

Evidence

Leaf litter in street sweepings: investigation
into collection and treatment

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Miranda Kavanagh

Director of Evidence

Executive summary

Some local authorities operate dedicated street sweeping rounds to collect leaf litter in the autumn and early winter. The street leaf sweepings waste collected is variable in nature and composition. As well as containing silt, grit, water, litter and leaves, it is also likely to contain a range of contaminants such as oil, diesel, dog faeces, small dead animals, heavy metals and salt. The exact amount of this waste collected is unknown, as the data for street leaf sweepings are not reported separately to general street sweepings, but evidence suggests a range from tens to thousands of tonnes per year. Current Environment Agency guidance, published in 2012, is that street leaf sweepings should not be composted.

In 2012 and 2013, the Environment Agency carried out a series of trials with local authorities involving the sampling and analysis of batches of seasonal street leaf sweepings waste, treatment of these batches (principally by commercial-scale composting with other wastes), and sampling and analysis of the treated outputs. The aim of these trials was to identify generic circumstances whereby seasonal street leaf sweepings could be collected and composted without compromising existing compost quality and environmental protection. It was based on an earlier, small-scale study in Wales in 2011 that detected the presence of potentially toxic elements including chromium, copper, molybdenum, nickel and zinc.

A total of 22 local authorities participated in the trials, providing more than 60 samples of street leaf sweepings and 30 samples of composted output. Greatest participation was from authorities in the south and south-west of England, with one authority in Wales and two in the north-west of England. As the local authorities were self-selecting and participation was low, it was challenging to draw reliable conclusions to give a national perspective on the composition of collected street leaf sweepings or the subsequent quality of their composted outputs but it was possible to make some important observations about the data collected.

Levels of physical and chemical contaminants were found to vary considerably within samples of compost from source-segregated green waste and street leaf sweepings. In general, the concentrations of potentially toxic elements such as arsenic, chromium, copper, lead, nickel and zinc were not too dissimilar to levels observed in green wastes and green and green/food composts. However, levels of polycyclic aromatic hydrocarbons (PAHs) were found to be much higher in street leaf sweepings and the composted output than in the other materials. PAH contamination may originate from a number of polluting sources including vehicle exhaust emissions, oil spills and leaks, and tyre wear. Some PAHs are known genotoxic carcinogens and pose a significant risk to human health and the environment.

The main conclusion from this current trial is that most samples of composted street leaf sweepings are not suitable for agricultural or horticultural use but may be suitable, as a compost-like output, for less sensitive uses such as in the redevelopment or reclamation of previously developed land. It is recommended that the current Environment Agency position on the use of street leaf sweepings to produce compost for agricultural or horticultural use remains unchanged.

Although this trial was larger than the previous Welsh study, the data were not always sufficient to draw reliable conclusions. Recommendations are made for further work that could be carried out to better understand the variability of contaminants within street leaf sweepings and the effects of different collection methods, and the routes and timing of collections, within individual authorities and on a national basis. Further studies on pretreatment and composting options to tackle the issue of PAHs might also be beneficial.

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Torridge District Council

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1 Introduction

This report presents the findings of a series of trials carried out by local authorities and commercial-scale composters between late 2012 and early 2013 involving:

- the collection, sampling and analysis of batches of seasonal street leaf sweepings waste
- the treatment of these batches, principally by commercial scale composting with other wastes
- the sampling and analysis of the treated outputs

The trials were managed by the Environment Agency and the Organics Recycling Group (ORG), the main trade body for commercial-scale composters. The main aim of the trials was to identify generic circumstances whereby seasonal street leaf sweepings could be collected and composted without compromising compost quality and safety.

The report was peer reviewed by an independent expert, Dr Graham Merrington of WCA Environment Limited. His findings are included as Appendix D.

1.1 Background to the trials

Street sweeping waste is very variable in nature. As well as containing grit, water, litter and leaves it is likely to contain a variety of physical, biological and chemical contaminants including glass, plastic and metal, dog faeces, small dead animals, salt and pollution from roads and vehicles. See Section 2 and Appendix A for more details on the sources of chemical pollution in street sweeping wastes.

The options for dealing with street sweeping waste are limited. Apart from being landfilled, one option is to treat the waste to separate out and recover some of the components, such as grit and sand. It is well established that street sweeping waste is not suitable for composting.

Some local authorities operate dedicated street sweeping rounds to collect leaf litter in the autumn and early winter. The leaf litter collected as street sweepings will contain variable amounts of the contaminants normally found in street sweepings. The composition of the collected leaf litter will depend on a number of factors such as:

- whether it is collected from rural or urban roads
- whether or not roads have recently been salted or gritted
- the presence of nearby roadworks or road traffic accidents

The amounts that individual local authorities collect annually vary from a few tonnes to hundreds of tonnes. Although local authorities do not typically report the amount of street leaf sweepings collected separately from total street sweepings and gully emptyings, they are considered to be only a small proportion of the total by weight. According to estimates from WasteDataFlow (WDF), one two-tier local authority¹ covering a mixed rural and urban area of around a quarter of a million hectares reported total waste arisings from street sweeping and gully emptying of around 5,000 tonnes for 2011 of which street leaf sweepings were about 1,700 tonnes

¹ A two-tier authority is one responsible for waste management, working closely with other local authorities that are responsible for recycling and waste collection. For example, a county council that works with district and borough councils.

(WasteDataFlow, 2013). More recently, the Environment Agency reviewed data from 352 English waste authorities, based on reported data to WDF for the period October 2012 to March 2013 (WasteDataFlow, 2013). Of these, 109 authorities reported street cleansing recovery totalling less than 70,500 tonnes. Of this figure, just over 9,200 tonnes was reported as being dedicated leaf litter from street cleansing by 19 of the waste authorities.

Commercial-scale composting plays an important role in diverting biodegradable waste from landfill and in recovering value from it. Most of the compost produced is spread on agricultural land, where it is a valuable source of stable organic matter and crop available nutrients, which can be used by farmers to help meet crop nutrient requirements and to maintain soil fertility. Composting, like all other waste treatments, is subject to regulation by the Environment Agency through the environmental permitting regime. The permits control the types and quantities of waste that can be handled and the conditions to be met to prevent any adverse impacts on health and the environment. These include possible adverse impacts from using contaminated or poor quality compost on land used for growing crops. Street leaf sweepings waste is not included as an acceptable waste type in either the exemptions for composting under the Environmental Permitting Regulations or in the Environment Agency's standard rules permits for composting.

Local authorities would like to be able to compost street leaf sweepings for spreading on agricultural land. If they could safely compost street leaf sweepings to produce an agriculturally acceptable product, the recycled material would count towards their recycling and composting targets. One issue that has increased the cost pressures on local authorities for the disposal of street leaf sweepings is the clarification by HM Revenue & Customs of landfill taxation around use of materials as protective layers at disposal sites (HMRC 2012).

In 2012 the Environment Agency published guidance which clarified the position on street sweepings, gully emptying and street leaf sweepings, specifying how and where these wastes could be dealt with (Environment Agency 2012a). The guidance on street leaf sweepings was informed by the findings of a small-scale trial looking at the composting of street leaf sweepings in Wales that indicated that the composted output was significantly contaminated with potentially toxic elements (PTE) including nickel, copper, chromium, molybdenum, zinc and petroleum hydrocarbons (Environment Agency 2012b). The guidance concluded that street leaf sweepings are not regarded as acceptable wastes for composting or anaerobic digestion, but that they could be accepted at appropriate mechanical biological treatment (MBT) facilities that produce a compost like output (CLO) for use on reclamation sites. CLO is classified as 'recovered' waste, which is not considered to be recycled or fully diverted from landfill.

Recognising that the Welsh trial was limited, the Environment Agency wanted to carry out a more detailed and extensive investigation. It therefore worked with ORG to devise the series of further trials described in this report.

1.2 Report structure

Section 2 summarises the findings of a literature review of potential sources of pollution in the roadside environment. These are described in more detail in Appendix A.

Section 3 outlines the trial methodology including aims and objectives, sampling and analysis methods, and assessment of results.

Section 4 sets out the analytical results from the trial and compares the levels found with other materials including green wastes and green and green/food composts.

Section 5 describes the assessment of output quality and suitability for agricultural use.

Section 6 presents the main conclusions and recommendations from the trial. The four appendices provide further information on the literature review, trial methodology, exposure modelling and the findings of the peer review.

2 Characterising leaf litter from roads

The literature on street leaf sweepings and roadside pollution was reviewed to:

- better understand the characteristics of leaf litter from roads
- identify the likely chemical and physical hazards to human health and the environment from its presence in composts

Below is a summary of the findings of the literature review. More details are given in Appendix A.

2.1 Bulk characterisation

Street sweepings (street cleansing materials) include leaves, grit, litter, glass, oils, paper, plastics and other materials (CDEP 2007, Environment Agency 2012a). Street leaf sweepings are a seasonal waste issue, most leaf fall occurring in the autumn and early winter.

Some local authorities operate dedicated leaf litter collections as part of their street cleaning programmes (Environment Agency 2012a). The precise amount of this waste collected is unknown as the data are not reported separately to the Environment Agency. In 2011 those authorities with dedicated collections for seasonal street leaf sweepings reported amounts varying from 10 tonnes to a maximum of 1,700 tonnes (based on WDF figures for all the local authorities in England).

Street leaf sweepings are likely to contain physical contaminants such as plastics, paper, foil and grits, as well dog faeces and animals killed on the roads.

2.2 Chemical characterisation

Street leaf sweepings are generally from:

- trees, hedges and bushes growing close to the road along verges and central reservations
- adjacent land uses including homes and offices

Chemical contamination arises as the plant takes up pollutants from soil and air, and from entrainment of soil, sediment, and dust within plant materials (Kabata-Pendias and Mukherjee 2007, Nicholson et al. 2003).

Important sources of contamination are:

- non-localised atmospheric deposition
- localised deposition from passing traffic and road management
- use of chemicals such as pesticides in landscape management

These sources are discussed in turn below.

2.3 Sources of chemical contamination

2.3.1 Non-localised atmospheric deposition

General atmospheric deposition of contaminants is a significant source of diffuse pollution in urban and rural soils (Nicholson et al. 2003). While atmospheric deposition is ubiquitous, deposition rates depend on the proximity to point sources of pollution such as heavy industry or heavy vehicle use on major roads (Defra 2010).

The most important sources of heavy metals are atmosphere depositions from energy production, mining, metal smelting and refining, manufacturing processes, transport and waste incineration (Environment Agency 2007b).

2.3.2 Localised deposition from road vehicles

Emissions from vehicles make a significant contribution to pollution close to the roadside. The main sources of contaminants from vehicles are (Pulles et al. 2012, Thorpe and Harrison 2008, Winther and Slento 2010):

- particulate and gaseous exhaust emissions
- particulate emissions from other parts of the car such as tyre and brake wear
- liquid leaks from the engine, transmission and coolant systems

Studies have shown that exhaust and non-exhaust sources contribute equally to total traffic-related emissions of particulate matter (Thorpe and Harrison 2008).

Vehicle exhaust emissions are the main source of air pollutants including carbon monoxide, carbon dioxide, nitrous oxides, methane, derived ozone and non-methane hydrocarbons (Colville et al. 2001, Maricq 2007, Tsai et al. 2012, Wang et al. 2013). However, these major gaseous contaminants are unlikely to accumulate in soil or plant matter. Of greater importance are emissions of particulate matter, including heavy metals and partial combustion products like polycyclic aromatic hydrocarbons (PAHs) and the wider range of volatile organic compounds (VOCs) and semi-volatile organic compounds (SVOCs). The main sources of metal emissions from vehicle exhausts are (Pulles et al. 2012, Winther and Slento 2010):

- combustion of fuel
- use of lubricants
- abrasion of the engine or after-treatment system including the catalyst

Engine lubricant oils are known to be high in zinc due to the use of zinc dialkyl dithiophosphate as an anti-wear agent (Pulles et al. 2012). Although engine oils contain a range of additives, metal contents in pure oils are generally kept low to minimise damage to catalytic convertors. In used oils, metal contents can be significantly increased due to engine wear. The increased use of catalytic convertors has also been linked to the release of platinum group elements (PGEs) including platinum, palladium and rhodium into the environment (Kalavrouziotis and Koukoulakis 2009).

Vehicle exhaust emissions are one of the most important sources of VOCs in urban areas and several studies have demonstrated that the concentration and type of VOCs depend on the car type, engine condition and other factors (Wang et al. 2013). The types of VOCs emitted from vehicles under a variety of conditions include:

- benzene, toluene, ethyl benzene and xylenes (BTEX)

- alkanes such as ethane, isopentane and *n*-undecane
- alkenes such as ethene and propene
- aromatics such as 1,2,4-trimethylbenzene and styrene
- carbonyl compounds such as formaldehyde and acetone

Organic compounds commonly found in exhaust particulate matter include PAHs and substituted nitrated PAHs, alkanes, steranes, hopanes and organic acids such as alkanolic and benzoic acids (Maricq 2007).

Brake and tyre wear are an important source of trace metals in the urban environment (Hulskotte et al. 2007, Thorpe and Harrison 2008). Modern brake lining materials are composites of many different ingredients, which are responsible for differing wear characteristics, physical and chemical properties of the bulk lining materials and emitted particles (Hulskotte et al. 2007). Important metal emissions from brake linings include antimony, barium, copper, magnesium and zinc (Gietl et al. 2010, Hulskotte et al. 2007). Rubber losses from tyres have been estimated to be around 50,000 tonnes per year in the UK (Environment Agency 1998). A range of tyre types, sizes and compositions are used in the motor vehicle industry according to required performance (Thorpe and Harrison 2008). Generally, tyres consist of a rubber compound reinforced with steel wires and textiles, and although tyre wear has been considered primarily a source of organic compounds, approximately 13% by mass is inorganic material (for example, zinc oxide), present in the form of various curing agents, accelerators and other additives (Environment Agency 1998, Thorpe and Harrison 2008). Typical emissions of PAH from tyre wear have been estimated to be 22 tonnes per year (Napier et al. 2008).

PAH concentrations in unused oil are generally low but are much higher (up to 1,000 times) in used engine oils (Napier et al. 2008). It has been estimated that around 16,000 tonnes per year of oil is lost as a result of vehicle spills and leaks, which may result in emissions of up to 320 tonnes of PAHs per year.

2.3.3 Localised deposition from roads

Most road surfaces in the UK have the same component materials – a mixture of aggregates of various grain size, bitumen and modifiers such as fillers and adhesives (Thorpe and Harrison 2008). Heavy use and adverse weather can result in significant road surface abrasion and the generation of road dust (Winther and Slento 2010). Although PAHs have been found in bitumen samples, the concentrations of individual compounds are very low due to their removal during the distillation process (Gadd and Kennedy 2003, Winther and Slento 2010). The major components of asphalt have been reported as silicon dioxide (sand), aluminium, iron, calcium, potassium, manganese, magnesium and titanium, with significant trace elements being barium, strontium, zinc, vanadium, lead and chromium. These levels are low relative to metal contamination incorporated from other sources such as brake linings and tyre wear (Thorpe and Harrison 2008).

Road surfaces can also accumulate particulate material from a wider variety of sources including vehicle emissions (as discussed above), salting and gritting activities, and the erosion and deposition of material from the roadside such as mud (Ramakrishna and Viraraghavan 2005, Thorpe and Harrison 2008). The resuspension of this deposited material is an important source of particulate matter. Its composition depends on elements and compounds typically found in rocks and soils, and therefore it will reflect local geology (Thorpe and Harrison 2008). Road dusts and roadside soils often contain significant concentrations of metals including cadmium, chromium, copper, iron, lead, manganese and zinc.

Large quantities of de-icing chemicals are applied to roads and pavements during the winter months to ensure the safety of drivers and pedestrians (Ramakrishna and Viraraghavan 2005). Approximately two million tonnes of salt (sodium chloride) are spread on the UK roads in each year with the majority spread on to motorways, trunk roads and main roads (Salt Union 2013). Less than a third of other roads are treated.

2.3.4 Localised application for landscape management

Management of roadside vegetation, including weed control, is an important part of maintenance for driver safety, to protect engineered formations, and for landscape aesthetics (Ramwell et al. 2002, Barker and Probst 2009). A significant amount of the herbicides applied to hard surfaces are washed away into surface drainage following rainfall. Amenity use of pesticides, including road and roadside applications, includes herbicides (mostly glyphosates) and a combination of algaecides, fungicides, insecticides and moss-killers (HSE 2011). The degradation rate of herbicides is variable and depends on environmental conditions.

2.4 Hazard characterisation

Soils and road dusts may be contaminated with heavy metals, which at high enough concentrations can cause harm to human health and the environment. Of particular concern are likely to be copper, lead and zinc because of a combination of their occurrence in soils and dusts, chemical mobility and toxicity. These elements are not treated by any subsequent composting process. Although much interest has been focused on PGEs, their toxicity to health is still uncertain (WHO 2000).

Soils and road dusts may also be contaminated by organic chemicals including pesticides, oils, PAHs and other compounds such as phthalates. Of particular concern are likely to be PAHs and pesticides. PAHs are likely to remain attached to dust particles rather than taken up by plants and the higher ring compounds are also resistant to biological treatment (Semple et al. 2001). Pesticides such as glyphosates are absorbed by plants through the leaves and are moderately degradable in soil and in compost systems. Both are toxic to health and the wider environment. Petroleum hydrocarbons may be found in street leaf sweepings, but their potential to transfer through the agricultural food chain is likely to be low. Higher toxicity is observed in low weight petroleum hydrocarbons, which are more volatile and biodegradable than heavier compounds, and these are therefore less likely to be found in soils, dusts and finished compost.

Sodium and electrical conductivity may be issues for street leaf sweepings collected during winter months when salting and gritting has taken place. Increased levels of sodium and chloride in soil have been associated with damage to soil quality and structure, and harm to soil microorganisms (Cernhlavkova et al. 2008, Ramakrishna and Viraraghavan 2005).

3 About the trial

3.1 Aims and objectives

The main aims and objectives of the trial were to:

- characterise the chemical and physical properties of street leaf sweepings from various local authority collections across the country to better understand the levels of, and variation in, contamination
- compare contamination levels in street leaf sweepings with other established compost feedstocks to understand the benefits and risks or potential harm that might result from using the output material on land
- establish whether the output material from composting street leaf sweepings is comparable with PAS 100 (BSI 2011) composts made from established source-segregated and well-characterised feedstocks
- establish whether the output material from composting street leaf sweepings is suitable for agricultural and horticultural uses without increasing risk to human health and the environment
- identify if any pretreatment techniques could be used to reduce or remove contamination loading

3.2 Trial options and methodology

The trial methodology is summarised here and set out in detail in Appendix B. It was developed by the Environment Agency and ORG working with local authorities and compost producers. Local authorities were invited to participate in the trial by subscribing to one of the following options:

- Option 1: Assessment of street leaf sweepings and composted sweepings
- Option 2: Assessment of street leaf sweepings only
- Option 3: Assessment of street leaf sweepings and pretreatment before composting.

Local authorities that applied to participate in the trial were self-selecting; no consideration was given as to whether participants were representative on a local or national basis (or part of any randomised sample).

Participants were asked to comply with the trial methodology. Compost operations carried out in relation to the trial were audited by the Environment Agency to ensure that they complied with the trial rules but there was no audit of street sweepings collections.

Street leaf sweepings were collected between October 2012 and February 2013. The street leaf sweepings for each collection round were sampled according to the study methodology and analysed by NRM Ltd at its laboratories in Berkshire for a range of physical and chemical parameters. Participants were also asked to complete a questionnaire that asked a series of questions about how, when and where each batch of street leaf sweepings had been collected. This collection route information included the types of roads and traffic density, and how the street leaf sweepings were collected such as the equipment used and which surfaces were generally swept.

Where local authorities had agreed to option 1, they also sent the collected street leaf sweepings for composting at various trial locations. Where the composting sites were PAS 100 accredited, the Environment Agency visited the sites to ensure the trial did not compromise the production of PAS 100 composts; in particular it insisted on separate drainage and precautions to avoid direct physical contact between the two types of compost. Street leaf sweepings were composted either on their own or after blending with green waste to adjust the carbon to nitrogen levels. The requirement was for batches of street leaf sweepings only and sweepings blended with other green wastes to be composted for a minimum of eight weeks but most were composted for 12 weeks, typically from February 2013 through to the end of April 2013. Each site operator was required to specify the proportions of materials in blended composts. All batches of composted trial outputs were kept separately for each collection round of street leaf sweepings provided. Each batch was sampled at the end of the composting period and the samples, along with samples of green waste if composted as a blending of input materials, were also sent to NRM for chemical and physical analysis.

In addition to the analysis undertaken by NRM, a selected subsample of street leaf sweepings and composted trial outputs were sent to the Environment Agency's laboratories in Nottingham for further chemical analysis for a range of PTEs. The additional elements included antimony, barium, silver, thallium and vanadium, which are known contaminants in roadside environments (see Appendix A for further information).

The analytical results for samples of street leaf sweepings, green wastes and composted trial outputs, and completed questionnaires were submitted to ORG, which acted as the main point of contact for the trial. Collated results from the trial were supplied to the Environment Agency, which subsequently carried out a review of the data supplied and an assessment of the suitability of the composted trial outputs for agricultural use.

One operator had been in discussions with the Environment Agency about their street sweepings treatment operation and possible composting of the organic fraction in autumn and early winter. The authority already operated a dedicated street leaf sweepings collection and therefore the trial was an opportunity to compare the organic output from the treatment process with outputs from composting of untreated street swept leaves. The treatment involved the removal of the leaves and other organic material from the street sweepings by screening and washing-out grit and fine materials. The organic fraction was then composted as described above for a minimum of eight weeks.

3.3 Trial limitations

The most significant limitation was that authority participation in the trial was voluntary and therefore the results were not randomly selected (since the authorities were self-selecting). It was hoped that high participation rates by local authorities would still generate a large and useful dataset but in the end the numbers of participants and samples collected was modest (see Table 4.1). Ultimately this restricted the Environment Agency's ability for statistical interpretation of the dataset (see Sections 4 and 5 for further information).

Only limited parameters for compost quality were analysed and assessed, notably a range of physical and chemical contaminants. A wide range of chemical contaminants may be present in the roadside environment, but chemical analysis was undertaken for only a proportion of these chemicals. While it would have been interesting to present data on platinum group elements (PGEs) and commonly used pesticides, they were not included in the trial on the grounds of cost.

In terms of compost quality, other factors such as the composting conditions were not reported or audited in the trial, which is a general requirement under PAS 100 (BSI 2011).

In addition, the composted trial outputs were not tested for maturity and stability. They were also not analysed for microbial contamination or for the presence of weeds or other pathogens. Although compost maturity and stability affect the availability of nutrients and the potential to cause local pollution in the forms of odour and leachate, for contaminants likely to accumulate in soil, the variability in maturity of the tested materials makes no fundamental difference to the outcomes of the trial. These factors should be taken into account by compost producers when deciding the appropriate use of the treated materials.

