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OPINION OF THE SCIENTIFIC COMMITTEE ON TOXICITY, ECOTOXICITY AND THE ENVIRONMENT (CSTEE) ON

"Questions to the CSTEE relating to scientific evidence of risk to health and the environment from polycyclic aromatic hydrocarbons in extender oils and tyres"

Adopted by the CSTEE during the 40th plenary meeting of 12-13 November 2003

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Terms of Reference

The Commission is considering a proposal for a Directive related to restrictions on the marketing and use of extender oils containing high levels of certain PAHs and of tires containing these oils. PAHs are not subject to the Regulation 793/93 or Directive 67/548/EEC so a careful scientific consideration of the risk is necessary. Industries producing extender oils apparently are ready to produce containing low levels of PAHs 2003-2005 whereas the European Association of the Rubber Industry (BLIC) considers it possible to phase out PAHs containing extender oils by 2009.

In order to decide on the appropriate risk reduction strategy the CSTEE is asked to answer the following questions:

1. Is it scientifically justified to conclude that certain PAHs could cause serious carcinogenic, mutagenic or reprotoxic effects for human?

2. Is it scientifically justified to conclude that PAHs are emitted into the environment as a consequence of the abrasion of the tyre tread?

3. Can a reduction of the concentration of PAHs in tyres reduce significantly the overall concentration of PAHs found in the environment?

4. Can benzo(a)pyrene be seen as a marker for the group of PAHs? Is it scientifically justified to conclude that a reduction of the concentration of benzo(a)pyrene would also lead to a reduction of the other PAHs?

5. How does the CSTEE evaluate the risks to human health or the environment from PAHs that are emitted into the environment as a consequence of the abrasion of the tyre tread?

Introduction

The rational for considering the proposal is based on reports submitted by the Swedish National Chemicals Inspectorate (KEMI Report of March 27, 2003) and the German Umweltbundesamt, which describe a significant contribution of tyre wear to the environmental contamination by PAHs. These 2 reports and a publication by BLIC provide the following information:

Tyre tread contains up to 28% extender oils with 17 to 357 mg/kg total PAHs (average 137 mg/kg). The benzo[a]pyrene (BaP)-concentration ranges between 1 and 16 mg/kg (average 5 mg/kg). These concentrations have been determined by

chemical analysis after Soxleth extraction of rubber slices. During use tyres of passenger cars lose up to 2 kg, tyres of trucks up to 12 kg of their mass by abrasion. The major part of the abraded material is dispersed on the ground near the roads and flushed away with the surface runoff and ends up in the water. From there it either ends up in the sludge of waste water treatment plants or in surface water where the particles sediment. The KEMI report indicates that it is not possible to provide a reliable estimate to what extend PAHs from tyre wear contribute to the PAHs contamination of the environment as compared to other sources.

Measurements in Stockholm suggest that the annual contribution of PAHs from tyre wear is about 4% of the emissions via exhaust gases of diesel and gasoline powered vehicles and that vehicle exhaust together with tyre wear account for 30 % of the PAHs levels in sediments.

The reports conclude that due to the rapid deposition of the relatively large tyre particles, they do not make a major contribution to the PAHs content of ambient air.

Most of the PAHs extracted from tyre wear are known carcinogens and the extracts induced severe effects in fish. A recent investigation (Stephensen et al., 2003) also proves that automotive tyres leak PAHs and that cytochrome P450 1A1 is induced in rainbow trout exposed to tires.

Due to the ongoing discussion tyre manufacturers increasingly use extender oils with low PAHs contents and the EC considers limiting the total PAHs content to 10 ppm and the content of BaP to 1 ppm.

CSTEE concludes that the major deficits in the documentation by KEMI and the Umweltbundesamt (Rauterberg-Wulff) are

- Insufficient quantification of the contribution of PAHs from tyre wear to soil and the aquatic environment as compared to other sources of pollution.

- Insufficient information on the bioavailability of PAHs from tyre wear particles in soil and the aquatic environment.

In order to respond to the questions addressed CSTEE has evaluated the contribution of PAHs from tyre wear particles to total PAHs emissions in ambient air and the aquatic environment. In these calculations, it is considered that for human health effects PAHs associated with respirable particles (PM10) are primarily of interest, whereas for environmental effects the total PAHs emissions are important.

