Analytical perspective on waste management for environmental remediation

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Increasing numbers of publications reveal researchers’ interest in waste management. After a short discussion on disposal strategies for wastes, we explore new ways of reusing wastes. Our main focuses are tannery waste, fly ash, food waste and compost. We also discuss the role of analytical chemistry in using waste for environmental remediation. Promoting clean methodologies in remediation is the best way to address future environmental challenges.

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1. Introduction

Waste is usually the product of industry or commerce, but it also comes from residential use, agriculture, medical facilities, radioactive sources, and light industries (e.g., dry-cleaning establishments). Wastes are mostly industrial by-products, whereas toxic wastes are poisons, even in very small or trace amounts. The growing concern for waste-management worldwide is reflected in the rise of publications (Fig. 1). A glance at waste-generation data [1] reveals that, compared to developed countries, developing countries generate much less waste. Whether developed or developing countries, we need wastetreatment strategies for environmental remediation.

Waste management is a pressing issue. The mass of waste produced throughout the world has been growing considerably for many decades, especially in affluent countries, as depicted in the data connecting national gross domestic product and waste generation per capita [2]. Human activity has become a major force in shaping the environment – a manifestation of the population explosion. Rapid urban growth and industrialization increase generation of solid waste. Many cities and towns in developing countries are in a miserable state of environmental degradation and face serious health risks, due to dumping of domestic refuse on streets and in public areas. The lack of adequate collection of wastes from roadsides and dustbins leads to severe contamination of water resources. Waste cannot be dumped without due concern and preparation, because not only is it unpleasant, unhygienic and potentially disastrous to our environment, but it also requires space and incurs expenditure to deal with the consequences of waste disposal. Our use of commodities challenges the capacity of air, water and land to cope with environmental problems. This situation fundamentally affects many other aspects of our society, such as the economy, energy and climate. We need to fulfill our human needs and prosper while taking into account and dealing with the problems of waste generation. The best way to tackle waste-related problems is to prevent waste generation, and to favor waste minimization, recovery, recycling and reuse.

2. Recovery, recycling, reuse and disposal

There are several waste-treatment strategies in use – all with the same goal in mind of using less material, reducing toxics and recovering more resources. Waste-treatment methodologies and technologies include landfill, open dumping, incineration, composting, recycling
vermicomposting, pyrolysis, gasification, combined pyrolysis and gasification, anaerobic digestion (biomethanation) and pelletization. Among these, landfill, open dumping, incineration, composting and recycling are traditional, whereas the others are comparatively new options.

Solid waste includes food residues, plastic and polyethylene, paper and cardboard, metals, glass, textiles, leather, rubber, synthetic materials and inert materials (stones, bricks, and ashes), which can be again classified into two groups – biodegradable and non-biodegradable. Natural recovery systems make use of food, organic and green waste (biodegradable) and then deal with in-vessel compost systems, while recycling is a logical option for materials not suitable for composting (non-biodegradable). Metals, plastics and glass are the most common of these materials. Automated and manual methods are used to sort out materials from construction sites (e.g., brick, tiles and concrete), which, after being sorted, may be re-used.

According to Mor et al. [3], solid-waste landfill is a common method of management in developing countries. Due to limited land availability, this is not a favored option [4]. In addition to lack of space, emission of leachates and gases to the environment needs to be taken into consideration.

Incineration is one of the attractive options for waste treatment in the South East Asian region [5]. Incineration transforms original waste into other forms (e.g., fly ash), which, in turn, requires landfill for disposal, so incurring cost additional to the operational cost of the incinerator, which also emits greenhouse gases into the atmosphere [6].

There is a noticeable contrast between the type of waste generated in developed and developing countries; developed countries generate more recyclable wastes, whereas developing countries generate more biodegradable wastes [7]. The waste composition of developing countries makes it clear that composting is the best possible option to deal with solid waste. Case studies show that developing countries are opting for composting [8].

Resource recovery is a major element in solid-waste management. Waste recycling can help to eliminate and thus to minimize waste. One of the main goals of sustainable waste management is to maximize recycling and reuse. Reuse of waste is important from many points of view:

- it helps to save and to sustain natural resources that cannot be replenished;
- it decreases pollution of the environment; and,
- it helps to save and to recycle energy in production processes.

According to the “waste-hierarchy” pyramid (Fig. 2), disposal of waste is the least favored option while prevention of waste is the most favored.