4 Trial results

In 2012, the emerging issue of Chalara dieback of ash (*Chalara fraxinea*), a serious fungal disease affecting ash trees, resulted in delays to the collecting, sampling and analysis of street leaf sweepings and the composting trials. In particular, local authorities and compost operators were delayed while the risk of compost contamination with fungal spores was assessed and new guidance issued.

A total of 22 local authorities in England and Wales provided data for the trials (Table 4.1). Greatest participation was from the south and south-west of England, with only one authority in Wales and two in the north-west of England. As the authorities were self-selecting and participation was low, it proved difficult to draw reliable conclusions to give a national perspective on either the composition of collected street leaf sweepings or the subsequent quality of their composts.

Table 4.1 Participation levels and number of material samples in the trials

| Option | Number of participating local authorities | | Number of samples |
|--------|---|--|-------------------|
| 1 | 12 | Street leaf sweepings | 65 |
| 2 | 9 | Green wastes | 7 |
| 3 | 1 | Pretreated street leaf sweepings | 2 |
| | | Composted street leaf sweepings output | 23 |
| | | Composted blended green waste and street leaf sweepings output | 8 |
| | | Pretreated sweepings composted output | 1 |

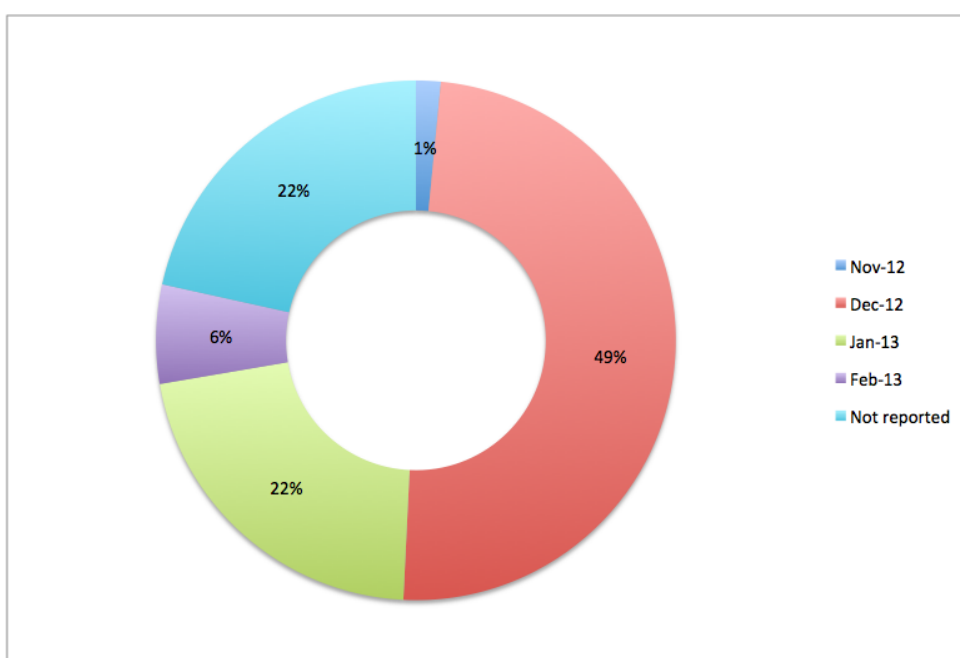
Statistical analysis of the sample data was also difficult because of the small sample size and skewed geographical datasets. For example, 18 of the 23 samples of composted street leaf sweepings analysed for option 1 came from a single authority. Although some duplicate chemical analysis was undertaken by Environment Agency laboratories, often only a single sample was taken for each batch of composted trial output, street leaf sweepings or green waste, and therefore understanding of within batch variability was limited. True randomisation of samples was not possible in this trial and therefore traditional methods of statistical analysis were not applicable. To allow a cautious interpretation of the summary data, a Bayesian approach was used to compare individual datasets (for example, street leaf sweepings and green wastes). Unlike other statistical methods, Bayesian tests do not require randomness in the underlying dataset in order to test or summarise the data.

In subsequent tables and discussion, the results for individual chemical and physical parameters are normally reported as the range and the median value. The median value represents the middle concentration observed in the analysed samples, that is, half the concentrations measured in the samples are greater than the median value and half the concentrations measured are less than the median value. The median value is a measure of central tendency within the sample data and is less affected by a skewed distribution than say an average value.

4.1 Street leaf sweepings

Some 21 authorities provided one or more samples of street leaf sweepings following option 1 or option 2 in the trial methodology. Around 70% of collections took place between December 2012 and the end of January 2013 (Figure 4.1), with almost half of samples collected in December. In total, 65 samples were collected with 22 samples provided by a single authority and five authorities submitting only a single sample each.

Figure 4.1 Collection date of samples of street leaf sweepings reported by participants as a percentage of all samples collected ($n = 65$)



Note: The sampling date was not reported for around 20% of samples.

4.1.1 Collection round characteristics

Local authorities were asked to complete a questionnaire to accompany each sample, which provided information about the collection route. Overall the return rate was good with 52 completed questionnaires submitted (about 80%). However, some inconsistencies were observed in the way the questionnaires were filled in and some gaps were identified. Many parameters were subjective and the overall dataset was too small to draw any strong statistical conclusions.

Not surprisingly, it was reported that most samples were collected from predominantly rural areas (71%) with only a minority from urban areas (21%). About 2% of respondents did not specify.

Collection rounds involving multiple road types were commonly reported. Very few collections took place on major roads, with most samples reportedly collected from minor roads (54%), B roads (31%) and residential streets (31%). Consistent with these types of road, most operators observed only low traffic volumes during sample collection (57%) with a minority (10%) finding high traffic volumes. About 2% of questionnaires did not specify.

Collections most commonly swept the road surface (75%) followed by the pavement (29%) and the roadside verge (23%). Sweeping of only a single surface during a collection round was the most common approach adopted for 66% of pavements and 69% of road surfaces.

4.1.2 Physical characteristics

Samples of street leaf sweepings were submitted for analysis for physical contaminants using the sampling and analytical methods used for the Compost Quality Protocol (CQP) and PAS 100 compliant composts (BSI 2011, WRAP and Environment Agency 2012a). Overall levels of physical contaminants were relatively low (Table 4.2).

Table 4.2 Physical contaminants found in leaf litter samples ($n = 65$)

| Physical contaminant | | Range | Median |
|-----------------------------|---------|---------|--------|
| % w/w of total sample >2 mm | Glass | 0–0.21 | 0 |
| | Metal | 0–0.02 | 0 |
| | Plastic | 0–0.80 | 0 |
| | Other | 0–0.34 | 0 |
| | Total | 0–0.94 | 0 |
| % w/w of total sample >4 mm | Stones | 0–17.16 | 1.66 |

4.1.3 Chemical characteristics

Samples of street leaf sweepings were submitted for chemical analysis using the sampling and analytical methods specified for Compost Quality Protocol and PAS 100 composts (BSI 2011, WRAP and Environment Agency 2012a).

Chemical analysis included:

- major crop nutrients
- organic contaminants including petroleum hydrocarbons and polycyclic aromatic hydrocarbons (PAHs)
- potentially toxic elements (PTEs) including metals and metalloids
- general chemical characteristics such as pH and organic matter content

The results are summarised in Table 4.3. In addition to the main analysis ($n = 65$), the Environment Agency analysed a subset (up to $n = 22$) for additional PTEs as noted in Table 4.3.

There was a high degree of variability in the results. Most contaminant levels were broadly similar to those found in green wastes but levels of PAHs appeared to be consistently high (see next section). Gasoline range organics including BTEX were not present in most samples, as would be expected by their volatility and biodegradability, but heavier molecular weight petroleum hydrocarbons were found in levels up to 2–3% by weight. However, these long-chain hydrocarbons are generally of low environmental concern because of their lack of mobility and toxicity.

Table 4.3 Chemical parameters for samples of street leaf sweepings

| Chemical parameter | | Range | Median |
|--------------------|----------------------------------|-------------|--------|
| Nutrients | Nitrogen (% w/w) | 0.12–1.98 | 1.01 |
| | Phosphorus | 438–2,463 | 966 |
| | Potassium | 441–6,573 | 1,793 |
| | Sulphur | 644–2,220 | 1,220 |
| | Organic matter (% w/w) | 8.3–94.1 | 65.2 |
| Organics | Gasoline range organics (C6–C10) | 5–184 | 5 |
| | EPH (C10–C25) | 245–8,973 | 1,189 |
| | EPH (C25–C40) | 506–21,941 | 2,639 |
| | EPH (C10–C40) | 868–30,913 | 4,233 |
| | PAHs (USEPA 16) | 1–234 | 8.6 |
| | Benzo[a]pyrene | 0.1–20.3 | 0.8 |
| Toxic elements | Antimony *** | 0.14–13.3 | 1.4 |
| | Arsenic | 0.11–77 | 3.2 |
| | Barium * | 39–80 | 62 |
| | Beryllium ** | 0.2–0.5 | 0.4 |
| | Boron * | 4.4–58.9 | 15.5 |
| | Cadmium | 0.04–1.47 | 0.2 |
| | Chromium | 0.26–95.7 | 11.4 |
| | Cobalt * | 2.6–10.6 | 5.8 |
| | Copper | 6–78.6 | 22.1 |
| | Fluoride | 10–74.9 | 18.7 |
| | Lead | 1.6–98.5 | 21.5 |
| | Manganese * | 386–899 | 651 |
| | Mercury | 0.05–0.18 | 0.05 |
| | Molybdenum | 0.02–5.32 | 0.75 |
| | Nickel | 0.75–51.5 | 8.9 |
| | Selenium | 0.02–0.5 | 0.16 |
| | Silver *** | 0.04–0.15 | 0.1 |
| | Thallium *** | 0.003–0.059 | 0.044 |
| | Tin * | 0.5–3.2 | 0.54 |
| | Vanadium * | 5.8–21.1 | 10.6 |
| Zinc | 25.8–296 | 113 | |
| Other | Dry matter (% w/w) | 14–72 | 30 |
| | pH | 4.9–8.4 | 6.9 |

Notes:

In mg/kg dry weight (DW) unless otherwise stated.

Sample size $n = 65$ unless marked * where $n = 22$, ** where $n = 9$ and *** where $n = 8$.

EPH = extractable petroleum hydrogen

USEPA 16 = US Environmental Protection Agency list of 16 PAHs considered priority pollutants – naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, dibenz[ah]anthracene, benzo[ghi]perylene and indeno[1,2,3-cd]pyrene

4.1.4 Comparison with other materials

To put the reported chemical levels in street leaf sweepings (see previous section) into some perspective, they were compared with analytical results from green wastes also collected during the trial. Seven samples of green waste were supplied by four authorities with more than half coming from the same authority. The statistical comparison was undertaken using a non-parametric test (Wilcoxon Rank Sum).² In general there were a lot of tied results in this statistical analysis, which combined with the limited sample size, led to difficulties in making a detailed interpretation of results.

For seven chemical parameters (potassium, phosphorous, lead, molybdenum, dry matter, pH and sulphur), the Wilcoxon test found that levels in green waste were significantly higher than leaf litter ($p \leq 0.05$). For two PAHs (benzo[a]anthracene and benzo[a]pyrene), the Wilcoxon test found levels in street leaf sweepings that were significantly higher than in green waste ($p \leq 0.05$). Levels of another PAH, indeno[123-cd]pyrene, was also found to be higher ($p \leq 0.06$).

Differences between the two wastes for a number of PTEs and PAHs, including benzo[a]pyrene, are illustrated in a series of box-and-whisker plots in Figures 4.2 and 4.3.³ Also shown in these figures is urban herbage data from the UK Soil and Herbage Survey (UKSHS), a comprehensive survey of the concentrations of major contaminants in soil and herbage at 122 rural, 28 urban and 50 industrial locations throughout the UK (Environment Agency 2007a, 2007b). Grasses were the principal component of the aboveground herbage samples collected. The box plots present data from England and Wales since the street leaf sweepings were collected only from these two countries. Urban herbage data were considered more likely to be representative of the plant matter in green wastes from residential, parks and open spaces. In general, urban herbage reported chemical levels between two and five times higher than herbage from rural locations in the UKSHS.

The box plots support the findings from the Wilcoxon tests; despite considerable variability, PAH levels in the street leaf sweepings appeared markedly higher than those found in either green wastes or urban herbage. Differences in PTE levels were not so apparent with high levels of variability demonstrated for all materials. In most cases, however, the levels measured in urban herbage were the lowest reported values.

Of the street leaf sweepings surveyed in this trial ($n = 65$), 22 samples (about a third) had total PAH concentrations (based on USEPA 16) less than the method detection limit of 1 mg/kg DW.⁴ The median concentration for all samples was 8.6 mg/kg DW and the maximum was 230 mg/kg DW. Of the green wastes in this trial ($n = 7$), five

² The Wilcoxon test ranks the results across groups, then sums the ranks in each group and tests to see if those summed values are the same. When results are ranked in this manner, there are often 'ties', and if there are many tied results, the conclusions need to be treated more cautiously.

³ A box plot is a useful way to visualise the distribution of a sample population and to display the differences between two or more populations without making any assumptions about the underlying statistical distribution. Box plots are non-parametric. The box represents the range from the first to third quartiles of the data (the values lying between the 25th and 75th percentiles), the line within the box is the second quartile (median value) with the vertical lines from the top and bottom of the box extending to the maximum and minimum values respectively. The vertical length of the box plot gives an indication of the variability (spread) of the data and any skewness is indicated by differences in the positioning of the median line and the different lengths of the top and bottom lines respectively.

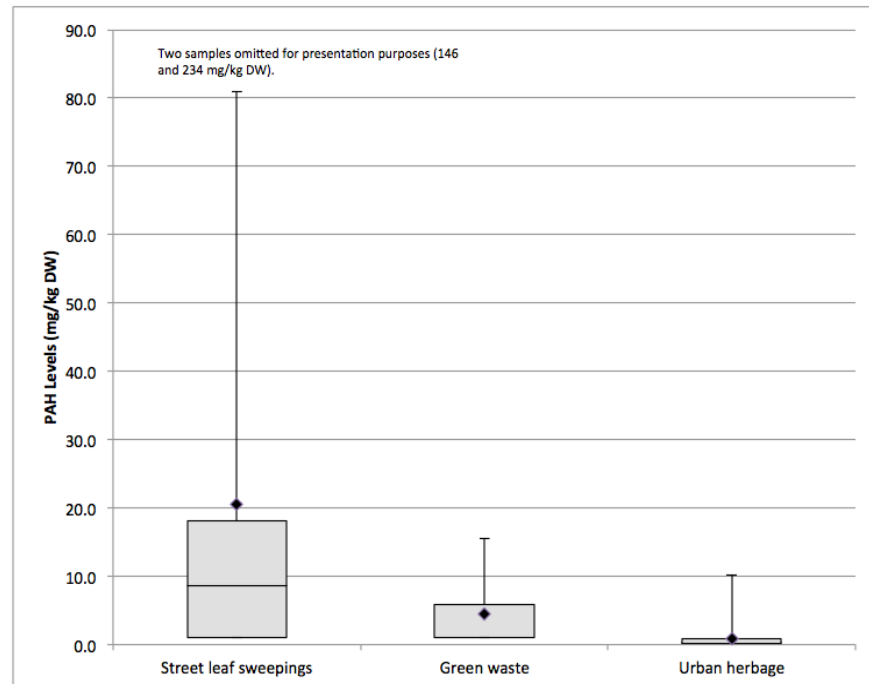
⁴ Five of the samples reported by the laboratory with a total PAH content less than the detection limit of 1 mg/kg DW for the sum of the USEPA 16 compounds were considered suspect because the sum of the measured levels of the individual PAH compounds in the same sample exceeded 1 mg/kg DW in total.

samples had total PAH concentrations (based on USEPA 16) less than the method detection limit of 1 mg/kg DW. However, two samples reported PAH levels markedly higher at 10.7 and 15.6 mg/kg DW respectively. Data on urban herbage from the UKSHS found much lower levels of PAH in plant matter than observed for the street leaf sweepings in this trial. Median total PAH levels (USEPA 16 without naphthalene) were only 0.24 mg/kg DW with a maximum level of 10 mg/kg DW. The median concentration in urban herbage was approximately 35 times lower than measured in the street leaf sweepings.

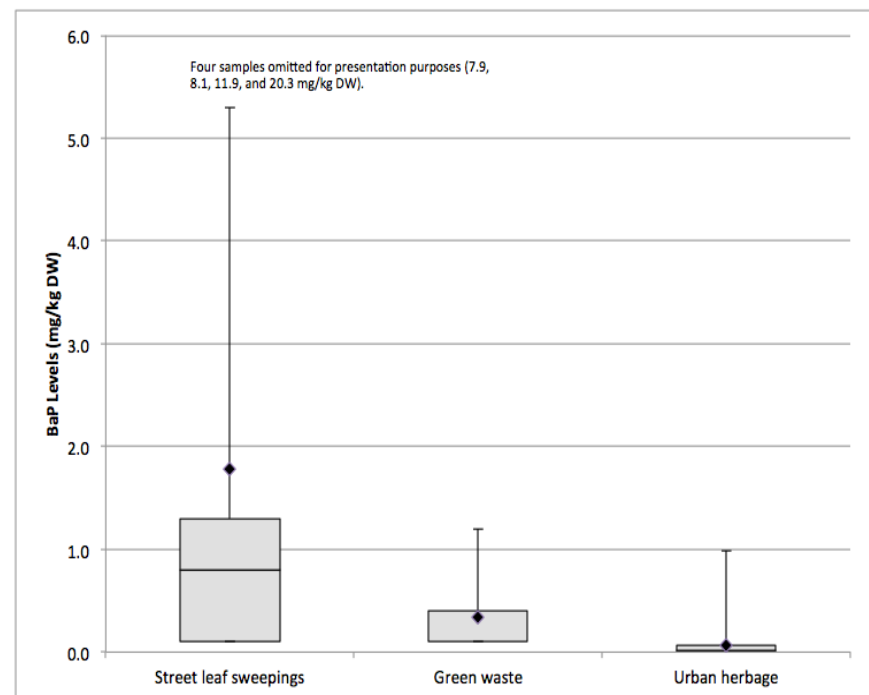
For benzo[a]pyrene, the street leaf sweepings in this trial had a median concentration of 0.8 mg/kg DW with a maximum level of 20 mg/kg DW, and with around a quarter of samples below the detection limit of 0.1 mg/kg DW. Of the green wastes, only two samples had levels above the same detection limit at 0.7 and 1.2 mg/kg DW respectively. Median levels in urban herbage were again much lower than observed for the street leaf sweepings with a median level of only 0.014 mg/kg DW for samples in England and Wales. The median concentration in urban herbage was approximately 60 times lower than measured in the street leaf sweepings.

Figure 4.2 Concentrations of total PAHs (a) and benzo[a]pyrene (b) in street leaf sweepings and green wastes collected and analysed for the trial

(a)



(b)



Notes: Diamond marker indicates mean value for the data presented. Several sample points of street leaf sweepings with very high PAH levels were omitted for presentational purposes. With the exception of the maximum value, all other presented statistical parameters are based on the full sample population. Also presented is the concentration found in urban herbage collected in England and Wales as reported by the UK Soil and Herbage Survey. Note that total PAH in urban herbage does not include naphthalene.

Figure 4.3 Concentrations of PTEs in street leaf sweepings and green wastes collected and analysed for the trial

(a) Chromium, copper, lead and zinc

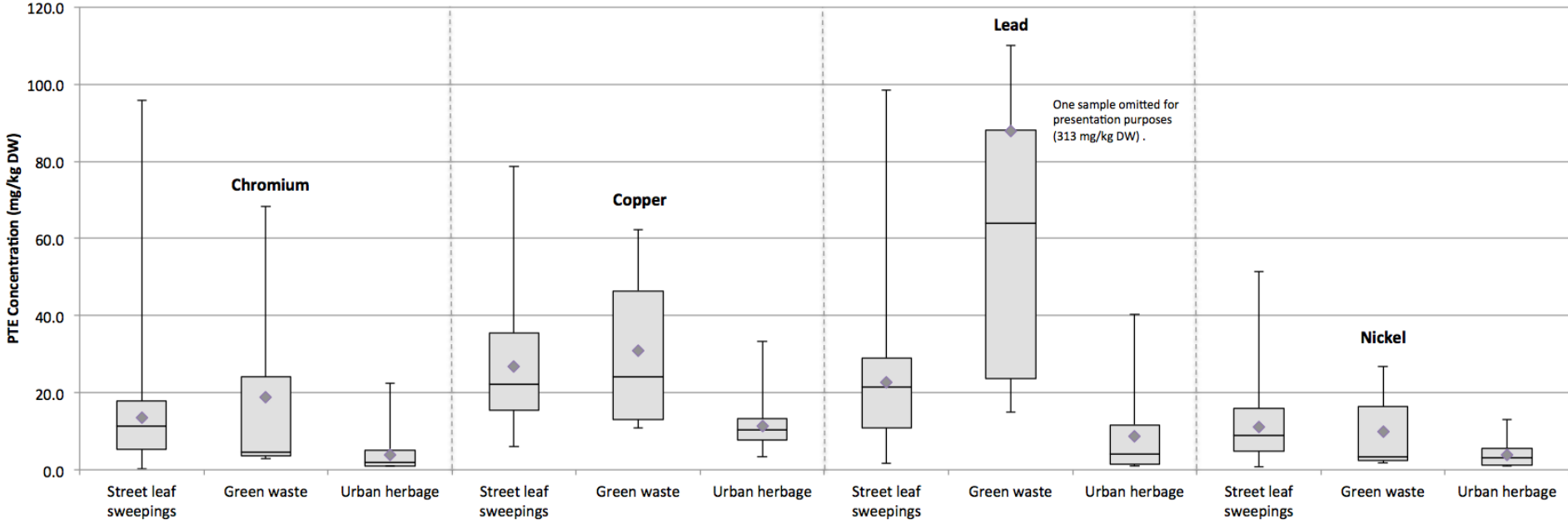
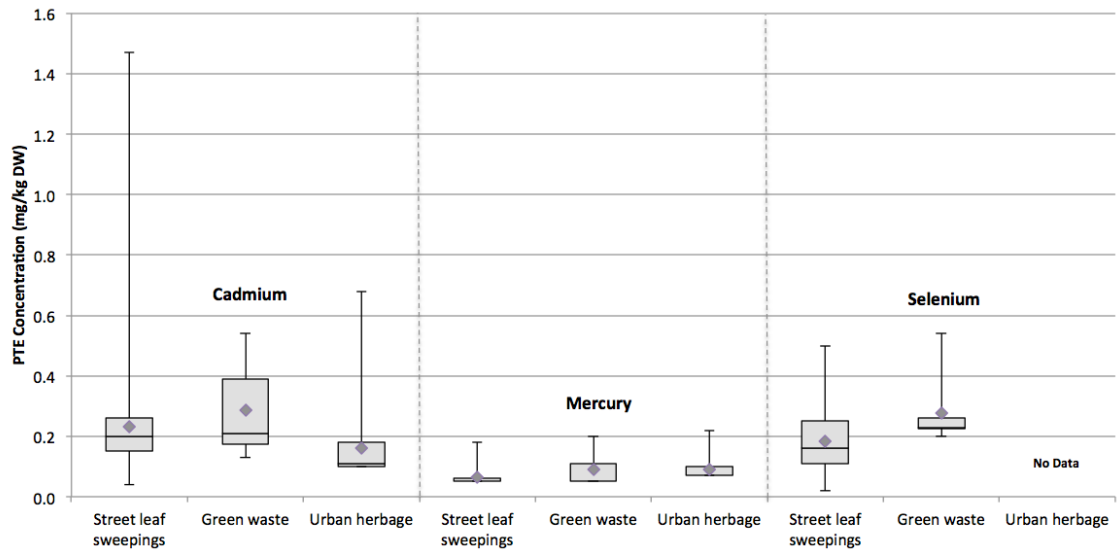


