

DISPOSAL OF WOOL SCOURING SLUDGE BY COMPOSTING

John Pearson, Fen Lu and Kimti Gandhi

Department of Textiles, University of Huddersfield, Queensgate, Huddersfield HD1 3DH, UK
Tel: 00 44 1484 472114
Fax: 00 44 1484 472826
E-mail: j.s.pearson@hud.ac.uk

Abstract

Disposal of wool scouring sludge is becoming more problematical, due to the need to satisfy environmental legislation concerning what is regarded as hazardous waste, and to the increasing cost of landfill and the scarcity of landfill sites. Composting the sludge to produce a safe, saleable product could be a viable alternative.

Key words:

wool scouring sludge, composting, environmental legislation

Introduction

Raw wool is the natural coat of the sheep when removed by shearing, and it contains a large quantity of impurities, around 35% by mass [1]. Typical mean values by mass of different impurities are: grease (15%); sand and dirt (10%); suint (8%); and vegetable matter (2%). An essential requirement for successful wool processing is the complete removal of these impurities during the scouring process.

The scouring effluent thus contains these impurities, which have high levels of COD (chemical oxygen demand), BOD (biochemical oxygen demand), suspended solids, organic matter and sheep dip chemicals. These levels exceed the limits for COD, BOD and suspended solids for aqueous discharge into surface water [2]. The discharge of wool scouring effluent to public sewers and sewage works can thus give rise to serious problems.

After treatment, the scouring effluent is separated into three categories: grease, liquor and sludge. The grease may go for sale, the liquor is discharged to the sewage works, and the sludge needs to be disposed of; in the United Kingdom, this has typically been done in landfills.

Allanach & Madden [3] have reported that the cost of disposal of spadeable wool scouring sludge in the United Kingdom is approximately £30 (€49) per tonne, including tax, whilst disposal of liquid sludges costs approximately £60 (€98) per tonne. It is expected that these costs will rise as landfill becomes scarcer and the landfill tax rises. A major reason for the high cost of disposal is that scouring sludges are prescribed as offensive wastes, since they contain high levels of organic material and potentially toxic sheep dip pesticides, such as organochlorines, organophosphates and synthetic pyrethroids. These can only be disposed of at prescribed waste disposal sites.

In addition, the European Community Landfill Directive [4] seeks to minimise the quantity of waste going to landfill and to eliminate hazardous waste from landfill. It clearly states that the proportion of biodegradable Municipal Solid Waste (MSW) disposed of to landfill must be reduced to 75% by 2002, to 50% by 2005 and to 25% by 2010. The Directive also defines MSW as household waste, together with commercial, industrial, institutional and other waste.

By 2010, therefore, landfill sites will only accept non-recoverable waste and inert waste, so that the disposal of wool scouring sludges to landfill will then be an untenable option. Incineration of such sludges has been investigated as a more environmentally friendly method for the disposal of wool scouring sludges [5]. Although the effectiveness of incineration in a rotary kiln incinerator has been demonstrated by Lu, in terms of the efficiency of the process, there are some problems with regard to the economics of the process, which are yet to be resolved. United Kingdom wool scouring companies alone produce over 100,000 tonnes of greasy wool, resulting in over 30,000 tonnes of sludge waste, while Woolmark Company data show that world production of greasy wool was approximately 2.25

Mtonnes in 2001, yielding over 930,000 tonnes of sludge waste. The problem of disposal of wool scouring sludge is therefore world-wide, and an economic and commercially viable solution to it will be of immense benefit to the wool scouring industry. This paper investigates the feasibility of disposing of wool scouring waste by composting.

The composting process

Composting as a waste management system

Composting has been defined as the production of a fertilising mixture, which has been subjected to controlled biological decomposition under aerobic and thermophilic conditions [6], or as a method of solid waste management whereby the organic component of the solid waste stream is biologically decomposed under controlled conditions into a state in which it can be handled, stored and/or applied to the land without adversely affecting the environment [7]. According to Walker [8], composting is an excellent tool for stabilising and facilitating the subsequent utilisation of organic waste materials, farm waste and certain industrial wastes.

