

# A review of vehicle related metals and polycyclic aromatic hydrocarbons (PAHs) in the UK environment

Fiona Napier\*<sup>1</sup>, Brian D'Arcy<sup>2</sup> and Chris Jefferies<sup>1</sup>

<sup>1</sup> Urban Water Technology Centre, University of Abertay Dundee, Bell Street, Dundee, Scotland, DD1 1HD

<sup>2</sup> Central Advisory Unit, Scottish Environmental Protection Agency, Perth, PH2 0PA

**Abstract** This paper considers the importance of automobiles as a continuing source of persistent pollutants in the environment, using the UK as a case study. The mass of lead, copper, zinc and polycyclic aromatic hydrocarbons (PAHs) being released by automobiles in the UK is estimated. It is shown that as other sources are reducing, automobiles are now major sources of copper, zinc and PAHs to the environment, and are trending upwards. The paper also discusses the measures available to reduce and control their impact on the environment. A number of options are available to policy makers that have the potential to reduce the environmental impact of motor vehicles, and technologies exist which can greatly reduce pollutant release from vehicles, but these are not yet widely used. Action focusing solely on one pollutant has the potential to deliver a reduction of one source, but with the possible effect of enhancing another. A holistic consideration of all impacts is desirable.

**Keywords** Automobiles; copper; PAH; pollution; traffic; zinc

## INTRODUCTION

There are currently 28 million cars on UK roads, accounting for 79% of road traffic. This is an 83% increase from 1980, and numbers are growing year on year (DfT, 2004). Figure 1 shows the increase in UK vehicle usage over the past 5 decades.

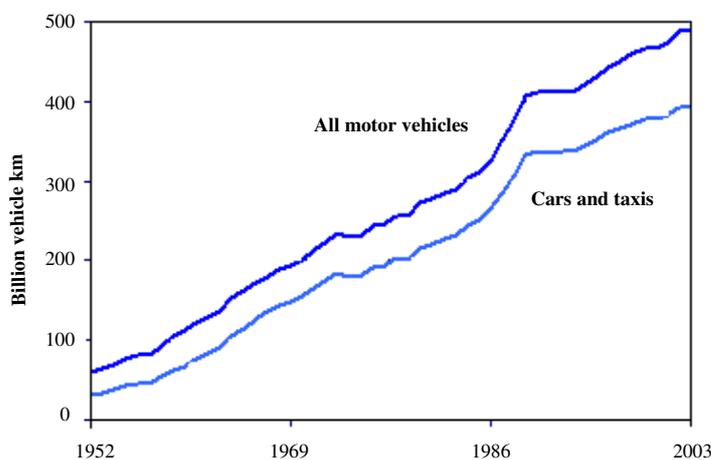


Fig 1. Increase in vehicle usage in UK 1952-2003 (National Statistics Online)

Urban runoff is a major cause of pollution, and is the source of many pollutants which can seriously affect the water quality and ecology of receiving waters (Wilson *et al*, 2005, Gray, 2004). In Scotland some 500km of Scottish rivers have been downgraded as a result of urban drainage (SEPA, 1999). Wilson *et al* 2005, suggested that for PAHs, combustion sources predominated from many of the urban catchments, implicating road traffic, since smokeless zones predominate in the UK. This paper considers the importance of automobiles as a

continuing source of persistent pollutants in the environment, using the UK as a case study, and discusses the measures available to reduce and control their impact on the environment.

## **METHOD**

This was a desk study, using published data from several sources to estimate the mass of heavy metals and polycyclic aromatic hydrocarbons (PAHs) being released by motor cars in the UK. Pollutant emissions from four vehicular sources were considered; oil spills and leaks, brake wear, tyre erosion and exhaust emissions. The values used were all from 2003, as this was the latest year for which there was a comprehensive range of data available, and data used are for passenger cars only (the only exception are the figures for oil, as the only data available for oil losses was for all vehicles). An estimated 495 billion vehicle kilometers were travelled in the UK in 2003 (National Statistics Online), and this figure was used in all relevant calculations.