Figure 4.3 Concentrations of PTEs in street leaf sweepings and green wastes collected and analysed for the trial

(b) Cadmium, mercury and selenium



(c) Arsenic and molybdenum

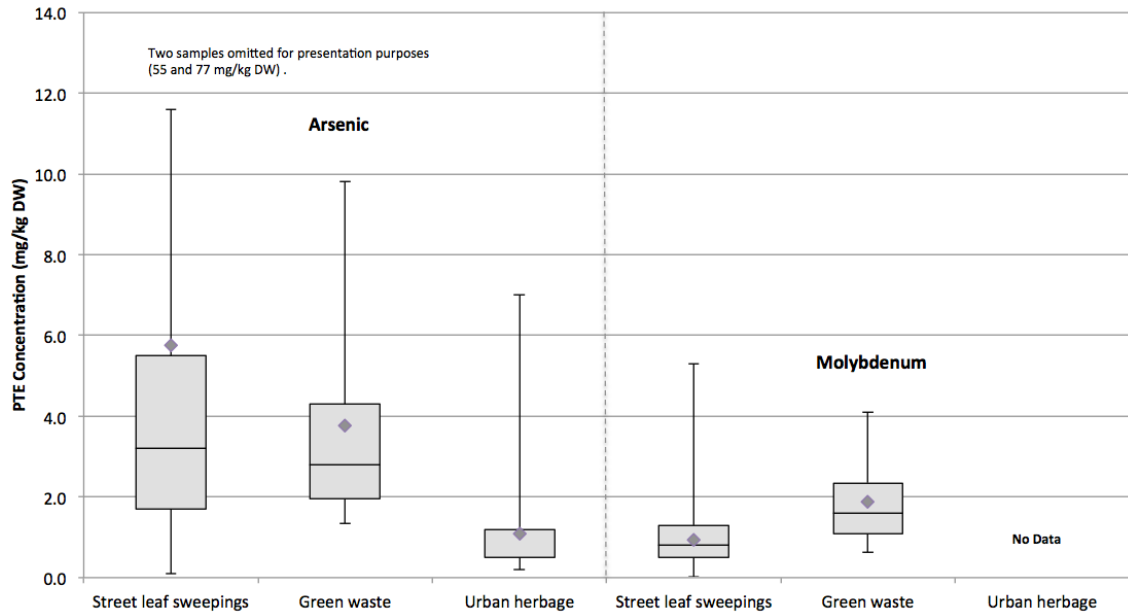
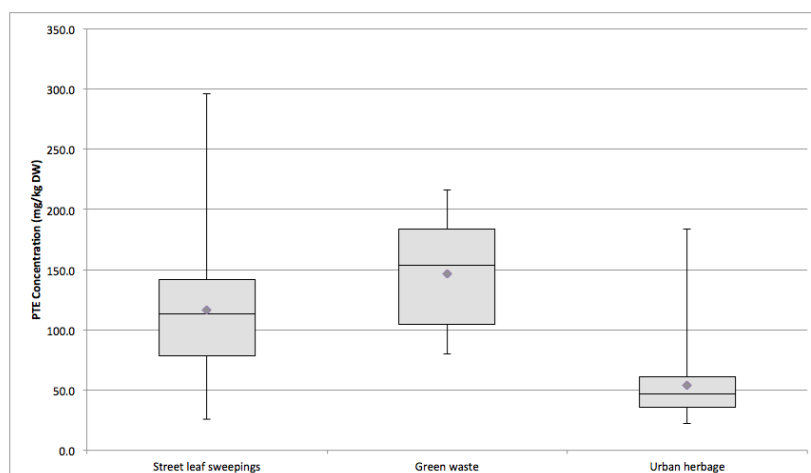


Figure 4.3 Concentrations of PTEs in street leaf sweepings and green wastes collected and analysed for the trial

(d) Zinc



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Notes: Diamond marker indicates mean value for the data presented. Several sample points of street leaf sweepings and one sample point of green waste with very high PTE levels were omitted for presentational purposes. With the exception of the maximum value, all other presented statistical parameters are based on the full sample population. Also presented is the concentration found in urban herbage collected in England and Wales as reported by the UK Soil and Herbage Survey.

4.2 Compost like outputs (CLO)

A total of 12 authorities provided one or more samples of composted street leaf sweepings and/or composted street leaf sweepings blended with green waste output for the trial. Most street leaf sweepings were collected between December 2012 and the end of January 2013 (Figure 4.1). Composting results were submitted between February and April 2013. Of the 31 composted samples collected and analysed, 18 samples were from a single authority and therefore these results were unlikely to be representative of a national picture. Most of the other participants submitted only one sample each.

4.2.1 Physical characteristics

Composted samples from the trial were submitted for analysis for physical contaminants using the sampling and analytical methods used for Compost Quality Protocol and PAS 100 composts (BSI 2011, WRAP and Environment Agency 2012a). Analytical data were available for only 24 of the 31 output samples (Table 4.4).

Overall levels of physical contaminants were found to be relatively low. Half the samples had physical contaminant levels of glass, metal and plastic below reporting limits for the fraction >2 mm. However, two samples (8%) from the same authority had glass levels in excess of the PAS 100 criteria of 0.25% for minimum quality compost.

Two samples had levels of stones below reporting limits for the fraction >4 mm. However, 12 samples (half of all samples with data) had stone levels in excess of the PAS 100 criteria of 10% for minimum quality compost suitable for all uses. These exceedances were reported across a wide range of authorities.

Table 4.4 Physical contaminants found in CLO samples (*n* = 24)

| Physical contaminant | | Range | Median |
|-----------------------------|---------|--------|--------|
| % w/w of total sample >2 mm | Glass | 0–0.49 | 0 |
| | Metal | 0–0.02 | 0 |
| | Plastic | 0–0.09 | 0 |
| | Other | 0–0.01 | 0 |
| | Total | 0–0.58 | 0.01 |
| % w/w of total sample >4 mm | Stones | 0–27.4 | 10 |

4.2.2 Chemical characteristics

Samples of composted outputs were submitted for chemical analysis using the sampling and analytical methods specified for Compost Quality Protocol and PAS 100 composts (BSI 2011, WRAP and Environment Agency 2012a). Chemical analysis included:

- major crop nutrients
- organic contaminants including petroleum hydrocarbons and PAHs
- PTEs including metals and metalloids
- general chemical characteristics such as pH and organic matter content

The results are summarised in Table 4.5. In addition to the main analysis (*n* = 31), the Environment Agency analysed a subset (up to *n* = 20) for additional PTEs as noted in Table 4.5.

As observed previously for street leaf sweepings (see section 4.1.3), there was a high degree of variability in the results. Cautious comparison of levels in street leaf sweepings before composting and in the final CLO suggested that the total petroleum hydrocarbon levels in the final output were much lower than in the input material. For example, the median measurement of the EPH C10–C25 fraction was reduced from 1,200 mg/kg DW to less than 450 mg/kg DW by composting and the EPH C10–C40 fraction was similarly reduced from 4,200 mg/kg DW to 2,000 mg/kg DW. This result was expected since many petroleum hydrocarbons are readily biodegradable in aerobic systems.

In the literature, lower weight PAHs were also shown to have been reduced by composting; while the potential accumulation of higher weight PAHs (especially the five- and six-ring compounds) was observed in green waste composts (Brandli et al. 2007). Five- and six-ring PAH compounds, including benzo[a]pyrene, are known to resist biological degradation and persist in soils (Semple et al. 2001). In a field-scale study of the composting of green wastes over a 16-week period, although the concentration of the low molecular weight PAHs in green waste compost fell by up to 90%, the accumulation ratios of the higher weight compounds were between 1.5 and 2.0 (Brandli et al. 2007).

Table 4.5 Chemical contaminants found in CLO samples

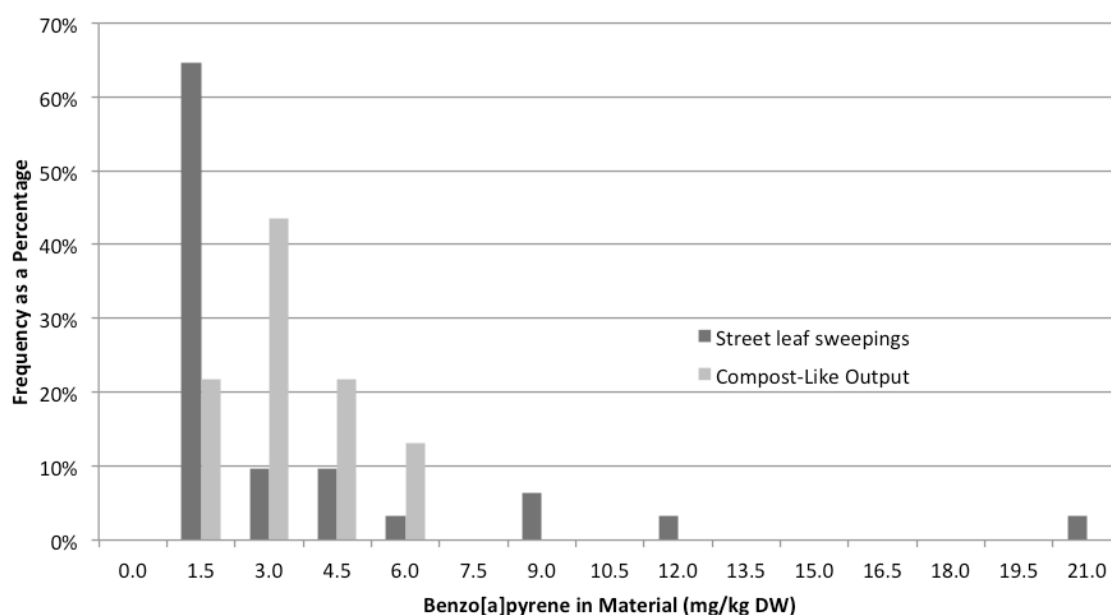
| Chemical parameters | | Range | Median |
|---------------------|----------------------------------|------------|--------|
| Nutrients | Nitrogen (% w/w) | 0.2–2.2 | 0.9 |
| | Phosphorus | 844–4,119 | 1,217 |
| | Potassium | 888–5,946 | 2,233 |
| | Sulphur | 712–3,167 | 1,284 |
| | Organic matter (% w/w) | 9.9–80 | 40.2 |
| Organics | Gasoline range organics (C6–C10) | 5–116 | 5 |
| | EPH (C10–C25) | 218–4,699 | 444 |
| | EPH (C25–C40) | 748–10,816 | 1,568 |
| | EPH (C10–C40) | 966–15,515 | 2,012 |
| | PAH (USEPA 16) | 1–52 | 18.5 |
| | Benzo[a]pyrene | 0.4–5.2 | 2.2 |
| Toxic elements | Antimony *** | 0.1–13.3 | 1.8 |
| | Arsenic | 0.6–404 | 4.3 |
| | Barium * | 43.1–80.2 | 63.3 |
| | Beryllium ** | 0.2–0.5 | 0.4 |
| | Boron * | 4.4–58.9 | 16.8 |
| | Cadmium | 0.06–0.62 | 0.23 |
| | Chromium | 6.8–152 | 52.9 |
| | Cobalt * | 2.6–10.6 | 5.6 |
| | Copper | 15.1–191 | 32.4 |
| | Fluoride | 10–106 | 28.7 |
| | Lead | 3.1–133 | 25.8 |
| | Manganese * | 386–899 | 666 |
| | Mercury | 0.05–0.19 | 0.05 |
| | Molybdenum | 0.03–8.7 | 3.6 |
| | Nickel | 9.1–79.4 | 29.1 |
| | Selenium | 0.04–0.57 | 0.23 |
| | Silver *** | 0.04–0.15 | 0.09 |
| Sodium | 151–7,542 | 740 | |

| Chemical parameters | | Range | Median |
|---------------------|--------------------|-------------|--------|
| | Thallium *** | 0.038–0.059 | 0.05 |
| | Tin ** | 0.6–3.2 | 1.2 |
| | Vanadium * | 5.8–21.1 | 10.6 |
| | Zinc | 54.4–304 | 115 |
| Other | Dry matter (% w/w) | 16.8–80.2 | 44 |
| | pH | 6.3–8.3 | 7.7 |

Notes: In mg/kg dry weight (DW) unless otherwise stated.
Sample size $n = 31$ unless marked * where $n = 20$, ** where $n = 8$ and *** where $n = 6$.
EPH = extractable petroleum hydrogen

This finding was also suggested in the present trials where median concentrations of benzo[a]pyrene were higher in output than in input materials. Figure 4.4 compares the levels of benzo[a]pyrene in composted street leaf sweepings ($n = 23$) with a subset of the untreated street leaf sweepings collected ($n = 31$) for the seven authorities that provided both input material and output samples for the trial. While the overall range of reported concentrations of benzo[a]pyrene in street leaf sweepings (0.1–20.3 mg/kg DW) appeared to have been reduced in the CLO (0.4–5.2 mg/kg DW), the median level in the output (2.2 mg/kg DW) was a factor of two higher than that found in the sweepings (0.8 mg/kg DW).

Figure 4.4 Benzo[a]pyrene levels in street leaf litter ($n = 31$) and CLO samples ($n = 23$) from the same participants



Note: Frequency has been converted into a percentage to allow for effect of different sample sizes.

Of the 23 composted street leaf sweeping samples analysed from the trial, 21 had benzo[a]pyrene levels >1 mg/kg DW. Samples from the composting of street leaf sweepings blended with green waste had the lowest levels of benzo[a]pyrene – of the eight samples only about half had benzo[a]pyrene levels >1 mg/kg DW.

Although the dataset was small and interpretation was difficult, the proportion of street leaf sweepings blended with green waste in the composting inputs did not appear to be a factor in the levels of PAHs and benzo[a]pyrene reported in the composted outputs. In the case of total PAHs, the two samples with the lowest levels had blended inputs at a 50:50 ratio. These ratios were in the middle of the range for the samples in this trial. Similarly for benzo[a]pyrene, three of the four samples with the lowest measured benzo[a]pyrene levels had the highest street leaf sweepings contents with blending ratios from 50:50 to 80:20. Three of the four samples with the greatest benzo[a]pyrene contents had the lowest mixing ratios of street leaf sweepings to green waste of around 30:70.

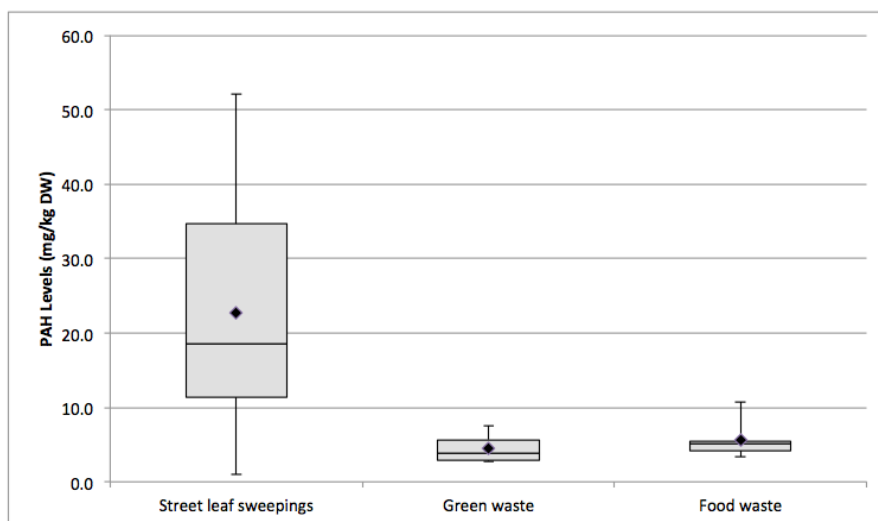
4.2.3 Comparison of trial outputs with other materials

To provide some perspective, the reported chemical levels in the composted outputs were compared with analytical results from other composts made from green and food wastes. There are few published datasets on composts, but WRAP (2011) measured levels of organic and inorganic contaminants in composts from six facilities in Wales in 2009 and 2010 as part of a larger study. WRAP took compost samples from windrows derived either from green wastes alone or mixed green and food wastes that had been subjected to a previous in-vessel composting stage to comply with the Animal By-Products Regulations, and were ready for land application. WRAP sampled batches of compost from each site in triplicate on two separate occasions.

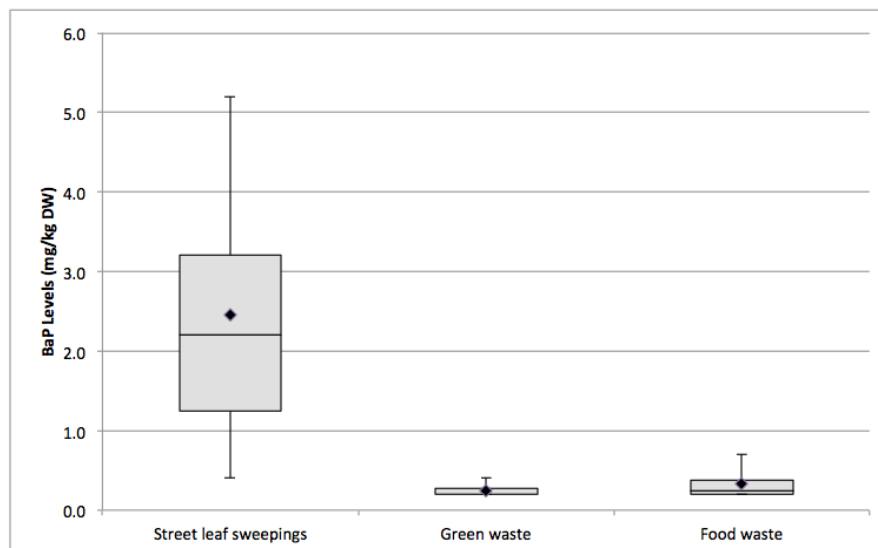
The differences between the different composts investigated by WRAP and composted street swept leaves for a number of PTEs and PAHs, including benzo[a]pyrene are illustrated in a series of box-and-whisker plots in Figures 4.5 and 4.6. The datasets are based on each compost batch, which is a single sample for the composts in this trial ($n = 31$) and the average of the three replicate samples taken by WRAP (2011) for each compost sampled in 2009 and 2010 ($n = 6$ for green compost and blended green/food composts respectively).

Figure 4.5 Concentrations of total PAHs (a) and benzo[a]pyrene (b) in outputs sampled during this trial and in green and green/food composts sampled by WRAP (2011)

(a)



(b)



Note: Diamond marker indicates mean value for the data presented.

Figure 4.6 Chemical concentrations of PTEs in outputs sampled during this trial and in green and green/food composts sampled by WRAP (2011)

(a) Chromium, copper, lead and zinc

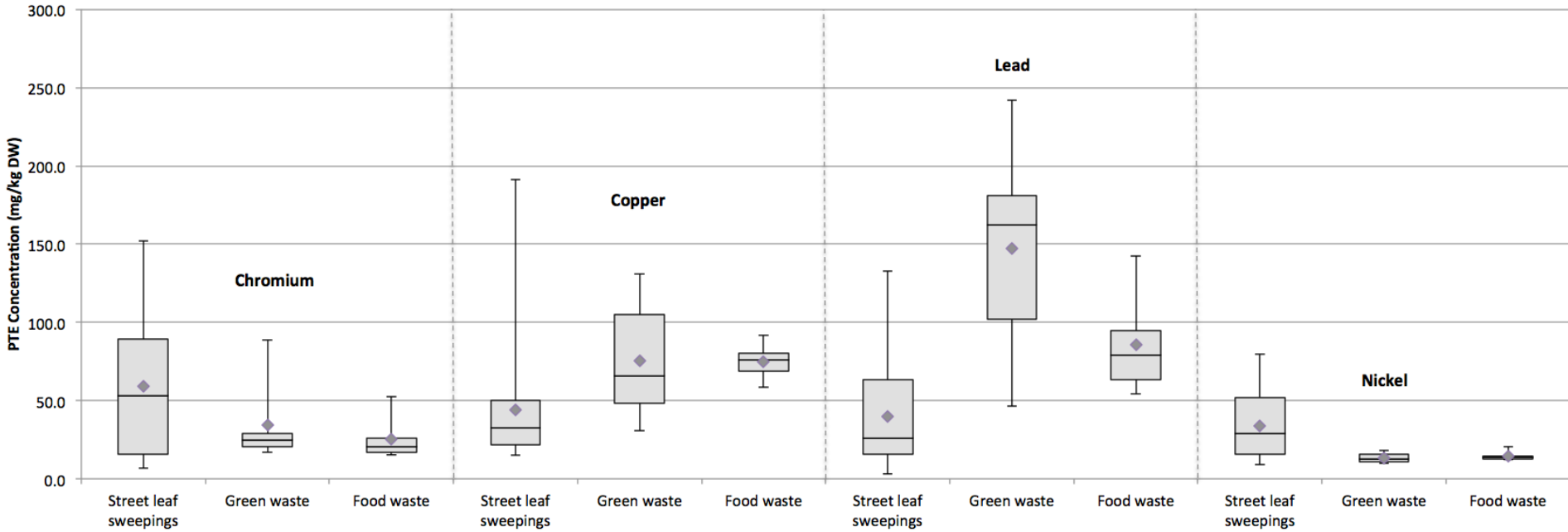
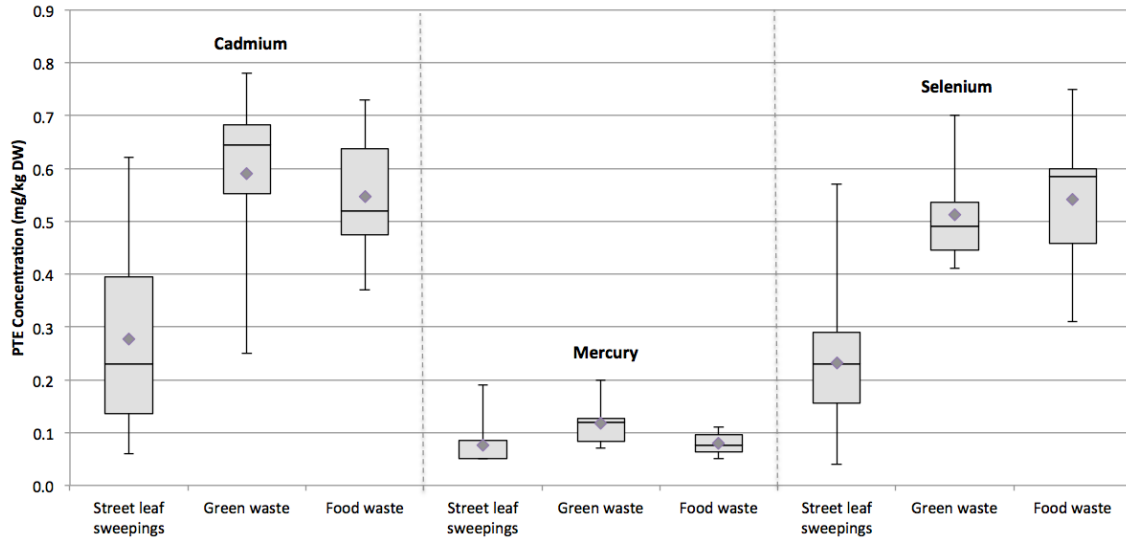


