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**ENVIRONMENTAL CRIME**

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## LIST OF ACRONYMS

AFO – animal feeding operation	LA-ICPMS – laser ablation inductively coupled mass spectrometry
ARF – advance recovery fee	LPEUR – Law for Promotion of Effective Utilization of Resources (Japan)
ASTER – Advanced Spaceborne Thermal Emission and Reflection Radiometer	LPME – liquid-phase microextraction
BAN – Basel Action Network	MODIS – moderate resolution imaging spectroradiometer
BTEX – benzene, toluene, ethylbenzene, and xylene	MS – mass spectrometry
CAA – Clean Air Act	MST – microbial source tracking
CAFO – concentrated animal feeding operation	MSW – municipal solid waste
CERCLA – Comprehensive Environmental Response, Compensation, and Liability Act	MTBE – methyl <i>tert</i> -butyl ether
CF-IRMS – continuous flow-isotope ratio mass spectrometry	NAPL – non-aqueous phase liquids
CPU – central processing unit	NCI – negative chemical ionization
CRT – cathode ray tube	NMP – Nutrient Management Plan
CWA – Clean Water Act	NPDES – National Pollutant Discharge Elimination System
DART – direct analysis in real time	NZEC – New Zealand Environmental Court
DIN S4 – extraction method of the Institut für Normung, Germany	OCP – organochlorine pesticide
DNA – deoxyribonucleic acid	OP-FTIR – open path Fourier transform infrared spectroscopy
DTSC – California Department of Toxic Substance Control	PAH – polycyclic aromatic hydrocarbon
EA NEN 7371/NEN 7371 – Dutch Environmental Agency Availability Test	PBDD – polybrominated dibenzo- <i>p</i> -dioxin
ECD – electron capture detector	PBDF – polybrominated dibenzofuran
EDXRF – energy-dispersive X-ray fluorescence	PCB – polychlorinated biphenyl
EEE – electrical and electronic equipment	PCE – tetrachloroethene
EIA – Environmental Investigation Agency	PCR – polymerase chain reaction
EPR – extended producer responsibility	PDA – portable digital assistant
ERI – electrical resistivity imaging	PDF – portable document format
ESA – Endangered Species Act	PFPD – pulsed flame photometric detectors
EU – European Union	RCRA – Resource Conservation and Recovery Act
e-waste – electronic waste	REACH – Registration, Evaluation, and Authorization of Chemicals
FID – flame ionization detector	RHA – Rivers and Harbors Act
FIFRA – Federal Insecticide, Fungicide, and Rodenticide Act	RoHS – Restriction on Hazardous Substances (EU)
FTIR – Fourier transform infrared spectroscopy	RT – reverse transcription
GC – gas chromatography	SDWA – Safe Drinking Water Act
GC-ECD – gas chromatography-electron capture detection	SEM – scanning electron microscope
GC-FID – gas chromatography-flame ionization detection	SPLP – Synthetic Precipitation Leaching Procedure
GC/HRMS – gas chromatography/high-resolution mass spectrometry	SPE – solid phase extraction
GC-IRMS – gas chromatography-isotope ratio mass spectrometry	SRM – standard reference material
GC-MS, GC/MS – gas chromatography-mass spectrometry	SW-846 – US EPA Test Methods for Evaluating Solid Waste, Physical/Chemical Methods
GC-NCI-MS – gas chromatography-negative chemical ionization mass spectrometry	TCA – trichloroethane
GIS – geographic information systems	TCE – trichloroethene
GPR – ground-penetrating radar	TCLP – Toxicity Characteristic Leaching Procedure
GPS – global positioning system	TSCA – Toxic Substances Control Act
HARL – Home Appliance Recycling Law (Japan)	TTLIC – Total Threshold Limit Concentrations
HF-LPME – hollow fiber based liquid-phase microextraction	TV – television
ICPMS – inductively coupled plasma-mass spectrometry	UEEE – used electrical and electronic equipment
IRMS – isotope ratio mass spectrometry	US – United States
LA – laser ablation	US EPA – United States Environmental Protection Agency
	UV – ultraviolet
	VTB – viral toolbox
	VOC – volatile organic compound
	WEEE – waste electrical and electronic equipment
	WET – Waste Extraction Test
	XRD – X-ray diffraction

## INTRODUCTION

This environmental crime review paper is a followup to the review prepared for the 15<sup>th</sup> Interpol International Forensic Science Symposium in October 2007. It is compiled primarily from a literature review of papers published since early 2007 on specific environmental crime and environmental forensics topics. Books, critical reviews, and other papers that were published before 2007 are also included when those may be of interest to the reader.

This paper differs in format from previous environmental crime review papers in that it does not present a field or crime scene examination component nor does it summarize the wide range of analytical instrumentation and methods useful to environmental analysis. For readers desiring some background knowledge for field investigations and sample analysis, the review papers prepared for the three prior Interpol International Forensic Science Symposia are available.<sup>1-3</sup>

Electronic waste (e-waste) and microbial source tracking are two topics highlighted in this review. Concerns over the hazardous components in electrical and electronic equipment (EEE) have led to regulations that seek changes in the components of the equipment and establish requirements for disposing of that equipment.<sup>4-10</sup> Initiatives for reuse, recycling, and proper disposal of e-waste are underway in many nations, but the success of the initiatives and the compliance with regulations vary.<sup>11-15</sup> Literature searches reveal many papers on the contamination and health effects from improper recycling of waste EEE (WEEE).<sup>16-20</sup> Many papers discuss the leachability of metal contaminants from e-waste with reference to the United States Environmental Protection Agency (US EPA) Toxicity Characteristic Leaching Procedure (TCLP),<sup>21, 22</sup> although metals are not the only contaminants of concern.<sup>23, 24</sup>

Microbial source tracking (MST) involves the various biological and chemical techniques that can be used to determine a source of fecal pollution. Both human and animal waste can contaminate water sources, and the origins and causes for contamination events must be determined in order to prevent repeat occurrences.<sup>25, 26</sup> Several groups are investigating methods that may help locate a single source or multiple contributing sources of a contamination.<sup>27-30</sup>

Environmental crime and environmental forensics are large, general search topics. The environmental crime search yielded papers with specific regulatory and enforcement reviews<sup>31-34</sup> and legal perspectives on environmental crime cases.<sup>35-37</sup>

An essential environmental forensics journal turns 10 years old; some of its history and fundamental papers are revisited.<sup>38</sup> Chemical characterization and determining sources of pollutants are key topics in environmental forensics papers.<sup>39-44</sup> A new area of environmental forensics investigations was proposed in response to problems with imported Chinese drywall.<sup>45</sup>

## ENVIRONMENTAL CRIME

For the past 5 years, Solow has reviewed United States (US) environmental crime enforcement actions from the preceding year.<sup>33, 46-49</sup> For the survey of enforcement actions of 2009, Solow and Carpenter summarized notable criminal cases and graphically presented the number of cases prosecuted under environmental statutes. Also included were summaries of the current US EPA National Enforcement Initiatives, the new US Department of Justice guidance on discovery, the increased oversight of chemicals in the Toxic Substances Control Act, and the emerging environmental issue of e-waste.<sup>33</sup> In addition to the usual summaries of selected cases, Solow's survey of 2008 developments included a note about criminal cases on illegal discharges from vessels and the decisions on the corporate criminal liability for acts of non-managerial employees involved in these illegal discharges.<sup>46</sup> The 2007 survey included an interview with Stacey Mitchell, chief of the US Department of Justice Environmental Crimes Section, a mention of a highly-rated environmental crimes blog, and discussions on Clean Water Act (CWA) prosecutions, voluntary disclosures, and attorney-client privilege.<sup>47</sup>