### **Pollutants chosen for case study**

Motor vehicles are responsible for the release of a range of contaminants to the environment, of which PAHs and heavy metals are among the most toxic. They also persist in sediments, with the potential for future release to the environment, for biological uptake, and with potential enhanced management costs for disposal of stormwater derived sediments. It was therefore decided to focus on these pollutants.

With literally hundreds of thousands of PAH isomers possible, the logistics make it impossible to monitor all compounds, and only a fraction of the total have even been identified (Harrison, 2001). The USEPA have chosen 16 to represent the family, and these compounds have become the standard suite for environmental studies. The PAH values given here are for total PAH content based on the USEPA 16.

Motor vehicles are a source of toxic metals. Of most concern in the aquatic environment are copper (Cu), zinc (Zn), cadmium (Cd), mercury (Hg) and lead (Pb). The EU End of Life Vehicles Directive addresses pollution from several metals, namely mercury, lead, chromium and cadmium, with a phasing out of their use in car manufacture. This means that traffic is unlikely to be a major source of these metals in the future. However, the Directive allows several exemptions, most notably for lead, and it was therefore decided to include lead in this study. Copper and zinc were also included in the study as their concentrations are likely to continue to rise with traffic volume.

### **Data sources**

#### *Oil losses*

Ellis and Chatfield (2000) estimated that 16000t of oil per annum are released to the environment as vehicles via spills and leaks, and this figure was used in the calculations. Average concentrations of 125 mg/l (Zn), 2mg/l (Cu) and 1.1mg/l (Pb) were assumed (Davis *et al*, 2001), and a conversion of 1142 litres of motor lubricating oil to one tonne (DTI 2005).

It is difficult to obtain an estimate for total PAH content of lubricating oil. A report prepared for the United Nations Environment Programme (INCHEM, 1998) reports levels of most PAHs in unused oils  $\leq$  1mg/kg, with phenanthrene, anthracene, and fluoranthene present  $\geq$  10mg/kg. Taking the USEPA 16 PAHs, this would give an estimate of 43 mg/kg total (although this could clearly be underestimating the total for all PAH compounds). Coupe *et al*

(2005) report much higher concentrations of PAHs in used oil compared to unused – up to 1000 fold. This indicates a PAH content of up to 43 000 mg/kg. For the purpose of this case study, it was decided to assume that only half of the oil released to the environment is used oil, and a PAH content of 20 000 mg/kg was used in the calculations.

### Brake wear

The quantity of copper released by brake erosion was assumed to be 75 µg/km/vehicle , zinc 89 µg/km/vehicle and lead 3 µg/km/vehicle (Davis et al., 2001).

### Tyre erosion

Values for tyre erosion ranged from 0.0012 to 0.2g/km/vehicle (National Air Emissions Inventory<sup>1</sup>; Councell *et al*, 2004; Davis *et al*, 2001; Novotny and Olem, 1994). Apart from the lowest value of 0.0012g/km/vehicle (NAEI), all the remaining values fell between 0.125-0.2g/km/vehicle, and a figure of 0.125 g/km/vehicle was used for the calculations (the discrepancy between values is discussed later). A PAH content of 350mg/kg was assumed (ENDS, 2005), Cu µg/g, Pb µg/g and Zn 3400 µg/g (metal values from Davis et al, 2001).

### Exhaust emissions

The 2003 values for exhaust emissions were all taken from the UK Air Emissions Inventory (National Air Emissions Inventory<sup>2</sup>); Cu 0.4t, Pb 1.1t, Zn 1.0t, PAHs 130t.

## RESULTS

Using the data described, the mass of each pollutant deriving from car sources in the UK was calculated. The results are shown in Table 1 and Figure 2.