Figure 4.6 Chemical concentrations of PTEs in outputs sampled during this trial and in green and green/food composts sampled by WRAP (2011)

(b) Cadmium, mercury and selenium



(c) Arsenic and molybdenum

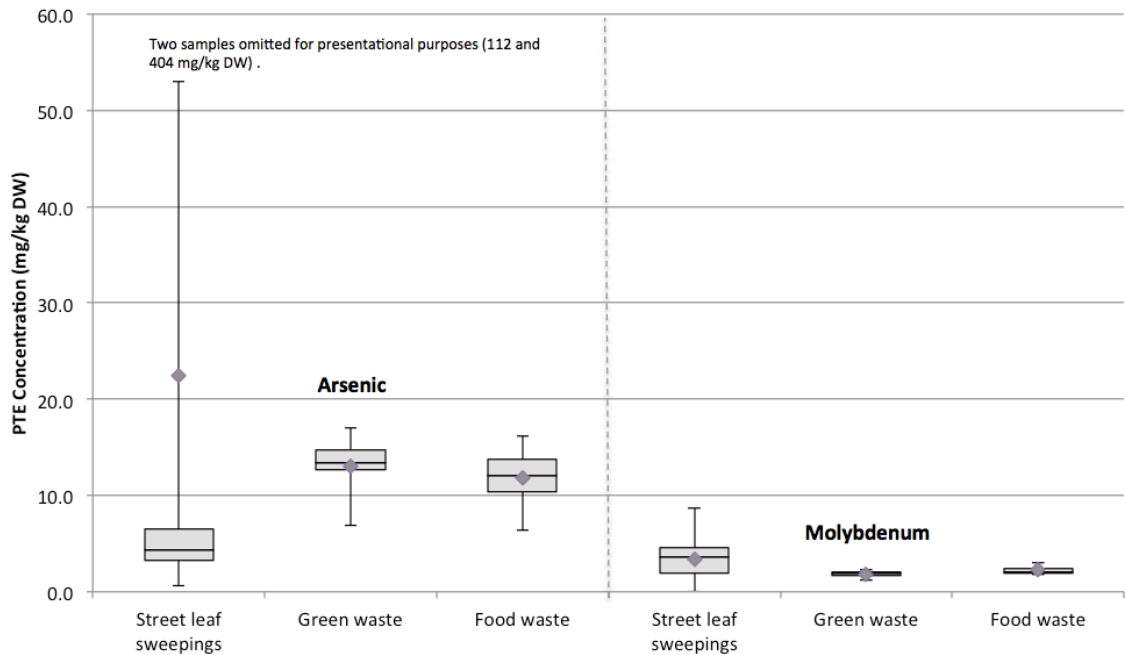
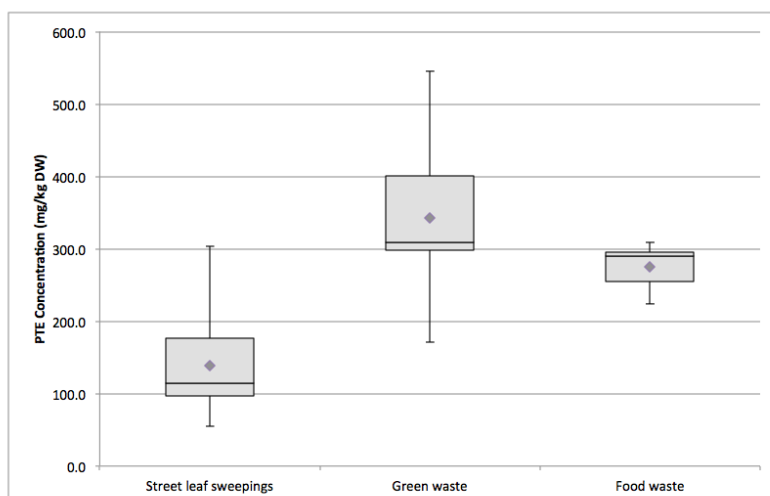


Figure 4.6 Chemical concentrations of PTEs in outputs sampled during this trial and in green and green/food composts sampled by WRAP (2011)

(d) Zinc



Notes: Diamond marker indicates mean value for the data presented. Several sample points of street leaf sweepings with very high levels of arsenic were omitted for presentational purposes. With the exception of the maximum value, all other presented statistical parameters are based on the full sample population.

PAH levels in the composted street leaf sweepings in this trial were much higher than those found in either of the two types of compost sampled by WRAP (2011). The median PAH level in the composted trial outputs of 18.5 mg/kg DW was much higher than the levels of 3.9 and 5.2 mg/kg DW found in the WRAP study in green and green/food composts respectively. A similar result was also found for benzo[a]pyrene; the median level in composted trial outputs was 2.2 mg/kg DW, which was around 10 times higher than the 0.2 and 0.3 mg/kg DW found in green and green/food composts respectively.

The European Commission's Joint Research Centre (JRC) sampled a wide range of composts from across Member States to establish an indicative baseline understanding of contamination levels (IPTS 2012). The survey included 24 samples of green composts produced from the biodegradation of garden and park waste only (most came from France and Belgium, none were from the UK). Over 90% of the samples had PAH levels (based on 12 compounds out of the USEPA 16⁵) slightly more than 2 mg/kg DW with a median level close to 1 mg/kg DW; the maximum value found was around 7 mg/kg DW. These values are broadly consistent with the levels of contamination observed by WRAP (2011). However, IPTS (2012) noted that samples from '... green waste compost, as well as sludge compost contain sometimes high PAH concentrations'.

In the trial reported here, the median PAH level reported in the samples of composted street leaf sweepings (18.5 mg/kg DW) was around 18 times higher than the median

⁵ The four compounds excluded in the JRC study from the USEPA 16 were acenaphthylene, acenaphlene, fluorene and naphthalene. In most output samples in this trial, these four compounds were reported only at their detection limits and, where measurable, they represented a maximum of 5% of the total PAH content.

value reported by the Institute for Prospective Technological Studies (IPTS) in Spain (IPTS 2012). One trial sample had reported PAH levels below the detection limit of 1 mg/kg DW, but this was suspect because the measured levels of individual PAH compounds in the same sample exceeded 7 mg/kg DW. No consistent relationship between PAH levels in untreated street leaf sweepings and CLO was found. Overall, only two composted trial samples reported total PAH levels less than the maximum value reported by IPTS (2012).

Differences in PTE levels were more difficult to interpret. Composted street leaf sweepings appeared to have higher levels of chromium, nickel and molybdenum than in the green and green/food composts sampled by WRAP (2011). Conversely, the composted trial outputs appeared to have much lower levels of lead, selenium and zinc compared with the levels found by WRAP. Although the levels of contaminants in composted street leaf sweepings, and to lesser extent the other types of compost, were highly variable, the box plots also suggested that levels of arsenic, cadmium and copper were higher in green and green/food composts than in the composted trial outputs. The link between contaminant levels in composted samples and possible roadside contamination was unclear.

4.3 Treatment options

Insufficient data were obtained by this trial to evaluate pretreatment options for street leaf sweepings before composting (option 3 in the trial methodology).

Two samples (one from each of two batches) were collected and only a single sample of composted output was analysed. The operator reported that there was so little organic matter left after the treatment process that both loads of treated leaf litter were combined before composting.

No data were provided relating to the location of the collected sweepings, as the collected material had been transported via two transfer stations prior to delivery to site.

There was no assessment of physical contaminants.

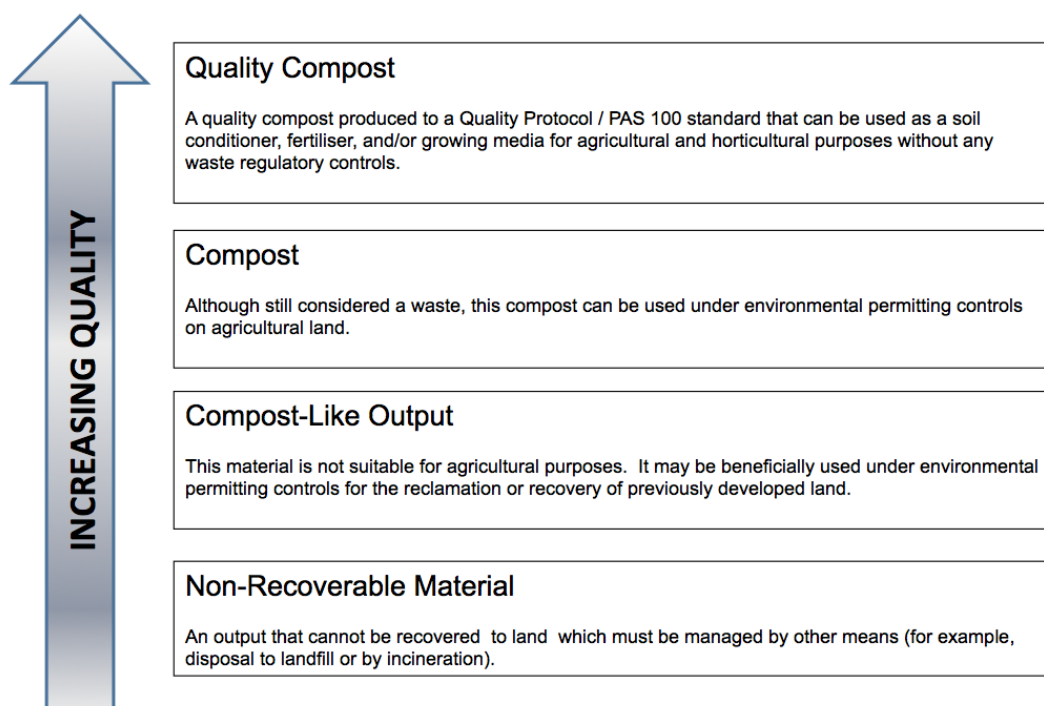
PAH and benzo[a]pyrene levels in the washed sweepings were reported to be high at 10.2–19.3 mg/kg DW and 1.1–1.4 mg/kg DW respectively. Sample variability meant it was impossible to compare these levels with those found in prewashed materials.

5 Assessment of quality and risk

One of the trial's main aims was to assess whether street leaf sweepings could be collected and composted for application to agricultural land without compromising compost quality or posing a greater risk to human health and the environment relative to existing composts from green and/or food wastes.

There are four main levels of compost quality in the UK (Figure 5.1).

Figure 5.1 Levels of compost quality



To determine whether the composted street leaf sweepings from this trial met the requirements for Quality Compost, physical and chemical contaminant concentrations in each sample ($n = 31$) were compared with the minimum quality requirements for PTEs and physical contaminants set out in PAS 100 (BSI 2011). Samples were compared individually with the quality criteria and a single sample failed if one or more of the measured levels exceeded the upper limits.

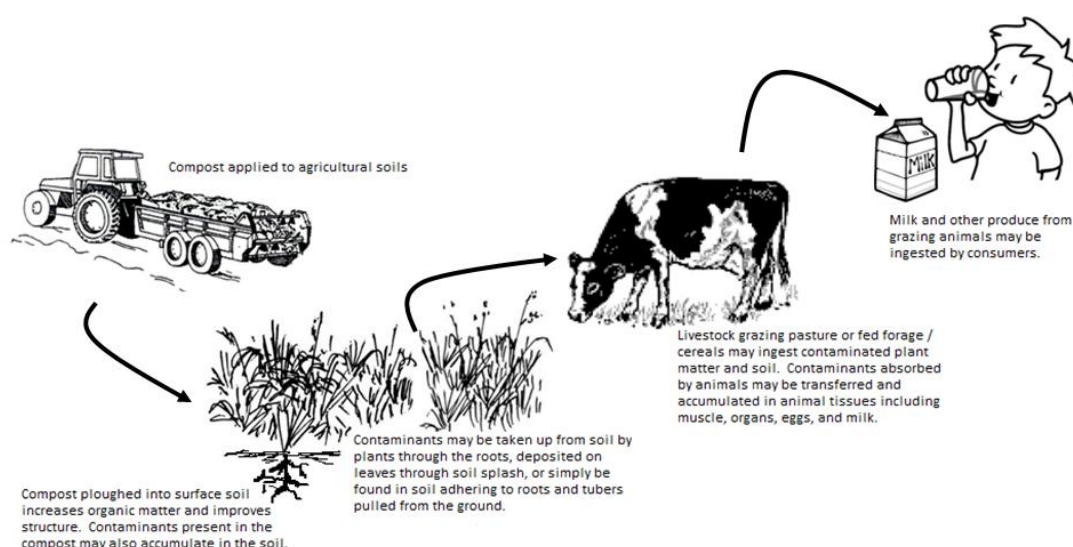
To consider the suitability of the composted samples from this trial for general agricultural and horticultural use (the next level down from Quality Compost), a generic risk assessment was carried out using the methodology developed by the Environment Agency for Quality Protocols and the tools used to assess site-specific risks for the landspreading of wastes under the environmental permitting regime.

The most important assumption made in the risk assessment was that application of composted street leaf sweepings to agricultural land resulted in an increase in contaminant concentrations in top soil with the potential for transfer of contaminants from soil through crops and livestock to consumers. This pathway is illustrated in Figure 5.2.

Emissions of contaminants to soil and their transfer through the food chain were based on an Environment Agency model called Waste Applied to Land: Tool for Environmental Risk Assessment (WALTER), which is described in Appendix C. Predicted chemical rates of emissions to land, concentrations in soil and foodstuffs, and consumer exposures were compared with guidelines that are protective of soil quality, crop, livestock and human health. Each sample of composted trial output was assessed separately for all relevant chemical parameters.

The third tier of compost quality shown in Figure 5.1, 'Compost-Like Outputs', was not assessed.

Figure 5.2 Pathway by which trace contaminants in compost applied to agricultural land migrate through the human food chain



5.1 Assessment for Quality Compost

Composts derived from source-segregated biodegradable materials and wastes are used in agricultural, horticultural, land restoration, soft landscaping, sports recreation, and other markets in the UK. The Compost Quality Protocol specifies a set of criteria for the product of a waste treatment process such that, if they are met, the product will no longer be regarded by the relevant regulatory authorities as waste (WRAP and Environment Agency 2012a).

5.1.1 Methodology

As an initial screen for suitability in this trial, only the minimum quality requirements in terms of physical and chemical contamination in PAS 100 were considered (BSI 2011). These requirements are summarised in Table 5.1 and apply to levels measured in compost directly.

PAS 100 does not set an upper limit for persistent organic pollutants (POPs) such as PAHs because it accepts only source-segregated biodegradable inputs from a restricted range of sources. To perform an indicative test for PAHs, the levels found in the composted trial outputs were compared with the maximum limit for PAH (USEPA

16) of 6 mg/kg DW proposed in the third working document on end-of-waste criteria for biodegradable waste subject to biological treatment proposed by the Institute for Prospective Technological Studies in Spain (IPTS 2012). The limits in the working document have not been adopted by the European Commission or any Member States and therefore it is essential their use is considered carefully.

The measured levels of each chemical parameter in each composted trial sample were compared with the upper limits in Table 5.1 and the indicative end-of-waste criterion for total PAHs. Samples of composted street leaf sweepings were reported separately from blended composted outputs. An individual composted sample failed the assessment when one or more of these chemical or physical criteria were exceeded.

Table 5.1 PAS 100 minimum compost quality requirements for general use

| Parameter | Upper limit | |
|---|-------------|------|
| Cadmium (mg/kg DW) | 1.5 | |
| Chromium (mg/kg DW) | 100 | |
| Copper (mg/kg DW) | 200 | |
| Lead (mg/kg DW) | 200 | |
| Mercury (mg/kg DW) | 1 | |
| Nickel (mg/kg DW) | 50 | |
| Zinc (mg/kg DW) | 400 | |
| Total glass, metal, plastic and any other 'non-stone' fragments in >2 mm fraction (% w/w DW) | Total | 0.25 |
| | Plastic | 0.12 |
| Stones >4 mm in grades other than 'mulch' (% w/w DW) | 8 | |
| Stones >4 mm in 'mulch grades' (% w/w DW) | 10 | |

Source: BSI (2011)

5.1.2 Results

Of the samples of composted street leaf sweepings ($n = 23$), 39% had measured PTE levels that exceeded the upper limits for minimum compost quality for general use as set out in PAS 100. Five samples failed on nickel content and four samples failed for both nickel and chromium content. This is consistent with Figure 4.6, which suggested that these two contaminants were elevated in the composted trial outputs compared with other compost types.

More than half the samples of composted street leaf sweepings, 12 out of 23, exceeded the upper limits for physical contamination with the most common problem being stones.

Taking the chemical and physical contaminants together, 13 samples out of 23 had parameters exceeding the PAS 100 upper limit values. Eight of these 13 samples failed on both physical and chemical parameters, four samples failed only for physical contaminants and one sample failed only on chemical parameters.

Of the samples of composted blended sweepings and green wastes ($n = 8$), none failed to meet the PTE upper limits for minimum compost quality. However, 25% of samples (2 out of 8) failed for physical contamination. One sample had elevated stones

(11.2% w/w) and the other glass (0.42% w/w). The sample size for the composted blended wastes was small and therefore results should be interpreted cautiously.

As shown in Figure 4.5, elevated PAH levels were found in the composted trial outputs. Of the 23 samples of composted street leaf sweepings, 96% (22 out of 23)⁶ had total PAH levels greater than the proposed end-of-waste criterion for total PAHs. The total PAH levels ranged from 1.0 to 52 mg/kg DW with a median level of 21.9 mg/kg DW (nearly four times higher than the proposed end-of-waste criterion).

Composts of the blended wastes also showed high PAH concentrations. Of the eight samples surveyed, 75% of the samples tested (6 out of 8), had total PAH levels greater than 6 mg/kg DW. The total PAH levels ranged from 4 to 49 mg/kg DW with a median level of 12.3 mg/kg DW.

These results are summarised in Table 5.2. Taken altogether, only one sample of the composted street leaf sweepings and one sample of the composted blended wastes collected in this trial met both the upper limit values for PAS 100 and the indicative end-of-waste criterion for total PAHs.

Table 5.2 Comparison of trial outputs with PAS 100 upper limit values (BSI 2011) and the indicative end-of-waste criterion for total PAHs (IPTS 2012)

| | Number of samples | Percentage of all samples |
|---|-------------------|---------------------------|
| Passed | 2 | 6 |
| Failed | 29 | 94 |
| Reasons for failure | | |
| Exceeded PAH criterion | 28 | 90 |
| Exceeded PTE upper limit value | 9 | 29 |
| Exceeded physical contamination upper limit value | 13 | 42 |

Note: Some samples failed for multiple reasons.

5.2 Assessment for general agricultural use

Composts are a valuable source of stable organic matter and crop available nutrients, which are used by growers to meet crop nutrient requirements and to maintain soil fertility (Defra 2011a). However, they also contain low levels of contaminants, such as PTEs and POPs, which may pose a risk to the fertility of soils and the health of crops, livestock and consumers.

Compost added to the soil breaks down over time to release valuable nutrients for uptake by crops but the same process also releases contaminants that may persist and accumulate in soil, resulting in negative impacts on soil ecology (microbiology, vertebrates and invertebrates), crops, livestock and wildlife. Soil contamination migrates through the agricultural food chain and may pose a risk to the health of consumers.

⁶ One compost sample had a PAH level less than 6 mg/kg DW but it is suspected that this is an analytical reporting error (the sum of individual PAH compounds measured above the detection limit was about 7 mg/kg DW) and that this sample also most likely failed.

5.2.1 Methodology

Even though nearly all samples failed the assessment for Quality Compost, it was decided to further consider the risks to human health and the environment that might be associated with their use on agricultural land. The generic risk assessment consisted of three main steps:

- **Hazard identification** – the specific chemical and physical parameters within the output to be risk assessed
- **Emission and exposure estimation** – the definition of a reasonable worst-case scenario for the application of the composted trial output and subsequent modelling of rates of chemical addition to soil, changes in soil concentration and transfer of chemicals through the food chain
- **Assessment of the significance of the risk** – comparison of modelled emissions and exposures with guidelines for soil, foodstuffs and consumer exposures that are protective of health and the wider environment

Each step in the risk assessment is described below and the results discussed further in the next section.

Hazard identification

An important first stage in risk assessment is to identify the hazards posed by an activity that, in particular circumstances, could lead to harm to human health and the wider environment (DETR et al. 2000).

In the context of this assessment, the hazards were those associated with the release of contaminants in composted materials into agricultural soil. The hazards were defined as:

- toxicity to soil organisms and higher ecosystem trophic levels
- toxicity to agricultural crops and livestock
- toxicity to human consumers of produce farmed from compost amended soils

More than 40 chemical and physical characteristics in the samples of composted trial outputs were determined by physical and chemical analysis (see section 4.2). However, not all these parameters were carried forward to the risk assessment. For example, nutrients such as nitrogen, phosphorus and potassium are beneficial to agricultural soils and increase crop growth and yield.⁷ Elements such as calcium, iron, magnesium and titanium have low toxicities and their accumulation in agricultural soils from use of compost-like materials is not typically a cause for concern. Although pH can have an adverse effect on soils if especially high or low, it was considered to pose little risk within the range of reported values (pH 6.3–8.3) in the trial composts.

Some chemical parameters were not taken forward because the evidence collected was insufficient to be analysed further. This is particularly the case for petroleum hydrocarbons, which occur in the environment in complex mixtures. Chemical analysis of gasoline range organics (GRO) covers hydrocarbons in the range C6 to C12 which have a wide range of mobilities and toxicity in soil and the food chain. GRO analysis is therefore only an indicator of petroleum contamination and of little use in quantitative

⁷ Notwithstanding the need to carefully manage nutrient applications to land since excess applications pose a risk to controlled waters and the wider environment.

risk assessment. In general, GRO analysis found these volatile and mobile hydrocarbons in only 10% of the output samples tested (that is, only 3 of 31 samples had GRO levels above the detection limit of 5 mg/kg DW).

