The impact of European legislation on the reduction of Environmental Tobacco Smoke and Polycyclic Aromatic Hydrocarbons between 1995 and 2019 in Germany

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Introduction

Environmental tobacco smoke (ETS)

- Complex mixture of more than 4500 compounds in the vapor phase and the particulate matter comprising amongst others tobacco alkaloids, amines, alkanes, numerous polycyclic aromatic hydrocarbons (PAHs), aldehydes, carbonyls, and metals
- Described to exhibit adverse health effects and some of its compounds are described to be toxic (e.g., acrolein) or carcinogenic (e.g., benzene)
- Growing awareness of health impairment from ETS required the need for comprehensive legislation in order to protect non-smokers from exposure
- Regulations and legislative amendments in Germany since the early 2000's, e.g. first workplace smoking bans (ArbStättV, 2002), tobacco tax increase, smoking ban in federal facilities and public transport (2007)

Human Biomonitoring (HBM)

Human biomonitoring as ideal tool to …

… assess the **actual exposure** at individual and population level by determining a set of specific biomarkers

..assess the **effects of these regulatory actions** on human exposure

… tailor further **pollutant reduction measures**

Objective

Evaluation whether enforced **non-smoking regulations and smoking bans affected the ETS exposure** of the general population in Germany since their implementation in 2002 until today.

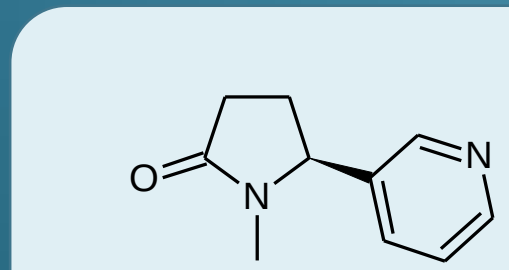
Biomarkers to assess ETS exposure

Cotinine

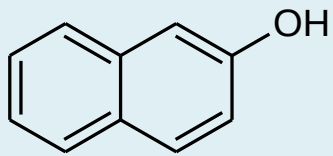
Major metabolite of nicotine
Exposure source: consumption of nicotine containing products, i.a. smoking

Monohydroxylated polycyclic aromatic hydrocarbons (OH-PAHs)

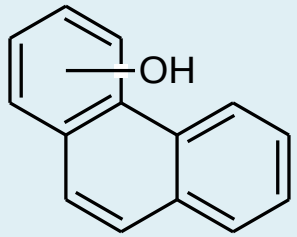
Metabolites of PAHs
Exposure source: vehicle exhaust, tobacco smoke, smoke from woodstoves and fireplaces, consumption of processed food (especially fried, grilled or smoked food)



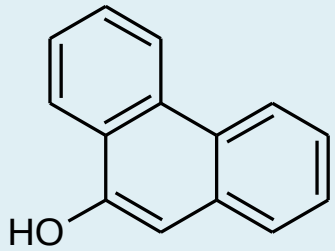
Cotinine



2-Naphthol

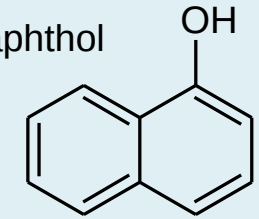


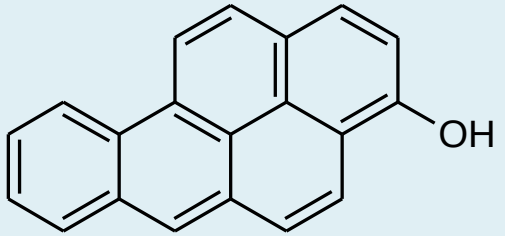
1-/2-/3-/4-OH-Phenanthrene



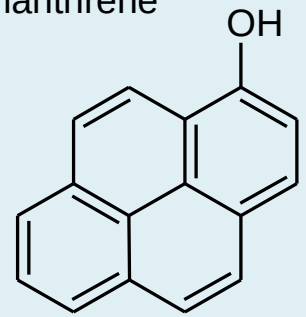
9-OH-Phenanthrene

1-Naphthol





3-Hydroxybenzo[a]pyrene



1-OH-Pyrene

Study Design

24-h-urine samples collected, stored in the Environmental Specimen Bank (**ESB**) and provided by the German Environmental Agency (Umweltbundesamt, **UBA**)