Table 1. Estimates of inputs to environment from vehicular use in 2003 (passenger cars)

	Tyre erosion (t)	Brake erosion (t)	Oil losses (t)	Exhaust (t)	Total vehicular (t)
Copper	0.3	24	0.038	0.4	37.74
Lead	1.0	1.5	0.02	1.1	3.62
Zinc	990	44	2.3	1	1037.30
PAHs	21.7		320*	130	471.70

\* value for all road vehicles

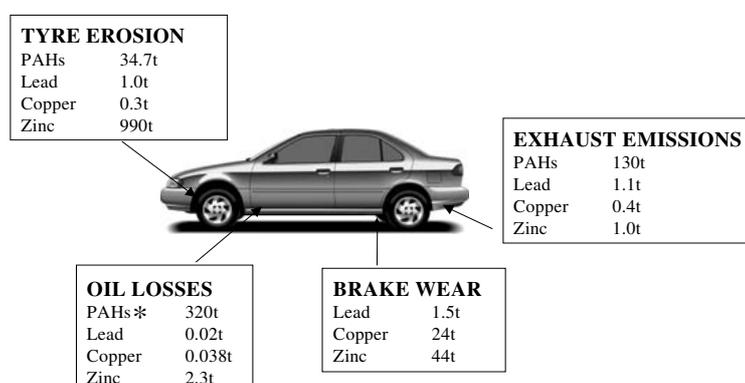


Fig 2. Derived values for estimated pollutant emissions from car components

## DISCUSSION

### Significance of car sources

Figure 3 compares the total car pollutant emissions calculated for the UK (2003) with total emissions from all sources for the same year (total values from National Air Emissions Inventory<sup>3</sup>).

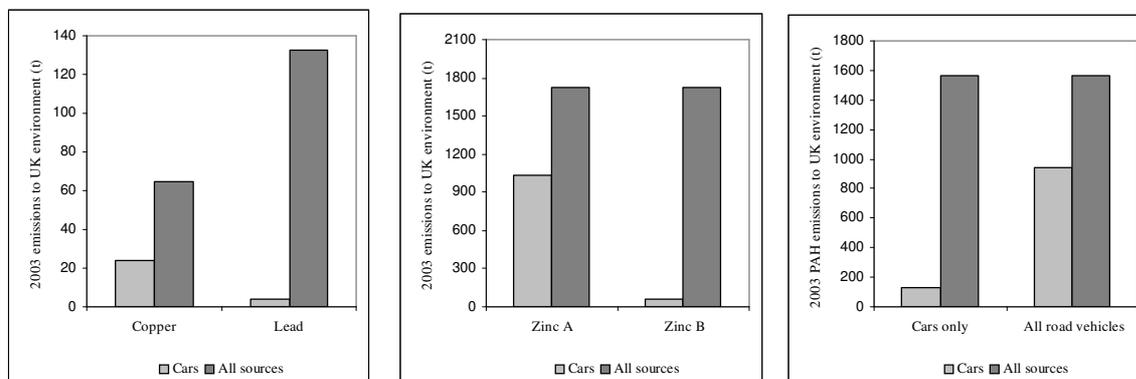


Fig. 3. Pollutant emissions from car sources vs. emissions from all sources

It can be seen that car-derived copper is now the largest single source of atmospheric copper pollution in the UK. This was not always the case, and Figure 4 shows that while total emissions are steadily reducing, the percentage from cars is increasing. This trend can be expected to continue as vehicle numbers continue to rise.

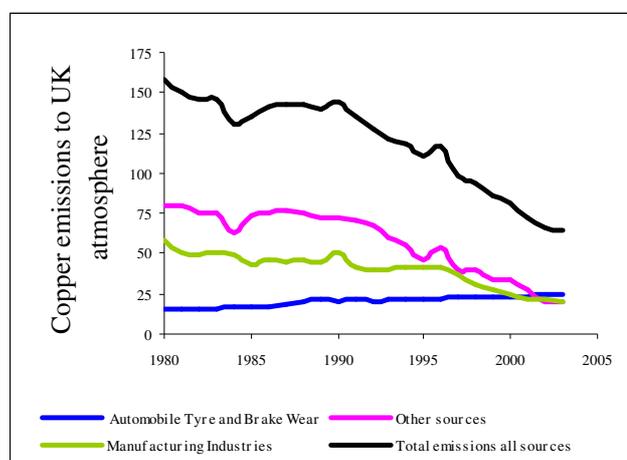


Fig. 4. UK copper emission trends

The drop in atmospheric lead due to reductions in fuel lead concentrations is well documented, and the banning of leaded fuel is a good example of effective environmental legislation. The effect on the UK atmosphere has been dramatic, and reflects similar trends in other countries that have banned leaded fuel. However, similar reductions have not been seen in all environmental compartments. It is clear from Figure 5 that the dramatic drop in air concentrations is not reflected in total riverine concentrations. A study of lead concentrations in riverine, marine and terrestrial systems in northwest Europe by Hagner (2002) reported a similar finding. This study also noted that lead concentrations have not declined in aquatic organisms, with freshwater species being more strongly influenced by the long-term

accumulation of lead in soil and aquatic systems, arguing strongly for minimising future inputs.