Fig. 3 shows the waste-management system for solid waste from waste source to final disposal. Waste at its source is sorted out into organic, recyclable and residual wastes. After bio-treatment of the organic fraction and recycling of the recyclables, the total residual waste is either incinerated or used for landfill.
3. Recent applications of wastes

In Table 1, we can see how wastes are utilized [9–41]. Waste fibers from the tanning industry have high sorption capacity with removal efficiency of pollutants from water [11]. Leather industries produce chromium-rich waste, which is employed in the ceramic-tile and building industries [12,13].

In spite of fly ash being a major environmental liability, it has been utilized in various fields. In conjunction with organic manure and microbial inoculants, fly ash can enhance plant-biomass production from degraded soils and can be used in agriculture [14–18]. Fly ash has also been used in the construction industry [19]. Fly-ash-waste materials were used with quicklime to immobilize lead, trivalent and hexavalent chromium present in artificially contaminated clayey sand soils. Fly ash has been converted into zeolites for ion-exchange applications [23,24] (e.g., recovery of metals [25,26]) and synthesis of novel materials (e.g., geopolymers [27]). Fly ash blended with lime is used for filling of abandoned coal mines [28].

Food waste and compost are used for production of biofuel [30], biofertilizer production and mitigation of green house gases [31,32], and as a source of clean energy [37].
4. Use of wastes for environmental remediation

Environmental remediation deals with the removal of pollution or contaminants from environmental media (e.g., soil, sediment, groundwater, or surface water) for the general protection of human health and the environment. The more traditional approach (used almost exclusively on contaminated sites in the 1970s to the 1990s) comprises primarily soil excavation and disposal to landfill and groundwater, but other technologies are also in use (e.g., solidification, stabilization, bioremediation and phytoremediation). The selection of the most appropriate method of remediation depends on the characteristics of the site, the concentrations and the types of pollutants to be removed, and the end use of the contaminated medium. The approaches include isolation and containment, mechanical and pyrometallurgical separation, chemical treatment, electrokinetics, biochemical processes, immobilization, toxicity reduction, physical separation and extraction. Where metal ions are involved, due to their non-degradable characteristics, it is impossible to decontaminate the site. However, it is possible to passivate wastes containing heavy metals and to reduce the volume of waste appreciably.

5. Specific cases of remediation through use of wastes

5.1. Tannery waste

Among the numerous wastes, chromium-containing solid wastes from the tannery are of major concern due to their high toxicity, so there is interest in the recovery of chromium. Studies carried out by Zhou et al. [9] showed the feasibility of applying the bioleaching process to tannery sludge with high chromium content. Inoculation of a mixture of iron- and sulfur-oxidizing bacteria and co-addition of ferrous salt and elemental sulfur accelerated acid production, and increased the oxidation-reduction potential originating from the bio-oxidation of ferrous salt and elemental sulfur. The concentration of dissolved chromium increased as the pH decreased in the sludge and reached its maximum removal of 95.6%. However, 20.4% of nitrogen, 24.5% of phosphorus and 14.3% organic matter were lost in the bioleaching process. The residual chromium content in the leached tannery sludge was acceptable for use in agriculture.

The high concentration of trivalent chromium along with organic/inorganic compounds in tannery sludge causes severe groundwater contamination in land disposal and chronic air pollution on incineration. The dried sludge was incinerated at 800°C under starved-air combustion to prevent the conversion of trivalent chromium to hexavalent chromium and subjected to a flow-through column test to evaluate the concentration of leachable organics and heavy metal ions present in it. The calcined sludge was then solidified and stabilized using fly ash and Portland cement and gypsum. The solidified bricks were tested for unconfined compressive strength and heavy-metal leaching. The stabilization of chromium (III) in the cement gel matrix was confirmed with scanning electron microscopy and X-ray energy dispersive spectroscopy [10].

Waste fibers from tanning industry have high sorption capacity. The tanned solid wastes are capable of
absorbing many times their weight in oil or hydrocarbons (6.5–7.6 g of oil and 6.3 g of hydrocarbons per gram of chrome shavings). Complete removal of pollutants from water is achieved [11].

Making lightweight ceramsite with sewage sludge is an effective new approach to disposal of sludge. However, there is a concern as to whether heavy metals (e.g., hexavalent chromium) in sewage sludge can be solidified in ceramsite after sintering. Leaching test results indicate that hexavalent chromium is stabilized in ceramsite and cannot be easily released to the environment again as secondary pollution, so eliminating concern for its application [12].