Size and concentration of tyre wear particles in the urban atmosphere

The first difficulty in assessing environmental exposure of PAHs from tyre wear particles, is the evaluation of PAHs content in tyres because the potential range is very wide (at least one order of magnitude).

According with the BLIC document “Tyre particulates as a source of PAHs in the environment”, the content of oils in tyres is very variable. Oil content in car tyres is 6-8% of the whole tyre mass, 11-16% of the tread; the content is lower in truck tyres (4.5% of whole tyre, very low in tread). The PAHs content in oils is in the range 300-700 mg/kg. From these values, the range of PAHs content in tyre particles can be calculated as follows:

Minimum (4.5 % of oil in tyre, 300 mg/kg PAHs in oil): 13.5 mg PAHs/kg

Maximum (16 % of oil in tyre, 700 mg/kg PAHs in oil): 112 mg PAHs/kg

The range is in some agreement with other literature data quoted in the BLIC document (original papers not available) indicating a range of 1-230 mg/kg, the values reported by the Umweltbundesamt (Rauterberg-Wulff) of 30-360 mg/kg, and with the measurements of 226 mg/kg tyre wear (Rogge et al., 1993). According to the UBA data, there is a significant correlation between BaP and total PAHs content.

The second step in assessing exposure is the evaluation of the amount of particles released. From different literature sources, particle emission data reported in the following scheme can be derived. Values are in mg of particles released per km of road. There is a reasonable agreement among various sources.

	Swiss Environ. Agency	US EPA	German exp. study	BLIC
	Per tyre	Per vehicle	Per vehicle	Per tyre
	mg/km			
Passenger car	16-50	120	53	20-30
Light duty vehicles (4 tyres)	69-150	210	107	25-40
Heavy duty vehicles (6-14 tyres)		410	539	50-70
Motorbikes		60		20-30

Particle size related emission factors are difficult to assess because they depend upon many factors such as tyre characteristics, environmental factors (road, temperature, etc), type of car, speed etc. Various studies, although differing in the way particles were collected and their size estimated, consistently suggest that airborne particles generated from the wear of tyres include particles with large size (aerodynamic diameters predominantly larger than 7 μm and ranging up to >100 μm), but also report a secondary population (less than 20% of the total, according to Cadle, (1978), of much smaller particles (<1 μm) (Cardina, 1974; Dannis, 1974; Pierson et al., 1974).

In the most recent and detailed study on the size distribution of tyre-derived particles, Fauser (1999) collected and fractionated particles with aerodynamic diameter <20 μm with pumps located 3 m above road surface and 1-15 m away from road. He used organic Zn and styrene-butadiene rubber as markers specific for tyre particles. By examining the size-distribution of particles containing these markers only, he concluded that they have a bimodal size distribution, with more than 90% by mass (in the range collected, i.e. <20 μm) smaller than 1 μm and the rest being larger than 7 μm. In the same study, the total concentration of tyre particles in a busy Copenhagen

road was estimated to be in the range 1-10 μg/m^3, with a mean value of 2.8 μg/m^3, which represented about 5% of the total airborne particles <20 μm. At a rural site the corresponding figures were 1.4 μg/m^3 and 2.4% of the total. There was no obvious variation in the fraction of tyre particles among different sizes.

The above study gives no information on the mass prevalence of particles larger than 20 μm. In another study which used 2-(4-morpholinyl)benzothiazole as a marker for tyre-derived particles, (Kumata et al. (1997) concluded that tyre particles made up approx. 3.6% of the total road dust particles >1 μm collected by vacuuming the road surface.

Taken in combination, the results of Kumata et al (1997) and Fauser (1999) study suggest that particles with diameter larger than 20 μm make up a substantial fraction of the total mass of tyre-derived particles. This is supported by data on tunnel measurements, given by Rauterberg-Wulff (UBA Report), which give emission factors for tyre-derived PM10 particles of 6.1 mg/km for passenger cars and 31 mg/km for heavy duty cars, while the emission factors for total tyre wear particles are given as 120 and 500 mg/km, respectively. From these figures it can be calculated that the 5-6% of these particles are in the PM10 size range. The BLIC Report gives similar figures, i.e. emission factors for PM10 of 7 mg/km for passenger cars, 50 mg/km for heavy vehicles and 5±2% of total tyre particles in the PM10 size range.