Cotinine determination according to Landmesser et al. (2019)

- Liquid-liquid extraction with ethyl acetate followed by LC-MS/MS analysis
- Calibration range: 0.2-1,000 µg/L

3-OH-BaP determination according to Rögner et al. (2021)

- LC-MS/MS after enzymatic hydrolysis and SPE
- Calibration range: 50-3,321 pg/L

OH-PAHs determination according to Ramsauer et al. (2011)

- LC-MS/MS after enzymatic hydrolysis and SPE
- Calibration range: 0.05-150 µg/L for 1-, 2-Nap,
  0.001-2.0 µg/L for 1-/2-/3-/4-/9-OH-Phe
  0.01-2.0 µg/L for 1-OH-Pyr

Study years:

1995 | 2000 | 2005 | 2010 | 2014 | 2015 | 2017 | 2018 | 2019

Male to female split 50/50

Exclusion of smokers: self-reporting, cotinine levels > 15 µg/L[1]

⇩

Evaluated study samples: 510

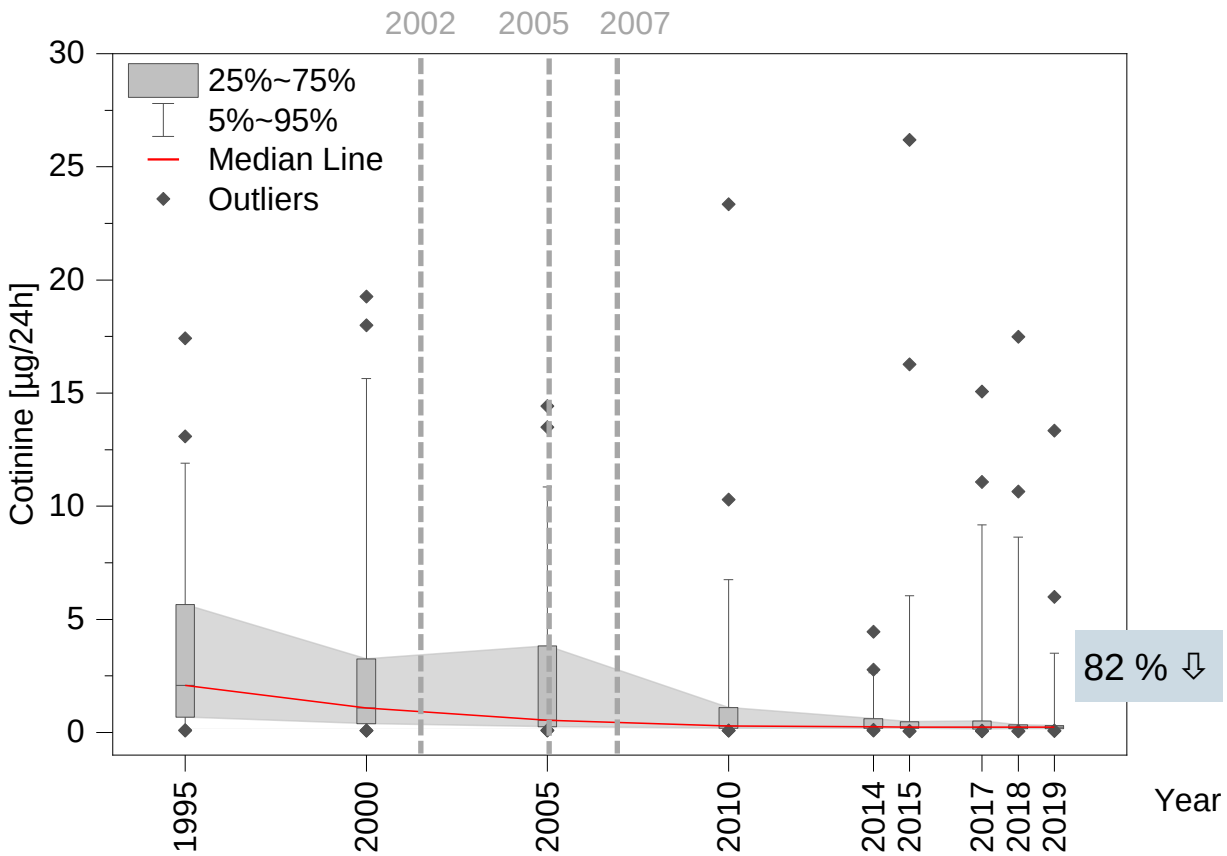
Normalization: 24-h-urine volume

Normal distribution: Shapiro-Wilk & D'Agostino-K squared test

Significance: Mann-Whitney U test, α = 0.05

[1]Benowitz, N. L., et al., 2009. Urine nicotine metabolite concentrations in relation to plasma cotinine during low-level nicotine exposure. Nicotine and Tobacco Research. 11, 954-60.

Cotinine



Legislation

2002: Regulation to ban smoking in the workplace (ArbStättV)

2002-2005: Significant tax increase

2005: Restricted advertising and sponsorship of tobacco products (Directive 2003/33/EC)

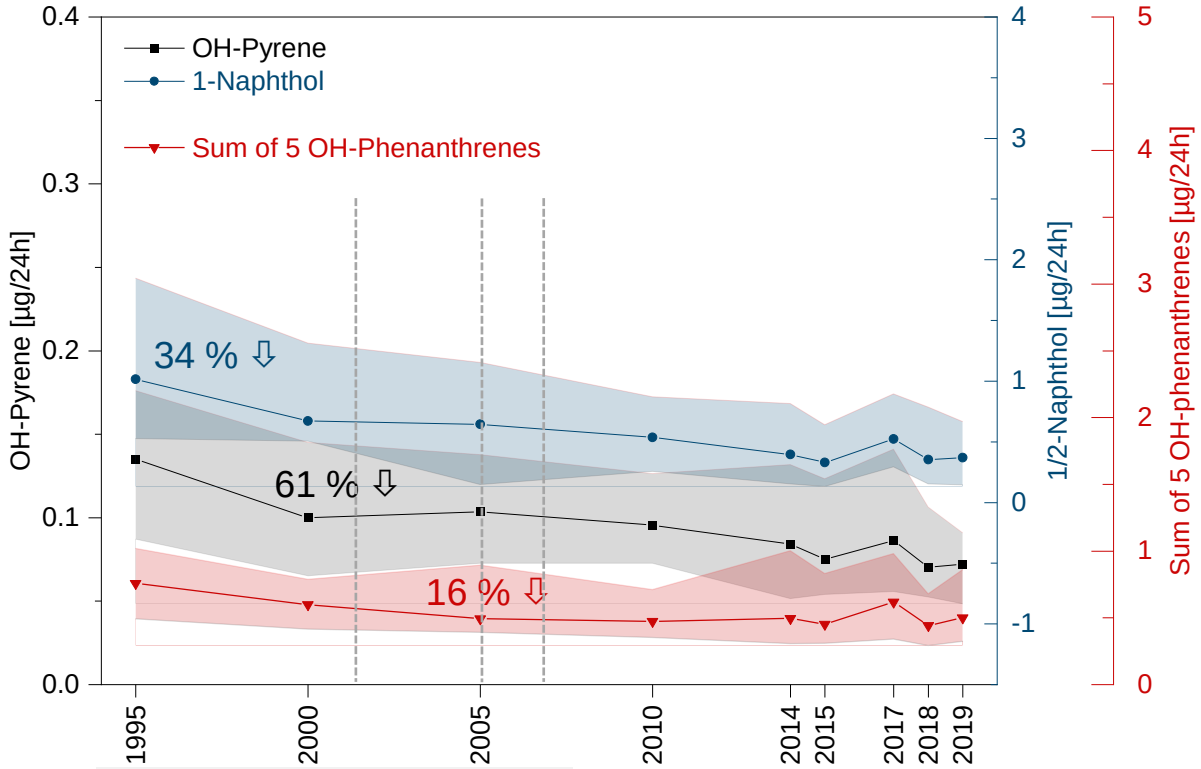
2007: Federal and State Laws for Protection from the Hazards of Passive Smoking, e.g. ban smoking in gastronomy, federal facilities, and public transport