EPH analysis reported levels of hydrocarbons in the C10–C40 fraction, with much higher levels observed in the C25–C40 fraction than in the C10–C25 fraction. As EPH analysis measures complex mixtures of petroleum hydrocarbons, it could not be taken forward to the risk assessment. Typically the heavier petroleum fraction (C25–C40) has a much lower environmental mobility and toxicity than the GRO range compounds, and at the levels found in the composted trial outputs the likely risks were considered low.

Table 5.3 Chemicals taken forward for risk assessment

| Chemical | Main hazard assessed |
|--|--|
| Antimony | Toxicity to human health |
| Arsenic | Toxicity to grazing livestock and human health |
| Barium | Toxicity to soil organisms and human health |
| Benzo[a]pyrene (as a marker for PAHs) | Toxicity to soil organisms and human health |
| Beryllium | Toxicity to human health |
| Boron | Toxicity to human health |
| Cadmium | Toxicity to human health |
| Chromium | Toxicity to crops |
| Cobalt | Toxicity to human health |
| Copper | Toxicity to crops and grazing livestock |
| Fluoride | Toxicity to grazing livestock |
| Lead | Toxicity to human health |
| Manganese | Toxicity to human health |
| Mercury | Toxicity to soil organisms, livestock and human health |
| Molybdenum | Toxicity to grazing livestock |
| Nickel | Toxicity to crops |
| Selenium | Toxicity to grazing livestock |
| Silver | Toxicity to human health |
| Sodium | Adverse effects on soil quality and structure |
| Thallium | Toxicity to human health |
| Tin | Toxicity to human health |
| Vanadium | Toxicity to human health |
| Zinc | Toxicity to crops |

Table 5.3 lists the chemical parameters taken forward in the risk assessment including the principal hazard(s) assessed. These chemicals were selected because:

- they were consistently found in the composted trial outputs at sufficient levels that might be of concern
- there is good evidence that they pose a hazard to soil quality and/or to crop, livestock or human health with expert evaluations of levels that are protective or pose a tolerable or minimal risk

Emission and exposure estimation

To complete the risk assessment, it was first necessary to define a scenario representative of a reasonable worst-case rate of application of the composted trial outputs to agricultural land.

Rates of use depend on many site-specific factors including existing soil conditions and the crop nutrient requirements, which vary considerably between different crop types and management schemes.

WRAP and the Environment Agency (2007) considered several different compost use scenarios in the technical assessment to support the development of the Compost Quality Protocol (WRAP and Environment Agency 2012a). Compost application rates varied from 9.4 tonnes dry weight (DW) per hectare (t DW/ha) for brassicas to 34 t DW/ha for main crop potatoes using Defra guidelines for crop requirements. The higher application rate for potatoes was considered likely to be excessive for Nitrate Vulnerable Zones (WRAP and Environment Agency 2007).

The updated analysis presented in Table 5.4 takes into account typical nutrient data reported in fertiliser recommendations for crops (RB209, Defra 2011a) for green and green/food composts. The risk assessment used the highest limiting rate for all scenarios of 33.3 tonnes fresh weight (FW) per hectare. This is equivalent to an annual application rate of about 20 t DW/ha based on a typical compost dry matter content of 60% w/w (Defra 2011a).

It is unlikely that risks to soil function and the transmission of contamination through the food chain will occur after a single application of compost at the annual rate of 20 tonnes DW per hectare (WRAP and Environment Agency 2007). However, it is important that the risk assessment demonstrates that use of composted street leaf sweepings will not result in harm to human health or the environment through repeated soil applications under a range of different conditions. In previous risk assessments for Quality Protocols such as the one for poultry litter ash (WRAP and Environment Agency 2012b), estimated differences in soil concentration of contaminants and their transfer through the food chain were evaluated after modelling 10 years of consecutive annual applications. This assumption was also used here.

Contaminant additions to land were estimated by multiplying the observed chemical concentration in each sample of composted trial outputs (see Table 4.5 for the range of values) by the application rate of 20 t DW/ha, taking into account the necessary unit conversions.

For example, if the sample was reported to contain 5 mg/kg DW arsenic, this was equivalent to a soil application rate of 100 g As per hectare ($5 \text{ mg/kg} \times 1000 \text{ kg/t} \times 20 \text{ t/ha} \times 0.001 \text{ g/mg}$).

Table 5.4 Reasonable worst-case annual application rates for crops based on RB209

| Crop scenario | Compost (t FW/ha) | | | Limiting rate |
|---|---|--------------------------------|---------|---------------|
| | P required as P ₂ O ₅ | K required as K ₂ O | N limit | |
| Winter wheat, straw removed (8 t/ha) | 41.7 | 26.4 | 33.3 | 26.4 |
| Winter wheat, straw incorporated (8 t/ha) | 40.0 | 19.1 | 33.3 | 19.1 |
| Potatoes (50 t/ha) | 83.3 | 65.5 | 33.3 | 33.3 |
| Brassicas (varies) | 66.7 | 54.5 | 33.3 | 33.3 |

Note: Assumes K Index or P Index is zero, typical compost nutrient and dry matter content as outlined in RB209 (Defra 2011a)

Soil concentrations were estimated using the WALTER model (see Appendix C), a simple mixing model which took into account during each year of landspreading:

- amount of a chemical added to the soil from applied composted trial outputs
- first-order losses from soil through chemical volatilisation, leaching, degradation and crop off-take

The estimated chemical concentration in soil at the end of the 10th year (that is, after 10 consecutive waste applications) was subsequently used to:

- predict chemical concentrations in foodstuffs
- estimate chemical exposures for the consumer from a diet of foodstuffs grown on compost trial outputs amended land

Figure 5.3 shows graphically all the food chain pathways considered.

WALTER used the following additional global assumptions.

- Starting soil concentrations before the first waste application were set to the median levels found in English soils (see Table 5.5).
- Wastes were assumed to be thoroughly ploughed in to a depth of 25 cm.
- Soil bulk density was assumed to be around 1,300 kg/m³.

Figure 5.3 Graphical representation of contaminant transport pathways through the food chain considered by the WALTER model

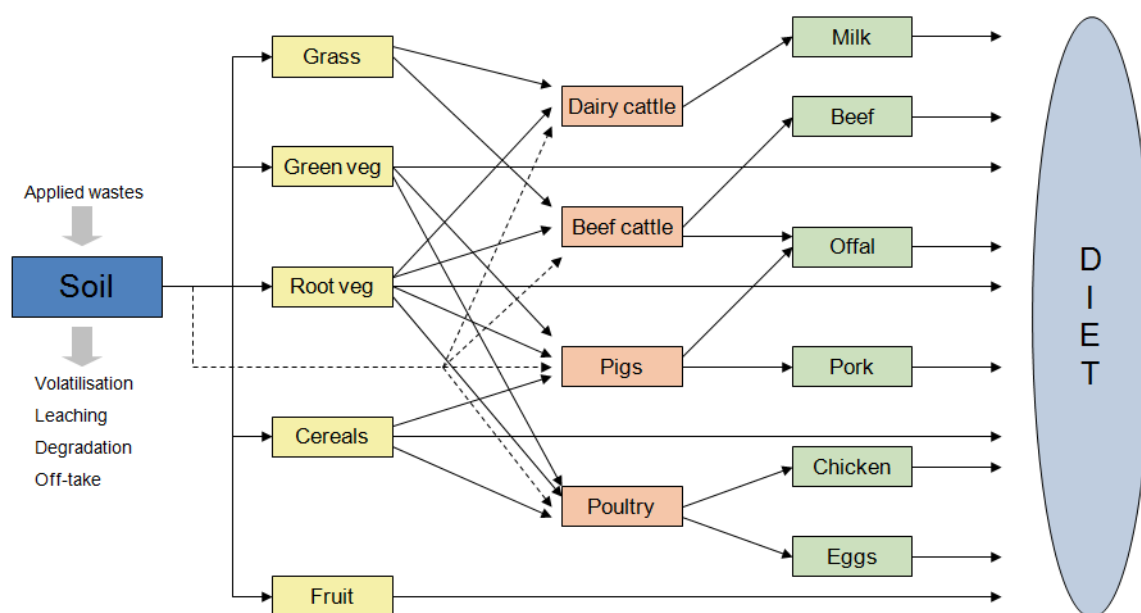


Table 5.5 Median values for rural soil background concentration of contaminants considered in risk assessment

| Chemical | Median concentration (mg/kg DW soil) | Reference |
|----------------|--------------------------------------|----------------------------|
| Arsenic | 8.3 | Environment Agency (2007a) |
| Barium | 60.5 | Salminen et al. (2005) |
| Benzo[a]pyrene | 0.067 | Environment Agency (2007b) |
| Boron | 34.0 | Archer and Hodgson (1987) |
| Cadmium | 0.3 | Environment Agency (2007a) |
| Chromium | 29.5 | Environment Agency (2007a) |
| Copper | 17.4 | Environment Agency (2007a) |
| Fluoride | No data | |
| Lead | 40.7 | Environment Agency (2007a) |
| Manganese | 397 | Salminen et al. (2005) |
| Mercury | 0.1 | Environment Agency (2007a) |
| Molybdenum | 0.9 | Salminen et al. (2005) |
| Nickel | 16.4 | Environment Agency (2007a) |
| Selenium | 0.5 | Archer and Hodgson (1987) |
| Zinc | 79.5 | Environment Agency (2007a) |

Risk evaluation

Chemical data from each individual sample of composted trial outputs were modelled to predict chemical addition rates to land, soil and foodstuff concentrations, and potential human exposures as outlined above. To consider the significance of these estimated chemical levels in the environment, they were compared with relevant benchmark criteria for the protection of the environment and human health. Where concentrations or levels of exposure exceeded these benchmark criteria, the result for the individual sample of composted trial outputs was recorded as 'failed'.

The different types of benchmark criteria used in the risk assessment are summarised in Table 5.6. Not every benchmark criterion was used for every chemical considered in the risk assessment and a specified type of criterion was not available for all chemicals. For example, the soil limits for sludge application (DoE 1996) were developed for only a short list of PTEs and other chemical contaminants such as PAHs could not be assessed using them. However, all the previously identified chemical hazards in the composted trial outputs were assessed against at least one benchmark criterion. Where there was more than one benchmark criterion for a chemical parameter such as for benzo[a]pyrene, the predicted environmental levels were compared against all respective criteria.

Table 5.6 Benchmarks for assessing the risks to health and the environment from chemical contaminants in soil, foodstuffs and consumer diets

| Benchmark | Measured in Unit | | Comments |
|--|------------------|----------------------|--|
| Soil limits for sludge application | Soil | mg/kg DW | Developed as a statutory or advisory limit for the application of sewage sludge to land to protect the health of crops, livestock and consumers. |
| Average annual application rates | | kg/ha per year | See above |
| Soil screening values | Soil | mg/kg DW | Screening tool to assess ecological risks from soil contamination |
| Maximum permissible levels of contaminants in animal feed and foodstuffs for human consumption | Foodstuffs | mg/kg FW | Statutory levels to protect animal and human health, and to minimise accumulation of contaminants in the human food chain |
| Dietary intake levels | Diet | mg/kg bodyweight/day | Acceptable, tolerable or minimal risk levels for chemical dietary intakes by consumers |

Soil limits for sewage sludge application (DoE 1996) are widely used to assess the risks to agricultural soils from other materials including industrial wastes and waste-derived composts. Where a chemical contaminant was covered by the *Code of Practice for Agricultural Use of Sewage Sludge* (DoE 1996), no subsequent comparison with any other benchmark criterion was deemed necessary. The benchmark criteria used in the risk assessment are listed in Table 5.7.

Table 5.7 Benchmark criteria used to assess risks to agriculture

| Contaminant | Maximum soil level (mg/kg DW) | Maximum application rate (kg/ha/year) |
|-------------|-------------------------------|---------------------------------------|
| Arsenic | 50 | 0.7 |
| Cadmium | 3 | 0.15 |
| Chromium | 400 | 15 |
| Copper | 100 | 7.5 |
| Fluoride | 500 | 20 |
| Lead | 300 | 15 |
| Mercury | 1 | 0.1 |
| Molybdenum | 4 | 0.2 |
| Nickel | 60 | 3 |
| Selenium | 3 | 0.15 |
| Zinc | 200 | 15 |

Source: DoE (1996)

As noted previously, not all chemicals identified as hazards were covered by the guidance on sewage sludge use in agriculture (DoE, 1996). It was therefore necessary to compare the estimated environmental emissions and concentrations for these chemicals with other relevant benchmark criteria protective of soil quality and human health. These other benchmark criteria are summarised in Table 5.8.

Predicted levels of soil contamination, and subsequent transport through the food chain, took into account existing levels in rural soils (that is, the chemical concentration in soil was the existing soil concentration plus the amount of chemical contaminants added from land application of the composted trial outputs). Where the data were available, existing levels were based on the median level of rural soils as measured by the UKSHS (Environment Agency 2007a, 2007b). In the absence of data from the UKSHS, median levels were estimated from UK soils data from the FOREGS study (Salminen et al. 2005) or from the older survey of England and Wales by Archer and Hodgson (1987). Table 5.5 lists values for the chemical contaminants in this assessment.

Table 5.8 Other benchmark criteria used to evaluate estimated chemical concentrations in soils and foodstuffs, and predicted dietary consumer exposures for individual samples of composted trial outputs

| Chemical | Benchmark criterion | Notes | Reference |
|----------------|---------------------|---|----------------------------|
| Antimony | 6 µg/kg BW/day | Tolerable intake from drinking-water | WHO (2003a) |
| | 78 mg/kg DW | Soil screening value for ecological receptors | USEPA (2010) |
| Barium | 20 µg/kg BW/day | Tolerable intake from drinking-water | WHO (2004) |
| | 890 mg/kg DW | Soil screening value for ecological receptors | RIVM (2001) |
| Benzo[a]pyrene | 0.01 µg/kg BW/day | Minimal risk value for dietary exposures | EFSA (2008) |
| | 1 µg/kg FW | Maximum level in processed cereal-based foods and baby foods for infants and young children | See note 1 |
| | 0.26 mg/kg DW | Soil screening value for ecological receptors (corrected for a soil organic matter content of 6%) | Environment Agency (2008a) |
| Beryllium | 2 µg/kg BW/day | Tolerable intake from drinking-water | WHO (2009a) |
| | 40 mg/kg DW | Soil screening value for ecological receptors | USEPA (2010) |
| Boron | 200 µg/kg BW/day | Tolerable intake from drinking-water | WHO (2009b) |
| Cobalt | 600 µg/kg BW/day | Tolerable intake for dietary exposures | EFSA (2009) |
| | 180 mg/kg DW | Soil screening value for ecological receptors | USEPA (2010) |
| Manganese | 200 µg/kg BW/day | Dietary exposure level unlikely to be a concern | WHO (2011) |
| Silver | 3 µg/kg BW/day | Tolerable intake from drinking-water | WHO (2003b) |
| Sodium | 150 kg/ha | Level unlikely to cause an adverse effect on physical condition of soil | Defra (2011a) |
| Thallium | 0.02 µg/kg BW/day | Dietary exposure level unlikely to be a concern | FSA (2009) |
| Tin | 2,000 µg/kg BW/day | Tolerable intake for dietary exposures | WHO (2006) |

| Chemical | Benchmark criterion | Notes | Reference |
|----------|---------------------|--|--------------|
| Vanadium | 10 µg/kg BW/day | Tolerable intake for dietary exposures | ATSDR (2012) |

Notes: ¹ European Commission Regulation (EC) No. 1881/2006 setting maximum levels for certain foodstuffs as amended by Regulation 1126/2007, 565/2008, 629/2008, 105/2010 and 165/2010
 BW = bodyweight

Before any evaluation of the risks using benchmark criteria other than the soil limits for sewage sludge application, predicted increases in soil levels after applying the composted trial outputs were assessed for their significance. This step ensured that trial outputs were not unduly penalised if the conservative nature of the risk assessment suggested problems close to existing ambient soil levels. Increased soil levels were considered trivial or insignificant where there was less than a 1% increase above median ambient soil concentration for each waste application. So for 10 consecutive applications of composted trial outputs, trivial increases were considered to be 10% or less. A chemical parameter found to cause an insignificant increase in predicted soil levels was not considered further and was deemed to have 'passed' the assessment. This approach was undertaken to be consistent with the policy approach adopted by the Environment Agency for the assessment of end-of-waste applications and landspreading deployments.

5.2.2 Results

Of the 31 samples of composted trial outputs only 13% (3) had PTE levels considered to pose a potential risk for agricultural use. All three samples failed because of the level of arsenic in the outputs (53, 112 and 404 mg/kg DW respectively); one sample additionally failed because of the level of sodium in the output (7,542 mg/kg DW). Interestingly these samples were taken from composted street leaf sweepings collected in south-west England, which is recognised as having elevated background levels of arsenic in soils (naturally occurring and as the result of historical mining activity). Plausible reasons for the elevated levels of arsenic in these composted trial outputs are the potential plant uptake of arsenic from naturally contaminated soils and a higher arsenic concentration in road sediments that become entrained within the sweepings during collection. However, without further evidence this is speculative.

Levels of the PAH, benzo[a]pyrene, in the samples of composted street leaf sweepings were of widespread concern. Of the 31 samples of composted trial outputs, 80% (25) had levels of benzo[a]pyrene considered to pose a risk for agricultural use. Most of these failures were due to risks to human consumption through the agricultural food chain, but 9 samples (29% of all samples) also resulted in soil concentrations that exceeded the benchmark criterion for the protection of soil ecology (see benchmarks in Table 5.8).

These results are summarised in Table 5.9. Taken altogether, 81% (25) of the samples of the composted trial outputs were considered to pose a risk when spread to agricultural land. The results relating to benzo[a]pyrene are discussed in more detail in the next section.

Table 5.9 Results of generic risk assessment from application of composted trial outputs to agricultural land

| | Number of samples | Percentage of all samples |
|---|-------------------|---------------------------|
| Passed | 6 | 19 |
| Failed | 25 | 81 |
| Reasons for failure | | |
| Risks from benzo[a]pyrene content | 25 | 81 |
| Risks from arsenic content | 3 | 10 |
| Risks from sodium content | 1 | 3 |
| Risks from all other PTE content (not reported above) | 0 | 0 |

Note: Some samples failed for multiple reasons.

5.3 Discussion

The biggest environmental concern raised by analysis of the composted trial outputs was the level of PAHs found in most samples. PAH levels were found to be much higher in street leaf sweepings and composted sweepings than in green wastes and typical green and green/food composts (see section 4). PAH and benzo[a]pyrene concentrations were the main reason why many composted trial samples were considered unsuitable as either a Quality Compost (see section 5.1) or as a compost suitable for general agricultural or horticultural use (see section 5.2).

Although there are potentially thousands of different PAH compounds in the environment, only a few hundred have been isolated and identified (IPCS 1998). Fewer still have been investigated for their impact on the environment and human health. In the 1970s, the USEPA selected 16 PAHs for routine chemical analysis and investigation in wastewater discharges at industrial point sources (Keith and Telliard 1979). Criteria for their selection included the availability of toxicological data, analytical methods and standards, and the frequency of their occurrence in analysed waters. Over the past 30 years, this list has been used worldwide as the basis for the development of guidelines and analytical suites for a range of environmental matrices. As a consequence, much of the research effort in environmental monitoring and assessment over this time has focused on these compounds.

Of greatest concern to human health are the carcinogenic PAHs including benz[a]anthracene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, chrysene, dibenzo[a,h]anthracene and indeno[1,2,3-c,d]pyrene (IPCS 1998, Environment Agency 2007b).

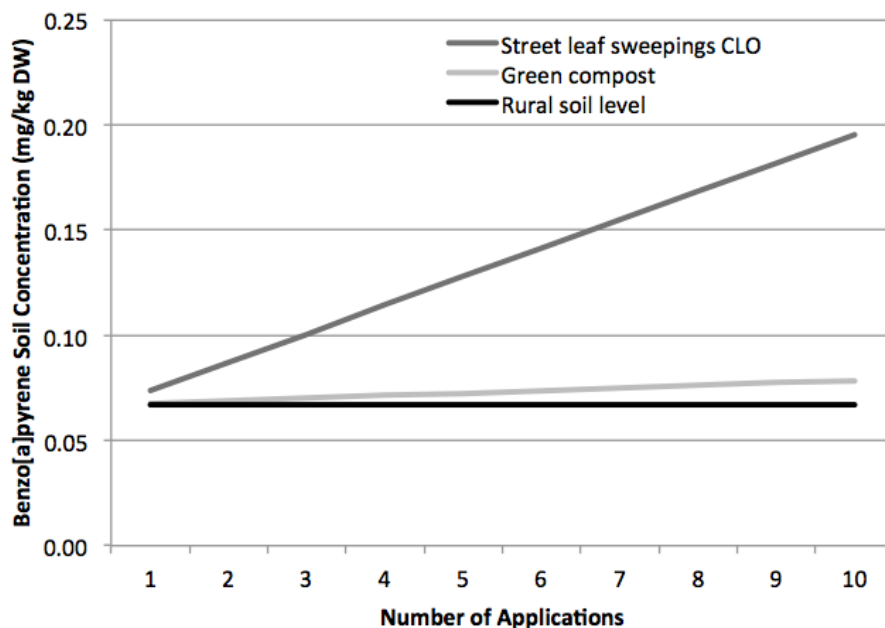
PAH contamination was identified as a potential problem in roadside environments by the literature review carried out to support the trial (see Appendix A). It is estimated that tyre wear and losses of used and unused motor oils could result in the emission of nearly 350 tonnes of PAHs per year into the UK environment (Napier et al. 2008). PAHs have been commonly found in diesel and petrol engine emissions in both gaseous and particulate forms.

Carcinogenicity is widely regarded as the main health concern from PAH exposure, individually or as a mixture. There is an extensive literature on the cancer epidemiology of workers exposed to complex PAH mixtures that conclusively demonstrates an elevated incidence of lung tumours on inhalation and of skin tumours from dermal contact. Animal studies have also shown that ingestion of coal tar mixtures may induce liver, lung and gastrointestinal tumours (EFSA 2008). Although investigations of the human carcinogenicity of individual PAHs has been dominated by studies involving benzo[a]pyrene (EFSA 2008, HPA 2009), there is clear evidence that many other PAHs induce tumours. To evaluate the cancer risks posed by exposure to PAH mixtures, HPA (2009) supported the use of a surrogate marker approach. Exposure to the surrogate marker, in this case benzo[a]pyrene, is assumed to represent exposure to all the PAHs in the environmental matrix. Thus the level of toxicity ascribed to the surrogate represents the toxicity of the PAH mixture (HPA 2009). A minimal risk level described for oral exposures to benzo[a]pyrene contaminants in soil would be around 0.01 µg/kg BW/day (Environment Agency 2009).