The data for car-related zinc are more variable than for copper and lead. The National Air Emissions Inventory (NAEI<sup>3</sup>) reports 89 tonnes of zinc released to atmosphere via car brake and tyre erosion combined (Zinc B in Fig.3). There is no breakdown of this figure into individual figure for brake and tyre erosion, but the tyre erosion rate assumed was 0.0012g/km/vehicle. This figure is at odds with values from other sources, which range from 0.125 – 0.2 g/km/vehicle. If the 0.125 value is used with the UK vehicle usage figure for

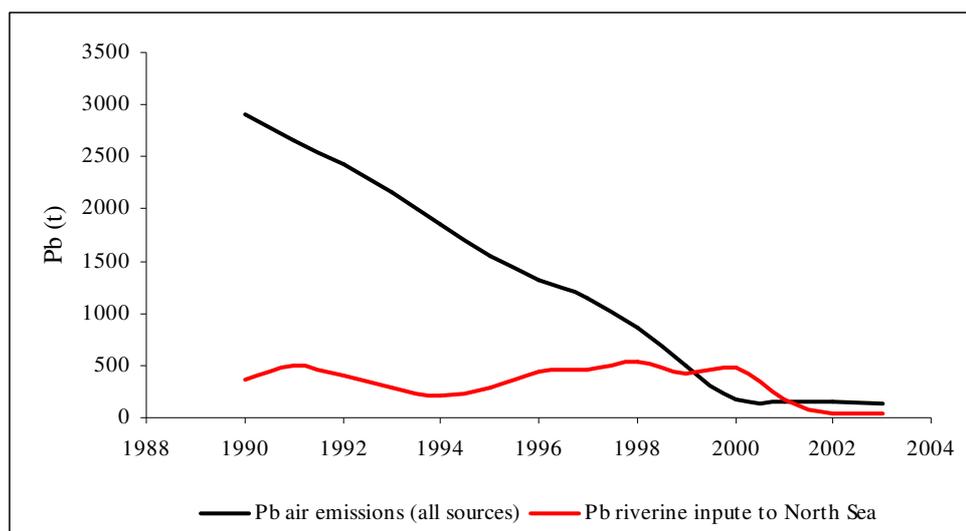


Figure 5. Comparison of lead in UK atmosphere and rivers (National Air Emissions Inventory<sup>3</sup>).

2003, a figure of 990t of zinc from car tyre erosion is derived (Zinc A in Fig.3).

As with copper, traffic has now become the major source of PAHs to the UK environment. Beasley and Kneale, 2002 reported that fuel and oil leaks from parked cars were a major contributor to high downstream PAH levels, and the figure for PAHs from oil losses derived in Table 1 support that. The estimated PAHs emissions from cars may look small compared to the total to atmosphere from all sources, but the data for total road vehicles is much higher, and is the dominant source, as shown in Figure 3. This is due to the fact that most cars are still petrol, while HGVs are diesel and produce far more PAHs. However, with numbers of diesel cars increasing, the contribution from car sources will also increase.

Even the conservative estimate of PAHs from oil losses given in Table 1 is considerable. When the contamination from both oil losses *and* exhausts is considered, it is apparent that cars are contributing large quantities of PAHs to both the air and aqueous environment. Without preventative action, this will continue.

### Controls

The aim of this study was to determine whether vehicles are a *continuing* source of metals and PAHs in the aquatic environment. To fulfil this aim as fully as possible, it is necessary to consider the efforts currently being made to curb the release of these pollutants, and also anticipate the impact of proposed future actions.

There are a number of methods available to policy makers for controlling the polluting effect of motor vehicles, and most fall into one of two categories; traffic management measures and automobile technologies.