Sanders writes an annual environmental law review for Water Environment Research. Each review includes regulatory changes and developments in wastewater treatment, ambient waters, hazardous wastes, and emerging issues. Also included are summaries of agreements, enforcement actions, and lawsuits from the previous year.<sup>50-52</sup>

*Natural Resources & Environment*, a journal published quarterly by the Section of Environment, Energy, and Resources of the American Bar Association, dedicated an issue to environmental crime (Winter 2009).<sup>53</sup> The issue included an interview with Granta Nakayama who was the Assistant Administrator for the US EPA Office of Enforcement and Compliance Assurance at that time.<sup>54</sup> Harrell et al. discussed the "knowing violations" aspect of criminal prosecutions, the variation in penalties among the US environmental statutes, and the complexities of prosecuting an environmental crime case. The authors explained that an environmental prosecutor would need to develop an understanding of statutes, programs, and the multiple layers of regulations at federal, state, and local levels and develop the skill to present the complex material to a jury.<sup>37</sup> Periconi provided a history of environmental crime prosecutions in the State of New York and added information on current trends and case law developments. The author described an overall decline in environmental crime prosecutions in New York state and suggested that one of the reasons given for this decline in prosecutions was the lack of jail time imposed for environmental crimes. Periconi noted that judges and juries seem more inclined to impose jail time for more threatening crimes (for example, street crime); civil enforcement, with its large fines and monitoring programs for environmental compliance, seems to have moved to the forefront of dealing with environmental violations.<sup>32</sup> Other topics from the environmental crime articles in the issue included the role of motive in environmental crimes, victims' rights, the prosecution of corporate officers, and new European Union (EU) chemical regulations.<sup>53</sup>

Burns et al. provided an overview of issues, history, and developments in the wide field of environmental crime in their book, *Environmental Law, Crime, and Justice*. The authors briefly reviewed types of environmental harm and detailed the US history of environmentalism and how it stimulated actions and policies of the federal government. The founding and history of the US Environmental Protection Agency, the development and enforcement of environmental

laws (including crime investigation and data sources), and the connection between environmental justice and enforcement are detailed. The authors also shared their perspectives on the future challenges and expectations regarding regulatory efforts and criminal prosecutions of environmental crimes.<sup>55</sup>

Shafer et al. authored a concise summary of environmental crime as defined by US statutes. The article starts with general introductions to criminal versus civil penalties, federal enforcement responsibilities, and links with other criminal violations. After a brief summary of the “knowing” element of criminal violations, the authors discuss the topics of liability, defenses, voluntary compliance, and sentencing. In the following sections, the authors examined the Clean Air Act (CAA), the Clean Water Act (CWA), the Rivers and Harbors Act (RHA), the Safe Drinking Water Act (SDWA), the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA), the Resource Conservation and Recovery Act (RCRA), the Toxic Substances Control Act (TSCA), the Federal Insecticide, Fungicide, and Rodenticide Act (FIFRA), and the Endangered Species Act (ESA). For each, the authors provided the purpose of the statute and the elements of an offense of that statute including violations, intent, and exceptions. Possible defenses and penalties for violation of the statute are also included. While each section in this review is brief, the authors provided more than 600 footnotes.<sup>36</sup>

Brickey wrote *Environmental Crime: Law, Policy, Prosecution*, a law school textbook dedicated to the study of environmental crime. The book, published in 2009, includes discussions on the environmental regulatory framework and the integration of criminal law principles with environmental law. Brickey focused on the criminal provisions for CWA, CAA, RCRA, and CERCLA and focused on the concept of knowing endangerment. Conventional criminal statutes often linked to environmental crime cases, such as conspiracy, mail fraud, false statements, and obstruction of justice, were discussed along with some enforcement issues. Also included were several excerpts from legal work by other authors.<sup>56</sup>

Sachs compared the chemical toxicity regulations of the US to those of the EU and found the US regulations lacking. The author stated that the Toxic Substances Control Act (TSCA) in the US has been hindered by the heavy information demand it places on the US EPA. Thousands of chemicals in production at the time TSCA was written were exempted from the Act, and, since then, the burden has been on the US EPA to prove that a chemical poses an unreasonable risk before it can compel testing by the manufacturer. In contrast to this, other US chemical regulations are more strict; manufacturers of pesticides and pharmaceuticals are required to prove the safety of their chemical products to regulators. Sachs described the EU Registration, Evaluation, and Authorization of Chemicals (REACH) legislation as the law that will buttress TSCA. REACH requires that chemicals manufactured or imported into the EU be registered and undergo an amount of toxicity testing in line with the volume sold. Manufacturers outside the EU must provide chemical registration and toxicity data on their chemicals prior to receiving approval to import into the EU. In this way, the toxicity data collected via REACH could be shared with the US EPA to improve the enforcement of TSCA.<sup>31</sup>

The International Network for Environmental Compliance and Enforcement published the second edition of the *Principles of Environmental Compliance and Enforcement Handbook* in April 2009.<sup>57, 58</sup> This handbook is available in portable document format (PDF) and can also

be accessed online at the International Police Expertise Platform Web site.<sup>58, 59</sup> This handbook gives the reader an overview of compliance and enforcement programs, the principles involved to make them effective, and information on how to build and manage compliance and enforcement programs.

*The International Lawyer*, a journal published quarterly by the American Bar Association Section of International Law, has an annual summary of selected developments, conferences, reports, and publications in international environmental law from the previous year. Review topics generally include atmosphere and climate, marine conservation, international hazard management, natural resources, and international economy and the environment. The 2009 review also included a section on international environmental litigation.<sup>35, 60, 61</sup>

The Environmental Investigation Agency (EIA) published “Environmental Crime—A Threat to Our Future” in October of 2008 and it is available online in PDF.<sup>62</sup> This report stated the definition of international environmental crime in five broad areas of offenses: illegal trade in wildlife, illegal trade in ozone-depleting substances, illegal transport and dumping of hazardous waste, illegal fishing, and illegal logging and trade in timber. Four case studies of crimes and five examples of successful enforcement models were included in the report.

With a strong emphasis on environmental harm and an international perspective on environmental issues, White wrote *Crimes Against Nature: Environmental Criminology and Ecological Justice*. Published in 2008, this book gave many examples of environmental harm from toxic waste dumping, airborne pollutants, and contamination of soil and water. The author included sections on environmental regulations and law enforcement.<sup>63</sup>

Tal et al. presented the interesting results of a year-long study of environmental enforcement actions in Israel. In the study, the authors compared the outcomes of criminal enforcement for environmental violations to the outcomes of administrative enforcement for environmental violations. Administrative enforcement actions were faster, required less detailed evidence, and were perceived as being less costly than criminal enforcement actions. The authors did warn that the use of administrative actions might lead a company to determine that the cost of the fines for violations could be less than the cost to improve environmental controls. “Rather than creating a ‘polluter pays’ dynamic, the message becomes: ‘it pays to pollute.’”<sup>64</sup> In contrast, the stigma of criminal conviction or incarceration might outweigh any financial calculation or monetary savings in avoiding environmental improvements. The authors came to three conclusions from their study of 100 criminal cases and 100 administrative actions. The first conclusion was that the criminal enforcement process was more effective than the administrative process in achieving present and future compliance. The administrative path did not have clear consequences and a clear timetable for delaying or ignoring compliance requests. The administrative system contained additional weaknesses, as pointed out in the authors’ second conclusion; it lacked a timely schedule and a formal process for followup site visits to determine compliance. This time delay could allow an environmental violation to continue too long before a decision is made to pursue a criminal enforcement action. With the third conclusion, the authors stated the advantage of a combined benefit and penalty approach to environmental enforcement. The authors discovered that when enforcement personnel could

offer something to the regulated community, such as funds to assist in making environmental improvements, compliance and cooperation was more likely to occur.<sup>64</sup>