Reduced smoking prevalence additionally contributes to reductions in ETS exposure

- additional tobacco tax increases
- prohibition of advertising for cigarette products
- sales ban to youths <18 years
- warning on tobacco products

Successful legislation minimized exposure of the non-smoking population from ETS

OH-PAHs



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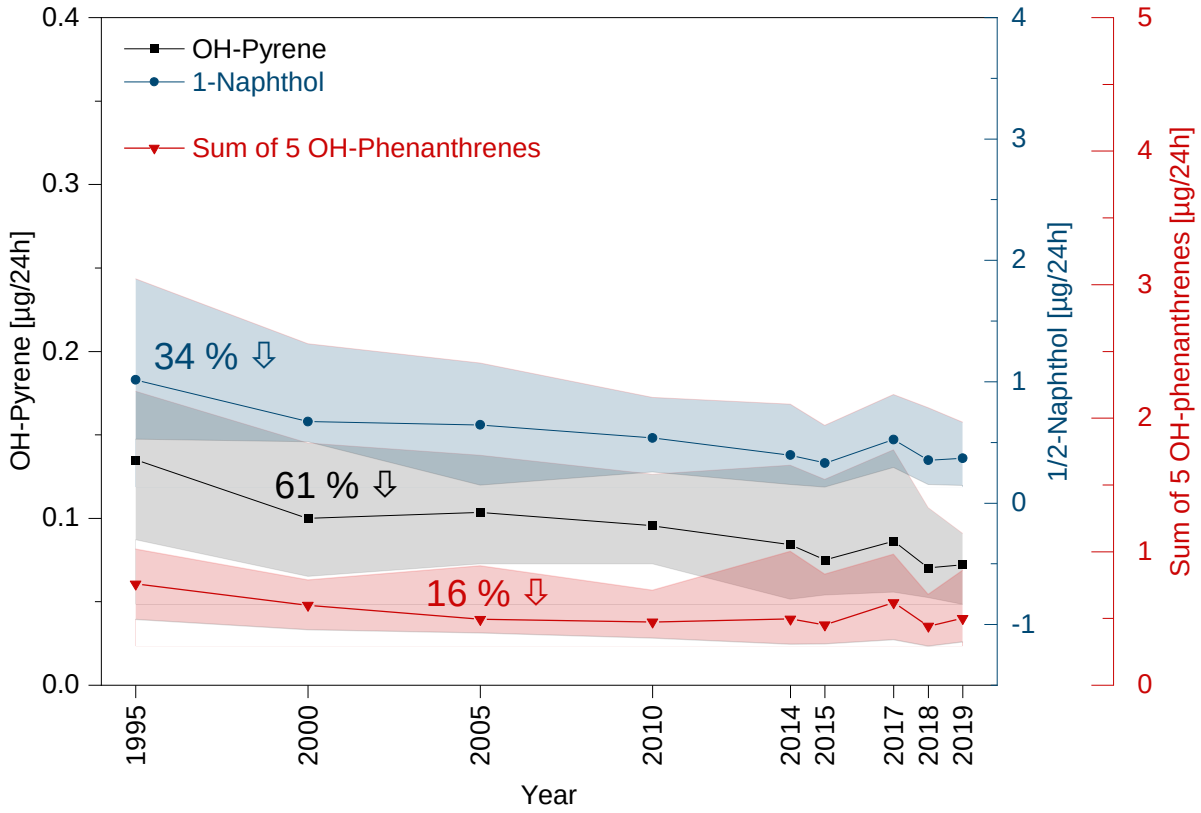
Significant decrease observed for **OH-Pyrene, 1-Naphthol, 1-OH-, 2-OH-, 3-OH-phenanthrene**