A case study of tannery sludge showed that it can be used in manufacture of bricks for up to 10% of their weight. Bricks containing sludge were higher in porosity than controls. Bending strength and frost resistance were acceptable in bricks made with 10% sludge, but decreased below acceptable levels in bricks made with 15% sludge. Increased emission of aromatic and chlorinated hydrocarbon vapors from the sludge in the pre-heating zone of the furnace could easily be taken care of by recycling this exhaust to the high-temperature zone [13].

5.2. Fly ash
Fly ash represents a potential major environmental liability, but, due to its bulk mineralogy and chemistry, it can serve as a source of materials for large-volume, low-tech applications.

After the removal of soluble salts, including chlorides, pretreated fly ash was used to simulate the cement-making process. It was found suitable for reuse in cement production due to its high ash content. The addition of fly ash in cement raw material did not affect the compressive strength of the cement. The compressive strengths of all cement samples were greater than the standard values of the ASTM (American Society for Testing and Materials) for Type II Portland cement [19].

For an annual cement production in the United Kingdom of the order of 1.5 × 10^7 tonnes, replacement of 20% of this quantity using waste materials and industrial by-products, such as fly ash, would direct some 3 × 10^8 tonnes away from landfill. Snelson et al. [20] have studied the development of compressive and tensile strength for concrete with replacement of cement with ash levels up to 40%. It was found that the combined use of tyre chips and fly ash offers a wide variety of low-strength types of concrete for applications such as blinding, footpaths, and car-park and building foundations. This would benefit the environment by reducing the amount of fly ash going to landfill worldwide.

Nohchaiya et al. [19] noted that the workability of Portland cement–fly ash–silica concrete in most cases remained higher than that of the control Portland cement concrete. Furthermore, utilization of silica with fly ash was found to increase the compressive strength of concrete at early ages (pre 28 days) up to 145% with the highest strength obtained when silica fume was used at 10 wt%. Moreover, scanning electron micrographs show that utilization of fly ash with silica fume resulted in a much denser microstructure, thereby increasing compressive strength.

Luna et al. [23] proposed use of this easily synthesized zeolitized fly ash in municipal treatment plants, as pre-treatment of a biological process or as a refining step for biologically-treated leachate. This method could be used for pre-landfill or post-landfill leachate treatment in combination with, or included within, other processes (i.e., a biological process), mainly to enhance the efficiency of ammonium-salt removal. There are other researchers [24], who have worked with zeolites synthesized from fly ash and used it for the removal of heavy and toxic metal ions.

Fly ash has also been utilized as low-cost adsorbent for various adsorption processes for removal of pollutants in air and water systems. A lot of work have been conducted using fly ash for adsorption of NOx, SOx, organic compounds, and mercury in air, and cations, anions, dyes and other organic matters in waters [25,26]. Fly ash has been used for the synthesis of the geopolymers and applications of geopolymer-based materials within new ceramics, cements, matrices for stabilizing hazardous waste, fire-resistant materials, asbestos-free materials and high-tech materials. Fly ash-based geopolymers were found to immobilize a number of trace metals (e.g., Be, Bi, Cd, Co, Cr, Cu, Nb, Ni, Pb, Sn, Th, U, Y, Zr and rare-earth elements). The leachable levels of elements (As, B, Mo, Se, V and W) occurring in their oxyanionic forms increased after geopolymerization [27].

5.3. Food residues
Food wastes can be generated in the production, processing, transportation, distribution, or consumption of food. Microbial inoculation enhances the degradation of food wastes, increases the total nitrogen and the germination rate of alfalfa seed, shortens the maturity period and improves the quality of biofertilizer. Microbial conversion of food waste to biofertilizer is a feasible, potential technology to maintain natural resources and to reduce the impact on environmental quality [31].

Food waste can recycle soil-nitrate nitrogen, which is prone to leaching after crops are harvested. Nitrogen recycling not only reduces the amount of nitrogen fertilizer applied in corn production but also mitigates greenhouse-gas emissions by saving energy to be used for the production from industrial fertilizer of the same amount of nitrogen required for growth of corn crop [32].