Thus different approaches lead to the conclusion that tyre particles

- are emitted in various sizes ranging up to >100 μm. aerodynamic diameter,
- have a bimodal size distribution, with about 5% by mass of an aerodynamic diameter <1 μm, the rest being in the non-respirable range, and
- make up approx. 5-6% of all respirable (PM10) in the urban atmosphere

The CSTEE notes that the relatively high temperatures in the contact surface between tyre and road may also give a vapour phase emission of the PAHs. So far, however, only low levels of gaseous PAHs have been observed (Rauterberg-Wulff).

Contribution of tyre wear to PAHs emissions from other sources, especially diesel

CSTEE is aware that the contribution of the different sources like traffic, industry, wood burning and others to the total PAH emissions varies between the different European regions. Despite these differences the contribution of traffic is usually in the range of 20 to 30% (KEMI, UBA, EEA CORINAIR 1997, Boström et al 2002).

Using

a) data in the UBA Report for tyre wear and diesel exhaust emissions in Germany,

b) the BaP content of tyres and diesel particles given in the same Report and

c) the above mentioned evidence that 5% of tyre particles are in the PM10 range

the following calculations can be made:

	Total tyre wear (kt)	BaP in total tyre wear (5 ppm)	BaP in PM_{10} from tyres (5%)	Total diesel PM_{10}	BaP in diesel PM_{10} (13 ppm)	Ratio tyre BaP/diesel BaP in PM_{10}
1990	53	265 kg	13.2 kg	42 kt	540 kg	0.02
2000	70	350 kg	17.5 kg	32 kt	420 kg	0.04
2020	88	440 kg	22.0 kg	10 kt	120 kg	0.18

These figures suggest that the contribution of tyre particles to BaP on respirable particles is currently < 5% relative to diesel exhaust, but may increase as diesel-associated emissions are reduced in the future. If diesel exhaust constitutes the main contributor to PM10 levels in urban regions, then the figure of < 5% represents the contribution of tyre wear particles to the total respirable BaP in the urban atmosphere. On the other hand, the urban atmosphere PM10 also contains contributions from other sources (gasoline exhausts, home heating, industrial sources etc). For example, according to the EU Position Paper on PAHs (Annex 2, Table 6), in the UK in 1995 diesel and gasoline vehicles contributed 2.0 and 5.7 t BaP, respectively. Thus the contribution of tyre particle-derived BaP to the total BaP present on respirable particles in the urban atmosphere is currently well under 2%.

On the other hand, the above calculation suggests that the contribution of tyre wear particles, relative to diesel exhaust, to total BaP emissions (and not just those associated with PM10) may be much higher. However, their contribution to the total BaP emissions, from all sources, will again be much lower, depending on the relative importance of diesel-derived vs. other sources. The UBA Report states that in the year 2000 BaP emissions from tyre wear amounted to 0.35 t (p. 7), while, according to the EU Position Paper on PAHs (Annex 2, Table 4), in 1995 total anthropogenic emissions of BaP in Germany were 26.4 t. Based on these figures, the contribution of tyre wear to total environmental emissions of BaP in Germany would be calculated to be < 2%. This conclusion is in reasonable agreement with the statement in the KEMI Report (p. 25) that tyre wear contributes only 4% of the emission of PAHs via petrol and diesel exhaust gases in the Stockholm area.

From this CSTEE concludes that, on a mass balance basis, tyre wear contributes probably less than 2% to BaP exposure associated with the respirable fraction of particles in the urban atmosphere (PM10) and represents a much smaller proportion of the total anthropogenic BaP emissions to this fraction.

For assessment of the biological significance of these estimates, the bioavailability of PAHs in tyre particles has to be considered, about which insufficient information is available. Due to this lack of information it is not possible to estimate the consequences of a reduced PAHs content in tyres for the existing PAHs concentration in the environment. However, taking into account a 4% contribution of tyre wear to the total automobile exhaust emissions that are considered to be 30% of the total PAHs contents in sediments a limitation of PAHs in tyres will at present not considerably affect the PAHs concentrations in sediments.

Environmental fate and bioavailability

About the fate of particles in the environment, precise quantitative figures are not available, but, in qualitative terms, it can be assumed that larger particles remain close to the road surface and can be transported by runoff. Small particles can be transported by wind. However, it is reasonable to assume that most of the particles are deposited relatively close to the emission sites and can be transported in surface water bodies by runoff. Thus, environmental systems at risk are surface soil and aquatic ecosystems.