Composting has been used as a waste disposal technique in the United States for over 50 years [9], and many composting sites have been established since then, both in Europe and the United States, to treat wastes from such diverse sources as agricultural (particularly farm slurries), sewage sludge, household refuse, single substrate wastes such as brewery waste and even animal carcasses (see for instance [10], [11]). Indeed, wool waste has been composted in the United States in the form of fibrous wool wastes [12] and fellmongering effluent [13], while the CSIRO (Australia) Scour Waste Integrated Management System (SWIMS) has been reported to produce a compostable sludge from scouring effluent [14]). Although several municipal composting plants were established in the United Kingdom in the 1960s, all of these have been closed for technical and economic reasons, and composting has thus not been widely used as a waste management technique in the United Kingdom [9].

The composting process

Composting is a biological process in which organic material is broken down by the action of micro-organisms. The degradation process takes place in the presence of air ('aerobic'), and results in elevated process temperatures and the production of carbon dioxide, water and a stabilised organic residue.

The key feature of the composting process is the generation of heat by biological activity during the decomposition of the substrate material. By forming the wastes into large masses under appropriate conditions, they will reach high temperatures, resulting in rapid degradation. More importantly, these temperatures have a sanitising effect upon the waste, reducing the numbers of pathogenic organisms [15]

There are three common methods of composting in use in the United Kingdom:

- ✓ *Windrowing* is where the waste is placed in elongated lines, about 2m high, and is mechanically turned to aerate the composting waste. In this method, it is necessary to have sufficient process control to guard against excessive odour generation and the release of aerosol and particulate emissions, which can lead to potential health hazards during the mechanical turning process.
- ✓ In the *aeration static pile* method, aeration, by means of blowing or sucking air through the material, is used to control oxygen levels and the temperature in the pile. The configuration of the pile is similar to that of the windrow, but the pile is built on top of a grid of perforated pipes, which serve as air ducts connecting the air manipulation system to the material.
- ✓ *In-vessel composting* uses an enclosed reactor system, which allows close control over temperature, moisture content and rate of aeration. Although a high capital cost is involved, this process is becoming increasingly popular.

The compost obtained by following any of these three processes may be in a stable condition, but it is not necessarily suitable for immediate use. The final stage of composting involves processes to remove non-composted material and other contaminants, such as glass, plastics and metals. The compost is then size-reduced and sieved, ready for use.

Materials and methods

Composting wool scouring sludge waste

In general, wool scouring sludge is an organic waste, and it contains significant colonies of micro-organisms which are able to survive the elevated temperatures used in the wool scouring process.

The presence of pesticides in the wool sludge is one of the main problems with regard to its safe disposal. As mentioned above, landfill is not a desirable option; converting the sludge into a reusable and (preferably) marketable product may be a much better waste management approach.

The aeration static pile process was the preferred composting system used in this work, on the advice of the Department of Civil Engineering at the University of Leeds, United Kingdom, who have themselves conducted trials for composting commercial waste materials. Aeration to control the temperature and oxygen levels within the composting material was controlled by blowing air through the material pile. Because the wool sludge consists of small particles and has a high moisture and grease content, it appears as a sticky, wet material; it was realised that it would be unlikely to compost without additional compostable materials. It was considered appropriate to use green waste, which would provide a carbon source and open structure for the composting material and would be a readily available material.

In order to carry out the trial, sludge from a commercial chemical flocculation treatment plant of a wool scouring company in West Yorkshire, United Kingdom, was used. The green waste, consisting of leaves, tree branches and grass mowings, was obtained from the Risley landfill site, United Kingdom, where the composting trial took place.

4.5 tonnes of wool sludge (approximately 4.34 m³) was thoroughly mixed with 3.4 tonnes of green waste (approximately 8.66 m³), giving ratios of sludge to green waste of 1:0.76 by mass and 1:2 by volume.