Stott gave a brief summary of how environmental enforcement is handled in the United Kingdom and provided a table of the acts of Parliament and the statutes used by the Environment Agency to protect the environment. Stott mentioned that, in general, the fines for water quality violations were higher than those for waste violations and speculated that the difference might be due to the inability to demonstrate the real cost of illegal waste dumping to the courts. One example he presented was a case in which a man abandoned nearly 200 drums of toxic waste. The fine for this violation was about half of what the man had been paid when he was hired to do the “disposal.” The authorities ended up spending more than five times the amount of the fine in cleanup costs. Stott pointed out that, with new regulations, the Environmental Damage (Prevention and Remediation) Regulations 2008, the courts could ensure that pollution will be cleaned up and the costs or damages would be recovered.<sup>65</sup>

Bostan et al. outlined how environmental crimes are categorized in Romania. The authors presented the punishment categories (range of time for imprisonment or range of criminal fines) and listed below each of the penalties descriptions of qualifying violations (for example, releasing pollutants into the water, atmosphere, or soil). Personal responsibility for environmental crimes and the classifications of crimes stipulated in environmental law in Romania also were presented in the paper.<sup>34</sup>

Two papers, both filled with references, discussed the problems and some possible solutions to environmental pollution issues faced by China. Sitaraman provided the history behind and development of environmental laws in China. Included in the paper was a table listing the major environmental laws of China from 1979 to 2004 and a table listing the major environmental treaties signed or ratified by China. Sitaraman observed that the environmental laws are not always specific enough to avoid variations in interpretation between different regions in China. The author suggested cleanup efforts, advanced technology, and citizen involvement as tools that could be used in improving environmental protection efforts.<sup>66</sup>

Goelz suggested that a special environmental court, modeled after the New Zealand Environmental Court (NZEC), could provide a good start to dealing with China’s environmental problems. Goelz compared this proposition to the current existence of China’s other specialized courts, including military courts, railway transport courts, maritime transport courts, and forestry courts. The author made some comparisons between the structure and operation of the NZEC and that of the Chinese maritime courts and outlined how an environmental court could be designed to align with the Chinese legal system and best serve the goal of environmental protection.<sup>67</sup>



# ENVIRONMENTAL FORENSICS

## 10 YEARS OF ENVIRONMENTAL FORENSICS

March 2000 hailed the first print issue of the journal *Environmental Forensics*. The first editorial traced the emergence of “environmental forensics” back nearly 20 years to the need to distinguish between different petroleum hydrocarbons in the environment.<sup>68</sup> Since that time, the editors noted, the field of environmental forensics had expanded to include a wide range of scientific techniques. An investigator was no longer just involved with identification of hydrocarbon products (often termed “chemical fingerprinting”) or the fate and transport modeling of hydrocarbons in the environment. Now, the editors pointed out, an investigator could use a toolbox of techniques from historical sources and scientific disciplines: historical aerial photography, industrial archeology, regulatory history, analytical chemistry, atmospheric chemistry, geochemistry, toxicology, hydrogeology, environmental fate and transport, computer modeling, and health risk assessment.<sup>68</sup>

That editorial also included the prediction that environmental forensics would take a larger role in regulatory and legal actions in the future. Environmental attorneys, regulatory agencies, and insurance companies seeking to establish or allocate liability for environmental contamination would need environmental forensics. Also, scientists and the courtroom would need the knowledge to distinguish between the evidence and expert opinions based on valid scientific methods and the unwanted intrusion of junk science.<sup>68</sup>

In the 10 years since that first editorial, *Environmental Forensics* has become a primary journal for developments in the field of environmental forensics. It also has been one of the best sources for reviews on the disciplines and techniques used in environmental forensics. A newcomer to this type of investigative work would find many excellent fundamental reviews throughout this journal. The legal and investigative emphasis of *Environmental Forensics* complements two other journals in the environmental arena, the Royal Society of Chemistry’s *Journal of Environmental Monitoring* and the *International Journal of Environmental Analytical Chemistry*, the official journal of the International Association of Environmental Analytical Chemistry.<sup>69,70</sup>

Morrison published a two-part critical review of environmental forensics techniques in the journal during its first year.<sup>71,72</sup> Morrison stated that when the results of environmental forensics techniques are introduced in the courtroom as evidence, the scientific validity of the data can be examined and challenged, often with success. His purpose for writing the review papers was to give an overview of the different techniques available and the benefits and purpose of each. The reader could then choose the single technique or combination of techniques needed to successfully investigate and prove the case.<sup>71,72</sup> Table 1 lists the topics from these two Morrison review papers. It also lists other important review papers published in *Environmental Forensics* and the key topics from those papers.

A wide variety of topics have been published in *Environmental Forensics* since 2007. Table 2 lists a survey of the topics in these categories. These topics have been organized in the general categories of contaminants, techniques, location or media, legal, or other.

**Table 1. Topics in Selected *Environmental Forensics* Review Papers**

Title, Author(s) and Year	Topics
<p><b>Critical Review of Environmental Forensic Techniques, Part I</b><sup>71</sup> RD Morrison 2000</p>	<ul style="list-style-type: none"> <li>• Aerial Photography</li> <li>• Underground Storage Tank Corrosion Models</li> <li>• Commercial Availability of a Chemical</li> <li>• Chemical Formulations and Applications Unique to a Manufacturing Activity</li> <li>• Polychlorinated Biphenyls (PCBs)</li> <li>• Age Dating Chlorinated Solvents</li> <li>• Presence of Non-Aqueous Phase Liquids (NAPLs)</li> </ul>
<p><b>Critical Review of Environmental Forensic Techniques, Part II</b><sup>72</sup> RD Morrison 2000</p>	<ul style="list-style-type: none"> <li>• Petroleum Hydrocarbons</li> <li>• Fingerprinting</li> <li>• Proprietary Additives</li> <li>• Alkyl-leads</li> <li>• Oxygenates</li> <li>• Dyes</li> <li>• Stable Isotope Analysis</li> <li>• Weathering and Biomarkers</li> <li>• Degradation Models</li> <li>• Pristane/Phytane Ratios</li> <li>• Benzene/Toluene/Ethylbenzene/Xylene (BTEX) Ratios</li> <li>• Contaminant Transport Models</li> </ul>
<p><b>Application of Forensic Techniques for Age Dating and Source Identification in Environmental Litigation</b><sup>73</sup> RD Morrison 2000</p>	<ul style="list-style-type: none"> <li>• Underground Storage Tank Corrosion Models</li> <li>• Commercial Availability of a Chemical</li> <li>• Chemical Applications Unique to a Manufacturing Activity</li> <li>• Age Dating and Source Identification of Chlorinated Solvents</li> <li>• Age Dating and Source Identification of Petroleum Hydrocarbons</li> <li>• Proprietary Additives</li> <li>• Alkyl-Leads</li> <li>• Lead Scavengers</li> <li>• Oxygenates</li> <li>• Stable Isotope Analysis</li> <li>• Biomarkers</li> <li>• Degradation Models</li> <li>• Contaminant Transport Models</li> <li>• Contaminant Transport through Pavement and Soil</li> <li>• Ground Water Models</li> </ul>
<p><b>Application of Aerial Photography and Photogrammetry in Environmental Forensic Investigations</b><sup>74</sup> WM Grip, RW Grip, and RD Morrison 2000</p>	<ul style="list-style-type: none"> <li>• Photograph Acquisition</li> <li>• Interpretation of Aerial Photography</li> <li>• Trial Presentation of Aerial Photography</li> </ul>
<p><b>Age Dating of Environmental Organic Residues</b><sup>75</sup> IR Kaplan 2003</p>	<ul style="list-style-type: none"> <li>• Long-Term Methods:             <ul style="list-style-type: none"> <li>○ Radiometric Carbon-14 (<sup>14</sup>C)</li> <li>○ Amino Acid Racemization</li> </ul> </li> <li>• Short-Term Method:             <ul style="list-style-type: none"> <li>○ Release Times of Crude Oil, Refined Petroleum Fuels, and Chlorinated Solvents</li> </ul> </li> </ul>