Human exposure to PAH contaminants in food is a widely recognised hazard with maximum limits introduced for the levels of benzo[a]pyrene in some foods intended for human consumption (EFSA 2009, EC Regulation (EC) No. 1881/2006 as amended). Action at European level has variously proposed to restrict potential PAH inputs into the agricultural food chain through limits in sewage sludge and recovered biodegradable wastes but many proposals have not been implemented. There is still considerable debate about whether PAH additions to land from sludge have any measurable impact on levels in food and the consumer diet (Smith 2009).

Figure 5.4 shows the predicted increase in benzo[a]pyrene concentration in soil from multiple applications of the composted street leaf sweeping outputs in this trial and the green composts sampled by WRAP (2011) using the median observed benzo[a]pyrene concentration. The ambient rural soil concentration of 0.067 mg/kg DW is shown as a baseline. After 10 applications of composted street leaf sweepings output, the estimated soil concentration has nearly doubled compared with the median ambient level.

Figure 5.4 Predicted levels of benzo[a]pyrene in agricultural soils after 10 applications of street leaf sweepings output or green composts contaminated at the observed median concentration from this trial or as reported by WRAP (2011)



The chemical properties used to estimate the transfer of benzo[a]pyrene through the food chain in this risk assessment are summarised in Table 5.10. In common with other organic compounds, the transport and partitioning of PAHs in the environment is determined to a large extent by their physical–chemical properties including water solubility, vapour pressure, and the partition coefficients between air and water (K_{aw}), octanol and water (K_{ow}) and soil organic carbon and water (K_{oc}) (IPCS 1998, Environment Agency 2008b). Higher molecular weight PAHs like benzo[a]pyrene are relatively immobile in the environment and bind strongly to organic matter. The half-life of benzo[a]pyrene in agricultural soils following sludge application has been estimated to range from 3 to 15 years, with an average of 8 years (Wild et al. 1991).

Field and pot experiments have generally reported low to very low soil-to-plant concentration factors (in the range 10^{-2} to 10^{-4} on a fresh weight basis) for PAHs in a range of different crops. Translocation of PAHs from roots to edible aboveground leaves and shoots is considered poor, with greater emphasis on contamination of below ground roots and tubers. OEHH (2012) observed that:

‘... although there are a considerable number of studies investigating PAH exposure in the environment, there are surprisingly few studies that provide reliable data for estimating transfer coefficients in food-producing animals’.

Despite their lipophilicity and potential for bioaccumulation in fatty tissues, a decline in bioaccumulation of PAHs with increasing molecular weight was noted by OEHH (2012), citing both lower gut assimilation efficiency and increased metabolism rates as possible reasons.

Figure 5.5 shows the breakdown of predicted adult and infant intakes by dietary food group. The critical groups for human consumption were milk and meat. PAH transfer into milk and meat is dependent on levels in animal diet, which typically includes cereals and grass, and incidental soil ingestion during grazing. In the modelling work for this risk assessment, almost 80% of livestock animal intakes of benzo[a]pyrene were predicted to derive from the direct ingestion of contaminated soil.

Figure 5.6 presents the relationship between predicted adult consumer dietary intakes and measured benzo[a]pyrene levels in composted street leaf sweepings after 10 applications to soil. For comparison the figure also shows the predicted adult intake (0.00076 mg/day for an adult with a bodyweight of 76 kg) associated with a minimal risk level. Of the 31 composted trial outputs, around 20% (7) had measured benzo[a]pyrene levels >4 mg/kg DW, which if applied repeatedly as a soil improver to agricultural land corresponded to a predicted adult intake twice the minimal risk level for adults.

Table 5.10 Chemical parameters used to estimate agricultural transfer of benzo[a]pyrene

| Parameter | Value (at 10°C unless stated) | Notes and reference |
|---|-------------------------------|--------------------------------------|
| Air–water partition coefficient (K_{aw}) | 1.76E-06 | Environment Agency (2008b) |
| Diffusion coefficient in air | 4.38E-06 m ² /s | Environment Agency (2008b) |
| Diffusion coefficient in water | 3.67E-10 m ² /s | Environment Agency (2008b) |
| Log (octanol–water partition coefficient) (K_{ow}) | 6.18 | Environment Agency (2008b) |
| Log (organic carbon–water partition coefficient) (K_{oc}) | 5.11 cm ³ /g | Environment Agency (2008b) |
| Molecular weight | 252.31 g/mol | Environment Agency (2008b) |
| Vapour pressure | 2.00E-08 Pa | Environment Agency (2008b) |
| Water solubility | 3.8E-03 mg/L (25°C) | Environment Agency (2008b) |
| Soil-to-plant transfer into cereals | 3.7E-07 kg/kg DW | Modelled (Environment Agency (2009)) |
| Soil-to-plant transfer into green vegetables | 1.6E-05 kg/kg DW | Modelled (Environment Agency (2009)) |
| Soil-to-plant transfer into root vegetables | 2.2E-02 kg/kg DW | Modelled (Environment Agency (2009)) |
| Soil-to-plant transfer into fruit | 8.9E-06 kg/kg DW | Modelled (Environment Agency (2009)) |
| Soil-to-plant transfer into grass | 7.8E-06 kg/kg DW | Modelled (Environment Agency (2009)) |
| Transfer coefficient into meat | 7.00E-02 day/kg FW | OEHH (2012) |
| Transfer coefficient into offal | 7.00E-02 day/kg FW | Assumed same as for meat. |
| Transfer coefficient into milk | 1.00E-02 day/kg FW | OEHH (2012) |
| Transfer coefficient into eggs | 3.00E-03 day/kg FW | OEHH (2012) |

Figure 5.5 Relative contributions of different food groups to predicted total diet exposure for adults and infants

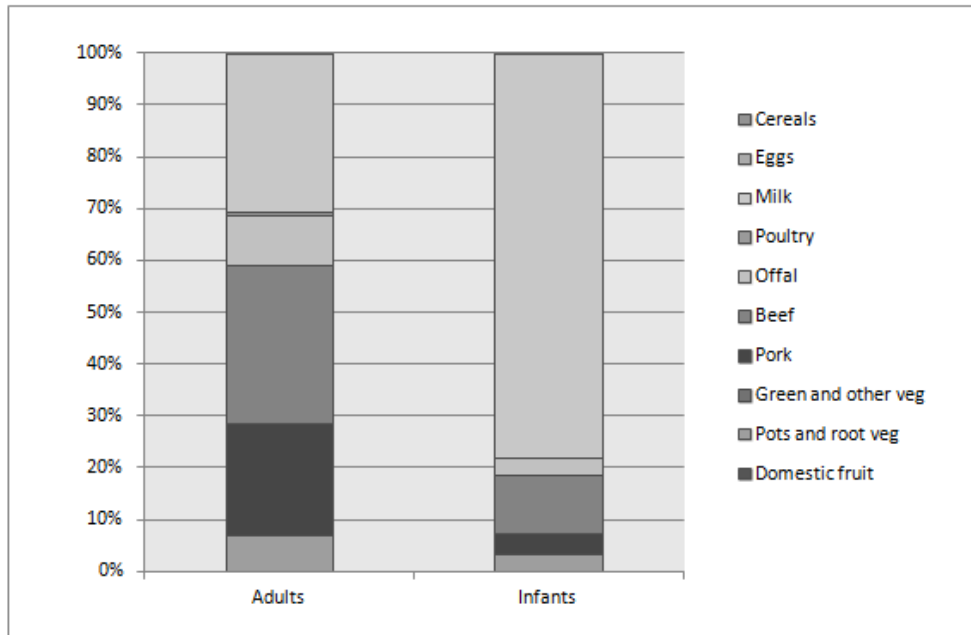
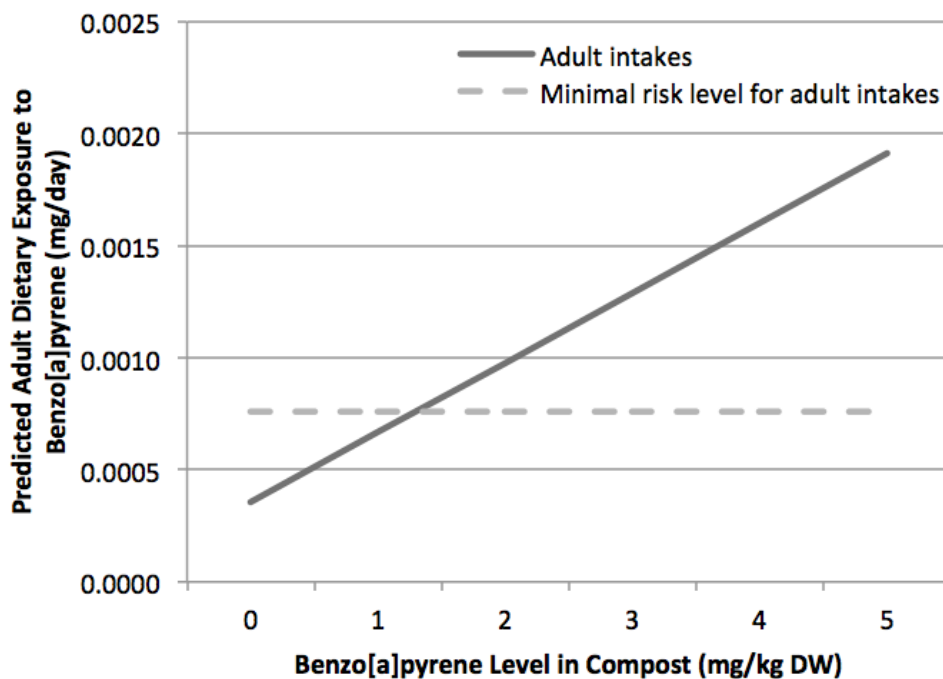


Figure 5.6 Predicted levels of adult consumer exposure after 10 applications according to benzo[a]pyrene levels in composted street leaf sweepings sampled in this trial



Note: Minimal risk level is shown for comparative purposes.

6 Conclusions

6.1 Conclusions from the trial

There are 230 waste collection authorities in England and a further 91 unitary authorities. Only 22 authorities (less than 10%) participated in the trial. The majority of the participants were located in the south and south-west of England. Taking into account the number of samples received, a significant proportion of the data collected came from just one authority (33% of street leaf sweepings and 56% of composted outputs).

PAHs were the most significant contaminants found in street leaf sweepings collected by these trials and at levels consistently higher than typically found in green and food wastes. Composted street leaf sweepings in this trial also had much higher PAH levels than those found in green and green/food composts in a study by WRAP (2011). PAHs made a major contribution to the poor results achieved by composted street leaf sweepings in the assessments for agricultural and horticultural use. Overall, only 6% of the samples of composted outputs in the trial met the criteria in this report for Quality Compost and only 19% of samples were considered to have passed the generic risk assessment.

The presence of PAHs in the roadside environment was not unexpected, but the scale of the differences between street leaf sweepings and green wastes (often from urban parks and gardens) was more surprising. Some PAHs are known and potent genotoxic carcinogens, and government advice is that exposure to such compounds should be kept as low as reasonably practicable (ALARP). Compared with existing green and green/food composts (WRAP 2011), agricultural use of the composted street leaf sweepings in this trial would result in much higher PAH emissions to soil, which would potentially lead to a relative increase in risks to health and the wider environment.

The small sample sizes and the inherent bias towards a few authorities meant that it was difficult to draw general conclusions or produce a national picture from the results of this trial. As noted specifically by the peer review (see Appendix D), the specific challenges identified in this investigation are common to these types of study. The dataset was too small to allow any detailed assessment of the potential links between methods of street leaf sweepings collection and contamination levels in the analysed materials, and limited the possibilities for statistical analysis. As expected from the nature of the materials, the trials found a considerable variation in the levels of contamination in samples of street leaf sweepings and their composted outputs, with large differences observed in samples collected by the same authority. However, it is clear that the dataset from this trial does not support any change to the current Environment Agency guidance on the use of street leaf sweepings to produce compost for agricultural or horticultural use. This view was endorsed by the peer review, which considered that “... *the current dataset would seem unlikely to support any other conclusions than those drawn here.*”

PAHs were investigated in this trial by following a waste streams approach to examine the inherent hazards present in street leaf sweepings. If the trial had considered only existing guidance on compost quality and risk (DoE 1996, BSI 2011), then these additional hazards posed by street leaf sweepings might have been overlooked.

6.2 Recommendations

The trial highlighted a number of gaps in the evidence base around the collection and biological treatment of street leaf sweepings, and their potential suitability for agriculture and other end uses. Recommendations for how these evidence gaps could be addressed are set out below.

Recommendation 1: Further work to examine the suitability of street leaf sweepings for producing CLO

One of the aims of this trial was to assess the suitability of composting street leaf sweepings for agricultural or horticultural use. An additional option for reuse of such materials is the restoration of previously developed land.

Site operators who participated in the trial were required to make provision for use or disposal of the composted outputs generated by the composting of street leaf sweepings. Before the trial began, the Environment Agency considered that the composted trial outputs were most likely to be classed as CLO, which could be used to restore non-agricultural land to beneficial use. However, this possible end-use depends on many different factors including location, specific use and consistency in controlling the variable levels of contamination. The suitability of the materials collected by this trial for CLO could be assessed in future work as an alternative to disposal.

Recommendation 2: Further work to better understand the variation in contamination in street leaf sweepings and trial outputs

A significant limitation of the trial was the small number of samples collected and the inherent bias within the results towards one or two authorities. This made it difficult to understand the reasons for the variation in levels of contamination between samples and the potential impacts of factors such as collection routes and methods. A smaller geographical study by one or more authorities with a more detailed and more experimentally sound sampling plan for street leaf sweepings collection and composting could provide a better understanding. Any such additional work could potentially consider contaminants that were not investigated in this trial including PGEs and pesticides.

Recommendation 3: Further work on the treatment options to improve compost quality

PAHs were the main contaminants affecting composted output suitability, in particular the four- and five-ring compounds including benzo[a]pyrene. Aerobic biological treatment of PAHs is known to be challenging but it has been used successfully to treat contaminated soils. It would be useful to understand whether a consistent and higher output quality could be achieved by revising the composting process such as by lengthening treatment time or through the use of novel inoculants.

In addition, particulate matter such as entrained dust and dirt may be a significant source of the heavier molecular weight PAHs investigated in this trial and pretreatment to remove particulates such as washing or physical separation may improve compost quality. Data from a commercial pretreatment process were available for the trial but were insufficient to allow any conclusions about its effectiveness. A well-designed and operated study to examine the treatment process in more detail may have merit. However, a number of operators already have environmental permits to treat street leaf sweepings and the data to demonstrate the effectiveness of pretreatment processes may already be available.

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List of abbreviations

| | |
|-------|---|
| BTEX | benzene, toluene, ethyl benzene and xylenes |
| BW | bodyweight |
| CLO | compost like output |
| DW | dry weight |
| EPH | extractable petroleum hydrocarbons |
| FW | fresh weight |
| GRO | gasoline range organics |
| IPTS | Institute for Prospective Technological Studies |
| MBT | mechanical biological treatment |
| ORG | Organics Recycling Group |
| PAH | polycyclic aromatic hydrocarbon |
| PGE | platinum group element |
| POP | persistent organic pollutant |
| PTE | potentially toxic elements |
| SVOC | semi-volatile organic compound |
| UKSHS | UK Soil and Herbage Survey |
| USEPA | United States Environment Protection Agency |
| VOC | volatile organic compound |
| WDF | WasteDataFlow |
| w/w | weight per weight |

Appendix A: Literature review – characterisation of street leaf sweepings

The Environment Agency is working with local authorities and ORG⁸ to better understand the risks of using street leaf sweepings in composting facilities. Its current position is that untreated street leaf sweepings are not regarded as acceptable wastes for composting or anaerobic digestion (Environment Agency 2012a).

This literature review summarises the available information on the characterisation of street leaf sweepings with the aim of identifying the likely chemical and physical hazards to human health and the environment from their presence in composted materials.

A.1 Bulk characterisation

Street sweepings (also known as street cleansing materials) include leaves, grit, litter, glass, oils, paper, plastics and other materials (CDEP 2007, Environment Agency 2012a). Street leaf sweepings are generally a seasonal issue, with most leaf fall occurring in the autumn and early winter. Considerable quantities can accumulate on pavements, verges, road surfaces and central reservations.

Some local authorities operate dedicated street leaf sweepings collections as part of their street cleaning programmes (Environment Agency 2012a), contributing an estimated collection of around 67,000 tonnes of street leaf sweepings per year (based on WDF figures to the Environment Agency for all the local authorities in England).

Although these wastes will mainly be leaf litter, they may also contain physical contaminants such as plastics, paper, foil and grits. In a small scale study in Wales, collected street leaf sweepings contained 0–10% soil and grit by volume and up to 2% other recyclables including glass and paper (Environment Agency 2012b). In addition to street leaf sweepings, the authorities in Portland, Oregon, USA, also recycled considerable quantities of sand, grit, and asphalt grindings from street sweeping collections (Anon 1998).

A.2 Chemical characterisation

Street leaf sweepings are generally from:

- trees, hedges and bushes growing close to the road network along managed and unmanaged verges, or in the central reservation
- residential, municipal and commercial parks, gardens and landscaped areas adjacent to roads and pavements

Chemical contamination may arise from plant uptake of pollutants through the soil or via the air followed by translocation within the plant to leaves and stems (Kabata-Pendias and Mukherjee 2007) and from the deposition of airborne dusts or the

⁸ The Organics Recycling Group was formed on 1 January 2013 by the merger of the Association for Organics Recycling and the Renewable Energy Association.

entrainment of soils and sediments on leaf surfaces and within fallen plant materials. There are several sources of this plant, soil and dust contamination:

- non-localised atmospheric deposition of dusts, gases and vapours
- localised deposition of dusts, gases, liquids and vapours from road vehicles
- localised deposition of dusts, gases and vapours from roads
- localised application of chemicals for landscape management

A.3 Sources of chemical contamination

A.3.1 Non-localised atmospheric deposition

General atmospheric deposition of contaminants is a significant source of diffuse pollution in urban and rural soils (Nicholson et al. 2003). While atmospheric deposition is ubiquitous, deposition rates depend on proximity to point sources of pollution such as heavy industry or major roads (Defra 2010). The most important sources of heavy metals in the atmosphere include energy production, mining, metal smelting and refining, manufacturing processes, transport and waste incineration (Environment Agency 2007b). Metals are deposited from air to soil and plant surfaces via several processes including wet, dry and occult deposition (Nicholson et al. 2003). Metals deposited on the soils will gradually become incorporated into the soil and will contribute to overall soil concentrations. Metals falling on to vegetation may remain on the leaf surface, be absorbed by the plant or be washed off into the soil by rainfall. Atmospheric deposition is a significant source of heavy metal inputs to agricultural land in England and Wales (Table A.1).

Table A.1 Estimated total annual metal inputs (tonnes/year) to agricultural soils in England and Wales from different sources for 2008

| Source | Zn | Cu | Ni | Pb | Cd | Cr | As | Hg |
|---------------------------------|--------------|--------------|------------|------------|-------------|------------|-----------|-----------|
| Atmospheric deposition | 1,009 | 333 | 72 | 99 | 6.3 | 20 | 8.1 | 9.0 |
| Livestock manures | 998- | 364 | 21 | 34 | 1.7 | 23 | 9.5 | 0.1 |
| Biosolids | 701 | 364 | 42 | 167 | 1.9 | 101 | 6.6 | 1.5 |
| Industrial wastes | 80 | 41 | 32 | 10 | 1.2 | 10 | 0.2 | 0.2 |
| Dredgings | 63 | 17 | 8 | 19 | 0.3 | 9 | 2 | 0.2 |
| Compost | 116 | 34 | 9 | 64 | 0.4 | 14 | <0.1 | 0.1 |
| Fertilisers and lime | 150 | 32 | 25 | 12 | 6.7 | 71 | 4.5 | <0.1 |
| Ash | 145 | 43 | <1 | <1 | <0.1 | <1 | <0.1 | <0.1 |
| Other sources | 74 | 19 | <1 | <1 | <0.1 | <1 | <0.1 | <0.1 |
| Total | 3,336 | 1,248 | 210 | 406 | 18.5 | 248 | 31 | 11 |
| % atmospheric deposition | 30 | 27 | 34 | 24 | 34 | 8 | 26 | 82 |

Notes: Defra (2010)

A.3.2 Localised deposition from road vehicles

Emissions from vehicles make a significant contribution to anthropogenic pollution close to the roadside. The main sources of contaminants from vehicles are particulate and gaseous exhaust emissions, particulate emissions from other parts of the car such as tyre and brake wear (Thorpe and Harrison 2008), and liquid leaks from the engine, transmission and coolant systems. Studies have shown that exhaust and non-exhaust sources contribute equally to total traffic related emissions of particulate matter (Thorpe and Harrison 2008).

Air pollutants

In urban areas, motor vehicle exhaust emissions are the main source of air pollutants including carbon monoxide, carbon dioxide, nitrous oxides, methane, derived ozone and non-methane hydrocarbons (Colvile et al. 2001, Maricq 2007, Tsai et al. 2012, Wang et al. 2013). The major gaseous contaminants are unlikely to accumulate in soil or plant matter and are not relevant to leaf litter. Of greater importance are emissions of particulate matter (including heavy metals and partial combustion products like PAHs) and the wider range of VOCs and SVOCs (Maricq 2007, Pulles et al. 2012, Tsai et al. 2012, Wang et al. 2013).

Metal emissions from exhausts

The three main sources of metal emissions from the exhaust are combustion of fuel, use of lubricants and abrasion of the engine or after-treatment system including the catalyst. Table A.2 presents summary data on the concentrations of metals found in petrol and diesel in Europe; sample variability was high for all metals measured.

Engine lubricant oils are known to be high in zinc (Pulles et al. 2012) due to the use of zinc dialkyl dithiophosphate as an anti-wear additive with levels in the range 750 to 1,100 mg/kg. Although engine oils contain a range of other additives including viscosity index improvers, corrosion inhibitors, detergents and dispersants, metal contents in pure oils are generally kept low to minimise damage to catalytic convertors. In used oils, metal contents can be significantly increased due to engine wear. Table A.2 shows typical levels of elements in used lubricating oils for diesel and petrol engines. Table A.3 summarises the most common sources of elements in engine lubricating oils. Mercury, selenium and arsenic are considered to be present at negligible levels in oils (Winther and Slento 2010).

The increased use of catalytic convertors has also been linked to the release of PGEs including platinum, palladium and rhodium into the environment. Platinum and palladium are used to oxidise carbon monoxide and other hydrocarbons in the exhaust gases, while rhodium is used to reduce nitrous oxides.

PGEs have been detected in urban air and roadside soils and dusts, although the reported levels, 10–50 pg/m³ air and 10–400 ng/g soil, are generally very low (Kalavrouziotis and Koukoulakis 2009). A significant difference in average platinum levels in road dusts between heavily trafficked roads (mean 132 ng/g) and lightly trafficked roads (mean 23 ng/g) was suggested to reflect the importance of exhaust emissions to roadside levels (Lee et al. 2012). Due to the rarity and value of PGEs, some serious consideration has been given to its economic recovery from street dusts (Derbyshire, 2011).