The aeration static pile used in this trial had a perforated pipe laid down on its base to act as an air distributor. One end of the pipe was connected, via a non-perforated hose of the same diameter, to an electric aeration fan (Secomak model 142; 372 W centrifugal fan), and the other end of the pipe was blocked off. The pipe was covered with a layer of straw to prevent blockage from overlaying composting material. The mixed waste was then laid down over the hose in a pile, and the surface of the pile was covered with a shallow layer of shredded green waste to insulate the mixed waste.

A temperature probe was inserted into the middle of the pile in order to read the temperature of the composting material. The probe was connected to a process controller unit, which operated at fixed intervals of 30 minutes to switch the aerator fan on and off. It was set to sense a waste temperature of 37°C; if the waste temperature went below this value, the fan would be switched off, and if the waste temperature exceeded this value, the fan would remain on until the temperature was brought down to the set-point value, or until the end of the 30-minute interval.

Assessment of Compost Stabilisation

Since composting is characterised by a high degree of microbiological activity, which ceases when all available nutrients have been consumed, a compost with a low biological activity is described as being stable. It is generally believed that compost should be well stabilised prior to application involving plant growth, since an unstable compost may contain substances harmful to plants. Assessment of stabilisation is not easy, since there is currently a lack of understanding of the processes involved. However, a number of methods have been used to this effect [16]:

- I. carbon-based analysis
- II. humification indicators
- III. molecular size determination
- IV. enzyme assays
- V. respiration measurements
- VI. phytotoxicity assays

According to the nature of wool-scouring sludge, and in relation to commercial suitability and appropriateness, the following two methods were chosen to assess compost stabilisation:

Carbon-based analysis

This method involves measuring the organic carbon and nitrogen levels in the compost and the carbon to nitrogen ratio (C:N). The C:N ratio of the solid phase is an index traditionally used to evaluate the maturity of a compost. Even though the C:N ratio in the solid phase cannot be used as an absolute indicator of compost maturation, a value of 20 or less can be considered satisfactory [17].

Respiration measurements

As composting proceeds, there is a fall in respiratory activity, because less substrate is available for the micro-organisms to digest. The respiration rate in a mature compost should therefore be significantly lower than that in a fresh material. Very low respiration rates signify a state of endogenous respiration [16].

Sludge samples

Samples were collected from each of three different positions in the composting pile, for testing. These were blended to make a single, composite sample, which was then sieved through a 10-mm mesh to remove coarse materials. The blended samples were analysed at weekly intervals for moisture content, volatile solids and pH value. The total nitrogen, nitrate nitrogen, ammonia nitrogen, total carbon, organic carbon and organic matter were analysed at the start of composting, and thereafter every four weeks. The pesticide concentration levels were also analysed, at the start of the composting process, and then after 42, 83 and 110 days of composting respectively. Once the compost had matured, the final compost quality was assessed.

The Composting Association [18] has proposed limit levels for specific contaminants in compost (Table 1):

Table 1. Limit levels for contaminants in compost [18]

contaminant		upper limit permitted
human pathogens	Salmonella SPP	absent in 25 g
	Escherichia coli	absent
	faecal coliforms	1000 CFU/g
potentially toxic elements (mg/kg dry matter)	cadmium	1.5
	chromium	100
	copper	200
	lead	150
	mercury	1
	nickel	50
	zinc	400
physical contaminants (% m/m of total air-dried sample)	total glass, metal, plastic >2 mm	1.5
	stones, minerals	5
weed contaminants	weed propagules	5 viable propagules/l
phytotoxins	plant tolerance	20% below control

Pesticide concentration

The levels of pesticides in the wool scouring sludge and in the composting samples were analysed at EnCo, Ilkley, West Yorkshire, United Kingdom. The samples were thoroughly mixed with hydrocarbon solvent, and were then allowed to stand until the solvent separated and floated to the top. The supernatant solvent was removed, placed in a clean, dry, tared flask and evaporated to dryness. The residue was re-dissolved in fresh solvent and the solution was passed through a High Performance Liquid Chromatography (HPLC) system, using the Perkin Elmer normal phase system, so that the fraction of residue containing the organochlorine, organophosphorus and synthetic pyrethroid

pesticides was separated from the rest of the residue and was collected separately. The pesticide residues were then analysed in a gas chromatography capillary column, using the Perkin Elmer Autosystem XL.