Correlation with cotinine

	Spearman r	p-value
1-OH-Pyr	0.38	2.2E-19
1-Nap	0.45	1.8E-26
1-OH-Phe	0.27	7.8E-10
2-OH-Phe	0.37	3.5E-18
3-OH-Phe	0.41	1.3E-22
4-OH-Phe	0.17	8.1E-05
9-OH-Phe	0.27	6.6E-10
Sum of 1-/2-/3-/4-/9-OH-Phe	0.31	5.3E-13

Correlation of OH-PAH excretion and ETS exposure

OH-PAHs



Confounding factors

- Indoor air pollution evoked by burning domestic fuels for heating or cooking, and traffic emissions infiltrating indoor air
- Ambient air from industries and traffic
- Intake of processed food (e.g., fried, smoked, barbequed food)

Legislation

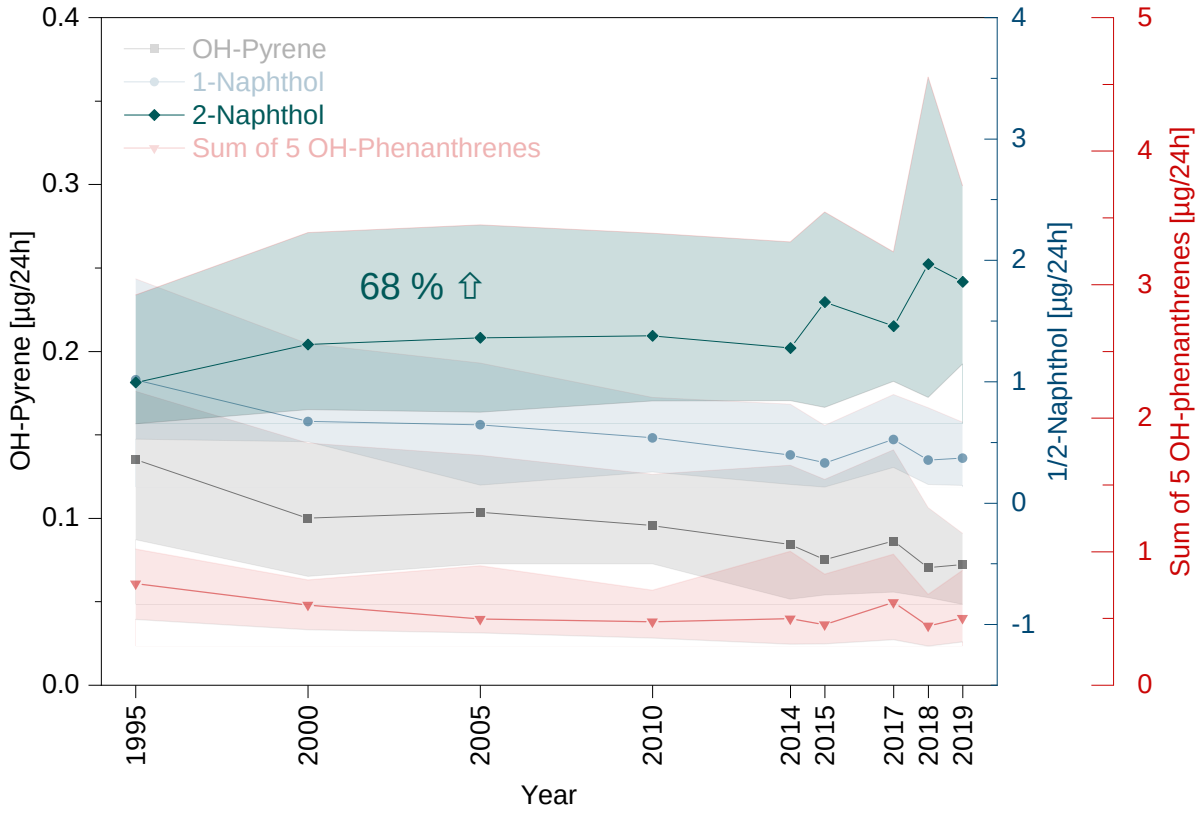
2001: EU regulations to minimize PAH emission from combustion plants and industrial installations (2001/80/EC)