**Table 1. Topics in Selected *Environmental Forensics* Review Papers**

Title, Author(s) and Year	Topics
<b>Fingerprinting of Hydrocarbon Fuel Contaminants: Literature Review</b> <sup>76</sup> H Alimi, T Ertel, and B Schug 2003	<ul style="list-style-type: none"><li>• Fingerprinting Techniques</li><li>• Characterization of Hydrocarbon Fuel Contaminants:<ul style="list-style-type: none"><li>○ Fingerprinting of Fuel Contaminants Using Polycyclic Aromatic Hydrocarbon (PAH) Compounds</li><li>○ Fingerprinting of Fuel Contaminants Using Biomarkers</li></ul></li><li>• Fate of Hydrocarbon Fuels in the Environment:<ul style="list-style-type: none"><li>○ Weathering of Volatile Aromatic Hydrocarbons</li><li>○ Weathering of n-Alkanes (or n-Paraffins)</li><li>○ Weathering of PAHs</li><li>○ Weathering of Biomarkers</li></ul></li></ul>
<b>Combustion-Derived Polycyclic Aromatic Hydrocarbons in the Environment—A Review</b> <sup>77</sup> ALC Lima, JW Farrington, and CM Reddy 2005	<ul style="list-style-type: none"><li>• Formation of PAHs</li><li>• Environmental Fate:<ul style="list-style-type: none"><li>○ Physicochemical Properties</li><li>○ Biodegradation</li><li>○ Photodegradation and Chemical Oxidation</li></ul></li><li>• Source Apportionment:<ul style="list-style-type: none"><li>○ Source Diagnostic Ratios</li><li>○ Historical Records</li><li>○ Stable Carbon Isotopic Composition</li><li>○ Radiocarbon Measurements</li></ul></li></ul>
<b>Forensic Fingerprinting of Biomarkers for Oil Spill Characterization and Source Identification</b> <sup>78</sup> Z Wang, SA Stout, and M Fingas 2006	<ul style="list-style-type: none"><li>• Biomarker Chemistry</li><li>• Biomarker Analysis Methodologies</li><li>• Characteristic Fragment Ions of Biomarkers</li><li>• Biomarker Distributions</li><li>• Diagnostic Ratios (Indices) and Cross-Plots of Biomarkers</li><li>• Unique Biomarker Compounds</li><li>• Effects of Weathering on Chemical Composition of Oil and Biomarkers</li><li>• Application of Multivariate Statistical Methods for Biomarker Fingerprinting</li></ul>
<b>Applied Dendroecology and Environmental Forensics. Characterizing and Age Dating Environmental Releases: Fundamentals and Case Studies</b> <sup>79</sup> J-C Balouet, G Oudijk, KT Smith, I Petrisor, H Grudd, and B Stocklassa 2007	<ul style="list-style-type: none"><li>• Fundamentals:<ul style="list-style-type: none"><li>○ Scientific Background</li><li>○ Dendroecological Principles</li><li>○ Field and Laboratory Procedures</li><li>○ Ring-Width Data and Principles</li><li>○ Characterizing the Environmental Release</li></ul></li><li>• Case Studies:<ul style="list-style-type: none"><li>○ Contamination by Heating Oil</li><li>○ Chlorinated Solvent Plume</li><li>○ Unknown Plumes</li><li>○ Leaking Underground Storage Tanks</li><li>○ Former Gasoline Service Station</li></ul></li></ul>

**Table 1. Topics in Selected *Environmental Forensics* Review Papers**

Title, Author(s) and Year	Topics
<b>Age Dating Heating Oil Releases, Part 1. Heating-Oil Composition and Subsurface Weathering</b> <sup>80</sup> G Oudijk 2009	<ul style="list-style-type: none"> <li>• Heating-Oil Composition</li> <li>• Heating Oil in the Subsurface</li> <li>• Assessing Heating-Oil Release</li> <li>• Factors Influencing Petroleum Weathering <ul style="list-style-type: none"> <li>○ Hydrocarbon Physical State, Petroleum Chemistry</li> <li>○ Temperature, Light</li> <li>○ Contact with Water, Hydrologic Conditions</li> <li>○ Soil Lithology, Texture, and Moisture</li> <li>○ Oxygen and Nutrients, Redox Conditions</li> <li>○ Vegetation, Bacteriocides</li> </ul> </li> <li>• Sequence of Biodegradation</li> <li>• Christensen and Larsen Method</li> </ul>
<b>Age Dating Heating Oil Releases, Part 2. Assessing Weathering and Release Time Frames Through Chemistry, Geology and Site History</b> <sup>81</sup> G Oudijk 2009	<ul style="list-style-type: none"> <li>• Age-Dating Methodology</li> <li>• Assessing Petroleum Weathering with Chromatograms</li> <li>• Sampling and Laboratory Analyses</li> <li>• Evaluating the Age Range</li> <li>• Critiquing the Matrix Age Range</li> <li>• Range of Error</li> <li>• Case Study</li> </ul>
<b>The Rise and Fall of Organometallic Additives in Automotive Gasoline</b> <sup>82</sup> G Oudijk 2010	<ul style="list-style-type: none"> <li>• Tetraethyl Lead-Based Additives</li> <li>• Lead Scavengers</li> <li>• Tetraethyl Lead Extenders</li> <li>• Mixed Lead Additives</li> <li>• Lead Phase-Out</li> <li>• Organometallic Anti-Knock Agents</li> </ul>
<b>Reconstructed Plume Method for Identifying Sources of Chlorinated Solvents</b> <sup>83</sup> BL Murphy and F Mohsen 2010	<ul style="list-style-type: none"> <li>• Degradation Pathways</li> <li>• Reconstructing Chlorinated Solvent Plumes</li> <li>• Factoring in Groundwater Flow Direction: A Case Study</li> <li>• Trichloroethane (TCA) Plume Age-Dating</li> </ul>

**Table 2. Survey of Recent Topics Published in *Environmental Forensics* (2007–May 2010)**

Category	Topics Include
Contaminant	Polychlorinated biphenyls, lead, benzene, chlorinated hydrocarbons, gasolines, fuels, pesticides, dioxins, methyl <i>tert</i> -butyl ether (MTBE), polycyclic aromatic hydrocarbons, air particulates, mercury
Technique	Fingerprinting (hydrocarbons, lead), ground-penetrating radar (GPR), environmental modeling, wipe samples and analysis, carbon aerosol measurements, microscopy, statistics, dendrochronology, isotopic characterization
Location or Media	Surface water, ground water, river water, public drinking water, coastal erosion, soils, air, nutrient concentrations, sewage overflow, mining waste, buried waste
Legal	Bias and testimony, Daubert, cost allocations, crime investigation
Other	Biological infections, climate change, greenhouse gases, chemical warfare agent surrogates, standard reference materials (SRM), radiological hazards

## RECENT BOOKS AND PAPERS

Hester and Harrison edited the 2008 book *Environmental Forensics*. This was volume 26 in the Royal Society of Chemistry Publishing series, “Issues in Environmental Science and Technology.” Contributors to this book tackled topics of source identification, microbial techniques, spatial considerations of stable isotope analysis, chemical fingerprinting of petroleum, chlorinated solvents, and ground water pollution.<sup>84</sup>

Wang and Stout compiled the expertise of more than 30 scientists into the 2007 book *Oil Spill Environmental Forensics: Fingerprinting and Source Identification*. Topics from the 17 chapters included methods and factors affecting petroleum fingerprinting, spill site investigation, petroleum biomarker fingerprinting, identification by comprehensive two-dimensional gas chromatography, quantitative chemical fingerprinting, chemical heterogeneity of modern marine fuel oils, biodegradation of oil hydrocarbons, oil spill remote sensing, and case studies.<sup>85</sup>