Electron capture detectors were used to identify the organochlorine and synthetic pyrethroid pesticides, whilst nitrogen-phosphorus detectors were used to detect organophosphorus pesticides. When a pesticide had been detected, confirmatory analysis was carried out in a second gas chromatograph, fitted with similar detectors, but with a different chromatography column. The results were down-loaded to a Turbochrom Automated Integrator System for calculation of pesticide concentrations.

Volatile solids

The proximate analysis tests designed for the testing of coal and coke were used, based on BS 1016 – 104.1 [19], 104.3 [20], 104.4 [21] and 106.7 [22] for the determination of moisture content, volatile matter, ash content and fixed carbon content respectively.

Electrical conductivity

Electrical conductivity was measured by mixing a 30 g sample of sludge with 300 ml distilled water in a 500 ml conical flask. The contents were thoroughly agitated for 15 minutes and then filtered. The conductivity of the filtrate was determined by using a conductivity meter and calibrated probe.

Respiration

Respiration of micro-organisms in the compost was assessed by using the Specific Oxygen Uptake Rate (SOUR) test developed by Lasaridi & Stentiford [23]. The analysis was carried out for dissolved oxygen concentration (mg/l) against real time (minutes). The SOUR was calculated according to this equation:

$$\text{SOUR} = (60 \times [S]_{\text{max}} \times V_s) / (m \times DS \times VS)$$

The maximum value of the slope $[S]_{\text{max}}$ (mg O₂/l/min) obtained from this calculation was used to evaluate the results, where the SOUR is expressed as mg O₂/gVS/h, and

60	=	conversion constant
V _s	=	volume of slurry in litres
M	=	total mass of compost sample in g
DS	=	fraction of dry solids in the total mass (0.0 – 1.0)
VS	=	fraction of volatile solids in the dry fraction (0.0 – 1.0)

Total nitrogen

The Kjeldahl method number 48 –(nitrogen in plant material, described in MAFF/ADAS [24]) was used for the analysis of total nitrogen.

Ammonium nitrogen, nitrate nitrogen and ammonium nitrogen:nitrate nitrogen ratio

Ammonium nitrogen and nitrate nitrogen were detected in accordance with Method 53, MAFF/ADAS [24]. For ammonium nitrogen determination, magnesium oxide was added to the distillation flask, trapping the ammonia in a boric acid indicator solution (1% m/V). Distillate was titrated against 0.005 M sulphuric acid, using a methyl red/methylene blue indicator. The concentration of ammonium nitrogen is given in mg/kg.

For nitrate nitrogen determination, Devarda's alloy was added to the sample residue (after distillation for ammonium nitrogen), trapping the nitrate in a boric acid indicator solution (1% m/V). The distillate was titrated against 0.005 M sulphuric acid, using a methyl red/methylene blue indicator. The concentration of nitrate nitrogen is given in mg/kg.

The ammonium nitrogen to nitrate nitrogen ratio = $\frac{\text{ammonium nitrogen (mg/kg)}}{\text{nitrate nitrogen (mg/kg)}}$

Organic matter

The organic matter content is the amount of organic matter lost after muffle furnace incineration at 550°C. The method used was the same as that for volatile solids, stated above.

