BACKGROUND // JANUARY 2016

Polycyclic Aromatic Hydrocarbons

Harmful to the Environment! Toxic! Inevitable?

German Environment Agency

Umwelt Bundesamt

Imprint

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Publications as a pdf:
www.umweltbundesamt.de/en/publikationen/polycyclic-aromatic-hydrocarbons

Photo credits:
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As at Januar 2016

ISSN: 2363-829X

Inhalt

1. Polycyclic aromatic hydrocarbons – problem chemicals?

Whether in mouse pads, toys, or bathing shoes – polycyclic aromatic hydrocarbons (PAHs) are ubiquitous. Most frequently, non-branded products at affordable prices get negative attention in this respect, but products from renowned companies may contain PAHs as well. We encounter PAHs as atmospheric pollutants produced by small combustion units (such as fireplaces and stoves in homes), traffic, industrial processes, and tobacco smoke. They can also be detected in foodstuffs, especially at summertime BBQ events or in smoked products.

What are polycyclic aromatic hydrocarbons and what risk do they represent? Why do we find PAHs time and again in objects of everyday use, and why do we encounter them on a daily basis? What is done to address the problem, and what can each individual do?

The German Environment Agency (UBA) would like to answer these and other questions in this publication.

2. Where do PAHs come from?

PAHs are produced in incomplete combustion of organic matter such as wood, coal, or oil. As a general rule, the lower the temperature of the fire and the less oxygen is available, the more incomplete do these materials burn and the more PAHs are produced. A large portion of the PAHs enters the atmosphere through natural processes that cannot be controlled by humans, such as wood fires or volcano eruptions. Man-made emissions mainly come from combustion processes as well: from small combustion units, industrial processes, fireplaces, or tobacco smoke. In addition, this group of substances is a natural component of fossil raw materials, specifically coal and petroleum. The latter contains between 0.2 % and 7 % PAHs (National Research Council, 2003). Refining processes[1] such as coking for coal and cracking for petroleum generate products such as coke, tar, petrols, waxes, or oils.

The slags generated in these processes are incinerated or used as a construction material in road building. If PAHs are not removed from slag or from coke oven and refinery products, they will enter the environment due to their persistence. Tar oils and specific oils from petroleum refining can be added as softeners to rubbers and plastics. The largest portion of the PAHs that reach consumers comes from these applications.

1 These processes treat a raw material at high temperatures, pressures under exclusion of air and/or in the presence of catalysts. All these reactions are incomplete since there is no sufficient amount of oxygen. The final products are gaseous and liquid distillation products; coking also produces solid residues that are rich in carbon.

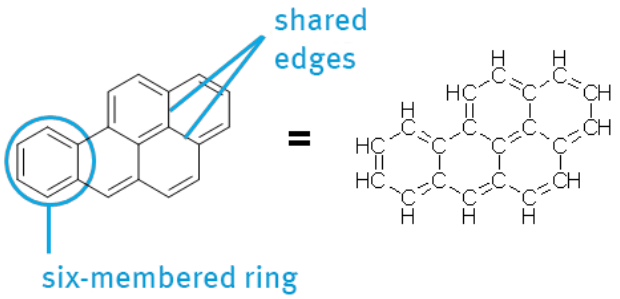
Also toys can contain polycyclic aromatic hydrocarbons (PAHs). Foto: © debramillet / www.fotolia.com

What are PAHs – the chemistry of a problem group of substances

The group of PAHs includes all compounds that are composed of two to seven rings of carbon and hydrogen atoms (see Figure 1).

Figure 1

The structure of PAHs, using the example of benzo[a]pyren
shown on the right with carbon and hydrogen atoms



Most of these rings consist of six carbon-hydrogen units and are joined along shared edges. The ring system results in a special array of electrons chemists call "aromatic", which is responsible for the special chemical properties of this group of substances. In addition to the variable structure of the ring system, the molecules may carry various side chains instead of hydrogen atoms. This group of substances is therefore very big and includes an estimated 10,000 compounds. The individual compounds often have very similar characteristics. PAHs almost always occur as mixtures due to the way they are generated. PAHs are solid at room temperature and strongly bind to soot, soil, or dust particles.

The properties of the individual PAHs depend on the number of hydrocarbon rings: PAHs are generally lipophilic, which means they dissolve poorly in water but well in fats and oils. This tendency increases with a growing number of rings, i.e. the more rings are present, the more fat-soluble is the substance and the better it accumulates in the fatty tissue of organisms.

3. Why are PAHs such a concern?

PAHs are an alarming group of substances for humans and environmental organisms. Many PAHs are carcinogenic, mutagenic and/or toxic for reproduction (Crone and Tolstoy, 2010). Some PAHs are at the same time persistent, bioaccumulative, and toxic for humans and other organisms. Persistent means that the substances remain in the environment for a long time and are hardly decomposed there. Bioaccumulative chemicals accumulate in organisms – including the human body.

Substances that combine these three characteristics represent a particular level of concern under an environmental aspect. Experts speak of PBT substances in this context (Persistent, Bioaccumulative, and Toxic substances). If such chemicals are released, they can no longer be removed from the environment due to their characteristics. On the contrary: They accumulate and can harm plants, animals, and ultimately humans.

PAHs bind to dust and soot particles and enter the atmosphere in this way as well. They can be transported over long distances there due to their persistence. The PAH-containing dusts return to the surface of the earth via rain, fog, or snow, are deposited on soil and on plants, and enter surface waters. Since PAHs can be transported to remote areas of the earth, they are also found far away from human sources of entry, such as in remote mountain lakes (Quiroz et al., 2010), in the Arctic and Antarctic regions.

Many PAHs always occur as variable mixtures. This is why often "representatives" of a group of substances are determined in chemical analyses. In 1977, the U.S. Environmental Protection Agency (EPA) added 16 PAHs to the list of "priority pollutants" of the U.S. Clean Water Act (nine of which are listed in Table 1). These 16 PAHs were selected because they are highly toxic and easily chemically detectable, include a wide range of potential structures and were frequently found in waters. Chemists mostly measure the sum total of these 16 compounds to determine the PAH content of products. Benzo[a]pyrene serves as the lead compound, which means it is considered representative of all other PAHs.

The idea is that PAHs always occur in mixtures: If benzo[a]pyrene is contained in a substance or product, this typically applies to all other PAHs of concern, which have very similar properties. Benzo[a]pyrene was selected because this compound is particularly carcinogenic. However, there has also been criticism of this reduction of the problem: For example, the Panel on Contaminants in the Food Chain at the European Food Safety Authority (EFSA) submitted a report on PAHs in foodstuffs in 2008 in which they conclude that benzo[a]pyrene alone is not a suitable marker for the presence of PAHs in foodstuffs. According to EFSA, a combination of four specific PAHs is best suited as an indicator of PAH content in foodstuffs. These are benzo(a)pyrene, benz(a)anthracene, benzo(b)fluoroanthene, and chrysene.