From the data provided, the assessment of particle input in surface water bodies is not easy. A document provided by BLIC (Environmental impact of tyres: ecotoxicological approach) reports an evaluation of tyre debris concentration in rainwater in France. The methodological assumptions for the evaluation are not clearly described, thus the reliability of the results cannot be independently verified. However, a maximum concentration of 34 mg/L of tyre debris in rain water was calculated. Assuming a concentration of 100 mg PAHs/kg, gives a concentration of 3.4 µg/L of particulate bound PAHs in rain water.

Therefore, PAHs enter into the aquatic environment as bound to organic particulate matter. They are highly hydrophobic chemicals, with log Kow values ranging from a minimum of about 3.5 for the lightest compounds (e.g. naphthalene) and a maximum of more than 6 for the heaviest ones (e.g. BaP).

No clear data have been provided about the release of PAHs in water from tyre debris. In the previously quoted BLIC document, a lixiviation experiment is mentioned, but the results are not clearly reported.

It follows, that for aquatic organisms living in the water column, water-borne exposure from dissolved PAHs from tire debris would be very low. However, the oral exposure of aquatic organisms using small particles as food source such as zooplankton and some mollusc species can be relevant.

In the quoted BLIC document, acute toxicity tests on aquatic organisms by measuring inhibition of algae growth, inhibition of mobility of chlocera crustacean and mortality of fish indicate an acute NOEL of some grams of tyre debris per liter. These data seem reasonable, but recent experiments (Stephensen et al., 2003) indicate induction of EROD activity on fish exposed to tyres. Exposure was made by submerging new car tyres in 400 L thanks one day before putting fish in the tanks. No PAHs analysis was performed during the experiments. A quantitative comparison between this kind of exposure and a realistic exposure of natural water bodies cannot be made.

Completely different is the situation of the sediment compartment. Particulate bound PAHs are transported to sediments and can, due to their persistence, accumulate in it. Again, useful information to properly assess exposure in sediments has not been provided. Seston (sedimenting material) sampled in central Stockholm have been extracted and fractionated. The fractions were injected into rainbow trout eggs, and the enzyme activities were then studied in the larvae. One of the studied fractions contained PAHs, and this fraction from all sampling stations increased EROD activity, and from some stations glutathione activity as well (Åkerman et al., 2002).

About bioavailability of PAHs in sedimented tyre debris, sediment-dwelling organisms, as well as detritophagous fishes, may ingest the sediment and it is reasonable to suppose that, in the conditions of the digestive system of vertebrates and invertebrates, PAHs release from particles may be relatively high. Therefore, a risk for sediment-eating organisms and a transfer in the aquatic trophic chain is likely to occur.

The possible risk for the sediment community of surface water bodies is confirmed by experimental field and laboratory studies on the effects of motorway runoff on freshwater ecosystems (Maltby et al., 1995a, 1995b). These studies indicate that the aquatic community exposed trough sediment shows significant effects, while the effects on aquatic organisms exposed trough water are low or negligible.

Similar comments can be made for the terrestrial compartment. Some experimental data reported in a Danish document (Fauser 1999), indicate a tyre debris concentration of 630 mg/kg and 50 mg/kg in top soil at 0 and 30 m from an highway respectively, showing a reduction of more than 90% in 30 m. Assuming again a PAHs concentration in tyres of 100 mg/kg, this led to a PAHs concentration of 63 and 5 µg/kg respectively. Taking into account the potentially high bioavailability for soil dwelling organisms, these concentrations are not negligible.

Conclusion

The information provided does not allow a precise quantitative assessment of the emissions of tyre debris into the different environmental compartments, nevertheless it is reasonable to assume that most of the debris remain on the top soil or are transported into surface water by runoff.

No precise information is provided on the release of PAHs in water from debris, although it is reasonable to assume that the concentration of PAHs in water and the bioavailability for water organisms should be low, even if some experimental data reported in the literature show controversial results.

Most of the debris in the aquatic environment is accumulated in sediments where it can potentially reach not negligible concentration, although a quantitative exposure assessment cannot be made. However, due to the increased bioavailablity of PAHs, a risk for sediment organisms is confirmed by experimental studies, and a transfer in the aquatic trophic chain cannot be excluded.

The only quantitative figures provided refer to top soil close to a highway, indicating a potential risk for soil-dwelling organisms, even if limited to a reduced area around the road, and the possibility of transfer in the terrestrial trophic chain.