Biodiesel fuel from edible oil waste [33–35], use as animal feed [36] and bio-hydrogen production as a source for clean energy [37] are some other remediation strategies.
5.4. Compost

Biological treatment of organic waste is an age-old practice that has had a strong revival in the past decade. Gil et al. [38] have studied the effects of compost produced from cattle manure and applied it as a replacement for the mineral fertilizer conventionally used for maize. Results show that this compost was a good substitute for the mineral fertilizer. Its application improved the chemical properties and nutrient status of the soil in relation to the mineral fertilization and it did not take the levels of heavy metals in soils over dangerous limits.

In order to dispose off sewage sludge, Pengcheng et al. [39] added sewage sludge compost to a silty-clay embankment soil. The results showed that addition of sewage-sludge compost increased available nitrogen, phosphorus, organic matter, cation-exchange capacity, and water content, decreased the bulk density, and improved ryegrass growth on a highway embankment in a semi-arid part of China. Its application not only enhanced ryegrass growth and reduced run-off and soil erosion but also improved soil-quality parameters, increased the growth of ryegrass, and reduced the total mass flux of sediments in run-off. Heavy-metal concentrations in sediment from surface run-off were similar to that in the control treatment for application rates for sewage-sludge compost up to 60 tons/ha.

Application of municipal solid-waste compost to agricultural soils is becoming an increasingly important global practice to enhance and to sustain organic matter and fertility levels of soil. To explore the economic feasibility of applications of municipal solid-waste compost, nutrient-management plans were developed for rice-wheat and cotton–wheat cropping systems in a developing country. Three-year field trials were conducted to measure yields and to determine the economic benefits of using three management strategies and two nutrient doses. Fertilizer doses were based on standard nitrogen, phosphorus and potassium recommendations or on site-specific soil measurements of phosphorus levels available to plants. Results showed that incorporating municipal solid-waste compost improved physical properties (e.g., bulk density and penetration resistance). No potential risks of heavy-metal accumulation were observed [40].

According to Moldes et al. [41], composting may be a safe, successful strategy for accelerating decomposition and stabilization of biodegradable components of biowaste from municipal solid waste, for sustainable complete recycling, thereby producing compost that can be used as soil conditioner and/or organic fertilizer. Here, compost was obtained on an industrial scale by anaerobic fermentation followed by aerobic stabilization. The dependent variables (shoot dry weight of Lepidium sativum and Hordeum vulgare) were adjusted to linear equations, and good statistical parameters for correlation and significance were obtained. The best results were obtained by mixing municipal solid-waste compost with composted pine bark, rather than mixing municipal solid-waste compost with peat.

6. Role of analytical methodologies in the field

Different experimental approaches have been suggested in the past few decades to determine metal species in complex matrices of unknown composition. Current remediation technologies use chelating agents for mobilization and removal of potentially toxic metals from contaminated soils. These processes can be done in situ, as enhanced phytoextraction, chelant-enhanced electrokinetic extraction and soil flushing, or ex situ, as the extraction of soil slurry and soil heap/column leaching. Biosurfactant foams are also applied for the removal of leachable metal ions and act as an effective bioremediation agent. Metals and organic wastes are treated simultaneously.

We should note that, in connection with waste-related problems, terms like green analytical chemistry and clean methodologies have emerged in the field of analytical chemistry. Solid-phase extraction, microwave-assisted extraction, supercritical fluid extraction, pressurized fluid extraction, gas chromatography-mass spectrometry (GC-MS), high performance liquid chromatography-mass spectrometry (HPLC-MS), and tandem mass spectrometry (MS²) are currently in use in recovery and removal of biodegradable and non-degradable wastes. Table 2 shows analytical methodologies in waste-treatment strategies.

Dutta et al. [42] synthesized dithiooxamide-functionalized chloromethylated polystyrene-divinylbenzene (PS-DVB) resin and used it in the separation of heavy-metal ions [e.g., Cu(II), Zn(II), Cd(II) and Pb(II)]. Parameters, such as the amount of the resin, effect of pH, equilibration rate, sorption and desorption of metal ions, and the effect of diverse ions have been studied, and the reliability of the method has been tested by analyzing certified samples.

Similarly, a PS-DVB (8%) copolymer was functionalized by coupling it through an —N=N— group with 2-naphthol-3,6-disulfonic acid (NDSA) [45]. This method was successfully applied for the speciation of chromium in natural-water samples.