Many PAHs enter the atmosphere through combustion processes and thus spread across wide areas. Photograph: © Fumes Rybson / www.sxc.hu

Table 1

Some select PAHs and their properties

Health hazard!  **Environmental hazard!**  **Warning:** 

Name (CAS No.)	Melting point in °C	Boiling point in °C	Molecular formula	Hazard designation	Hazard symbol
Benz[a] anthracene (56-55-3)	160	435	$C_{18}H_{12}$	Hazard: May cause cancer, very toxic to aquatic organisms, with long-term effect	
Benzo[b]fluoroanthene (205-99-2)	168	481	$C_{20}H_{12}$	Hazard: May cause cancer, very toxic to aquatic organisms, with long-term effect	
Benzo[j]fluoroanthene (205-82-3)	166	480	$C_{20}H_{12}$	Hazard: May cause cancer, very toxic to aquatic organisms, with long-term effect	
Benzo[k]fluoroanthene (207-08-9)	217	481	$C_{20}H_{12}$	Hazard: May cause cancer, very toxic to aquatic organisms, with long-term effect	
Benzo[a] pyrene (50-32-8)	175	495	$C_{20}H_{12}$	Hazard: May cause cancer, may cause genetic defects, impair fertility, and cause harm to the unborn child, very toxic to aquatic organisms, with long-term effect	
Benzo[e] pyrene (192-97-2)	178	493	$C_{20}H_{12}$	Hazard: May cause cancer, very toxic to aquatic organisms, with long-term effect	
Chrysene (218-01-9)	255	448	$C_{18}H_{12}$	Hazard: May cause cancer, very toxic to aquatic organisms, with long-term effect	
Dibenz[a,h] anthracene (53-70-3)	267	524	$C_{22}H_{14}$	Hazard: May cause cancer, very toxic to aquatic organisms, with long-term effect	
Naphthaline (91-20-3)	80.5	218	$C_{10}H_{8}$	Warning: May presumably cause cancer, harmful if swallowed, very toxic to aquatic organisms, with long-term effect	

4. The path of PAHs into the environment and to the consumer

PAHs can enter the environment and reach consumers in manifold ways. Not all paths described here are relevant for the emission situation in Germany because the input of PAHs into the air differs based on a country's level of economic development. The main sources of PAHs in economically less developed countries include domestic combustion of wood, coal, or straw as well as forest and steppe fires. Emissions

from coal combustion for power generation are predominant in emerging countries. Air pollution by PAHs in industrialized nations mainly comes from small combustion units in households.

Production, transport, and processing of petroleum and coal

Large quantities of PAHs can be released into the environment by leakages or accidents in the extraction, transport, or refinery of petroleum. The environmental load remains local when oil is spilled into the soil. Entire ecosystems collapse if large quantities of oil are spilled into rivers or lakes, and the load from PAHs contained in petroleum contributes to such collapse. The Niger Delta region in Nigeria, the most populous country in Africa, is a particularly dramatic example. Large international oil companies have operated drilling rigs here since 1958. Environmental experts estimate that about 1.5 million tons of petroleum have inadvertently been released into the environment until 2006 (FME Nigeria et al., 2006). This is equivalent to about 3,000 to 105,000 tons of toxic PAHs. The adverse effects on the nature and humans are dramatic.

Accidental oil spills into the seas also destroy ecosystems in vast areas. In 2010, BP's oil rig Deepwater Horizon exploded in the Gulf of Mexico and caused an oil slick. An estimated 600,000 tons of oil were spilled into the sea (Crone and Tolstoy, 2010). This is equivalent to about 1,200 to 45,000 tons of highly toxic PAHs that will harm the Gulf of Mexico for decades.

In coal mining, PAHs mainly enter the environment through dusts. The material in stock piles and tailings also contains PAHs and pollutes soil, waters, and the groundwater.

When coal is processed in coking plants, PAHs can enter the environment through exhaust gases and waste water from processes. The soil and groundwater on the premises of former gas works and coking plants are often highly contaminated with PAHs, especially around tar pits and the areas where coal and waste were stored. Extensive protective measures or an expensive remediation of contaminated sites are required where PAHs pollute the soil and groundwater.

Combustion processes

The World Health Organization (WHO) most of all considers air pollution (by combustion systems and traffic), smoke from open fireplaces and tobacco smoke risks for humans of coming into contact with PAHs (WHO, 2010).

In 2004 alone, 530,000 tons of the 16 EPA PAHs were emitted into the atmosphere worldwide. China has the lead with 114,000 tons, followed by India with 90,000 tons, and the United States with 32,000 tons (Zhang and Tao, 2009).

In 2010, Germany emitted 191.5 tons of the four PAHs benzo[a]pyrene, benzo[b]fluoroanthene, benzo[k]fluoroanthene and indeno[1,2,3-cd]pyrene into the atmosphere[2], approx. 93 % of which came from small and medium-size combustion units in households and businesses, roughly 5 % from industrial processes, the rest from large combustion plants and traffic (less than 1 %).

PAH emissions through combustion.
Photograph: © Bernd-von-Dahlen / www.pixelio.de

PAHs from industrial sources in Germany

PAHs belong to the reportable substances in the German Pollutant Release and Transfer Register (PRTR). In 2010, a total of 21 operating facilities reported releases of a total of 4,170 kg into the air and 50.4 kg into the water. The table below assigns the reports to the various industries in the PRTR.

2 http://www.uba.de/emissionen/publikationen.htm

Table 2

Analysis of PAH releases in the PRTR

(2010 reporting period)

Medium	Industry	Substance	Number	Release [kg p.a.]
Air	Chemical industry	Naphthaline	1	651.0
Air	Energy sector	Naphthaline	1	242.0
Air	Energy sector	PAH	1	514.0
Air	Metal industry	Naphthaline	1	121.0
Air	Metal industry	PAH	1	117.0
Air	Mineral industry	Naphthaline	7	1,441.0
Air	Mineral industry	PAH	1	188.0
Air	Other industries (production of carbon and graphite)	PAH	2	896.0
Water	Waste and wastewater management	Fluoroanthene	3	12.2
Water	Waste and wastewater management	PAH	1	5.4
Water	Energy sector	PAH	1	8.8
Water	Metal industry	PAH	1	24.0

Input into waters

PAHs enter the waters via sewage treatment plants and from many diffuse sources. The Federal Environmental Agency has studied the inputs of PAHs (Fuchs et al., 2010): Emissions into the atmosphere are of the greatest significance. In addition to direct deposits onto water surfaces, substances first deposited onto urban ground are washed into the waters via erosion and surface run-off. More than 80 % of the PAH input into waters are thus influenced by atmospheric deposition.

PAHs in our daily lives

PAHs enter the ambient air through dusts to which they are bound and abrasion from rubber products such as car tyres.

It can happen that we inhale them. PAHs are contained, inter alia, in the soot from diesel engine exhaust gases, for example from cars and lorries, but also from diesel locomotives, vessels or large machinery[3]. Tobacco smoke is another significant source of PAHs. Foodstuffs also contain them, for example smoked

PAH emissions from traffic. Not only the exhaust gases can be relevant PAH sources but also abrasion from rubber products.
Photograph: © Rainer-Sturm / www.pixelio.de

3 According to the most recent analysis by the experts at the IARC research centre in Lyon, soot particles from diesel exhaust gases have a carcinogenic effect on humans: https://www.iarc.fr/en/media-centre/pr/2012/pdfs/pr213_E.pdf

and barbecued meals, cocoa, and chocolate. Tar oils, petroleum-based extender oils and industrial soots are used to some extent in products made of rubber or soft PVC. Therefore these products also contain PAHs. They can be absorbed through the skin when these products are used.