### *Traffic management*

The simplest and most effective solution is to reduce vehicle numbers. Figure 6 shows that as vehicle usage continues to rise, vehicle-related pollution also rises if no action is taken to prevent it.

The UK Government White Paper *The Future of Transport* (2004) (DfT, 2004) sets out Government strategy towards transport and the environment. The emphasis is focused on reducing CO<sub>2</sub> emissions and meeting EU objectives for air quality, reflecting the current focus on climate change. The clear indications are that this will

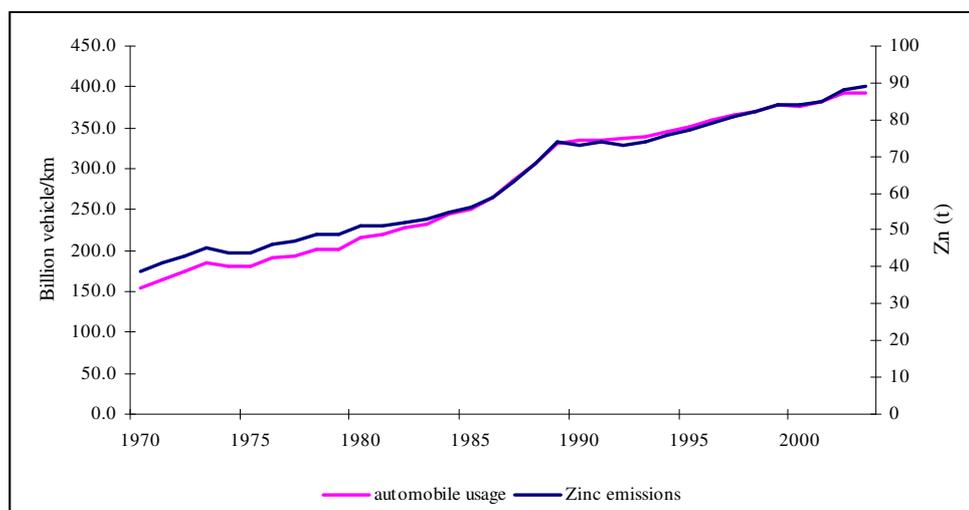


Fig 6. UK trends in vehicular emissions of particulate zinc, and annual vehicle usage.

be the focus of policy, certainly in the short-term. However, this has implications for the control of other pollutants, especially PAHs. Diesel vehicles are becoming more numerous on European roads, for a number of reasons. Diesel engines are more fuel-efficient than petrol, a strong driver for individual motorists to choose a diesel vehicle, and also commercial fleets. Diesels also produce less CO<sub>2</sub> than petrol vehicles, which is necessary to slow climate change. However, PAH concentrations in diesel vehicle exhausts are higher than in petrol vehicle exhausts (Miguel et al, 1998; Rogge et al, 1993; Van Metre et al, 2000) and as diesel numbers increase, a corresponding rise in PAH emissions will follow if preventative action is not taken.

Traffic flow characteristics greatly affect emission of pollutants, and traffic management can reduce or increase vehicle related pollution. Free flowing traffic allows more efficient engine operation, less erosion of brakes and less build up of contaminants on road surfaces at traffic “hot-spots”. For example, it has been estimated that speed bumps could increase emissions by up to 117% for carbon monoxide (CO), 195% for nitrous oxides (NO<sub>x</sub>) and 148% for total hydrocarbons (Daham *et al*, 2005). Since PAHs are well correlated to both CO and NO<sub>x</sub> (Lim *et al*, 1999), it is reasonable to assume that PAH emissions would be increased also, as well as metals from brake erosion. The study concluded that this effect could be reduced by the use of cameras to reduce speed rather than bumps, as drivers would maintain a constant low speed instead of following a pattern of braking and accelerating. This example shows that by

taking a more holistic approach to traffic management, a single action can achieve multiple goals.

#### *Automobile technologies*

Reducing toxic metals used in automobile construction would reduce pollution. The European ELV Directive goes some way to achieving this, but car manufacturers can do more. For example, it is possible to use ceramic brakes in cars rather than copper. A simple solution, but one that is not currently in use.