Organic carbon and carbon:nitrogen ratio

Organic carbon and the carbon:nitrogen ratio were determined by following the standard operating procedure (SOP) based on Method 56, MAFF/ADAS [24] according to the following equations:

$$\text{organic carbon} = \frac{1.2 \text{ (mg)} \times (V - FY) \times 10}{\text{mass of sample (g)}}$$

where

- V** = volume of 66.7 mM potassium dichromate (ml)
- Y** = volume of 0.4 M ferrous sulphate titrated (ml)
- F** = standardisation factor of 0.4 M ferrous sulphate

$$\text{C : N ratio} = \frac{\text{organic carbon (\% m/m)}}{\text{total nitrogen (\% m/m)}}$$

Results

Analyses of composted wool scouring sludge

Figure 1 shows the temperature profile of the composting pile. As the air temperature during the composting period was between 15 and 25°C, it is clear from the initial temperature of the composting pile that the composting process started almost immediately, without any need for acclimatising the composting micro-organisms. The temperature rose to 52.7°C in seven days, and then fluctuated between about 42 to 53°C in response to changes in ambient temperature. The temperature fell steadily after about 90 days' composting, indicating that micro-organism action was declining and that the compost was maturing and stabilising.

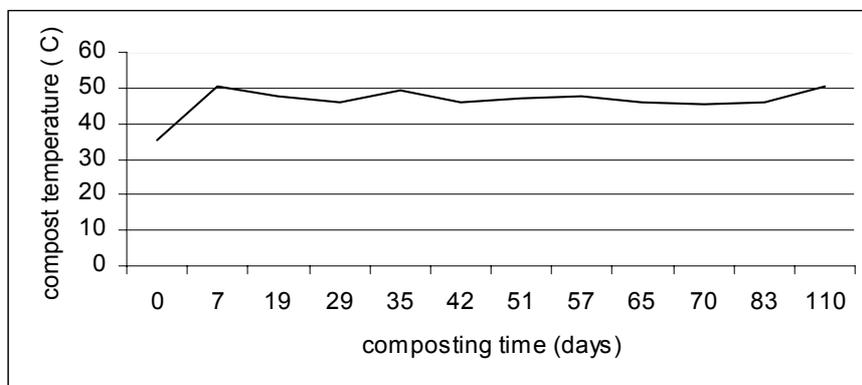


Figure 1. Temperature of the composting pile during the composting period

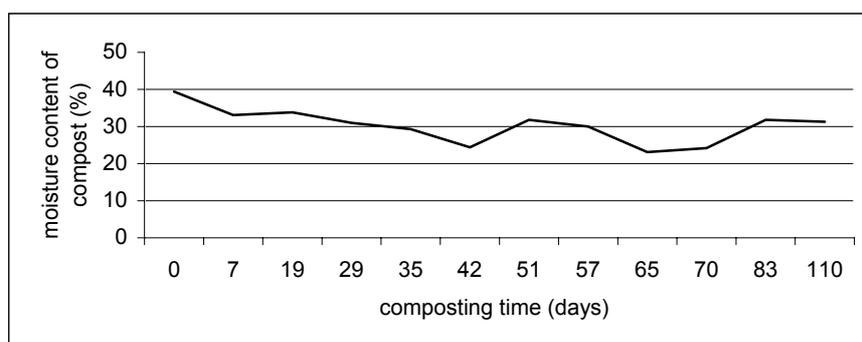


Figure 2. Moisture content of the composting pile during the composting period

Moisture is necessary for the micro-organisms to remain active. Figure 2 shows that the moisture content in the composting pile varied from 23 to 39%, depending on changes in air humidity, but was always at a level sufficient to promote biodegradation.

Figure 3 shows that the pH value of the composting pile varied during the composting period. At the start, the compost had a low pH value, probably due to acid generation in the sludge waste, representing the early stages of anaerobic biodegradation. The pH returned to a value of between 7 and 8 within seven days, and then remained between 6.6 and 8.4 throughout the composting period.