Hand (slightly creamed with moisturizer) after one-time touching a rubber coated hammer shaft. The spots where PAHs were transmitted due to contact with the rubber fluoresce in UV light. Holes in the coating become visible.

Photograph: TÜV Rheinland Group

At the beginning of the 20th century, drinking water pipes used to be protected against corrosion in some areas by an inside layer of tar, which allowed PAHs to enter the drinking water. This practice has been abandoned in Germany for many decades, and PAH-contaminated drinking water is highly unlikely to be found today. The WHO has advised against the use of such coatings for health reasons for decades, but they are still in extensive use in some countries (WHO, 2011). The EU has defined maximum concentrations of PAH in drinking water (see Section 6).

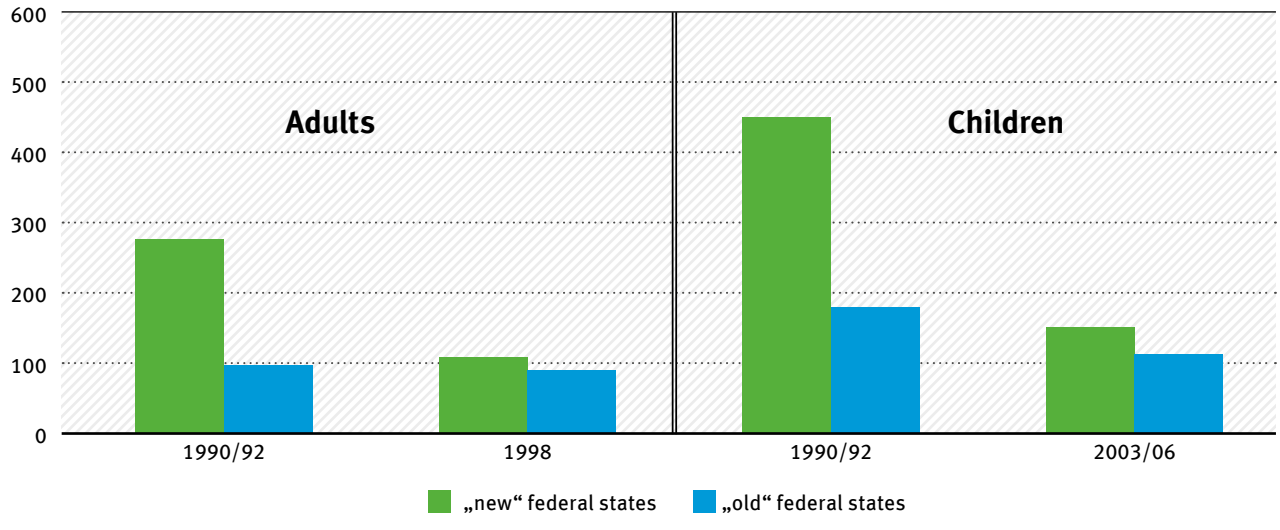
The German Environment Agency has determined the exposure of the population to PAHs in Germany on a representative scale in environmental surveys (Schulz et al., 2007). These surveys were based on detecting metabolites in the urine. Figure 6 shows a comparison of the findings for the lead substance, 1-hydroxypyrene, from the second (1990/92), third (1998), and fourth environmental surveys (2003/06) in Germany's "old" (Western German) and "new" (Eastern German) federal states.

Exposure to PAHs in adults has clearly dropped between 1990 and 1998, especially in the "new" federal states. The major reason should be less contaminated outside air, since PAH emissions were dramatically reduced in the new federal states after the reunification. Figure 2 also shows that children are more exposed to PAHs than adults. This finding can also be shown for other harmful substances; the reason is that children absorb more harmful substances in relation to their body weight than adults do.

Figure 2

1-Hydroxypyrene concentration in the urine of adults and children (non-smokers only) in Germany

The unit of measurement is nanograms per litre



Source: Deutsche Umweltstudie zur Gesundheit, GerES 2003-2006 (ehemals Kinder-Umwelt-Survey)

5. Which products may contain PAHs?

Products made of rubber or plastic

Independent laboratories keep detecting elevated PAH contents in consumer products. These include tool and bicycle handles (see Figure 7), shoes, or sports items (BfR, 2009); (Umweltbundesamt, 2010); (TÜV Rheinland, 2009).

This hammer shaft is an example of a rubber-containing product
Photograph: © Rainer-Sturm / www.pixelio.de

The reason for this is extender oils that are added to the rubber, typically together with fillers, to achieve the desired elasticity. Extender oils are used in some products to make brittle synthetic materials such as PVC soft and flexible. These extender oils, e. g. tar oil, are produced as by-products or waste products in coal and petroleum processing (see Section 2). PAH-containing extender oils are inexpensive and make the products affordable. While extender oils with reduced PAH content and elastic rubber and plastic materials without extender oils are available, these products are often more expensive due to the greater manufacturing effort required. Therefore, it is most of all products from the low-cost and import segments that contain extender oils with PAHs. Items that look like a bargain are in reality often contaminated with harmful substances.

Producers also often use industrial soot to dye plastics black. But regular soot contains PAHs as well. It would often be possible to switch to PAH-free alternatives. Where this is not feasible for technological reasons, producers should use soot with a low PAH content.

Unfortunately, one cannot tell products that contain PAHs by their appearance, and there is also no rapid test for PAH-contaminated products. A strong oil-like odour as known from petrol stations can be a hint. Some products still exude this odour long after they have been purchased.

Another example of a potentially PAH-containing product are bathing shoes
Photograph: © Rainer-Sturm / www.pixelio.de

Table 3 summarizes the PAH concentrations in examples of bathing shoes that UBA had measured in a study (Kalberlah et al., 2011). These concentrations are clearly higher than permitted, for example, by the GS quality mark for tested safety.

Table 3

Measuring results in bathing shoes

PAH determination in PVC	
Naphthaline	82 mg/kg
Acenaphthylene	‹0.1 mg/kg
Acenaphthene	‹0.1 mg/kg
Fluorene	170 mg/kg
Phenanthrene	120 mg/kg
Anthracene	23 mg/kg
Fluoroanthene	31 mg/kg
Pyrene	36 mg/kg
Benzo[a]anthracene	21 mg/kg
Chrysene/triphenylene	32 mg/kg
Benzo[b]fluoroanthene	7.5 mg/kg
Benzo[a]pyrene	8.7 mg/kg
Benzo[j/k]fluoroanthene	4.0 mg/kg
Indeno[123-cd]pyrene	3.9 mg/kg
Dibenzo[ah]anthracene	1.3 mg/kg
Benzo[ghi]perylene	5.9 mg/kg
Total PAHs	**546 mg/kg**

Quelle: Kalberlah et al., 2011

Tyres

PAH-containing extender oils have been used legally in car tyres until 2009. An EU-wide threshold value for PAH-containing extender oils in car tyres has been in effect since January 1, 2010.