Table A.2 Geometric mean element concentrations ($\mu\text{g}/\text{kg}$) in petrol and diesel from several European countries (Pulles et al. 2012) and metal contents in used engine oils (Winther and Slento 2010)

| Element | Petrol | Diesel | Petrol engine oil | Diesel engine oil |
|---------|--------|--------|-------------------|-------------------|
| Ag | nd | nd | 3,000 | 3,000 |
| Al | nd | nd | 5,000–20,000 | 10,000 |
| As | 0.27 | <0.05 | nd | nd |
| Cd | 0.26 | <0.025 | 5,000 | 5000 |
| Cr | 5.3 | 8.7 | 5,000–20,000 | 1,000–8,000 |
| Cu | 4.3 | 6.3 | 5,000–30,000 | 3,000–15,000 |
| Fe | nd | nd | 5,000–25,000 | 10,000–40,000 |
| Hg | 7.5 | 2.4 | nd | nd |
| Ni | 2.3 | 0.12 | 5,000 | 5,000 |
| Pb | 1.6 | 0.4 | 30,000 | 15,000 |
| Na | nd | nd | 20,000 | 25,000 |
| Se | 0.18 | <0.05 | nd | nd |
| Sn | nd | nd | 20,000 | 15,000 |
| Zn | 33 | 18 | 1,000,000 | 1,000,000 |

Note: nd = not detected

Organic emissions from exhausts

Vehicle exhaust emissions are one of the most important sources of VOCs in urban areas and several studies have demonstrated that the concentration and type of VOCs depend on the car type, engine condition and other factors (Wang et al. 2013). Measurements of VOCs emitted from petrol cars driven under a variety of conditions have identified more than 50 individual compounds. The 10 highest compounds by weight fraction were:

- ethylene
- toluene
- benzene
- isopentane
- ethyne and propylene
- 1,3-dimethylbenzene and 1,2,4-trimethylbenzene
- ethane
- 2-methylpentane

Together these 10 compounds amounted to more than 50% of the mass of compounds present (Wang et al. 2013).

Table A.3 Most common sources of elements in lubricating oils

| Element | Sources |
|---------|---|
| Ag | Trace element |
| Al | Pistons, bearings, cases (heads and blocks) |
| B | Detergent/dispersant additive, antifreeze inhibitors |
| Ba | Detergent/dispersant additive |
| Ca | Detergent/dispersant additive |
| Cr | Rings, a trace element in steel |
| Cu | Brass or bronze parts, copper brushings, bearings, oil coolers, also an additive in some oils |
| Fe | Cylinders, rotating shafts, the valve train and any steel part sharing the oil |
| K | Antifreeze inhibitor, additive in some engine oils |
| Mg | Detergent/dispersant additive |
| Mn | Trace element, additive in petrol |
| Mo | Anti-wear additive, coating on some new rings (washes off during running in) |
| Na | Antifreeze inhibitors, additive in some engine oils |
| Ni | Trace element in steel |
| P | Anti-wear additive |
| Pb | Bearings |
| Si | Airborne dirt, sealers, gaskets, antifreeze inhibitors |
| Sn | Bearings, bronze parts, piston coatings |
| Ti | Trace element |
| Zinc | Anti-wear additive |

Note: After Winther and Slento (2010)

A study of light-duty diesel vehicles found the dominant VOCs to include styrene, *n*-propylbenzene, *n*-undecane, ethyltoluene, 1,2,4-trimethylbenzene, toluene, xylene, isopropylbenzene and ethylbenzene, with emission factors between 10 and 60 mg/km. The major carbonyl species from diesel exhausts were found to be formaldehyde, acetaldehyde, acetone, butyraldehyde and *m*-tolualdehyde, with emission factors of between 1 and 10 mg/km (Tsai et al. 2012).

The chemical composition of exhaust particulates has been extensively reviewed (Maricq 2007). Commonly found hydrocarbons include PAHs (including substituted nitrated PAHs), alkanes, steranes, hopanes and various organic acids – notably alkanolic and benzoic acids. Many of these compounds originate from unburned fuels and lubricating oil components, their partially oxidised products, and metallic ash from fuel and oil additives (Maricq 2007). Although well over 100 hydrocarbons have been individually identified in exhaust gases, these typically constitute less than 5% by

weight of the organic material identified in particulate matter. Most organic chemicals in particulate matter remain challenging to extract, resolve and identify.

In a survey in California, naphthalene was the most abundant PAH in petrol and in diesel, along with phenanthrene and fluorene (Marr et al. 1999). The total PAH content of diesel was reported to be 1–4% by weight. The low molecular weight PAHs such as pyrene and fluoranthene are predominantly emitted by diesel vehicles, while higher molecular weight PAHs like benzo[ghi]perylene are used as tracers for petrol engine contamination (Phuleria et al. 2007, Liu et al. 2008, Liacos et al. 2012). An analysis of emissions from different diesel cars and vans found that emission rates were highest for the low molecular weight PAHs including naphthalene, phenanthrene, acenaphthylene and fluorene (Nelson et al. 2008). An investigation of particulate emissions from a heavy goods vehicle found the highest emission rates for pyrene and fluoranthene, which together accounted for 90% of the PAHs measured (Mabilia et al. 2004). The main difference between these two studies appears to be that two- and three-ring PAH compounds are almost exclusively emitted in the gas phase, while the four-ring PAHs are mostly found as particulates (ATSDR 1995). Unlike other VOCs, the PAH profile in vehicle emissions has been shown to reflect fuel composition (Marr et al. 1999, Nelson et al. 2008). Road vehicle exhausts in the UK have been estimated to emit around 130 tonnes of PAHs annually (Napier et al. 2008).

Emissions from brake wear

Brake and tyre wear are an important source of trace metals in the urban environment (Hulskotte et al. 2007, Thorpe and Harrison 2008). Frictional contact between brake system components results in wear of the brake lining materials and the brake disc/drum. Investigation of the physical and chemical properties of brake wear particles is hindered by the wide range of brake lining materials currently used (Thorpe and Harrison 2008). These include:

- fibres providing mechanical strength and constituting up to 35% of brake lining mass made from metals, carbon, glass and synthetic materials such as Kevlar
- abrasives to increase friction and keep surfaces clean make up around 10% of the lining and are made from aluminium oxides, iron oxides, quartz and zircon
- lubricants to stabilise frictional properties constitute up to 30% of the brake lining made from graphite and various metal sulphides such as antimony trisulphide
- fillers to reduce manufacturing costs constitute up to 70% of the brake lining and include barite and calcite
- binders to maintain structural integrity of the brake linings under extreme mechanical and thermal stresses and include phenolic resins

The proportion of the different components varies depending on the type of lining. Modern brake lining materials are composites of many different ingredients, which are responsible for differing wear characteristics, physical and chemical properties of the bulk lining materials and emitted particles. Experimental studies noted by Hulskotte et al. (2007) suggested around 50% of wear was emitted to the atmosphere, up to 30% fell on to the road surface, and the remainder was retained on the car. Table A.4 lists metal concentrations from a range of sources. Hulskotte et al. (2007) noted the importance of brake linings to the emission of copper to the environment. Gietl et al. (2010) noted that the metals with highest abundances were barium, magnesium,

antimony and zinc, and that at traffic-influenced sites it seemed likely that brake wear was the main source of these elements.

Although numerous organic substances are utilised in brake lining manufacture, there is limited information on the presence of specific organic compounds because brake linings have to be able to resist extreme thermal and mechanical stress, as well as refractory attack by solvents in case of brake fluid leakages. Consequently, only a small proportion of the organic content of brake linings is extractable (Thorpe and Harrison 2008).

Table A.4 Metal concentrations present in brake linings and emitted brake dust

| Metal | Concentration (mg/kg) | |
|-------|-----------------------|----------------|
| | Car brake linings | Car brake dust |
| Al | 3,765 | 330–2,500 |
| As | <2–18 | <2–11 |
| Ba | 2,638 | 5,900–74,400 |
| Ca | 14,300 | 920–8,600 |
| Cd | <1–41.4 | <0.06–2.6 |
| Co | 6.4–45.8 | 12–42.4 |
| Cr | <10–411 | 135–1320 |
| Cu | 11–23,4000 | 70–39,400 |
| Fe | 1.2–63.7 % | 1.1–53.7% |
| K | 857 | 190–5,100 |
| Li | 55.6 | – |
| Mg | 6,140 | 83,000 |
| Mn | 181–3,220 | 620–5,640 |
| Mo | 0.4–215 | 5–740 |
| Na | 15,400 | 80 |
| Ni | 3.6–660 | 80–730 |
| Pb | 1.3–119,000 | 4–1,290 |
| Sb | 0.07–201 | 4–16,900 |
| Se | <1–15 | 4.5–115 |
| Sr | 81.4 | 300–990 |
| Zn | 25–188,000 | 120–27,300 |

Notes: Thorpe and Harrison (2008)

Emissions from tyre wear

Tyre wear is an important contributor to PM₁₀ emissions to the atmosphere, with annual losses of rubber from tyres in the UK estimated to be 53,000 tonnes (Environment Agency 1998). Frictional contact between the road surface and tyre tread results in abrasion and emission of particles into the atmosphere. As is the case for brake lining materials, a range of tyre types, sizes and compositions are used in the motor vehicle industry according to required performance (Thorpe and Harrison 2008). In general, tyres consist of a rubber compound reinforced with steel wires and textiles (Environment Agency 1998).

Although tyre wear has been considered primarily a source of organic compounds, approximately 13% by mass is inorganic material, present in the form of various curing agents, accelerators and other additives (Thorpe and Harrison 2008). Around 22 tonnes of PAH are estimated to be emitted annually to the environment from tyre erosion using a typical PAH tyre concentration of 350 mg/kg (Napier et al. 2008). Tyre wear has been identified as a significant source of zinc in the environment; zinc oxide is used as an activator in the vulcanisation process. A summary of metal concentrations in tyres from a range of sources is shown in Table A.5.

Table A.5 Metal concentrations in passenger car tyre tread

| Metal | Concentration (mg/kg) |
|-------|-----------------------|
| Al | 7–470 |
| Ba | 10.4–370 |
| Ca | 100–2,000 |
| Cd | <0.05–2.6 |
| Co | 0.5–4.1 |
| Cr | <1–30 |
| Cu | <1–490 |
| Fe | 40–4,600 |
| K | 110–410 |
| Mg | 6,140 |
| Mn | 0.8–100 |
| Na | 120–530 |
| Ni | <1–50 |
| Pb | 1–160 |
| Sb | <0.2–0.9 |
| Sr | <0.5–40 |
| Ti | 560 |
| Zn | 430–10,250 |

Notes: After Thorpe and Harrison (2008)

Vehicle spills and leaks

It has been estimated that around 16,000 tonnes per year of oil is released to the environment as a result of vehicle spills and leaks (Napier et al. 2008). PAH concentrations in unused oil are generally low with only phenanthrene, anthracene and fluoranthene present at levels greater than 10 mg/kg. However, much higher levels (up to a thousand times) have been reported in used engine oils. Estimates suggest that losses of used and unused oils may result in as much as 320 tonnes PAHs per year entering the environment (Napier et al. 2008).

A.3.3 Localised deposition from roads

Most road surfaces in the UK have the same component materials – a mixture of aggregates of various grain size, bitumen and modifiers such as fillers and adhesives (Thorpe and Harrison 2008). Heavy use and adverse weather can result in significant road surface abrasion and the generation of road dust (Winther and Slento 2010).

Road surface materials

Road surface materials may be broadly categorised as concrete or asphalt (Thorpe and Harrison 2008). Concrete road materials are a mixture of mineral aggregate, sand and cement. Asphalt consists primarily of mineral aggregates (around 95% by weight) combined with a mostly bituminous binder. The most commonly used binder, petroleum bitumen, is obtained from the distillation of crude oils and is made up of many thousands of different high molecular weight aliphatic and aromatic hydrocarbons (Nicholls 1998). A summary of the broad composition of bitumen is shown in Table A.6.

Table A.6 Composition of bitumen

| % Weight | Components |
|----------|---|
| 5–25 | Asphaltenes – brown or black amorphous solids with molecular weights in the 1,000 to 50,000 range |
| 5–50 | Resins – dark brown solids or semi-solids with molecular weights between 900 and 1,300 |
| 40–60 | Aromatics with molecular weights between 500 and 900 |
| 1–25 | Saturates with molecular weights in the range 500 to 800 |

Notes: After Nicholls (1998)

PAHs have been found in bitumen samples, but the concentrations of individual compounds are very low due to their removal during the distillation process (Gadd and Kennedy 2003, Winther and Slento 2010). The major components of asphalt have been reported as silicon dioxide (sand), aluminium, iron, calcium, potassium, manganese, magnesium and titanium, with significant trace elements being barium, strontium, zirconium, zinc, vanadium, lead and chromium (Winther and Slento 2010). A summary of trace elements in asphalt is shown in Table A.7. These natural levels are low relative to metal contamination incorporated from other sources such as brake linings and tyre wear (Thorpe and Harrison 2008).

Various fillers and fibres may be added to reinforce binder–aggregate bonding including glass, coal fly ash and shredded rubber tyres (Thorpe and Harrison 2008). Sulphur may be added to harden the binder along with other modifiers including polymers, epoxy resins, and metal complexes. Van Metre et al. (2009) highlighted the

potential for PAH-contaminated dust particles to be washed from coal-tar based sealants which are used to protect pavements and car parks from weathering and aging. They observed PAH concentrations up to 65 times greater from treated surfaces compared with bare concrete or asphalt.

Table A.7 Metal concentrations in asphalt binder

| Metal | Metal content (mg/kg) |
|-------|-----------------------|
| Cr | 1–35 |
| Cu | 0.3–17 |
| Fe | 12–30 |
| Ni | 15–100 |
| V | 50–600 |
| Zn | 10–546 |

Notes: After Winther and Slento (2010)

Road surfaces can also accumulate particulate material from a wider variety of sources including vehicle emissions (as discussed previously), salting and gritting activities, and the erosion and deposition of material from the roadside such as mud (Ramakrishna and Viraraghavan 2005, Thorpe and Harrison 2008). This deposited material is often called road dust or road-deposited sediment, and its resuspension is an important source of particulate matter emissions.

Road dusts

The composition of road dust has been found to be dominated by elements and compounds typically associated with rocks and soils (Thorpe and Harrison 2008). It will therefore often reflect the local geology and vary greatly between locations. Road dusts and roadside soils often contain significant concentrations of metals including cadmium, chromium, copper, iron, lead, manganese and zinc.

Road dust collected from the streets of Manchester consisted of a wide range of grain types including (Taylor and Robertson, 2009):

- silicate and alumina-silicate minerals
- iron oxides from corrosion of steel
- iron-rich glass derived from slags and waste processing activities
- spherical iron oxides and glasses from high temperature combustion processes
- minor amounts of pure metallic grains such as copper and lead

Most particles were in the medium to fine sand size fraction (63–300 µm) with, on average, only 8% being in the finest grain-size fraction (<63 µm) (that is, silt and clay fraction) with no marked temporal variation over a nine-month period (Robertson and Taylor 2007). The authors also observed that the iron oxide and iron-rich glass particles were the major hosts for heavy metal contaminants. Although contaminant enrichment in finer particles has been reported elsewhere, most metals are found in the coarser fractions because of their dominant contribution to mass (Robertson and Taylor

2007). Several authors including CDEP (2007) have noted that zinc and copper has surpassed lead as being the most common metals found in road dusts and sediments.

Temporal variability in contaminant loadings in road dusts will depend on several factors including source variability (both type and magnitude) and also contaminant mobility. Robertson and Taylor (2007) found that the lack of temporal variability in copper concentrations observed in Manchester road dusts may be due to copper's preferential and strong adsorption to organic matter particles, which lessened the likely effect of seasonal weather on its dissolution and leaching. Levels of lead and zinc were shown to vary throughout the year with the authors suggesting this might be linked to traffic density.

De-icing chemicals

Large quantities of de-icing chemicals are applied to roads and pavements during the winter months to ensure the safety of drivers and pedestrians (Ramakrishna and Viraraghavan 2005). Approximately two million tonnes of salt (sodium chloride) are spread on the UK roads in each year with the majority spread on to motorways, trunk roads and main roads (Salt Union 2013). Less than a third of other roads are treated. Crushed rock salt has a tendency to cake quickly in storage, which means that stockpiling for a winter maintenance programme is impossible without the use of an anti-caking agent (Salt Association 2013). UK salt producers generally use sodium hexacyanoferrate (II) at about 100 mg/kg or less for this purpose.

A.3.4 Localised application for landscape management

Management of roadside vegetation including weed control is an important part of maintenance for driver safety, to protect engineered formations and for landscape aesthetics (Ramwell et al. 2002, Barker and Probst 2009). A significant amount of the herbicides applied to hard surfaces are washed away into surface drainage following rainfall. In a roadside study, Ramwell and Probst (2002) found that total percentage losses of applied active ingredients were 75, 66 and 35% for atrazine, diuron (no longer authorised) and glyphosate respectively after 25 mm of accumulated rainfall. Others such as those based on oryzalin and oxadiazon had losses much less than 10%.

A survey of amenity use of pesticides, including road and roadside applications, in the UK found that around 750 tonnes of pesticides were sold in 2006 for amenity and industrial use with 91% being herbicides – mostly glyphosates with much smaller proportions of diuron and 2,4-dichlorophenoxyacetic acid (2,4-D) – and the remainder a combination of algaecides, fungicides, insecticides and moss-killers (HSE 2011). Diuron products were used primarily to treat hard surfaces such as roads and pavements but it was anticipated that, following its withdrawal, there was likely to be a significant rise in those using glyphosate chemicals (HSE 2011). In 2006, fungicide treatment by local authorities was dominated by thiophanate-methyl (almost 25% of non-herbicide use) and dichlorophen, while for moss-killing it was ferrous sulphate and dichlorophen. Dichlorophen has since been withdrawn.

A.4 Contaminants in roadside soils and plants

A number of studies have looked at levels of pollutants in soil, air, dust and plants close to roads in the UK and internationally. Pollutants from vehicle emissions can be deposited on soils and plants by dry and wet aerial deposition, and by run-off water from road and pavement surfaces (Pagotto et al. 2001, Napier et al. 2008).

Air pollutants have been extensively monitored at roadside sites in the UK. Metal levels have been associated with organic carbon and iron oxide particles, which are formed by various roadside sources (Kim et al. 2009). Air quality monitoring adjacent to a busy trunk road into Birmingham found reasonable correlations between measures of traffic activity and pollutants including copper, zinc, molybdenum, barium and lead, and the PAHs fluoranthene, pyrene and methylphenanthrene (Harrison et al. 2003). Kawanka et al. (2008) measured a range of nitro-PAHs including 1-nitropyrene in air samples collected from the side of a busy highway in Tokyo, finding that concentrations in ultrafine airborne particles (<2.5 µm) were typically much higher than observed in coarser particles. Riddle et al. (2008) measured the size distribution of particles of hopanes, steranes and PAHs upwind and downwind of a freeway in California. They concluded that diesel fuel appeared to make the largest contribution to ultrafine particle mass.

Summary data on concentrations of soil contaminants along roadsides and in street dusts found in the literature review are shown in Tables A.8 and A.9 respectively. The G-BASE survey of Wolverhampton in the 1990s, one of the first extensive datasets for the UK, showed urban soils contaminated with heavy metals including lead along the major routes into and out of the city (Bridge et al. 1997). Kalavrouziotis and Koukoulakis (2009) reported a number of studies showing elevated levels of PGEs, concluding that this provided 'substantial and undisputed evidence' that automobile catalysts are the predominant source of PGE pollution. Lee et al. (2012) supported this assertion, observing a difference in average platinum levels between heavily trafficked roads (mean 132 ng/g) and lightly trafficked roads (mean 22.8 ng/g). Pagotto et al. (2001) measured various metal contaminants in soils adjacent to a rural highway and observed a sharp increase in concentrations within 2.5 m of the road. The authors also noted that in many cases the mobility of the metal contaminants was low with the exception of cadmium. PAH levels have also been extensively studied. Measurements of soil concentrations in three European cities, including Glasgow, found that phenanthrene, fluoranthene and pyrene accounted for 40% of the total PAH content (Morillo et al. 2007). The authors concluded that motor vehicle exhausts was likely to be the major source of these compounds in urban soils.

Robertson and Taylor (2007) examined spatial and temporal variation in metal contents of road dusts collected in Manchester. Temporal and spatial variability was observed in most cases but not all samples. Jung et al. (2009) characterised the pollutants in street sweepings in Florida and found that geometric mean levels of copper and zinc were statistically greater than levels found in ambient soils. Of the trace organic contaminants examined, which included VOCs, PAHs, phthalates and pesticides, they detected only di-*n*-butyl phthalate, pyrene, 4,4'-DDT and endosulfan II in more than 5% of samples. Clark et al. (2000) examined the metal and PAH content of road dust collected from towns in the west of Scotland including trunk, distributor and residential roads, and two car parks. They reported significantly elevated concentrations of copper, iron, lead and zinc with mean levels of 106, 35,000, 150 and 287 mg/kg respectively. Mean PAH and oil/grease levels were 167 and 15,000 mg/kg respectively, with fluoranthene, pyrene, benzo[k]fluoranthene, dibenzo[ah]anthracene and benzo[ghi]perylene accounting for 60% of the PAHs present.

Table A.8 Pollutant levels measured in roadside and urban soils

| Pollutant | Concentration |
|-----------|---------------------|
| Al | 10,800–33,400 mg/kg |
| Cd | 0.1–2.7 mg/kg |
| Cr | 21–80 mg/kg |
| Cu | 6.7–201 mg/kg |
| Fe | 5,800–36,400 mg/kg |
| Mn | 149–1,406 mg/kg |
| Na | 19–295 mg/kg |
| Ni | 5.9–26 mg/kg |
| Pb | 20–3,784 mg/kg |
| Pd | 1.1–440 ng/g |
| Pt | 0.3–444 ng/g |
| Zn | 149–1,406 mg/kg |
| PAHs | 148–51,822 µg/kg |

Notes: Pagotto et al. (2001), Bryson and Barker (2002), Morillo et al. (2007), Kalavrouziotis and Koukoulakis (2009), Lee et al. (2012)

Table A.9 Pollutant levels measured in street dusts and road sweepings

| Pollutant | Concentration |
|------------|--------------------|
| As | 0.5–14 mg/kg |
| Ba | 1.4–130 mg/kg |
| Cd | <0.1–1.5 mg/kg |
| Cr | 1.3–552 mg/kg |
| Cu | 28–3,721 mg/kg |
| Fe | 2,955–65,000 mg/kg |
| Mn | 70–294 mg/kg |
| Ni | 1.7–78 mg/kg |
| Pb | 1.4–660 mg/kg |
| Zn | 4.3–796 mg/kg |
| Phthalates | 5.4–29 µg/kg |
| PAHs | 0.005–167 mg/kg |
| Pesticides | 25–2,410 µg/kg |

Notes: Clark et al. (2000), Robertson and Taylor (2007), Jung et al. (2009)

Several authors have also looked at sodium levels in roadside soils to examine the impact of de-icing management practices (Bryson and Barker 2002, Marosz 2011). In a study in Massachusetts, Bryson and Barker (2002) found roadside sodium concentrations decreased markedly around 5 m from the road. In contrast, Marosz (2011) found no significant differences between leaves taken from roadside trees and a control sample from parkland.