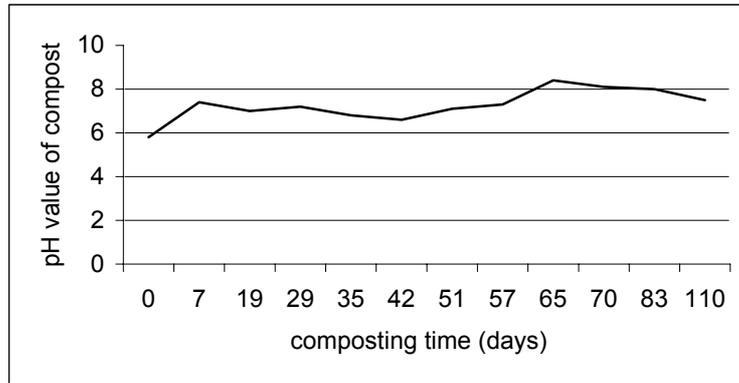


Figure 3. pH value of the composting pile during the composting period

Figure 4 shows that the percentage of volatile solids reduced gradually throughout the composting period from a maximum of 37% to about 26% at the end of the composting period, again indicative of the degradation process.

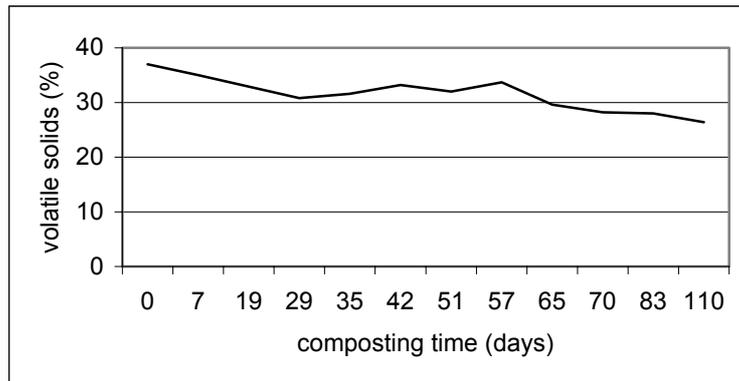


Figure 4. Concentration of volatile solids in the composting pile during the composting period

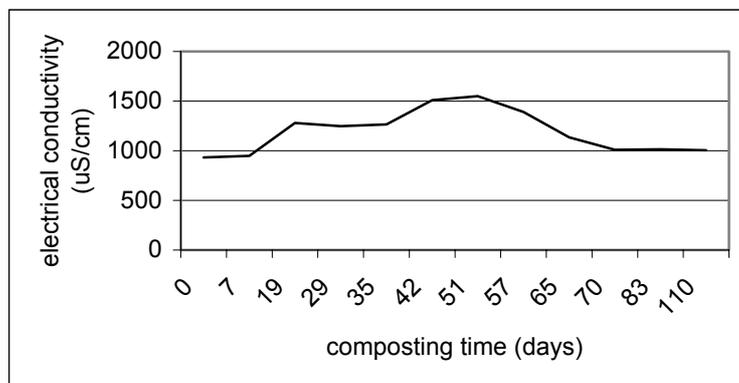


Figure 5. Electrical conductivity of the composting pile during the composting period

The electrical conductivity of the composting pile rose to a maximum after 51 days of composting (Figure 5) and then reduced, levelling off after about 70 days of composting. The initial rise could be due to the release of soluble material as the waste biodegraded; the subsequent fall could be due to the degradation of some of this soluble matter. The levelling-off could indicate that biodegradation was slowing down and that the compost was maturing.

Table 2 shows the results of the SOUR tests carried out on three samples, taken from the composting pile on the 19th, 42nd and 83rd days respectively of the composting period. The table clearly shows that oxygen consumption dropped significantly during this period, demonstrating the steady decline in aerobic degradation as the material in the composting pile was used up.

Table 2. SOUR tests carried out on sludge during the composting period

sample number	composting time (days)	respiration: SOUR (mgO ₂ /gVS/h)
1	0	-
2	7	-
3	19	7.31
4	29	-
5	35	-
6	42	2.93
7	51	-
8	57	-
9	65	-
10	70	-
11	83	1.98
12	110	-

Table 3 shows that the concentration of nitrogen declined slowly during the composting period, while that of carbon declined more rapidly. The overall reduction in the C:N ratio indicated that the compost was maturing and stabilising.

