diffusion of oxidants and inaccessibility of PAHs in the bulk particle.

The gas phase reaction of ANT with NO_3 radicals and the formation of 9-NANT either in the gas phase or adsorbed on particles still unclear. Future studies might help us to better understand the chemical transformation of anthracene in the atmosphere, this suggest that probably both pathways contribute to the formation of 9-NANT on particles. Accordingly, this explain why 9-NANT is the most abundant NPAH in this study and in parallel makes ANT one the lowest concentrations in particle phase.

References:
Jinzhu. M. et al., Atmospheric Environment 45, 917-924, 2011.
Wenyuan, C. et al., Environ. Sci. Technol., 48, 8671–8678, doi:10.1021/es501543g, 2014.
Yang, Z. et al., Journal of Environmental Sciences, 25, 1817–1823, 2013.
Arey, J. et al., Env. Sci. Tech, 22, 457-462, 1988
Dimashki, M. et al., Atmospheric Environment 34, 2459-2469, 2000.

Line 318: For all this part you should cite the previous articles where this approach has been described and used:

-A. Albinet, E. Leoz-Garziandia, H. Budzinski, E. Villenave, J.-L. Jaffrezo Nitrated and oxygenated derivatives of polycyclic aromatic hydrocarbons in the ambient air of two French alpine valleys: part 1: concentrations, sources and gas/particle partitioning Atmos. Environ., 42 (2008), pp. 43-54,

-B.A.M. Bandowe, H. Meusel, R. Huang, K. Ho, J. Cao, T. Hoffmann, W. Wilcke PM2.5-bound oxygenated PAHs, nitro-PAHs and parent-PAHs from the atmosphere of a Chinese megacity: seasonal variation, sources and cancer risk assessment Sci. Total Environ., 473-474 (2014), pp. 77-87, 10.1016/j.scitotenv.2013.11.108

-W. Huang, B. Huang, X. Bi, Q. Lin, M. Liu, Z. Ren, G. Zhang, X. Wang, G. Sheng, J. Fu. Emission of PAHs, nitro-PAHs and oxy-PAHs, from residential honeycomb coal briquette combustion. Energy Fuel, 28 (2013), pp. 636-642, 10.1021/ef401901d

-Tomaz, S., Shahpoury, P., Jaffrezo, J.-L., Lammel, G., Perraudin, E., Villenave, E. and Albinet, A.: One-year study of polycyclic aromatic compounds at an urban site in Grenoble (France): Seasonal variations, gas/particle partitioning and cancer risk estimation, Science of The Total Environment, 565, 1071–1083, doi:10.1016/j.scitotenv.2016.05.137, 2016.

Two references added (Albinet et al., 2008; Tomaz et al., 2016), Bandowe et al. 2014 already cited.

Line 321-322: That are not the references showing that

References moved beside "commonly used"

Author's changes in manuscript (L.321-322)

For PAHs, Benzo[a]pyrene was chosen as the reference chemical because it is known as the most carcinogenic PAH (OEHHA., 1994, 2002) and is commonly used (Albinet et al., 2008; Tomaz et al., 2016; Alves et al., 2017; Bandowe et al., 2014; Ramírez et al., 2011) as an indicator of carcinogenicity of total PAHs.

Line 336-337: Please specify in a Table which TEF values have benne used and the references

The toxicity Equivalency Factor (TEFs) and references used for individual PAHs, OPAHs and Nitro-PAHs are shown in supporting information on Table S3.

Line 375-379:

-Albinet, A., Leoz-Garziandia, E., Budzinski, H., Villenave, E. and Jaffrezo, J.-L.: Nitrated and oxygenated derivatives of polycyclic aromatic hydrocarbons in the ambient air of two French alpine valleys: Part 1: Concentrations, sources and gas/particle partitioning, Atmospheric Environment, 42(1), 43–54, doi:10.1016/j.atmosenv.2007.10.009, 2008.

-Tomaz, S., Shahpoury, P., Jaffrezo, J.-L., Lammel, G., Perraudin, E., Villenave, E. and Albinet, A.: One-year study of polycyclic aromatic compounds at an urban site in Grenoble (France): Seasonal variations, gas/particle partitioning and cancer risk estimation, Science of The Total Environment, 565, 1071–1083, doi:10.1016/j.scitotenv.2016.05.137, 2016.