A few studies have examined the level of pollutants, mostly heavy metals, found in plants with a focus on trees because they tend to collect particulate matter and to assimilate and accumulate pollutants through their leaves (Singh et al. 1995, Moreno et al., 2003, Maher et al., 2008, Power et al., 2009). Summary data on plant concentrations found in this literature review is shown in Table A.10.

A study of the levels of particulate matter and the leachable metal contents from leaves from birch trees adjacent to a busy main road in Norwich found that zinc and iron had the highest roadside concentrations, with manganese, barium and lead at lower levels, and cadmium and chromium below detection limits (Maher et al. 2008). Due to the association between pollutant loading and road gradient, the authors concluded that the pattern of exposure was due to exhaust emissions and would not have arisen from resuspension of roadside dusts. It was concluded that urban roadside tree leaves exhibited much higher accumulations of particulate pollutants including iron and lead than compared with leaves growing on a parkland site. Kalavrouziotis and Koukoulakis (2009) reported a number of studies showing elevated levels of PGEs in plants including grass. Bryson and Barker (2002) reported sodium concentrations in leaves from trees growing alongside roads treated with salt.

The study of gaseous pollutants has tended to focus on their impacts on vegetation in terms of growth, visible damage to leaves and other biochemical changes. For example, leaves are known to rapidly absorb sulphur dioxide which may in turn form phytotoxic species such as sulphite and bisulphite but it is also oxidised to sulphate and stored within plant tissues (Singh et al. 1995). Nakajima et al. (1994) measured levels of 1-nitropyrene, one of the most prevalent nitroarenes emitted from vehicle exhausts, in azalea leaves from various roadside locations.

Table A.10 Pollutant levels measured in roadside plants

| Pollutant | Concentration |
|---------------|-------------------------|
| Na | 0–4,131 mg/kg |
| Pd | 1.5–6.8 ng/g dry matter |
| Pt | 0.3–30 ng/g dry matter |
| 1-nitropyrene | 0.17–2.5 ng/g |

Notes: Nakajima *et al.* (1994), Bryson and Barker (2002), Kalavrouziotis and Koukoulakis (2009)

Appendix B: Methodology

B.1 Appropriate composting sites

Proposed facilities where composting was to take place were assessed to ensure their suitability and to confirm they could comply with the trial's rules. Composting was required to be undertaken on an impermeable surface with a designated wastewater and liquor storage. Exempt activities were excluded from the trial.

The trial applied only to seasonal leaf litter from street sweepings (EWC code 20 03 03). This excluded leaf litter from parks and gardens as this is not considered to be street sweeping waste. Trials could only be carried out at facilities permitted to accept waste code 20 03 03 in one of the following scenarios:

- a non-CQP composting process allowed to treat 20 03 03 waste as specified by the facility's environmental permit or management system
- a CQP composting process allowed to treat 20 03 03 waste as specified by the facility's environmental permit or management system under the derogation set out in clause 4.2.5 of ORG's Compost Certification Scheme Rules (ORG 2012a)

B.2 Collection of leaf litter in street sweepings

Initially the trial was set up to look at collection of seasonal leaf litter in street sweepings in rural areas and to subsequently compost the collected material – this is termed option 1. A request made to assess the quality of leaf litter in urban and semi-urban areas without composting led to development of option 2.

B.2.1 Option 1 input wastes

Option 1 involved the treatment by aerobic composting of separately collected leaf litter consisting of autumn leaf fall or plant material swept up from verges.

The local authority or operator was required to record how, when and where the clean leaf litter was collected (for example, reference/name or route, type of road if collected from the pavement or from the road side and so on). This was documented alongside individual batches to help the project team to understand differences with locations, collection methods and so on. It was envisaged that collection of road sweeping was a planned activity and that routes were clearly designated allowing information to be clearly auditable.

Pre-screening to remove identifiable litter such as cans and plastic items was acceptable. Where this was done the proportion of contamination was recorded and kept with route data.

It was permitted to blend a proportion of the leaf litter with green waste to balance nitrogen requirements. Operators were required to produce evidence of blending rates by weight at a minimum of 30% leaf litter (for example, 30 tonnes leaf litter to 70 tonnes green waste). The leaf litter feedstock, as well as the untreated garden waste blended with the leaf litter, was analysed in advance of treatment for a number of contaminants. Compost operators were expected to obtain the results of this analysis before combining street sweepings with green waste. It was expected that the resultant compost would be analysed using the same suite of tests.

B.3 Option 2 collection only

To allow urban areas to sample their leaf litter but without composting it, the project team agreed that some local authorities could just sample the leaf litter. Details required where samples were collected included:

- collection round
- specific road
- which surface it was collected from: road, pavement or verge
- what type of collection equipment was used

B.4 Composting process control

The following criteria were followed for the process of composting.

- Leaf litter was either composted by itself to produce a leaf mulch or blended with green wastes to the correct carbon to nitrogen ratio to produce a compost (minimum of 30% leaf litter: 70% green waste).
- Batch control was required to be carried out and to be auditable through all stages of the process. This included leaf litter blended with green waste, which was required to be in separate batches that could be clearly identifiable and traceable at all times.
- Batch control was matched to allow for the variability and homogeneity of the collection routes. This meant that routes needed to be similar when forming a batch to allow meaningful information. For example, urban collection with low route use was matched to a similar collection area with 50 vehicle movements or less per day.
- The proportion of leaf litter had to be a minimum of 30% by weight. Compost operators were required to analyse the untreated leaf litter feedstock, the untreated garden waste feedstock and the resultant compost.
- Each site/operator was required to state whether or not the leaf litter was blended and to ensure this information was clear on the submitted results. Where possible PTEs and organic contaminants in the blend should have been given as specified in PAS 100 by weight not volume. Where the operator could not access a weighbridge, the operator was required to demonstrate how the waste was blended.
- Local authorities were encouraged to also test the green waste feedstock before blending with leaf litter to understand how compost derived from leaf litter compares with compost from other green waste feedstock.
- Operators were required to comply with any other relevant conditions of their permit.
- The compost was required to be batch controlled and tested to assess contamination.
- There was a minimum composting time of eight weeks to assess how any contaminants were treated over a specified time period. This aimed to decrease variability of the results.

- All data was submitted on completion of the trial.

Under the CQP certification scheme derogation, the CQP operator was expected to either not recirculate any liquor from the leaf litter batches into the CQP batches, or if they wanted to recirculate the liquor into any of the CQP batches, to test the liquor before and after the trial to see whether the quality of the liquor was affected by the leaf litter and whether the liquor could be safely applied.

B.5 Physical and chemical analysis

The sampling methodology followed the ORG guidelines of sampling compost as used in the PAS 100 sampling requirements (ORG, 2012b).

Samples were analysed for PAHs, PTEs, pH, heavy metals, nitrogen, phosphorus, potassium and organic matter.

Appendix C: Waste Applied to Land: Tool for Environmental Risk

The framework described in this appendix forms part of an assessment and monitoring process. This develops health protective estimates of exposure as an early screen for the risk posed by a wide range of organic and inorganic chemicals that may be present in trace levels in waste materials. The framework has been codified in a Microsoft® Excel based model called 'Waste Applied to Land: Tool for Environmental Risk' (WALTER).

C.1 Mixing model

WALTER uses a simple mixing model that assumes that waste is incorporated to a maximum depth of 25 cm and that the residual volume of added waste to soil is no greater than 10% per hectare over 30 years (about 0.3% per hectare per year). Residual volume is defined as the proportion of added waste that consists of non-degradable solids.

The chemical concentration in soil (mg/kg DW or ng/kg DW for dioxins) is equal to the existing level in soil at the start of a given year plus that added from waste during the year minus that lost by volatilisation to air, leaching to groundwater, by soil biodegradation and crop off-take. Chemical losses are estimated using pseudo first-order rates that take into account chemical and soil properties. Chemical losses are estimated from the time that waste is first applied and subsequent applications up to a maximum period of 30 years are treated independently via a time offset. The annual average chemical concentration in soil in any given year is simply the sum of the chemical concentration at the beginning and end of the year divided by two.

In WALTER, the user specifies:

- waste application rate in tonnes per hectare per year
- incorporation depth – either surface dressing to a depth of 5 cm or ploughed in to a depth of 25 cm
- application frequency from a one-time application up to once a year for 30 years

The user can also select from a predefined list of waste types to specify its dry bulk density and non-degradable fraction. The user can also choose default soil properties from a pre-defined list of soil types from the Contaminated Land Exposure Assessment (CLEA) software (Environment Agency 2009).

C.2 Chemical losses

WALTER assumes pseudo first-order losses of chemicals from waste amended soils from four processes:

- volatilisation
- leaching
- biodegradation

- crop off-take (not included for dioxins and furans)

Volatilisation and leaching are based on soil and chemical properties using algorithms presented in *Technical Guidance Document on Risk Assessment* (ECB 2003).

Default biodegradation rates are based on a matrix of biodegradability classification and the soil-water partition coefficient (ECB 2003). Chemicals are classified as being readily biodegradable (lowest default half-life is 90 days), inherently biodegradable (lowest default half-life is 300 days), or not appreciably biodegradable within the timescales of the risk assessment.

Chemicals are classified according to authoritative information on their biodegradation half-lives in aerobic soils (Mackay et al. 2006). Chemicals considered readily biodegradable have consistently demonstrated half-lives of less than 14 days and those that are inherently biodegradable consistently demonstrate half-lives of less than 90 days. Chemicals are not considered to appreciably degrade if their half-lives are greater than 90 days.

Off-take is estimated by adapting an approach proposed by the European Commission to consider risks from cadmium in fertilisers (ERM 2000, National Chemicals Inspectorate 2000). It relies on estimated plant uptake (see below) and average crop yields. An average crop yield for five produce groups (cereals, fruits, green vegetables, root vegetables and grass) was estimated from UK agricultural statistics (Defra 2010, 2011b, 2011c).

C.3 Transport via the food chain

Chemical transfer and accumulation in crops, meat and dairy produce are calculated for each year by the use of generic chemical concentration factors between soil and plants, and between soil/feed and the edible parts of livestock.

WALTER considers the following crops:

- cereals
- fruits
- green vegetables
- root vegetables
- forage (for estimating animal feed concentrations)

Generic chemical concentration factors for metals and other inorganic elements into plants are taken primarily from the literature on assessment of risks from radionuclide transfer in the environment and from the assessment of risks from land contamination. Important references include:

- *Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments* (IAEA 2010)
- *Review and Analysis of Parameters for Assessing Transport of Environmentally Radionuclides through Agriculture* (Baes et al. 1984)
- *Human Health Risk Assessment Protocol (HHRAP) Companion Database* (USEPA 2005)
- substance-specific reviews undertaken for the development of Soil Guideline Values

Generic chemical concentration factors for organic chemicals are estimated using screening models according to plant type which are based on chemical properties including octanol–water partition coefficient, air–water partition coefficient and chemical diffusivity (Trapp and Matthies 1995, Trapp and Schwartz 2000, Trapp 2002, Trapp 2007, Legrind and Trapp 2009).

Whether the waste is applied as a surface dressing or ploughed in, WALTER uses the average soil concentration to a depth of 25 cm to estimate plant uptake of contaminants. This simplification assumes that:

- plants passively uptake contaminants over the whole rhizosphere
- the screening risk assessment is unlikely to be sensitive to a chemical gradient resulting from surface dressing

WALTER considers the following meat and dairy types:

- chicken
- beef
- pork
- edible offal (liver and kidney)
- milk
- eggs

Chemical concentrations in meat and dairy produce are estimated from generic concentration or transfer factors derived from an extensive review of the scientific literature on the relationship between animal feed and edible produce (Hendriks et al. 2007, Leeman et al. 2007, RTI 2005, TNO 2011, OEHHA 2012). The chemical concentration in animal feed is a weighted average of chemical levels in forage, green vegetables, root vegetables and cereals according to typical diets for different livestock (ECB 1996). Inadvertent soil ingestion is also added for different animals based on literature values for cattle, pigs and chickens (Fries et al., 1982, Thornton and Abrahams 1983, van der Meulen et al. 2008).

Livestock are much more sensitive to the application of waste as a surface dressing than plants. This is primarily a result of indirect soil ingestion during grazing activities. When a waste is applied as a surface dressing, WALTER uses the contaminant concentration in the top 5 cm of soil to estimate the chemical intake via indirect soil ingestion as well as the contributions from other sources including forage and feed.

C.4 Dietary exposure assessment

Generalised food intake rates are estimated for adults (16–64 years) and infants (6–12 months) in WALTER using data derived from UK dietary surveys (Smith and Jones 2003). These generalised food intakes for UK consumers were recommended by the National Radiological Protection Board (now part of Public Health England) for use in radiological risk assessments.

Smith and Jones (2003) based their generalised intakes for adults on an older survey carried out in 1990. This was superseded by the National Dietary and Nutrition Survey carried out in 2000 and 2001 (ONS 2002), which was used by the Pesticide Safety Directorate to assess risks due to pesticide residues in food (PSD 2006). However, the current survey information is not in a format that can be easily integrated with estimates of environmental contamination. Smith and Jones (2003) noted in their preliminary

analysis that differences between the adult survey in 2001 and the results from earlier surveys were considered unlikely to have a significant impact on the generalised datasets.

The food intakes used in WALTER are the mean rates for consumers. These rates are higher than for the general population, which includes consumers and non-consumers (Smith and Jones 2003). However, they are lower than the 95th or 97.5th percentile data that are sometimes used to represent the upper level of 'normal dietary behaviour' (PSD 2006). This critical group data are not used in WALTER because other factors including the choice of soil-to-produce concentration factors and the assumption of a broad spectrum of potentially contaminated foods ensure that the overall exposure assessment represents a reasonable worst case.

WALTER includes the following food groups in the assessment of dietary exposure:

- cereals – wheat, oatmeal, buckwheat, semolina and so on
- green and other domestic vegetables – beans, broccoli, cabbage, cauliflower, courgettes, lettuce, marrow, peas, peppers, tomatoes and so on
- potatoes and root vegetables – carrots, leeks, onions, radishes and so on
- fruit – apples, cherries, pears, plums and so on
- beef, pork – including bacon and ham
- edible offal – liver, kidney and so on
- poultry – including chicken and turkey
- milk
- eggs

C.5 Risk evaluation criteria

WALTER is a generic risk assessment framework for screening the risks to health from application of waste materials to agricultural land. It considers the risks to health from consumers of food produced by waste-amended land for a period of up to 30 years. It estimates potential health risks by:

- identifying where chronic dietary exposures to a chemical may exceed acceptable daily intakes either in the short-term (the first five years after initial amendment) and in the longer term – these scenarios protect against front-loaded risks from initial waste deposition and also from the potential for longer term soil accumulation
- identifying where chemical levels in crops and livestock exceed feed and/or food safety limits in any given year
- comparing estimated dietary intakes for adults and children with levels of acceptable or tolerable intakes derived from relevant toxicological studies

In the case of genotoxic carcinogens, assessment of acceptable or tolerable intakes is against benchmarks indicative of minimal risk. These benchmarks are taken from a range of authoritative UK and international sources.

In addition to the assessment of dietary intakes, WALTER also checks the predicted concentrations of chemicals in selected produce against Maximum Levels established

for animal feeds and for food stuffs for human consumption by The Arsenic in Food Regulations 1959 (as amended), Directive 2002/32/EC on undesirable substances in animal feed (as amended), Regulation (EC) No. 1881/2006 (as amended) and Verstraete (2013).

This is a generic risk assessment and therefore it is possible for reasonable worst-case assumptions to suggest that existing ambient soil levels may pose a risk to health which may or may not actually be the case in some circumstances. To consider the contribution to soil levels from addition of composted trial outputs, the increase in contaminant soil concentrations after 10 applications at the annual applied rate are also reported from the modelling. As a rule-of-thumb, insignificant increases in contaminant levels are assumed where the median ambient soil concentration has increased by <10% after 10 applications (that is, the increase is <1% per application). Where increases are <10%, it is assumed that the contribution of composted trial outputs to the overall risk posed by ambient soil contamination is low.

C.6 Background levels in rural soil

Many contaminants identified as being of concern in the composted trial outputs are also ubiquitous in the environment and found in rural soils. It is necessary to take this into account in the risk assessment.

The data used are derived primarily from the UK Soil and Herbage Survey, a comprehensive survey of the levels of major contaminants in soil and herbage from across the UK (Environment Agency 2007a, 2007b). The median concentration is the preferred measure of central tendency in these data because it is typically log normally distributed. While a high-end concentration might provide a more conservative estimate of exposure, it will underestimate the contribution from added waste and vice versa.

Appendix D: Peer review

The scope of this peer review is to:

- provide a brief commentary on the study aims, methods and results to ensure they are clearly explained
- review the quality and quantity of data used in the risk assessment
- focus on the methodology followed
- assess the robustness of inferences and conclusions
- comment on the recommendations

The ideal assessment of potential environmental and human health risks from the application of an organic material to land would be one based on a full and statistically robust physico-chemical characterisation dataset of the organic material. This is rarely, if ever, available. Therefore, risk assessors need to work within a framework that accepts uncertainties in a structured, realistic, but generally precautionary way, so that evidence-based management decisions can still be made. This classic risk assessment paradigm has been followed in the investigation presented in this report.

The study aims are clear and are well written in an accessible, uncomplicated style. The footnotes explaining technical terms are a useful addition for the technical 'non-expert'.

The approach taken to the collection of samples of leaf litter is a reasonable one and the investigators have been hampered by the (un)willingness of the local authorities to provide data. The limitations of the trial are well set out (in section 3.3) as are the implications of the reduced dataset. The statistical treatment of the data is reasonable in light of the inherent spatial bias and relatively small number of samples.

It is unfortunate, but wholly realistic, that all of the potentially hazardous chemicals identified in Section 2, could not be determined in the samples. Conclusions drawn from this investigation have been contextualised in light of this, but this remains a considerable uncertainty.

Section 4 gives the trial results and comparison with other material. Again, this is well set out and the results from the laboratory analysis of the leaf litter street sweepings are shown to be relatively variable. This is especially true for the PAHs, arsenic and cadmium when compared to green waste and urban herbage. This chemical variability of the leaf litter sweepings is critical to understand in order to be able to draw robust conclusions from generic assessments. The relatively high concentrations and variable nature of some of the chemical determinands in organic materials being risk assessed often result in the need to take relatively precautionary assumptions in order to be confident that all scenarios are accounted for with a very low likelihood of type II errors (false negatives). The limited data available for consideration of treatment options also limits the conclusions that can be drawn from this investigation. The report authors have contextualised the results in an appropriate and balanced way.

An appropriate list of pollutants was selected for detailed consideration and the assessment of quality and risk presented in Section 5 follows a reasonable and logical approach in light of the limitations of the dataset. It is difficult to envisage what other approaches might have been taken in light of the data limitations. Appropriate caveats, for example in the cautious interpretation of PAH data in the fresh leaf sweepings and the composted leaf samples, are given. The tentative identification of an increase in the median concentration of the relatively recalcitrant (and hazardous) PAHs (specifically

BaP) after composting highlights a situation also identified in the composting and digesting of other organic materials (MBT CLO)

It is clear that the PAHs are the issue of greatest concern with these materials (>90% of samples exceed the selected assessment criteria in composted material). The value of 6 mg/kg from IPTS has been used to characterise the potential risks for total PAHs. The source is referenced in the text but more context would be helpful here as to the genesis of this value and its standing in relation to other values used latterly in the assessment of terrestrial risks from PAHs.

In relation to the section on hazard identification in section 5.2.1 and Table 5.3 silver is more likely an eco-risk than a risk to human health, perhaps too cadmium, for which secondary poisoning of wildlife and human food chain transfer (especially at these relatively high zinc levels) might be the main hazard.

The risk evaluation section provides a pragmatic, and sadly unavoidable, mix and match approach in the identification of benchmark criteria values. This is a situation very familiar to those assessing risks of chemicals in materials going to land in the UK. The use of a 10% increase in a chemical concentration above median background with 10 years of loadings is a useful line of evidence to consider in order to contextualise very conservatively derived limits. Furthermore, the actual risk characterisation results of the exposure assessment are not shown, it would be helpful if they were.

In section 5.2.2, it is not clear as to whether any other loadings exceeded the 10% in 10 years of the original soil level. The risk assessment section would undoubtedly benefit from presenting results of the exposure assessment, i.e. in terms of the concentrations of PTE in soils (and possibly selected foodstuffs) following 10 years application of leaf sweeping derived compost to agricultural land (and subsequent food chain transfer). These need to be clearly presented in the context of the soil quality and human health criteria presented in considerable detail in the report – currently we only see summary results for BaP in Figures 5.4a–c).

The risk assessment model for food chain transfer developed by the Environment Agency (WALTER, detailed in Appendix C) is judged to be appropriate as it addresses plant uptake, biotransfer and relevant dietary exposure pathways using parameter values from authoritative sources. The conclusions drawn from this assessment are well supported by the data and the methodology undertaken to deliver this assessment. Limitations in the dataset upon which the risk assessment is based are well acknowledged and many of these originate from the relatively small number of samples and bias to a very limited number of Local Authorities participating in the study.

The specific challenges identified in this investigation in relation to variability of outputs and bias from too few samples, from too few locations, are common in end of waste work. In order to resolve uncertainties and more clearly characterise the risks identified would require considerably more data. The current dataset would seem unlikely to support any other conclusions than those drawn here. The overall conclusion stating that the trial does not support any change to the Environment Agency's guidance is supported by the data and assessment given in this investigation.

The recommendations for further work are sound. There are a range of market factors (transport, perceived risks, consumer preferences) that influence recommendation 1 and these should be borne in mind. For recommendation 2, perhaps some additional statistical work based on the variability in the data presented here (from the one local authority that provided most the samples) might be used to determine the sampling intensity required to deliver the greater certainty on a reduced scale? In previous exercises looking at meeting a quality protocol, the assessment of the number of samples required to deliver an understanding of inter- and intra-sample variability has often been a sobering and ultimately prohibitive venture.

The inclusion of additional contaminants in the determinand suite is a very sensible recommendation. Perhaps it would be useful to contact Industry leads representing those chemicals in Europe who may be in a position to provide technical assistance and collaborative support (e.g. The European Precious Metals Federation, on going studies at the University of Wisconsin and by the USEPA). This can be important, especially in relation to the analytical experience needed to determine some non-routine chemicals in unusual matrices. Recommendation 3 may perhaps be already answered, or at least be readily achieved through identification of those operators already permitted to treat street leaf sweepings.

Graham Merrington and Ed Stutt

15 October 2013

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