Murphy and Morrison released a second edition of their book, *Introduction to Environmental Forensics*, in 2007. This book was designed to give the reader an organized presentation of the forensic tools available to use in environmental forensics. Updates from the first edition included chapters on laser ablation inductively coupled mass spectrometry (LA-ICPMS), scanning electron microscope (SEM) techniques, X-ray diffraction (XRD), pattern recognition methods, and emerging forensic techniques. The chapters on sampling techniques and statistical methods were also expanded for the second edition.<sup>86</sup>

Mudge was the editor of *Methods in Environmental Forensics*, published in 2009. Mudge also authored or co-authored four of the chapters. Topics in this book included radionuclides, chemical fingerprinting of petroleum hydrocarbons, biomarkers and stable isotopes, volatile organic compound (VOC) analysis, molecular microbiology, statistical methods, air pollution monitoring, and evidence and expert witnesses.<sup>87</sup>

Mudge also contributed a chapter in *Criminal and Environmental Soil Forensics*.<sup>88</sup> Even though this book had a heavy emphasis on soil forensics specific to the effects from cadavers and footwear, it contained some chapters that could be useful for the analysis of pollutants in soils. These chapters included discussions on tracing soil and ground water pollution, current and future spectroscopic methods of analysis for soils, and a novel handheld sensor using anodic stripping voltammetry for real-time detection of heavy metals.<sup>89</sup>

Selected papers published on environmental forensics topics are listed and briefly described in Table 3. Most of these papers are from the past three years, but a few older review papers are included as an aid to the interested reader.

**Table 3. Recent Papers in the Field of Environmental Forensics**

Category	Description
Contaminant	<p>Use of three methods (ground water modeling, subsurface environmental conditions, and isotopic fingerprinting) to evaluate the extent of dissolved perchlorate from multiple sources<sup>90</sup></p> <p>Discussion and case study on analysis and attribution of polycyclic aromatic hydrocarbons in sediments; emphasized need for analysis that includes both parent (non-alkylated) and alkylated compounds<sup>91</sup></p> <p>Investigation of polychlorinated biphenyl concentrations in gas and particles from ambient air samples taken in a suburb in Bursa, Turkey; temperature, evaporation, and atmospheric transport were factored into the study<sup>92</sup></p> <p>Study of the differences in chemical composition of used and unused motor oils; used motor oils showed the presence of gasoline combustion residues and PAHs<sup>93</sup></p> <p>Testing at an oil and gas processing facility for oil characteristics that would distinguish between naturally-occurring crude oil seepage and oils present on the site due to anthropogenic causes; use of gas chromatography/mass spectrometry (GC/MS) and isotopic techniques to determine chemical signatures<sup>94</sup></p> <p>Characterization of ambient air particulate mass and ionic species near industrial zones and the Taiwan Strait in central Taiwan; major components of fine (PM<sub>2.5</sub>) and coarse (PM<sub>2.5-10</sub>) particulate matter were identified along with possible sources<sup>95</sup></p> <p>Characterization and identification problems of two oil spills (off-road diesel and crude oil) undergoing methanogenic biodegradation; concerns expressed about fingerprinting during this degradation process<sup>96</sup></p> <p>Physical and chemical characterization of samples of “slag” and “tar-like” materials; physical analysis included density, microscopic character, magnetic properties, and float-sink behavior; chemical analysis included the chemical fingerprinting of the total extractable hydrocarbons, polycyclic aromatic hydrocarbons, and petroleum biomarkers<sup>97</sup></p> <p>Study on methods to identify sources of PAHs in soils; analyzed for 45 PAHs, measured PAH ratios, and measured <i>n</i>-alkanes; recommended the extended 45 PAH analysis before pursuing additional methods for source identification<sup>98</sup></p> <p>Total alkylated PAH characterization using Ion Signature software<sup>99</sup></p>
Technique	<p>Use of satellite images to detect sources of pollution on the coast of Lebanon; more than 80 major sources of pollution were detected using the Advanced Spaceborne Thermal Emission and Reflection Radiometer (ASTER) and Landsat 7 ETM+ satellite images<sup>100</sup></p> <p>Use of moderate resolution imaging spectroradiometer (MODIS) remote sensing technique to take spectral measurements of five oil types (kerosene, lubricating oil, light diesel oil, heavy diesel oil, and crude oil); MODIS also used to monitor Jeyeh, Lebanon, storage tank leaking event<sup>40</sup></p> <p>Distinguishing oil spill patterns on water and determining the size of a major oil spill on the coast of Lebanon using satellite imagery<sup>101</sup></p> <p>Use of ground-penetrating radar and electrical resistivity imaging (ERI) to assess the geometry and extent of illegally buried toxic waste<sup>102</sup></p>

**Table 3. Recent Papers in the Field of Environmental Forensics**

Category	Description
	<p>Two case studies in using geological materials to exclude or link suspects to illegal waste disposal in Northern Ireland<sup>103</sup></p> <p>Study on soil-sampling techniques using soil treated with fipronil insecticide and placed in either opaque bags that block ultraviolet (UV) light or clear glass jars; factors of time, temperature, humidity, and UV light exposure were considered in the study<sup>104</sup></p> <p>Use of marker compounds and analysis by gas chromatography-mass spectrometry (GC-MS) and gas chromatography with flame ionization detection (GC-FID) to identify the oil contamination in the Bohai Sea of China<sup>105</sup></p> <p>Use of hollow fiber based liquid-phase microextraction (HF-LPME) coupled with GC, GC-MS, and gas chromatography-isotope ratio mass spectrometry (GC-IRMS) to identify hydrocarbons in oil spills<sup>44</sup></p> <p>Geochemical study of the tailings after processing and disposal of the Agios Filippou high sulfidation epithermal deposit (located in Kirki in northeast Greece); assessed the toxicity and potential for acid generation from the tailings<sup>106</sup></p> <p>Use of gas chromatography-combustion-isotope ratio mass spectrometry to determine stable carbon isotopes for ethenes in ground water samples to understand history of a trichloroethene (TCE) ground water plume; some results pointed to a tetrachloroethene (PCE) contamination<sup>107</sup></p> <p>Use of gas chromatography/high-resolution mass spectrometry (GC/HRMS) to identify compounds from water samples taken above a pollutant plume<sup>108</sup></p> <p>Fingerprinting five types of commercial oils (heavy diesel, crude oil, diesel, gasoline, and lubricant) using three dimensional fluorescence spectroscopy and GC-MS<sup>39</sup></p> <p>Use of compound-specific stable carbon isotope analysis and consideration of physical site characteristics responsible for petroleum weathering to understand the age of methyl <i>tert</i>-butyl ether releases<sup>109</sup></p> <p>Characterization of PAH sources in rivers and coastal environments of Louisiana using principal components analysis<sup>110</sup></p> <p>Study of arsenic contamination in Bangladesh using geographic information systems (GIS) and univariate and bivariate statistical analysis<sup>111</sup></p> <p>Use of carbon and hydrogen isotopes to create a two-dimensional isotope fingerprint to differentiate gasoline sources in environmental assessments of gasoline-contaminated sites<sup>112</sup></p> <p>Study on the potential for rapid semi-quantitative surface mapping and analysis of “contaminants” (tested on powdered aspirin and caffeine) using a direct analysis in real time (DART)/time-of-flight mass spectrometer<sup>113</sup></p> <p>New GC-MS method to distinguish between anthropogenic petroleum hydrocarbons and biogenic organic compounds in order to properly assess contamination sites and toxicity risks<sup>43</sup></p>