The limits for tyres are lower than for children's toys
Photograph: © Rainer-Sturm / www.pixelio.de

It was introduced by a restriction in the European Chemicals Regulation REACH (Regulation (EC) No. 1907/2006). This restriction bans the use of extender oils for producing car tyres or tyre parts if these contain more than 1 mg/kg of benzo[a]pyrene or if the overall contents of all PAHs listed is more than 10 mg/kg. If the manufactured tyres and tyre treads exceed the specified limits, they may no longer be marketed since the beginning of 2010, which includes re-treaded tyres. However, this restriction does not apply to tyres of bicycles, children's scooters, or Kettcars.

It is the purpose of this regulation to reduce air pollution with PAH-containing dusts produced by abrasion. A test programme of European tyre manufacturers of 2011 has shown that some manufacturers and importers did not comply with the existing legal provisions or check their compliance (European Tyre & Rubber Manufacturers' Association, 2011). Checks by the competent state authorities in 2010 and 2011 however did not discover a violation of this regulation in another study[4,5].

Reecycling products from used tyres, e.g. surfaces for sports fields

The introduction of strict limits for PAH-containing extender oils in tyres solves the problem of PAHs produced by abrasion in road traffic. But up to 20 % of used tyres are recycled. Recycling is actually desirable but it also keeps material produced before 2010 that contains harmful substances and may even contain higher concentrations of PAHs in circulation. For example, used tyres are also processed into floor coverings by mixing the recycled granulate with the individual ingredients.

But there have been approval requirements for floor coverings in lounges, corridors, and recreation rooms for several years now, issued by the German Institute for Civil Engineering (DIBt) that exclude such use at least in many indoor spaces or allow it under surface layers made of other materials only. Surfaces of sports fields have been made of recycled granulate for several years. One common design are rubber granulate filled synthetic turf fields. A rubber granulate of recycled material is sprinkled among the 5 cm long synthetic fibres. Studies conducted in the

4 https://um.baden-wuerttemberg.de/de/presse-service/presse/pressemitteilung/pid/ergebnisse-der-marktueberwachung-2010-im-bereich-chemikaliensicherheit-1/
5 http://mulewf.rlp.de/de/themen/umweltschutz-umwelt-und-gesundheit/chemikaliensicherheit/stoffliche-marktueberwachung/pak-in-autoreifen/

United States (Zhang et al., 2008) have shown that the synthetic turf fields are not resistant to abrasion and that athletically active individuals, especially young people, may be exposed to PAHs via skin contact with abrasions from dusts. In the construction industry, recycled tyres are contained in building protection strips and mats used to protect the waterproofing or as covers of tunnels.

Protective coats, coatings, and adhesives

Coal tar pitch that is produced as a waste product in coal processing and has a particularly high PAH content is used in many paints or coatings. These are primarily corrosion protection coats. Many steel structures in industry such as hydraulic equipment, pipework, steel pilings in ports, or vessels are treated with such paints to protect them from water and to prevent rust formation (European Chemicals Agency, 2009).

Since regular coal tar pitch is too brittle, hard pitch is used and mixed with tar oils that can be heated to high temperatures and the mixture is then combined into a paint with mineral substances, ash, ground coal or polymers. Use of pitch-containing corrosion protection coatings is on the decline, all the more so since coal tar pitch in paints for vessels or port facilities has been banned throughout Europe (European Chemicals Agency, 2009).

Since PAHs are toxic to organisms such as fungi, PAHs are also used as active ingredients in wood preservatives. Creosote, which is obtained from coal tar, is particularly common. As a wood preservative, it is toxic to wood parasites, and it prevents the wood from drying out. Use of woods treated with creosote in gardens, parks, and playgrounds is prohibited. But it may still be used for treating railway sleepers, telephone and power line poles and fences for agricultural purposes (e. g. tree supports, hop and vineyard poles).

PAH emissions from wood treated with creosote can be considerable, especially in hot climatic conditions. The volatile fraction of the tar oils can enter the atmosphere through evaporation or drying of the wood. Railway sleepers treated with creosote also release about one third of the creosote into the environment during their average service life of 26 years, and this substance is not decomposed (Kohler et al., 2000).

Used railway sleepers and wood waste treated with creosote should therefore not be used for other purposes and thermally utilized (see below).

Hardwood flooring used to be glued with tar-based adhesives (containing up to 8000 mg of benzo[a] pyrene per kilogramme) into the 1950s. This resulted in increased indoor exposure. Buildings have to be refurbished still today if they include such contaminated wood flooring.

Road building materials and roof coverings

Bitumen or tar are used as binding agents for the minerals used in road surfaces made of asphalt, pavings, and roof coverings. They serve as an independent protective layer. While bitumen is produced during petroleum refining, tar is obtained in coal processing. Asphalt with PAH-containing tar was produced in the "old" federal states until 1970, in the "new" federal states until 1990. In the meantime, most European countries use bitumen instead of tar because it contains considerably less PAHs.

Tar can still be frequently found in old or repaired road surfaces. Tar is also still used in special surfaces, for example, of filling stations or parking lots in the United States. Roofs were frequently sealed with tar from 1970 to 1980 since tar provides good protection against moisture and ultraviolet radiation from the sun due to its chemical properties. The tar boards were also easy to attach and durable. Roof tars are mostly mixtures of pitch and filtered anthracene oil. Tar and tar boards are used in roofing still today, though to a much lesser extent since the carcinogenic effect of PAH-containing tar has become known (European Chemicals Agency, 2009).

Coal briquettes

Coal briquettes are still used for heating in industry and in residential buildings. Briquettes consist of various PAH-containing types of coal such as coke, peat (not permitted in Europe), or charcoal. Manufacturers typically add binding agents such as tar, pitch, or bitumen to their coal to keep the briquettes in shape. The binding agents themselves contain PAHs and can make up between five and twelve percent by weight of the briquettes. The carcinogenic tars can be substituted by other binding agents such as starch or molasses (European Chemicals Agency, 2009). Use of coal tar

pitch is banned in some countries, e. g. in Scandinavia or Germany. Lignite briquettes can be formed in a cold process without binding agents (European Chemicals Agency, 2009).

Clay pigeons

Marksmen use clay pigeons as targets. They must be stable since they are hurled into the air for shooting exercises. At the same time, they are to disintegrate when hit. Clay pigeons are mostly burnt from lime and a binding agent like coal tar pitch. Since this pitch can make up to 30 %, clay pigeons are a PAH source as well. According to EWU information by ECHA, about 200 million clay pigeons are produced and used per year. The marksmen typically do not dispose of their clay pigeons after the shooting. They remain in the environment. There is a law in the Netherlands that stipulates specific limits for PAH in clay pigeons. There are also less problematic alternatives in which various types of clay or petroleum pitch are used. But these are comparatively expensive (European Chemicals Agency, 2009).

6. What legislation is in place with respect to PAHs?

There are various regulations that stipulate limitation of PAHs in specific products and in the environment. There are also requirements for specific technical processes aimed at limiting PAH emissions. The purpose of these regulations is to replace PAH-containing products in the long term and to optimize technological combustion processes to protect humans and the environment.