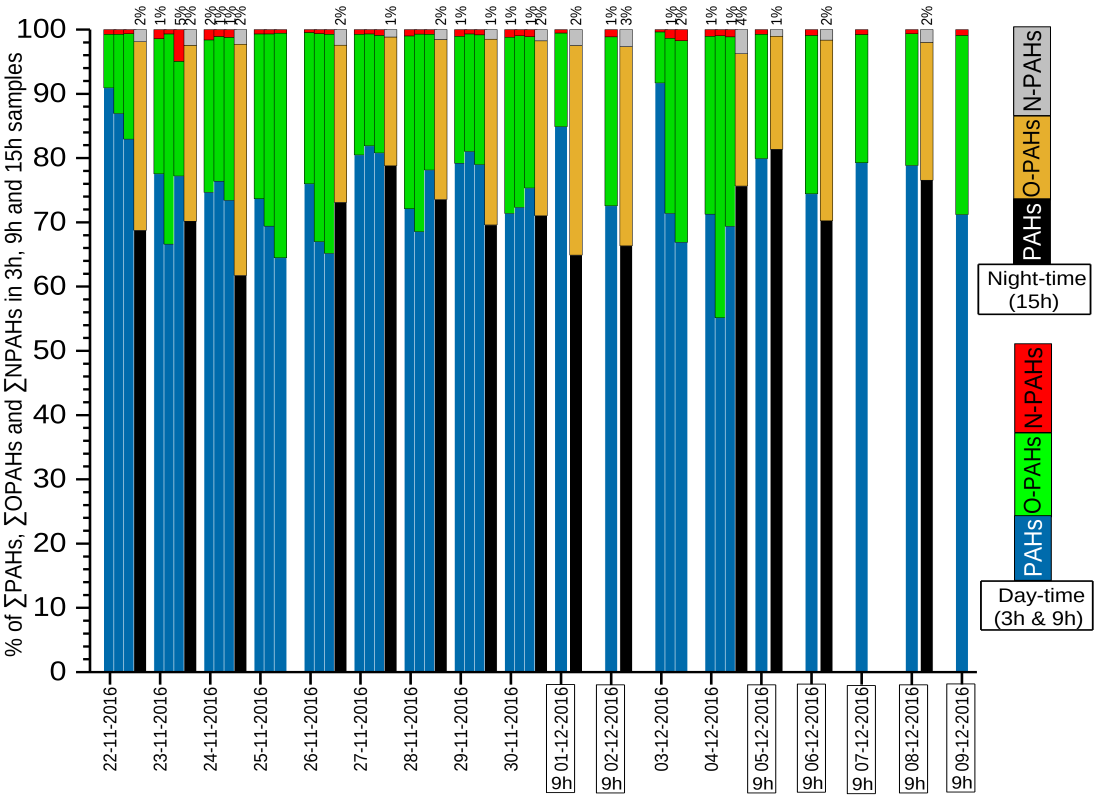
Tomaz et al., 2016 cited. Text modified.

Author's changes in manuscript (L.375)

Our results were considerably higher than those estimated for western European cities during the winter, such as Grenoble: 1.4 ng m^{-3} (Tomaz et al., 2016), Oporto: 3.56 ng m^{-3}, Florence: 1.39 ng m^{-3} and Athens: 0.43 ng m^{-3} (Alves et al., 2017). ECR values estimated for each city were 31 (Grenoble), 6.6 (Oporto), 17 (Florence) and 54 (Athens) times lower than our ECR estimation.

Line 672: Figure 2 Really difficult to read

Slightly modified and re-arranged. Please see below


% of ∑PAHs, ∑OPAHs and ∑NPAHs in 3h, 9h and 15h samples
0
10
20
30
40
50
60
70
80
90
100
2%
1%
5%
2%
2%
1%
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22-11-2016
23-11-2016
24-11-2016
25-11-2016
26-11-2016
27-11-2016
28-11-2016
29-11-2016
30-11-2016
01-12-2016
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9h
PAHs
O-PAHs
N-PAHs
Night-time
(15h)
PAHs
O-PAHs
N-PAHs
Day-time
(3h & 9h)