**Table 3. Recent Papers in the Field of Environmental Forensics**

Category	Description
	<p>Study to correlate odor index to pollutants from a pharmaceutical plant using open path Fourier transform infrared (OP-FTIR) and dispersion modeling; two volatile organic compounds emitted by the plant were ethyl acetate and acetone<sup>114</sup></p> <p>Comparison between two different sampling media, quartz filters and overhead projection film, for ambient air particulates and particulate mercury at a site in central Taiwan<sup>115</sup></p> <p>Development of new method for compound-specific chlorine stable isotope analysis for vinyl chloride using a continuous flow-isotope ratio mass spectrometer (CF-IRMS); samples were taken from a confined sandy aquifer contaminated with vinyl chloride and located near Ferrara in northern Italy<sup>116</sup></p> <p>Investigation of sources of benzene and chlorobenzene in a shallow and deep aquifer using the combination of compound-specific isotope analyses, hydrogeologic data, contaminant concentrations, and site history<sup>117</sup></p> <p>Analysis of soil samples to determine concentrations of residual organochlorine pesticides (OCP): used gas chromatograph-electron capture detection (GC-ECD) and gas chromatograph-negative chemical ionization mass spectrometry (GC-NCI-MS) after Soxhlet extraction and Florisil solid phase extraction (SPE) column cleanup<sup>118</sup></p> <p>Critical review of compound-specific stable isotope analysis of organic contaminants using gas chromatography-isotope ratio mass spectrometry; 2004 paper with biodegradation table and over 100 references<sup>119</sup></p> <p>General review of forensic applications of isotope ratio mass spectrometry, including references to the determination of the origin of contaminants in the environment, analysis of environmental contaminants, and carbon and chlorine isotopes in chlorinated organic compounds<sup>120</sup></p> <p>Review of the applications of stable isotopes to environmental studies including discussions on BTEX, PAHs, PCBs, and MTBE<sup>121</sup></p> <p>Critical review on the current and future uses of enriched stable isotope analysis in biological systems; includes tables listing tracers for aquatic ecosystems, terrestrial ecosystems, animals, and humans<sup>122</sup></p>
Location or Media	<p>Nutrient analysis of the Ria Formosa Lagoon in Portugal; nitrogen sources traced to agricultural areas, and phosphates traced to a golf complex and sewage discharges<sup>123</sup></p> <p>Analysis of sediment samples from eight locations in the Ria Formosa Lagoon for PAH concentrations; the most significant source of PAHs in the summer was determined to be boat traffic<sup>124</sup></p> <p>Determining water quality and sources of pollution in the Ljubljana River in Slovenia; input of pollution was categorized as coming from point sources (tannery and municipal effluents) or diffuse sources<sup>125</sup></p> <p>Assessment of the metal contamination in the Riou Mort River watershed, an area affected by mining and ore-treatment activities, using hydrological and</p>



**Table 3. Recent Papers in the Field of Environmental Forensics**

Category	Description
	<p>geochemical monitoring<sup>126</sup></p> <p>Sampling and analysis for organic pollutants along a portion of the Iloilo River in the Philippines; analysis focused on three major groups: PAHs, branched and cyclic saturated hydrocarbons, and sterols<sup>41</sup></p> <p>Site assessment of a section of floodplain used for the disposal of tailings and waste from multiple milling and smelting operations; historical documents, aerial photographs, geochemical and stratigraphic data were used to develop mixing equations to quantify contributions from the different mining sources<sup>127</sup></p> <p>Use of a methodology to predict the nitrogen concentration in the ground water beneath unsewered areas; field measurements matched predictions of nitrogen concentration; methodology was applied to the Liman Region in Antalya, Turkey<sup>128</sup></p> <p>Study of PAHs and hopanes in atmospheric aerosols from samples taken in nine locations in peninsular Malaysia; biomass burning and vehicular emissions were determined to be the main sources of PAHs, and the significant source of hopanes was determined to be crankcase oil in street dust<sup>129</sup></p> <p>Case investigation of a subsurface plume of non-aqueous phase liquid (NAPL) at an automotive service station; four different gasoline formulations were identified from analysis with the conclusion that four separate gasoline releases occurred<sup>130</sup></p> <p>Investigation of a subsurface plume of NAPL at a truck fueling facility; two separate sources of diesel contamination were determined from the distribution patterns of diesel-range alkane and aromatic hydrocarbons and from an evaluation of source and weathering diagnostic ratios<sup>131</sup></p> <p>Study of a large coastal area to assign sources to PAH contamination; sources were classified as petrogenic, pyrogenic, diagenetic, or biogenic<sup>42</sup></p> <p>Compilation of data from ambient air studies in Asia from 1997 to 2006, including sampling techniques, sampling instruments, sources, and average concentrations for dry deposition and related metallic elements<sup>132</sup></p> <p>Study of total gaseous mercury concentrations in indoor and outdoor air in residential locations in downtown Chongqing, China; results indicated higher concentrations at night and during the summer, but gaseous mercury concentrations were highly variable overall due to anthropogenic emissions, atmospheric changes, and unique events<sup>133</sup></p> <p>Review of methyl <i>tert</i>-butyl ether water monitoring data from drinking water supplies across the United States<sup>134</sup></p> <p>Characterization of street sediments in Kolkata, India; samples were taken from roadsides in residential, industrial, and inner city areas and from five sites along a highway; results of energy-dispersive X-ray fluorescence (EDXRF) analysis showed higher concentrations of copper, zinc, tin, lead, antimony, chromium, and nickel than from ambient or control soils; Kolkata metals concentrations were compared to reported analyses of street sediments from other major cities<sup>135</sup></p>

**Table 3. Recent Papers in the Field of Environmental Forensics**

Category	Description
Legal	<p>Advice for expert witnesses on presenting scientific findings in a Daubert hearing<sup>136</sup></p> <p>Calculating cost allocations to account for plume scenarios and petroleum contamination in soil and ground water<sup>137</sup></p>
Other	<p>Discussion on background values in environmental forensics, including references for general forensic techniques (historical document review, fingerprinting, statistical analysis) and specific forensic techniques (isotopic fingerprinting, geophysical method for evaluating metal backgrounds, dendroecology, and deoxyribonucleic acid [DNA] fingerprinting)<sup>138</sup></p> <p>Discussion of an error in the 1,1,1-trichloroethane (1,1,1-TCA) degradation pathway; reports the literature history with both correct and incorrect degradation pathways<sup>139</sup></p> <p>Study of natural spring water from the Eastern Black Sea region of Turkey of the activity concentrations of four radionuclides (<sup>226</sup>Ra, <sup>232</sup>Th, <sup>40</sup>K, and <sup>137</sup>Cs); concentrations of the elements in water were measured using inductively coupled plasma-optical emission spectroscopy<sup>140</sup></p> <p>Review of developments in standard reference materials (SRMs) for environmental forensics, including details on recertified and reissued standards and new standards for organic contaminants and biological and environmental matrices<sup>141</sup></p> <p>Discussion of the defective Chinese drywall used in thousands of homes in the United States that off-gas sulfur compounds; overview of the scientific and legal aspects of the problem<sup>45</sup></p> <p>Editorial on age-dating gasoline spills when insufficient or inadequate sampling has already occurred; includes table on various plume observations that can give an indication of the age of the plume<sup>142</sup></p> <p>Crime investigation of a landfill site containing hazardous waste from tannery and shoe factories located in Arroio do Meio, Rio Grande do Sul, Brazil; traditional crime scene examination conducted, along with a detailed description of the area; examination of possible pollutant sources; historical data gathering and sampling; and analysis of water, sediment, and animal tissues<sup>143</sup></p>

## ELECTRONIC WASTE

Electronic waste or “e-waste” has been the fastest-growing part of the solid waste stream since the mid-1990s.<sup>144</sup> In 2004, an estimated 100 million personal computers worldwide entered the waste stream for either recycling or disposal.<sup>145</sup> In 2005, more than 40 million computers were discarded in the US alone.<sup>146</sup> Governmental agencies, multinational committees, and environmental conservation groups internationally are on alert to find solutions to this immense waste problem.<sup>4, 5, 11, 147-153</sup>