The use of particulate traps and hydrocarbon absorbers greatly reduces tailpipe emissions of hydrocarbons and particulate matter (and consequently PAHs), but again, these are not yet widely used. Particulate traps will be required by EU legislation on all new diesel vehicles by 2008, but older vehicles will continue to pollute if owners are not forced to retrofit the devices.

It should be noted that abatement technologies, while reducing one type of pollution, can themselves cause pollution of another sort. Catalytic converters are very effective at reducing tailpipe emissions of nitrous oxides, carbon monoxide and hydrocarbons, but environmental concentrations of the platinum group metals used in the converters are rising as a result of catalyst erosion.

#### *Drainage infrastructure*

Conventional drainage infrastructure allows pollutants accumulated on hard surfaces to be washed directly into water courses. Sustainable urban drainage structures (SUDS), however, are increasingly being used to mitigate the negative impact of diffuse sources of pollution, including vehicle-derived pollutants in runoff. As they function primarily by trapping pollutants by filtration or sedimentation processes, persistent pollutants accumulate in the SUDS sediments. So while effective at preventing persistent pollutants reaching water bodies, SUDS technology should be viewed as a partial solution to managing the pollution from urban runoff – prevention measures will reduce the significance of sediment contamination, as other pollutants degrade in the systems and flows are attenuated.

## **CONCLUSIONS**

1. As vehicle usage continues to rise, vehicle-related pollution can also be expected to rise, if no action is taken to prevent it
2. Vehicles continue to be a source of persistent, toxic pollutants to the UK environment. As other sources have been reduced, vehicle usage has become the predominant source of copper and PAHs to the UK environment.
3. Technologies exist which can greatly reduce pollutant release from vehicles, but these are not yet widely used.
4. A number of options are available to policy makers and manufacturers that have the potential to reduce the environmental impact of motor vehicles. Action focusing solely on one pollutant has the potential to deliver a reduction of one source, but with the possible effect of enhancing another. A holistic consideration of all impacts is desirable.