Chemicals legislation (tyres, wood preservatives, mixtures for ultimate consumers)

The EU REACH regulation contains provisions on the handling of PAHs. According to this regulation it is generally forbidden to sell carcinogens, mutagens and substances toxic to reproduction (CMR substances) to ultimate consumers[6]. This regulation includes the eight PAHs that are already classified as CMR substances. But it only relates to substances or mixtures, such as paints and dyes. Products such as toys or shoes are not affected. Furthermore, creosote and other tar oil-based distillates are banned as wood preservatives. Creosote may not be used at all for indoor woods, for playgrounds, gardens and parks. But there are exceptions: In industrial processes (such as pressure impregnation), creosote may be used for treating railway sleepers, tree supports for agriculture, and vineyard poles[7]. REACH also stipulates limits for PAH-containing extender oils in car tyres[8].

Directives and regulations on PAHs in environmental media (air, soil, drinking water)

European directives and regulations are also aimed at minimizing PAH input into the environmental media air, soil, and water. For example, PAHs are regulated in the protocol on POPs (POP = persistent organic pollutants) for long-range, trans-boundary air pollution and in EU POP regulation (Regulation (EC) No. 850/2004).

To reduce the harmful effects of PAHs in the air, on human health and on the environment, the EU also has determined a target value to be achieved by December 31, 2012 (Directive 2004/107/EC relating to arsenic, cadmium, mercury, nickel, and polycyclic aromatic hydrocarbons in the air).

Existing directives are meant to reduce PAH emissions from combustion plants and other industrial installations. The EU Large Combustion Plant Directive 2001/80/EG (2001) stipulates limits for PAHs produced in combustion processes. The German Technical Instruction for Protection of the Air (TA Luft, 2002 version) limits emissions of carcinogens such as benzo[a]pyrene to 0.15 g/h or 0.05 mg/m3. The scope of the regulation on small and medium-sized combustion plants (1.BlmSchV) includes stoves and boilers in households. While it does not contain limits for PAHs, it sets limits for other harmful substances that are produced in an incomplete combustion process. Better combustion also reduces PAH emissions.

PAK in the air

Benzo[a]pyrene, to which a target value of 1 nanogram/m^3 applies, serves once again as a marker for PAHs in the air. To monitor air quality, the entire territory of Germany is divided into zones and ag-

6 Entries 28-30 of Annex XVII of the REACH regulation
7 Entry 31 of Annex XVII of the REACH regulation
8 Entry 50 of Annex XVII of the REACH regulation

The REACH Regulation (EC) No. 1907/2006

Every day, we come into contact with numerous chemicals: Individual substances (e. g. solvents), mixtures (e. g. varnishes), or products (e. g. textiles). But only for very few of the 130,000 chemicals used in Europe did the public and government authorities have sufficient information about health and environmental effects.

The EU enacted the REACH Regulation on the registration, evaluation, authorization and restriction of chemicals on June 1, 2007 to put an end to this shortcoming. One goal of the regulation is to identify chemicals of very high concern (SVHC), to adequately control and replace them with alternative substances. This is meant to improve the protection of human health and the environment when handling chemical substances.

All chemical manufacturers that produce more than one ton of a substance per year are obliged to submit specific information to the European Chemicals Agency (ECHA) in Helsinki. This includes information on substance properties, utilization processes and impacts on humans and the environment. The same applies to importers of chemical substances who import more than one ton per year. In a reversal of the burden of proof, REACH transfers responsibility for chemical safety from the national authorities who used to be responsible to the manufacturers and importers of substances: They will have to show convincingly that their products can be handled safely and are not unacceptably harmful to the health of users or consumers nor to the environment.

The manufacturers provide information about safe conditions of use in form of the safety data sheet to all their purchasers, the so-called downstream users, in the supply chain. The substances that are considered dangerous or even substances of special concern can be subjected to other regulatory measures besides registration, such as restrictions for specific uses or a general authorization requirement for all uses. Annex XVII of the REACH Regulation already contains a list of chemicals that are subject to restrictions of manufacture, use, or marketing.

glomerations in which the benzo[a]pyrene content is measured in PM_{10}[9] at approx. 115 stations (see Figure 3). Incidents of exceeding the target value have been reported to the European Commission since 2008: They occurred at isolated stations (max. 6 %) that were located close to road traffic or industrial facilities in the period 2008 - 2010. The target value was also exceeded in urban areas caused by wood burning in private households. No incidents of exceeding the target value were reported in the years 2011 to 2014 in Germany. In 2013 13 EU-Member States 3 registered incidents of exceeding the target value, primarily in urban and suburban areas (European Environment Ageny 2015).

PAHs in soil and water

The Federal Soil Protection and Contaminated Sites Ordinance (BBodSchV, July 12, 1999) stipulates three values to protect the soil from contamination: Precautionary, test, and action values. The precautionary values are to prevent the occurrence of adverse soil alterations. If test values are exceeded, there are specific indications of an adverse soil alteration. The action values are meant to prevent risks in soils. There are values for the sum total of the 16 PAHs listed by the U.S. EPA and for the individual substances benzo[a]pyrene and naphthaline.

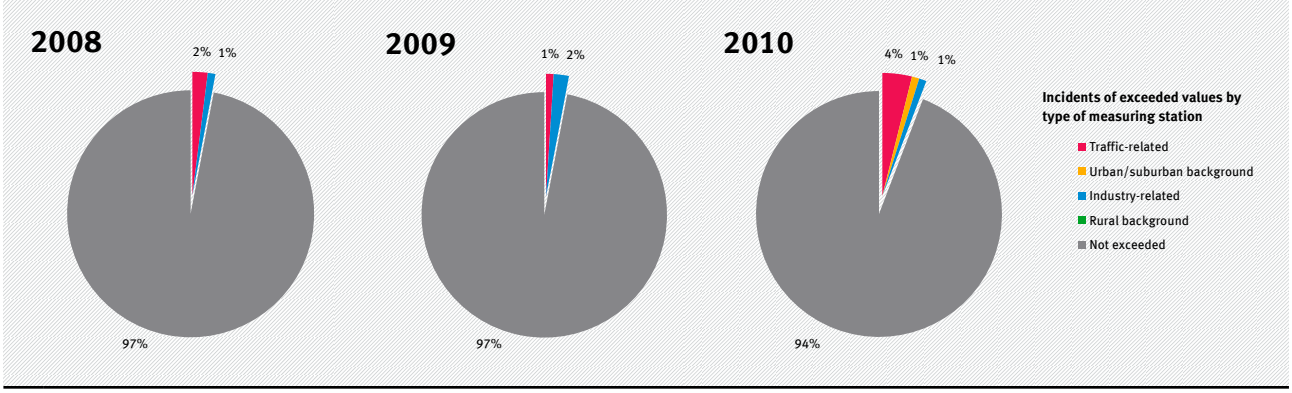
The test value of 1 mg/kg of fine soil dry matter (DM) is applicable for benzo[a]pyrene for the soil - crop path of action. The action values for benzo[a]pyrene are 2 mg/kg DM for children's play areas, 4 mg/kg DM for residential areas, 10 mg/kg DM for parks and recreational facilities, and 12 mg/kg DM for industrial and commercial plots of land. Precautionary values for benzo[a]pyrene in soils with a humus content > 8 % are 1 mg/kg DM, in soils with < 8 % humus content 0.3 mg/kg DM.

9 PPM (particulate matter): Particles that do not immediately drop to the ground but linger in the atmosphere for specific period of time. The fine dust particles are divided into fractions by grain size. PM_{10} are all dust particles with an aerodynamic diameter smaller than 10 microns.