Personal computers (including central processing units, monitors, keyboards and peripherals) are not the only type of waste electrical and electronic equipment (WEEE) of concern. Electrical and electronic equipment (EEE) that might be discarded into the waste stream includes large and small household appliances, cellular and regular telephones, portable digital assistants (PDAs), video and audio equipment (televisions, stereos, cameras, video recorders), electrical tools, and other computer equipment (scanners, printers, computer game units).<sup>9, 154, 155</sup>

Some discarded EEE is classified as used or UEEE for the purpose of reuse and/or recycling. WEEE or e-waste may be banned from importation into a country, but the same country may allow the importation of UEEE as part of a reuse or recycling operation. Li et al. listed the criteria to distinguish between WEEE and UEEE for 11 Asian countries, but the criteria are not uniform. The authors stated that the development of consistent standards could help restrict illegal transboundary movement and processing of e-waste.<sup>156</sup>

Trade shipments of UEEE frequently include some WEEE. The quantity of WEEE in the shipment of UEEE can be an amount negotiated between the exporter and the recycler/importer. As part of a deal for desirable UEEE, the recycler/importer agrees to deal with disposing of the useless WEEE. Or, the addition and amount of WEEE included in a shipment of UEEE can be an unfortunate surprise. Either way, the WEEE must be disposed of somehow in the importing country. Schmidt presented the findings of a Basel Action Network (BAN) investigation in Africa, with examples of the unregulated EEE market at work.<sup>157</sup> BAN representatives observed thousands of vendors in the Ikeja Computer Village, near Lagos, Nigeria, selling repaired and refurbished electronics such as computers, fax machines, and cell phones; these were products from the UEEE imports. They also observed piles of e-wastes filling swamps; these were the EEE junk items, the WEEE. When the piles of e-waste grew too high, the piles were burned, producing fumes that affected the local residents. BAN estimated that, of the purported UEEE shipped to Nigeria, between 25 to 75 percent was just waste or WEEE.<sup>157</sup>

Williams et al. presented the multiple facets to the reuse and recycling of computers in the global market and described some contrasting points of view about the e-waste issue. The authors used published research to make the argument that the hazards of leaching toxic materials out of landfills are minimal and that disposing of old computers in landfills could be environmentally preferable to recycling. In contrast, though, a lack of old, reusable computers would decrease the ability for poorer, developing markets to access information technology through low-priced, refurbished computers. Economically, the authors stated, the reuse and

recycling market provides employment, but unregulated recycling efforts have caused great harm to the environment and human health.<sup>158</sup>

Ogunseitan et al. described a major difficulty in dealing with the international e-waste problem in their 2009 policy brief. The difficulty is the worldwide patchwork of regulations and standards dealing with three parts of the e-waste problem: the movement of hazardous wastes, the use of hazardous substances, and the recycling responsibilities of the EEE producers. The authors explained that uneven implementation of standards had created “risk holes” in poor and developing communities. These are areas where markets trading in second-hand electronics and the recycling of illegally imported or domestic e-waste thrive.<sup>144</sup> McKenna expressed concerns about illegal e-waste in developing areas and discussed the application of the Basel Convention and WEEE Directive to the e-waste problem.<sup>159</sup> The Basel Convention set into action a multi-national agreement on the control of transboundary movements and disposal of hazardous wastes.<sup>6</sup> The WEEE Directive of the European Parliament (Directive 2002/96/EC) set an objective to reduce the quantity of waste electrical and electronic equipment and the harm to the environment.<sup>9</sup> Together, these European initiatives have the potential to decrease illegal e-waste.

## **REGULATORY AND LEGISLATIVE CONSIDERATIONS**

Ladou and Lovegrove briefly outlined the e-waste problem and presented the regulations and initiatives that might assist countries in dealing with e-wastes. The Basel Convention was described first, followed by the European Restriction on Hazardous Substances (RoHS) and the WEEE legislation. The authors mentioned the US EPA RCRA regulations but indicated that, even with the openness of the US EPA to consider improvements to these regulations, many impediments exist to actually revising the regulations. It was noted that several US states have passed or developed regulations similar to the RoHS legislation of Europe. Regulations developed in Asia such as the Home Appliance Recycling Law (HARL, Japan) and the Law for Promotion of Effective Utilization of Resources (LPEUR, Japan) have promoted the recycling of millions of appliances per year. The Environmental Protection Agency of Taiwan assigned the responsibility of recycling waste personal computers to producers in 1997. The authors added that several Asian countries are actively working on their own e-waste recycling or RoHS-type regulations.<sup>160</sup>

The US Government Accountability Office produced a report in 2008 on the weakness of the US regulations to address e-waste exports. The report explained that cathode ray tubes (CRT) are the only electronic device regulated by the US EPA. In order for CRTs to be exported for reuse, the US EPA must receive a notification of the intent to export CRTs and approval from the importing country. The report gave examples of the need for increased enforcement of the RCRA regulations limiting CRT exports, and it recommended expanding the scope of the CRT rule to include other e-waste types.<sup>13-15</sup>

Kutz provided a quick review of the e-waste problem, including a section on examples of what initiatives are being taken to correct the problem. The author described the efforts of businesses, the restrictions on hazardous chemicals as directed by the EU, and the possibilities of biodegradable and eco-friendly (non-plastic) case designs. Programs in Japan, Canada, and Taiwan were listed as examples that the US could observe in order to design a program that

could best address e-waste issues. Two programs were emphasized as potential e-waste solutions: advance recovery fees (ARFs) and extended producer responsibility (EPR). With ARFs, consumers pay a recycling or recovery fee for the electronic item at the time the item is purchased with the purpose of collecting enough funds to support e-recycling programs. EPR programs can place responsibilities on manufacturers (or producers) with the purpose of having the manufacturer cover the costs of managing the waste at the end of the product life. Kutz recommended an EPR program for the US and provided guidance for structuring that system.<sup>161</sup>

EPR and WEEE management initiatives were discussed by Nnorom and Osibanjo. The authors listed examples of the administrative, economic, and informative policy instruments that could be used to implement EPR. Administrative instruments included implementing product take-back programs, setting emission limits, and setting reuse and recycling targets. Economic approaches included advance disposal fees and deposit-refund systems. Environmental reports and labeling were two of the informative instruments listed. WEEE management initiatives in the EU, Finland, Japan, Switzerland, and Taiwan were also described.<sup>162</sup>

Krishna and Kulshrestha discussed the problem of e-waste in developing nations and focused their attention on Indian and Chinese directives. The authors mentioned the Basel Convention and the EU WEEE and RoHS Directives and also described the Bamako Convention. The Bamako Convention was adopted by 51 African countries and banned the import and trafficking of hazardous waste into Africa.<sup>163</sup>

E-waste initiatives for China, Nigeria, and India were the topics of several other papers. Ye et al. discussed the elements of the current legislative framework for e-waste management in China and analyzed the challenges that still exist.<sup>164</sup> Ni and Zeng presented a set of recommendations to address the e-waste crisis that included enhancing law enforcement, establishing national or local e-waste disposal centers, and coordinating with other countries on e-waste.<sup>164</sup> Umesi and Onyia suggested solutions for e-waste problems in Nigeria and lamented the fact that the Basel Convention provisions are not being enforced in originating countries. The authors recommended establishing adequate material collection systems and additional systems for the proper reuse, refurbishment, recovery, and recycling of materials; this plan would create employment and provide environmentally sound management.<sup>165</sup> Nnorom and Osibanjo reviewed the flow of secondhand and scrap electronic devices into Nigeria and noted that much of the growth in the information and communications technology in Nigeria had come from secondhand electronics. Management practices for e-waste were discussed, and the recommendation was made for formal recycling facilities and the implementation of a plan to check secondhand EEE for functionality before importing the devices.<sup>165</sup> Bandyopadhyay reviewed the e-waste management initiatives in India and included tables on the economics of the e-waste trade, estimates of e-waste in Mumbai, and the Indian WEEE policy framework.<sup>166</sup> Dwivedy and Mittal estimated current and future quantities of e-waste generation in India. Estimates of e-waste quantities could help recyclers make decisions in planning recycling infrastructure and capacity building. Total WEEE estimates for 2007-2011 were given at approximately 2.5 million metric tons, about 30 percent of which would be from personal computers.<sup>167</sup>