2004: EU regulations to minimize environmental PAH input to media air, soil, and water (850/2004/EC)

2005: EU regulations specifying maximum levels of PAHs in air (2004/107/EC)

2006/2011: EU regulations specifying maximum levels of PAHs in foodstuff (1881/2006/EC, 835/2011/EU)

2-Naphthol



Reasons for the trend are under discussion:

- Different routes on naphthalene metabolism based on level of exposure (Thai et al., 2020, Scherer et al., 2022)
- Increase of exposure specifically to 2-Naphthol or compounds specifically metabolizing to 2-Naphthol

Increase also observed in the U.S. (National Health and Nutrition Examination Survey, NHANES) and Canadian population (Canadian Health Measures Survey, CHMS)

Indication for an increased exposure to chemicals of concerns ?

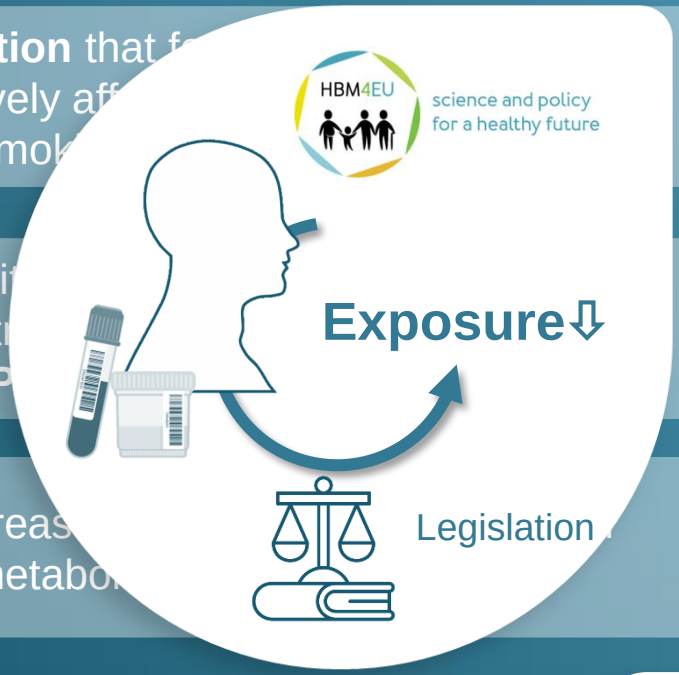
Results underline the importance and necessity of human biomonitoring! !

Thai, P. K., et al., 2020. Analysis of urinary metabolites of polycyclic aromatic hydrocarbons and cotinine in pooled urine samples to determine the exposure to PAHs in an Australian population. Environmental Research. 182.
Scherer, G., et al., 2022. Assessment of the exposure to polycyclic aromatic hydrocarbons in users of various tobacco/nicotine products by suitable urinary biomarkers. Archives of Toxicology.

Conclusion

First HBM assessing the urinary excretion of Cot and nine different OH-PAHs, including 3-OH-BaP, in Germany over a time period of 24 years

Study underlines the **importance of nationwide HBM** to monitor the exposure to chemicals of concern and to **assess the effects of regulatory measures** with respect to public health as well as Europe wide HBM to cover also exposure by transboundary air pollution.