Table 3. Nitrogen and carbon concentrations and C:N ratio in composting samples during the composting period

	Composting time (days)				
	0	29	57	83	110
total nitrogen (%m/m dry matter)	1.32	1.22	1.27	1.27	1.17
nitrate nitrogen (mg/kg)	23	31	7.3	18	22
ammonium nitrogen (mg/kg)	564	2670	2220	1430	1050
total carbon (% m/m dry matter)	23.25	18.46	19.80	17.20	15.55
organic matter (% m/m dry matter)	37.00	30.80	33.70	28.00	26.40
organic carbon (% m/m dry matter)	23.10	18.40	19.50	16.90	15.10
organic C:N ratio	17.50	15.08	15.35	13.31	12.91

Figure 6 shows that the concentration levels of various pesticides in the composting pile during the composting period significantly decreased during the composting period: the total pesticide concentration was reduced by 86.6% from that initially found in the scouring sludge, and by 76.5% from that initially found in the composting pile.

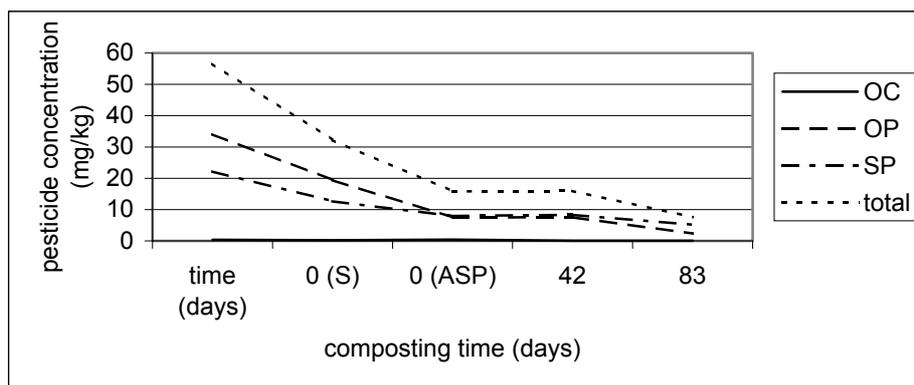


Figure 6. Pesticide concentration levels in sludge and in composting samples during the composting period

- ASP = aerated static pile (scouring sludge + green waste)
- OC = organochlorine pesticides
- OP = organophosphate pesticides
- SP = synthetic pyrethroid pesticides
- total = total pesticides

From the rates of pesticide degradation observed, it appears that if the composting process had been allowed to continue, the amount of pesticides would have further decreased, and the level of pesticides remaining would not have posed any immediate environmental threat. It is probable that these amounts would have continued to decrease even further, as the compost was being used for horticultural and/or agricultural purposes. However, the practical significance of the pesticide concentrations present in the compost after 110 days is unclear, since there are no identifiable guidelines available with regard to maximum levels of pesticide permissible in composts, even in the Compost Association's Quality Standard for Compost (proposal) produced in 2000.

Table 4 shows the levels of potentially toxic elements and contaminants in the mature compost, compared with the proposed limits suggested by the Composting Association [18].

Table 4. Potential toxic element and physical contaminant content of mature compost compared with the Composting Association's suggested limits

parameter		standard upper limit for compost	test result for mature compost in this work
potentially toxic elements (mg/kg)	cadmium	1.5	0.27
	chromium	100	24.1
	copper	100	60.3
	lead	150	102
	mercury	1	0.27
	nickel	50	18.1
	zinc	400	212
physical contaminants (%m/m)	total glass, metal, plastic >2 mm (%m/m)	1	glass: 0.22 metal: nil plastic: nil
	stones and other consolidated mineral contaminants >2 mm (%m/m)	5	11.9

Table 4 shows that, in mature compost, concentrations of all listed potentially toxic elements were within the suggested limits, and that the mature compost was suitable for use from this perspective. Levels of glass, metal and plastic in the mature compost were within suggested limits; stone contamination was higher than the permitted level, but this can be rectified by using a 5-mm sieve on the compost.