Figure 3

Percentage of air measuring stations in Germany with incidents of exceeding B(a)P

Note: Since no excess was measured in rural areas, there are no green sectors in the three diagrams. Source: Information extracted from German Air Monitoring Network.



Source: Information extracted from German Air Monitoring Network

Annex X of the EC Water Framework Directive (WFD 2000/60/EC (2000)) lists "priority substances." There are environmental quality standards at European level for 33 priority substances, including eight PAHs. The environmental quality standard for benzo[a] pyrene (annual average) is 0.05 µg/L. In addition, PAHs are classified as "priority substances" for which an extra phasing-out obligation is in place. These substances should no longer be input into the waters of the Community at a point in time to be determined.

There is no limit in Germany for the use of sewage sludge in agriculture. An amendment to the Sewage Sludge Regulation is being prepared that will stipulate a limit for benzo[a]pyrene of one milligram per kilogram of sewage sludge. This value would correspond to the value of the Federal Soil Protection and Contaminated Sites Ordinance.

Foodstuffs

"The Regulation for Setting Maximum Levels of Certain Contaminants in Foodstuffs" (Regulation (EC) No. 1881/2006) stipulates the highest levels of specific contaminants in foodstuffs. Benzo[a]pyrene, benzo[a]anthracene, benzo[b]fluoroanthene, and chrysene are used as lead substances for PAHs, for the sum total of which limits between one microgram per kilogram for infant formula and follow-on formula and thirty five micrograms per kilogram for smoked mussels have been determined.

The benzo[a]pyrene limit for drinking water set in the Drinking Water Ordinance is 10 nanograms per litre; one nanogram is one millionth of one milligram. Four other, less carcinogenic representatives of PAHs (WHO, 2011) may not exceed 100 nanograms per litre in total in drinking water. The rate of exceeding critical levels was less than one hundredth of a percentage point in Germany from 2005 to 2007 (Umweltbundesamt, 2011).

Toys

The Toy Safety Directive (2009/48/EC) applies to all toys. Toys may not threaten the safety or health of their users when used as intended and in a foreseeable way, bearing in mind the behaviour of children. This directive expressly prohibits CMR substances above a threshold value. The concentration limits of 100 mg/kg apply to the eight PAHs that are classified as carcinogens. Theoretically, PAHs may be contained in toys at levels below these limits. For benzo[a]pyrene, the limit is thus one hundred times higher than the one for extender oils in car tyres . This is a clear inconsistency in legislation that must be corrected by the EU.

Fuels

The EU Directive on the Quality of Petrol and Diesel Fuels (98/70/EC) stipulates that the PAH concentration may not exceed eight percent by weight in diesel fuels.

10 http://www.bfr.bund.de/cm/343/polyzyklische_aromatische_kohlenwasserstoffe_pak_in_spielzeug.pdf

Cosmetics

The use of all PAHs with CMR properties in cosmetics is prohibited. This is provided in Article 15 of the Cosmetic Products Regulation (Regulation (EC) No. 1223/2009).

Wood preservatives

DThe PAH-containing wood preservative creosote is carcinogenic and a substance of concern in the environment because it is persistent, accumulative, and toxic. Studies point to a high risk for organisms when used for woods that are in direct contact with soil or water.

Therefore, creosote as a wood preservative (see Section 5 – PROTECTIVE COATS, COATINGS, AND ADHESIVES) is regulated in the REACH Regulation (EC/1907/2006) and the Biocide Regulation. The REACH Regulation prohibits the use of creosote for consumers; use in industrial facilities and for commercial purposes is allowed under certain conditions. It was decided in 2011 to include creosote in the list of active ingredients of biocidal products permitted throughout the EU (Annex I of the Biocide Directive) (Directive 2011/71/EU), but with restrictions: The approval of creosote as a permissible active ingredient of biocidal products throughout the EU is limited to five years, and creosote may only be used in biocidal products where no suitable alternatives are available.

Currently, creosote-containing biocidal products are still on the market without a permit as part of transitional regulations. Each creosote-containing biocidal product has to receive a permit by April 30, 2015; creosote-containing biocidal products without a permit may no longer be marketed as per May 1, 2015. Member states that authorize creosote-containing biocidal products have to justify their decision in a report before the EU Commission by July 2016 and explain, inter alia, how they encourage the development of alternatives. Before the EU-wide approval as an active ingredient is renewed, creosote will be subjected to a comparative assessment with other wood preservatives.

Used wood that has been treated with tar oils as wood preservative has to be classified in Germany as waste wood category A IV in accordance with the Waste Wood Ordinance. It may only be energetically recycled or used to produce synthetic gas, activated carbon, and industrial charcoal; recycling into a wood-based material is excluded.

Construction

Various regulations in construction in Germany address PAHs. For example, there is a directive that regulates the environmentally compatible utilization of tar/pitch-containing finishing materials and the utilization of recycled asphalt in road building (RuVA-StB 01, 2005). Broken-up road surface materials having a PAH-content of less than 25 milligrams/kilogram are called recycled asphalt and can be utilized unbound under a waterproof layer. Recycled asphalt may even be used without meeting special requirements regarding safety, soil, and water protection if it contains less than 10 milligrams of PAH per kilogram. Broken-up road material is to be classified as pitch-containing if it is contaminated with more than 25 milligrams of PAH per kilogram. In this case, stricter requirements apply to utilization and installation methods. Classification as waste requiring special monitoring/harmful waste starts at 1000 milligrams of PAH per kilogram of broken-up road surface.

The "Suggestions for the evaluation of, and measures to reduce, PAH contamination by hardwood floors with tar adhesives in buildings" (DIBt-Mitteilungen, 2000) contain a guideline for evaluating PAHs that are used in tar adhesives for hardwood floors. This guideline also describes how PAH contamination in buildings in which such floors were installed can be reduced by renovation.

Floor coverings that are to be permanently installed in lounges, corridors, and break rooms require building inspectorate approval issued by the German Institute for Civil Engineering and may contain granulate from used tyres in the base layer of the flooring in exceptional cases only. The upper limit for the recycled material is 50 milligrams of PAH per kilogram and 5 milligram of benzo[a]pyrene per kilogram. If the PAH contents in granulate from used tyres decline over time as a result of the EU restriction, it will also be possible to lower the limits for floor coverings. Today, the average PAH content in granulate from used tyres still is about 40 milligrams/kilogram. When a building is demolished, floor coverings made of granulate from used tyres should be disposed of separately to prevent PAH-containing components from mixing with building rubble.

How do you benefit from REACH?

REACH for the first time gives potential buyers an opportunity to obtain information about substances of very high concern (SVHC) in products. You may ask their suppliers, home improvement centres or department stores if PAHs are contained in specific products. These have to answer your question within 45 days – whether you purchase the product or not.

You can find the substances of very high concern (SVHC) on the so-called candidate list. This list is available on the Internet at http://echa.europa.eu/chem_data/authorisation_process/candidate_list_table_en.asp. It contains one PAH so far (anthracene) and several substances that contain PAHs (five anthracene oils and "pitch, coal tar, high temperature"). This list and the people's right to information helps people to deliberately choose more sustainable alternatives and products without substances of very high concern when they decide to buy or not to buy a product.