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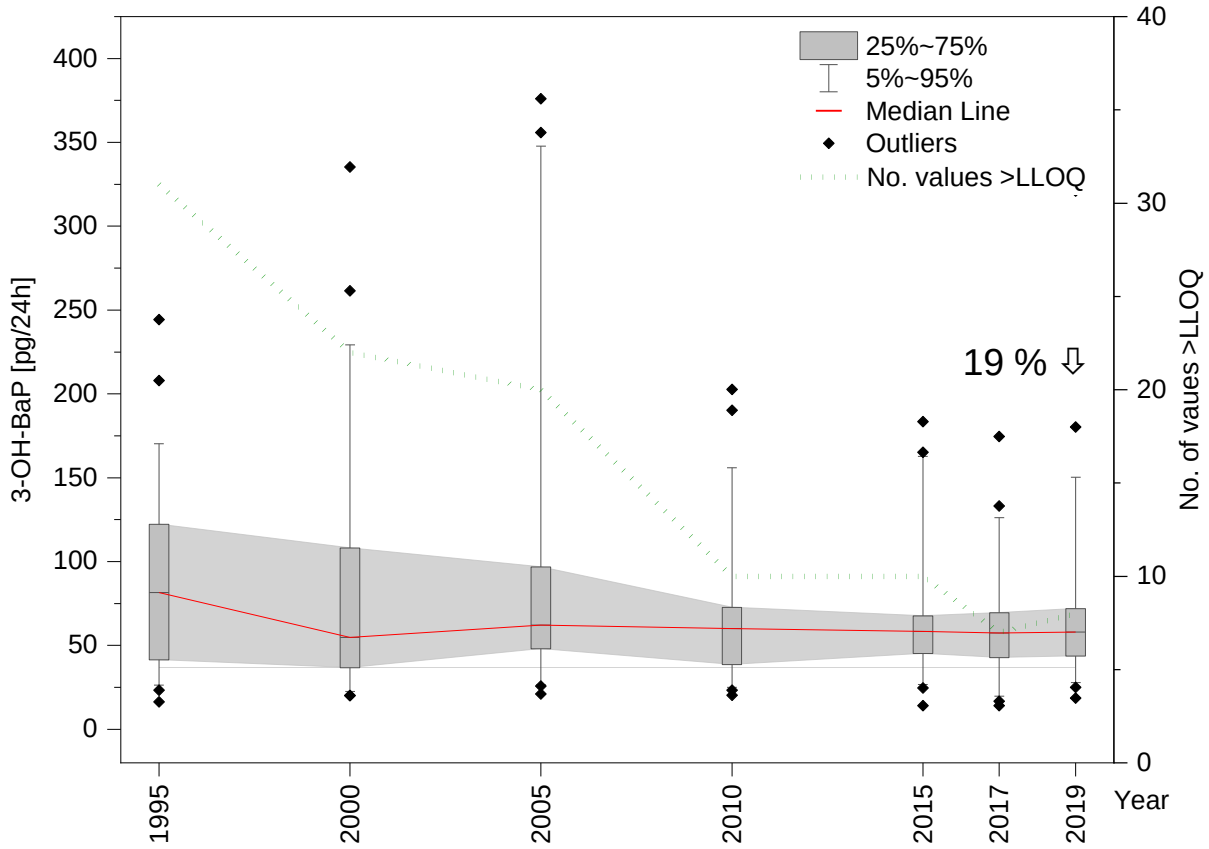
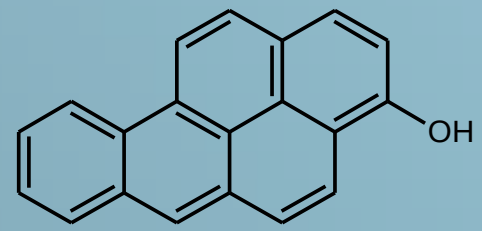
3-OH-BaP





3-Hydroxybenzo[a]pyrene

- First HBM study systematically assessing the urinary excretion of 3-OH-BaP in Germany
- Only 28 % of the 24-h urine samples > LLOQ (50 pg/L)
- 19 % decrease of 3-OH-BaP from 1995 until 2019