Conclusion

Overall, it is clear that composting an intractable, polluting wool scouring sludge waste by the aeration static pile process produced a safe, useful and marketable product.

References

1. Christoe, J. R., and Bateup, B. O., (1996). *Scouring simplified*. *Textile Asia*, **27**, 79.
2. Hewson, M. J. C., (1999). *Textiles and water: are they finishing the environment?* In 'Proceedings of the International Conference on Water and Textiles', University of Huddersfield, 1.
3. Allanach, D., and Madde, M., 1999. *The achievements of industry-funded environmental networks in developing cost effective pollution abatement strategies*, in 'Proceedings of the International Conference on Water and Textiles', University of Huddersfield, 331.
4. European Community, (1999). *Council Directive 1999/31/EC on the landfill of waste*.
5. Lu, X. F., (2001). *Wool scouring sludge incineration*, Ph D thesis, University of Huddersfield.
6. The Composting Association (1998). *Standards for compost*, in 'A feasibility study of the introduction of process and quality standards for compost in the United Kingdom.
7. Zucconi, F., and de Bertoldi, M., (1987). *Composting specifications for the production and characterisation of compost from municipal solid waste*, in 'Compost: production, quality and use', ed de Bertoldi, M., Ferranti, M. P., Hermite, P. L., and Zucconi, F., Elsevier Applied Science, 30.
8. Walker, J. M., (1996). *United States Environmental Protection Agency regulations for compost*.
9. The Composting Association, (1997). *The state of composting in the United Kingdom, a blueprint for action*.
10. Goldstein, N., Riggle, D., and Steuteville, R., (1992). *Sludge composting maintains growth*, *BioCycle*, **33**, 49.
11. Spencer, R., (1992). *Sludge composting takes town out of the landfill*, *BioCycle*, **33**, 52.
12. Das, K., Tollner, E. W., and Annis, P. A., (1997). *Bioconversion of textile industry wastes to value-added products*, in 'Book of papers, International conference and exhibition', AATCC, 426.
13. Plat, J. Y., Sayag, D., and Andre, L., (1984). *High-rate composting of wool industry wastes*, *BioCycle*, **25**, 39.
14. Anon (2000). *Turning scour effluent into valuable compost*, *Wool Record*, **159**, 35.
15. Carroll, B. A., (1993). *Composting sewage sludge: basic principles and opportunities in the UK*, *Journal of the Institute of Water and Environmental Management*, **7**, part 2, 175.
16. Inbar, Y., Chen, Y., Hodar, Y., and Hoitink, H. A. J., (1990). *New approaches to compost maturity*, *BioCycle*, **31**, 64.
17. Hirai, M. F., Chanyasak, V., and Kubota, H., (1983) *A standard measurement for compost maturity*, *BioCycle*, **24**, 54.
18. The Composting Association, (2000). *Standards for compost (proposal)*.
19. British Standard 1016-104.1, (1999). *Methods for analysis and testing of coal and coke. Proximate analysis. Determination of moisture content of the general analysis test sample*.
20. British Standard 1016-104.3, (1998). *Methods for analysis and testing of coal and coke. Proximate analysis. Determination of volatile matter*.
21. British Standard 1016-104.4, (1998). *Methods for analysis and testing of coal and coke. Proximate analysis. Determination of ash content*.
22. British Standard 1016-106.7, (1997). *Methods for analysis and testing of coal and coke. Proximate analysis. Determination of carbonate carbon content*.
23. Lasaridi, K. E., and Stentiford, E. I., (1996). *Respirometric techniques in the context of compost stability assessment*, in 'The science of composting', ed de Bertoldi, M., Sequi, B., Lemmes, B., and Papi, T., Blackie Academic and Professional (Chapman and Hall), 274.
24. MAFF/ADAS, (1986). *The analysis of agricultural materials*, Reference book, 3rd edition, HMSO.

▽△