## REFERENCES

- Baekken, T. 1994 Effects of highway pollution on a small Norwegian lake *Science of the Total Environment* 146/147 131-139
- Beasley, G., and Kneale, P., 2002 Reviewing the impact of metals and PAHs on macroinvertebrates in urban watercourses. *Progress in Physical Geography* 26, 2 pp 236-270
- Callender, E and Rice, K 2000 The urban environment gradient: anthropogenic influences on the spatial and temporal distributions of lead and zinc in sediments *Environmental Science and Technology* 34(2) 232-238
- Councell, T.B., Duckenfield, K. U., Landa, E.R. and Callender, E. 2004 Tire-wear particles as a source of zinc to the environment *Environmental Science and Technology* 38, 4206-4214
- Coupe, S., Lowe, T., Smith, H. and Eastwood, I 2005 The effects of Nutrients on the Biodegradation of Used and Unused Oil in Permeable Pavement Systems. In Proceedings of the 3<sup>rd</sup> National Conference on Sustainable Drainage, Coventry, UK. 222-230
- Daham, B., Andrews, G., Li, H., Partridge, M., Bell, M. and Tate, J. 2005 Quantifying the effects of traffic calming on emissions using on-road measurements. Conference paper presented at Society of Automotive Engineers (SAE) 2005 World Congress and Exhibition, Detroit
- Davis, A., Shokouhian, M. and Ni, S., 2001 Loading estimates of lead, copper, cadmium and zinc in urban runoff from specific sources *Chemosphere* 44 997-1009
- Defra: e-Digest of Environmental Statistics available online at ;  
<http://www.defra.gov.uk/environment/statistics/supp/spkf04.htm> (re-checked 17.7.06)
- Department for Transport: Government White Paper *The Future of Transport* 2004. Available at; [http://www.dft.gov.uk/stellent/groups/dft\\_about/documents/divisionhomepage/031259.hcsp](http://www.dft.gov.uk/stellent/groups/dft_about/documents/divisionhomepage/031259.hcsp) (re-checked 17.7.06)
- DTI <sup>(1)</sup> Digest of United Kingdom Energy Statistics 2005, available from DTI at;  
<http://www.dti.gov.uk/energy/inform/dukes/dukes2005/index.shtml> (re-checked 17.7.06)
- Ellis, J.B. and Chatfield, P. 2000 Oil and Hydrocarbons. In D'Arcy, B.J., Ellis, J.B., Ferrier, R.C., Jenkins, A. and Dils, R. (Eds) 2000 Diffuse Pollution Impacts. Terrence Dalton: Suffolk
- ENDS 2005 DEFRA consults on PAHs in tyres *ENDS Report* 360 2005 p42. Available at;  
[http://www.endsreport.com/index.cfm?action=report.article\\_printable&articleID=13605](http://www.endsreport.com/index.cfm?action=report.article_printable&articleID=13605) (re-checked 17.7.06)
- Gray, L. 2004 Changes in water quality and macroinvertebrate communities resulting from urban stormflows in the Provo River, Utah, USA *Hydrobiologia* 518 33-46
- Hagner, C. 2002 Regional and long-term patterns of lead concentrations in riverine, marine and terrestrial systems and humans in Northwest Europe. *Water, Air and Soil Pollution* 134 pp1-39
- INCHEM 1998 Environmental Health Criteria 202: Selected non-heterocyclic polycyclic hydrocarbons. Available online at;  
<http://www.inchem.org/documents/ehc/ehc/ehc202.htm#SubSectionNumber:3.2.1> (re-checked 17.7.06)
- Lim, L. H., Harrison, R.M., Harrad, S., 1999. The contribution of traffic to atmospheric concentrations of polycyclic aromatic hydrocarbons *Environmental Science and Technology* 33, 3538-3542
- Miguel, A., Kirchstetter, T., and Harley, R. 1998 On-road emissions of particulate polycyclic aromatic hydrocarbons and black carbon from gasoline and diesel vehicles *Environmental Science and Technology* 32: 450-455
- National Air Emissions Inventory<sup>1</sup>, available at;  
[http://www.aeat.co.uk/netcen/airqual/naei/annreport/annrep99/app1\\_29.html](http://www.aeat.co.uk/netcen/airqual/naei/annreport/annrep99/app1_29.html) (re-checked 17.7.06)
- National Air Emissions Inventory<sup>2</sup>, available at; <http://www.naei.org.uk/pollutantdetail.php> (re-checked 17.7.06)
- National Air Emissions Inventory<sup>3</sup>, available at;  
[http://www.naei.org.uk/emissions/emissions\\_2003.php?action=unece&options=1PAY](http://www.naei.org.uk/emissions/emissions_2003.php?action=unece&options=1PAY) (re-checked 17.7.06)
- National Statistics Online. Available at;  
[http://www.statistics.gov.uk/cci/nugget\\_print.asp?ID=1096](http://www.statistics.gov.uk/cci/nugget_print.asp?ID=1096) (re-checked 17.7.06)
- Novotny, V. and Olem, H 1994 *Water Quality: Prevention, Identification and Management of Diffuse Pollution* 1<sup>st</sup> ed. Van Nostrand Reinhold:New York
- Rogge, W.F., Hildemann, L. M., Mazurek, M. A., Cass, G. R., and Simoneit, B. R. T. 1993 Sources of fine organic aerosol. 2. Noncatalyst and catalyst-equipped automobiles and heavy-duty diesel trucks. *Environmental Science and Technology* 27: 636-651
- SEPA (1999) *Improving Scotland's Water Environment. SEPA State of the Environment Report*. Scottish Environment Protection Agency, Stirling. Available online at;  
[http://www.sepa.org.uk/publications/state\\_of/index.htm](http://www.sepa.org.uk/publications/state_of/index.htm) (re-checked 17.7.06)
- SEPA: OSPAR data provided by Judy Dobson.
- Van Metre, P., Mahler, B., Furlong, E., 2000. Urban sprawl leaves its PAH signature *Environmental Science and Technology* 34:4064-4070
- Wilson, C., Clarke, R., D'Arcy BJ, Heal KV and Wright PW (2005) Persistent pollutants urban rivers sediment survey: implications for pollution control. *Water Science & Technology* Vol 51 No. 3-4 pp 217-224.