In addition, it is the duty of every supplier under REACH to check his or her products for any SVHC in it whenever the candidate list is updated. If the extended list includes new SVHC, a supplier is obliged to actively inform the European Chemicals Agency and his or her commercial customers. Retailers have to inform us all, the ultimate consumers, accordingly upon request. You can ask your question in a fast and simple manner using an online form. All you need is the number under the bar code of the product. Alternatively, you can use the form letter by the Federal Environmental Agency. Exercise your right.

The form for your online-query and the form letters are available at the UBA-website www.reach-info.de. There, more information about the potential uses of SVHC are available, too. UBA thinks that companies should already take a more responsible approach and refrain from substances of very high concern or substances with comparable properties.

In addition to the candidate list, REACH provides other regulatory measures that help protect consumers, employees, and the environment from problem chemicals better than before. The approval of substances of very high concern allows their use on application only. Restrictions throughout Europe protect the people from uses of harmful substances that constitute a risk

7. A German initiative: restricted use of PAHs in rubber and plastic products

To provide better protection for humans and the environment, Germany launched an initiative in the summer of 2010 aimed at limiting the content of PAHs in consumer products throughout the EU. As a result, in 2013, an amendment of the REACH regulation came into force. Since 27th of December 2015, consumer articles may not exceed a total amount of 1 mg/kg of the eight carcinogenic PAHs. For toys and baby toys a threshold of 0.5 mg/kg applies. The restriction also applies for imported goods. Manufacturers and importers have to ensure that their products and articles are complient with the restriction.

The precautionary principle in European environmental policy

The precautionary principle is a pillar of environmental policy. Precautionary measures are meant to contribute to the maintenance, protection, and improvement of the quality of the environment. The declaration of the U.N. Conference on Environment and Development (UNCED) 1992 in Rio de Janeiro explains the precautionary principle in Chapter 35 (3) of Agenda 21:

"In the face of threats of irreversible environmental damage, lack of full scientific understanding should not be an excuse for postponing actions which are justified in their own right. The precautionary approach could provide a basis for policies relating to complex systems that are not yet fully understood and whose consequences of disturbances cannot yet be predicted."

REACH contributes to ensuring a high level of protection for human health and the environment and to making the handling of chemical substances as safe as possible. Unlike in the past, there is no need to provide the ultimate scientific proof that a substance or product is not safe before it can be regulated.

Emissions of PAHs should be restricted to expose humans and the environment as little as possible to PAHs. Since PAHs are also natural substances and combustion products that cannot be fully controlled, zero exposure cannot be reached. Therefore we will have to deal with this group of substances in the future, and more steps will be necessary. The first applicable regulations represent first steps towards this goal. Binding restriction of PAHs in consumer products throughout Europe, as it is envisaged now, constitutes another important step in the protection of humans and the environment.

8. What can each individual do?

Humans come into contact with PAHs. For example, through exhaust gases, smoked or sooted foods, or tobacco smoke. Everyone can reduce his or her personal exposure to PAH. When barbecuing, care should be taken that the food to be barbecued is not blackened, that the charcoal is thoroughly glowing, and that the food is not seared. Use of a grilling tray prevents the formation of unnecessary smoke from fat dripping into the fire. Avoiding (active and passive) tobacco smoke is another way besides avoiding PAH-containing products.

Rubber and soft PVC can indicate PAH content by its strong, oil-like odour. Products made of black rubber or plastic may contain untreated PAH-containing industrial soot. Quality symbols such as the GS mark or the Blue Angel provide some guidance. Germany has the GS mark for tested safety. Products made of rubber or plastic have been tested for their PAH content since 2007 as part of the GS testing. For granting the GS label 18 PAHs are tested, which may not exceed an amount of 50 mg/kg in sum. The threshold for the 18 PAHs is even more stringent for materials which can be a subject to mouthing or with a skin contact of more than 30 seconds; The total content of the 18 PAHs analysed may not exceed 1 mg/kg (AfPS, 2014). However: The GS mark is a voluntary sign. Manufacturers must decide if they wish to apply for the GS mark. If in doubt, avoid skin and mouth contact or use alternative products.

If people are insecure, they can use the new right to information under the REACH Regulation to obtain information about substances of very high concern. But it only applies to substances that are already listed on the candidate list. The only PAHs on this list so far are anthracene and five anthracene-containing oils as well as "pitch, coal tar, high temperature" (as per January, 2016). It is the responsibility of companies to develop strategies aimed at reducing PAHs in products and industrial exhaust gases beyond legal requirements. You can influence the policy of these companies by making informed consumer decisions.

9. Summary

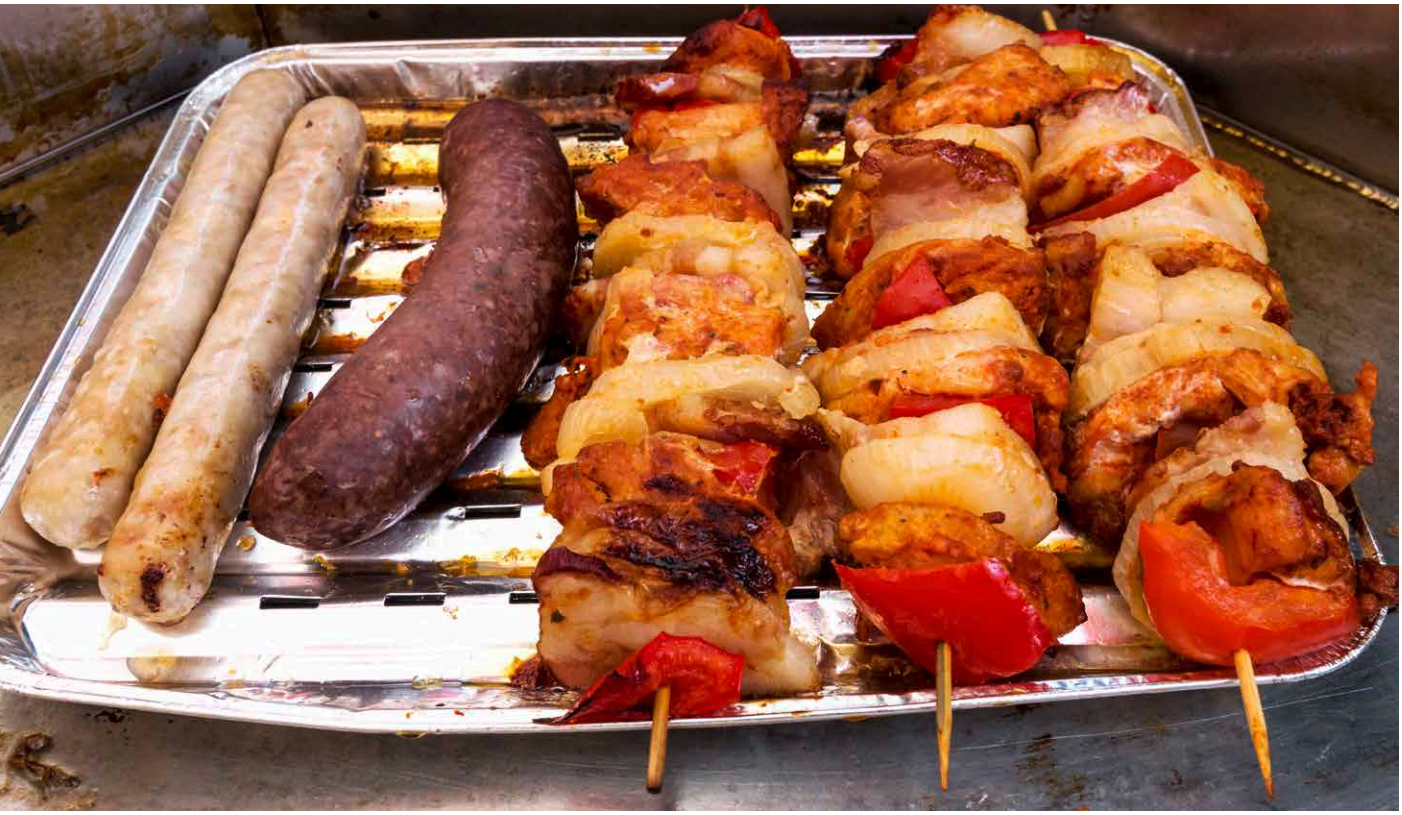
Polycyclic aromatic hydrocarbons (PAH) originate as by-products of incomplete combustion of organic materials, such as wood, petroleum and coal, when these materials are strongly heated in the absence of air. PAHs are also contained in refinery or coking plant products. In chemical terms, PAHs are a diverse group of aromatic substances composed of two to seven hydrocarbon rings. PAHs dissolve well in fats, bind to particles, and accumulate in organisms and the environment.