Question 1

> *Is it scientifically justified to conclude that certain PAHs could cause serious carcinogenic, mutagenic or reprotoxic effects for human?*

In its opinion expressed at the 24th CSTEE plenary meeting, (2001) CSTEE has recommended to consider PAHs as likely animal carcinogens. Of the 33 PAHs which have been evaluated by IPCS (1998) 17 are or have been suspected of being carcinogenic in laboratory animals. More recently RIVM evaluated 17 PAHs and

considered that only 4 have not to be regarded as carcinogenic, the others are either accepted or suspected carcinogens.

The carcinogens beyond reasonable doubt are benz[a]anthracene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, chrysene, dibenz[a,h]anthracene, and indeno[1,2,3-cd]pyrene and have been classified by IARC. Acenaphtene, acenaphtylene, fluoranthene, phenanthrene, and pyrene have been considered suspected carcinogens. Only naphthalene, anthracene, benzo[ghi]perylene, and fluorene have been considered non-carcinogenic.

Recently naphthalene has been shown to be carcinogenic in rats and mice (NTP 2000). Other PAHs like dibenzo[a,l]pyrene which may have a 100 times higher carcinogenic potency than B[a]P have not been evaluated so far. Several PAHs containing mixtures like coal tars and some exposure circumstances related to PAHs like coal production have been investigated in adequately designed epidemiological studies and proved to be carcinogenic to humans.

Thus, the CSTEE considers PAHs as likely carcinogens for man. This evaluation is based on the fact that many individual PAHs proved to be carcinogenic in a variety of experimental systems, whereas for no individual PAHs the conditions have ever existed for carrying out an epidemiological study allowing for the control of any confounding effect from any other PAHs. This is why no individual PAHs - not even BaP - is evaluated by IARC as "sufficient evidence of carcinogenicity to man“.

Question 2

Is it scientifically justified to conclude that PAHs are emitted into the environment as a consequence of the abrasion of the tyre tread?

Although the information provided does not allow a quantitative assessment of the emissions of tyre debris into the different environmental compartments, it is scientifically justified to conclude that PAHs are emitted into the environment as a consequence of the abrasion of the tyre tread.

Question 3

Can a reduction of the concentration of PAHs in tyres reduce significantly the overall concentration of PAHs found in the environment?

A reduction of the concentration of PAHs in tyres will insignificantly reduce the overall concentration of PAHs found in the environment. CSTEE concludes that, on a mass balance basis, tyre wear contributes less than 2% to BaP exposure associated with respirable particles in the urban atmosphere and represents less than 2% of the total anthropogenic BaP emissions. Since at least a semiquantitative correlation exists between BaP and total PAHs content of the material contribution of total PAHs in tyre wear will represent a similar percentage to PAHs in ambient air.

In sediments a 4% contribution of tyre wear to the total automobile exhaust emissions is estimated. Automobile emissions represent about 30% of the total PAHs contents in sediments so that PAHs in tyres amount to about 1% to the total PAHs concentrations in sediments. Consequently, at present a limitation of PAHs in tyres will not considerably affect the PAHs concentrations in ambient air and in sediments.

Question 4

Can benzo(a)pyrene be seen as a marker for the group of PAHs? Is it scientifically justified to conclude that a reduction of the concentration of benzo(a)pyrene would also lead to a reduction of the other PAHs?

According the data provided by UBA, there is a significant correlation between BaP and total PAHs content. This is in agreement with the previously released CSTEE opinion of June 12, 2001, wherein CSTEE concludes that B[a]P may be used as a semi-quantitative marker for the presence of carcinogenic PAHs in ambient air or other environmental matter. However, its application for the quantitative evaluation of the carcinogenic potency of these matrices is questionable because there are also other carcinogenic PAHs present in varying concentrations with differing mutagenic and/or carcinogenic potencies. Adequate information has not been made available by the different reports submitted to CSTEE.

Question 5

How does the CSTEE evaluate the risks to human health or the environment from PAHs that are emitted into the environment as a consequence of the abrasion of the tyre tread?

As described under Question 3, PAHs from tyre wear contribute less than 2% to PAHs exposure associated with respirable particles in the urban atmosphere. PAHs are genotoxic carcinogens and current knowledge considers such mechanisms non-thresholded and a linear extrapolation is justified. Accordingly, a 2% contribution to the total PAHs is associated with 2% of the cancer risk resulting from human PAHs exposure.

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