Again, a study investigated the detailed metal speciation/fractionations of a Cu-contaminated soil before and after electrokinetic remediation as well as their relationships with the soil microbial and enzyme activities. The findings suggested that the bioavailability of heavy metals in soil and their ecotoxicological effects on the soil biota before and after electromediation can be better understood in terms of their chemical speciation and fractionations [47].
A remediation agent containing a biosurfactant was prepared by spray drying the sterilized culture broth of Gordonia sp. strain JE-1058, and the agent was designated as JE1058BS. It showed a strong potential to be applied as an oil-spill dispersant even in the absence of solvent and also proved to be an effective bioremediation agent for oil spills at sea. These results indicate that biosurfactant-containing JE1058BS has great potential to a remediation agent for the clean-up of oil spills at sea and on shorelines [48].

A large part of treatment of process waste streams in practice is aerobic biotreatment in wastewater-treatment plants, as it is cost effective and generally more environmentally friendly than harsher chemical and physical treatments. Pharmaceutical syntheses use a range of halogenated compounds (either as reagents, solvents or intermediates) that pose particular challenges to microbial degradation. This is especially so for some fluorinated compounds due to resistance to enzymatic cleavage of the C–F bond in some cases. The data were obtained from a case study involving the monitoring of the biodegradation of 4-fluorocinnamic acid by chromatographic techniques. These methods were used to monitor not only disappearance of the compound but

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also formation of degradation products in order to confirm mineralization. Also, MS was used to elucidate the metabolic pathway [55].

The in vitro reductive degradation of a chlorinated herbicide by iron powder was investigated and the substrate studied was triallate (S-2,3,3-trichloroallyl di-isopropyl thiocarbamate), which contains a trichloroethylene moiety potentially reducible by zero-valent iron. Herbicide decay, corresponding evolution of total organic carbon (TOC), total organic halide (TOX) and chloride-ion release were regularly monitored throughout the reactions and the main degradation by-products were identified by HPLC-MS. The results showed that, after 5 days, the extent of herbicide degradation was about 97%, and that the reaction proceeded through the formation of a dechlorinated alkyne by-product (S-2-propynyl di-isopropyl thiocarbamate) resulting from the complete dechlorination of triallate [56].

GC-MS, GC-MS², LC-MS and LC-MS² have become indispensable tools for investigating pharmaceuticals and personal-care products (PPCPs) in environmental matrices. MS also allows use of isotope-labeled compounds (ILCs), if available, to correct for matrix effects and the uncertainty introduced during sample preparation and analysis, further enhancing data quality and the quality of the analysis when used for chromatographic detection. Sewage influents and effluents of different urban areas of Greece were analyzed for polar pharmaceutical residues, used in human medicine. Analysis was carried out using capillary GC-MS with selected-ion monitoring [57].

Mechanical thermal expression (MTE) is a developing process for the non-evaporative removal of water from low-rank coals prior to combustion. The water removed in this process contains both organic and inorganic components that would prevent its direct disposal to the environment. In a study [58], under appropriate conditions the concentration of organic impurities, as determined by biological oxygen demand (BOD), was reduced to be below guideline limits for release to surface waters. Evidence of the effectiveness of remediation for organic carbon removal was also demonstrated by Fourier-transform infrared (FTIR) and pyrolysis-GC-MS (Py-GC-MS). Py-GC-MS revealed that the volatile organic components (aliphatic and aromatic hydrocarbons) were reduced by ~95% by anaerobic treatment.

Degradation of picloram, a widely-used herbicide, was undertaken by the electrochemical advanced oxidation process, namely electro-Fenton in aqueous solution. This process generates catalytically hydroxyl radicals that are strong oxidizing reagents for the oxidation of organic substances. Mineralization of picloram was followed by the total organic carbon (TOC) analysis. At the end of 8 h of electrolysis, 95% of the initial TOC was removed. Several degradation products were identified by using HPLC, LC-MS, GC-MS and ion-chromatography analysis [59].

A preliminary investigation has been carried out on the occurrence and effects of antibiotics used in Italian aquaculture [60]. The presence of flumequine and oxytetracycline in sediments sampled from two trout farms and three sea-bass farms and in their surrounding environments was selected for an analytical investigation using MS².

7. Conclusions

The ultimate goal of waste management lies in its applicability to environmental remediation, so the challenge is to reduce, recycle and reuse waste.

Further developments and new techniques must continue to be introduced in using wastes for environmental remediation. Waste remediation may be useful and necessary as a short-term measure but it does not constitute green chemistry. Green chemistry plays a significant role in facilitating a holistic approach that eliminates waste at source. Promoting clean methodologies is the best way to address future environmental challenges. Green chemistry and sustainability of resources are essential for our future world.

References