PAHs enter the environment mainly through the air as a result of industrial and natural thermal processes. Consumers come into contact with PAHs in the form of contaminated rubber or plastic products and abrasion from rubber products, floorings, or wood preservatives. PAHs are absorbed via the air, tobacco smoke, and consumption of specific contaminated foodstuffs, such as smoked foods. Many PAHs are carcinogenic, mutagenic, or toxic for reproduction. Due to their chemical and biological stability and their potential for bioaccumulation, they are persistent in the environment and accumulate in organisms.

There are many individual regulations aimed at limiting the occurrence of PAHs in the environment and in products. For example, limits are in place for PAHs in the air, water, and soil, in foodstuffs and drinking water, for PAHs in tyres and certain wood preservatives, in fuels, toys, and there are some regulations in the construction industry.

The environmental quality objectives for PAHs in surface waters are not achieved everywhere in German surface waters. Contaminated soils (abandoned sites) frequently show higher values than the predetermined test values. There are no binding limits for consumer products in general. Meanwhile, consumers who wish to avoid PAH-containing products made of rubber or soft PVC can first and foremost rely on quality marks or independent product tests, which frequently include PAHs.

An initiative of German agencies, including UBA, aimed at banning PAHs in consumer products on a European level. Germany proposed a restriction procedure in conjunction with the new REACH Regulation that



Photograph: © Ascada / www.pixelio.de

the EU Commission will address in fast-track proceedings. The sum of the eight PAHs classified as carcinogenic may not exceed a concentration of 1 milligrams per kilogram in products since 27.12.2015. For toys and baby toys the threshold of 0.5 mg/kg applies.

The German Environment Agency is planning further steps aimed at limiting the risks for humans and the environment. This primarily includes the identification of other PAHs as substances of very high concern as defined in the European REACH Regulation. If PAHs are classified as substances of very high concern, all citizens have a right to ask retailers which products contain PAHs and at what levels. They can find out about the concentrations at which these substances are present. In addition, use of PAH-containing substances may be subjected to authorization. This means that only such applications will be permissible that are harmless or for which there is no alternative.

Private households can contribute to the reduction of PAH emissions by operating their wood or coal-fired stoves and boilers low in emissions – information how to do this can be found in the respective operating instructions.

Companies are urged to develop strategies aimed at minimizing PAH content in industrial exhaust gases and products that exceed legal requirements. They are asked to reduce PAH contamination as much as possible.

10. Glossary and List of Abbreviations

BAuA: Federal Agency for Industrial Safety and Occupational Medicine (Bundesanstalt für Arbeitsschutz und Arbeitsmedizin)

Restriction: A restriction relates to a specific use of a chemical, e. g. in the case of PAHs the use of PAHs in consumer products above a predetermined concentration. All other uses of the chemical are generally permitted. The restriction of a chemical can be related to its manufacture, use, or introduction to the market, including in mixtures and products. This allows the regulation of chemicals in imported products.

Substances of very high concern (SVHC): To identify a substance as a SVHC, ECHA or a member state of the European Union must prepare a dossier that meets the requirements of Annex XV of the REACH Regulation. This so-called Annex XV dossier lists substance properties (e. g. solubility in water, flammability, degradability, etc.), manufacturing processes, uses, and effects on organisms and assesses the risk for humans and the environment based on this data. If all member states agree on the evaluation of the substance, the chemical is added to the so-called candidate list.

BfC: Federal Chemicals Office (Bundesstelle für Chemikalien)

BfR: Federal Institute for Risk Assessment (Bundesinstitut für Risikobewertung), one of three federal German authorities responsible for REACH, based in Berlin.

CAS-No.: Chemical Abstracts Service Registry Number, an international designation standard for chemical substances

CMR: Carcinogenic, mutagenic or toxic to reproduction

ECHA: European Chemicals Agency based in Helsinki. All information collected about the approximately 100,000 chemicals used in the EU is pooled here.

Product: An object that receives its specific form, surface finish, or shape that determines its function to a greater extent than its chemical composition.

EFSA: European Food Safety Authority. European authority for food safety based in Parma. It is responsible for risk assessment of foods and feedstuffs in the EU.

Exposure: Technical term for contact with, or exposure to, a harmful substance in toxicology

Mixture: Blends, mixtures, or solutions consisting of two or more substances.

GS mark: Quality symbol confirming "tested safety"

Candidate list: A list of chemicals considered substances of very high concern by the EU. Manufacturers and importers are subject to special information obligations to their customers and end users with respect to substances that are on the candidate list.

PRTR: Pollutant Release and Transfer Register. An online register that provides information about harmful substances that are released by large industrial plants in your region. The European PRTR Regulation and the German PRTR Act provide the basis for the PRTR.

SVHC: Substance of very high concern (see this entry)

REACH: Registration, Evaluation and Authorisation of CHemicals. Regulation No. 1907/2006.

PBT: Persistent, bioakkumulative, and toxic

PVC: Polyvinyl chloride, a plastic that is actually hard and brittle. It can be made soft and elastic by adding softeners. Such softeners or extender oils often contain PAHs.

TA-Luft: German Technical Instruction for protection of the air

UBA: German Federal Environmental Agency (Umweltbundesamt), one of the three federal authorities responsible for the protection of health and the environment, with headquarters in Dessau-Rosslau

WHO: World Health Organization with headquarters in Geneva/Switzerland

Authorization requirement: all uses of this chemical are banned in the EU unless someone files an application for approval of a specific use and the EU Commission approves it. Use of this substance as an intermediate and its import into the EU as an ingredient of products are still exempt from authorization and therefore do not require approval.

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