## CONTAMINATION AND HEALTH EFFECTS

Materials used to manufacture electrical and electronic equipment include both valuable and hazardous components. Unregulated recovery of the valuable materials, such as copper wire or gold from UEEE or WEEE, can lead to situations in which very hazardous components are released, which may endanger human health and the environment. In his recent review on e-waste, Robinson made the point that the chemical composition of e-waste varied by the age and the type of the discarded item. The author stated that the composition continually changes to meet the demands of technological developments, the regulatory requirements of environmental agencies, and the requests of environmental organizations.<sup>168</sup> A table of environmental contaminants found in e-waste was provided in Robinson's review, and the health effects of many of these contaminants were listed by Schmidt.<sup>169</sup>

Basel Action Network representatives visited a city in China that was, and still is, a center for e-waste metals recovery.<sup>169</sup> Residents of Guiyu, according to BAN, had been receiving e-waste to process since 1995. The processes used for recovering metals included no protection for human health or the environment. Residents heated circuit boards in woks to melt lead solder to remove computer chips; the molten lead and burning plastic created toxic fumes. The molten lead was then poured on the ground, contaminating the soil and nearby river area. Pits filled with "aqua regia," a strong combination of hydrochloric and nitric acid that can produce smoke and fumes, were used to dissolve the computer chips. Used acid and sludge were dumped into the local river.<sup>169</sup>

Guiyu was visited in 2008 by BAN and a crew from the CBS network's primetime investigative show, 60 Minutes. The show obtained footage of smoldering piles of e-waste; residents heating circuit boards in the open air and pooling molten lead; residents using a large acid bath for gold recovery; and views of the smoky air and the dark, polluted river water.<sup>170-172</sup>

Fresh water samples from inside and outside of Guiyu were analyzed for metals by Wong et al. High metal concentrations attributable to the acid leaching of e-wastes (silver, cadmium, copper, and nickel) were found. Lead isotope composition from river samples was studied and indicated that lead from multiple sources was present in the two local rivers.<sup>19</sup> Air, soil, and sediment samples from locations in Guiyu were analyzed for persistent organic pollutants (flame retardants, dioxins and furans, polycyclic aromatic hydrocarbons, and polychlorinated biphenyls) and heavy metals in another study. Open combustion of e-waste and the dumping of materials at the end of the recycling process were suspected as major sources of toxic chemicals in the local environment. As expected, results from air analyses indicated high levels of the persistent organic pollutants and heavy metals when compared to samples from other cities. Soil and sediment analysis results also indicated extremely high levels of pollution.<sup>173</sup>

Lead and cadmium levels in the blood of the children living in Guiyu were the focus of work by Zheng et al. The authors sampled blood from children in Guiyu and from children in nearby Chendian, a town with no e-waste processing but with clothing manufacturing as the primary industry. Analyses found that children in Guiyu had higher levels of lead and cadmium in the blood when compared to the control children of Chendian.<sup>174</sup> Xing et al. tested for PCB levels in local river fish, air, and human milk samples in Guiyu. Results indicated that inhalation

exposure was a more significant contributor to PCB levels in humans than dietary exposure (fish).<sup>20</sup>

Analyses were performed on soil samples taken in e-waste recycling areas in the province of Taizhou, China. Jun-hui and Hang sampled the rice paddy soils from seven different sites, all with past or present e-waste recycling operations. The authors found soil samples from one of the active e-waste recycling sites to be high in cadmium.<sup>16</sup> Tang et al. analyzed soil samples taken from Wenling, an emerging e-waste recycling city in Taizhou. The samples were analyzed for heavy metals (copper, chromium, cadmium, lead, zinc, mercury, and arsenic) and persistent organic compounds, including polycyclic aromatic hydrocarbons and polychlorinated biphenyls. Results indicated that most heavy metals exceeded the Grade II value of soil quality standards set by the State Environmental Protection Administration of China. PCB levels ranged from 2 to more than 200 times higher in the e-waste area soil samples than in samples taken from reference sites. PAH concentrations were higher in the samples taken near small, household workshops. Overall, the authors found the small, household e-waste recycling workshops to be greater contributors to local soil contamination than large-scale plants.<sup>18</sup>

Samples from an e-waste recycling facility, agricultural land, and a chemical-industrial complex in Taizhou were analyzed for polybrominated dibenzo-*p*-dioxins and dibenzofurans (PBDD/Fs) by Ma et al. The matrices included workshop-floor dust, electronic shredder residues, leaves from trees and shrubs, and surface soils. Estimated daily human intakes of PBDD/Fs were calculated.<sup>175</sup> Chen et al. sampled surface sediment of the Nanguan River running through Taizhou. The authors determined the concentrations of PCBs, PAHs, and heavy metals in the samples. Toxicity studies were also performed. The authors noted that pollutant and toxicity levels near industrial recycling parks were much lower than areas near household workshops. This was attributed to the management and recycling technologies at use in the industrial parks.<sup>176</sup>

Wang analyzed scalp hair samples to determine heavy metal exposure to residents in Taizhou and used samples from two control sites for comparison. Lead, copper, manganese, and cadmium were significantly higher in concentration in the samples from Taizhou than in the control samples.<sup>177</sup>

Contamination by e-waste recycling in Bangalore, India, was studied by Ha et al. The authors sampled soil, airborne particulates, and human hair from e-waste recycling sites and reference sites in Bangalore and Chennai and analyzed the samples for trace elements. Concentrations were reported for 16 elements (vanadium, chromium, manganese, cobalt, copper, zinc, molybdenum, silver, cadmium, indium, tin, antimony, mercury, thallium, lead, and bismuth). In each results table, the analytical data from this study were compared to metals analysis data from other published studies. The authors concluded that a link could be made to metal contamination and exposure from e-waste recycling. But, the authors conceded, since not all sample types were taken from every site and no lifestyle considerations were factored into the hair analysis, this study was only a preliminary start for further investigations.<sup>17</sup>

## ANALYTICAL METHODS

Testing for the hazards posed by e-waste in the US has centered on the Toxicity Characteristic Leaching Procedure in the US EPA Test Methods for Evaluating Solid Waste, Physical/Chemical Methods (SW-846).<sup>178</sup> This test attempts to mimic conditions for waste items in a landfill. Liquid in a landfill may contact waste and may cause the leaching of metals or organic chemicals from that waste into the liquid. A breach in the landfill structure could allow this liquid to exit and contact soils, surface waters, or ground water. If one or more of 40 contaminants exceed toxicity limits, then the waste may be considered to be hazardous and issues of waste treatment and land disposal restrictions may need to be addressed.<sup>179</sup> Questions about the metals that might leach from e-waste disposed in landfills have led to several studies using computer parts and the TCLP method.

The primary researcher and research center for e-waste and TCLP during the past 10 years has been Townsend at the University of Florida in Gainesville.<sup>169</sup> The work published by his research group has shown that lead is the main concern for toxic, leachable metals in e-waste. Publications by the group are listed and described in Table 4. Townsend and his group also produced two reports, one for the Florida Center for Solid and Hazardous Waste Management on lead leachability from CRTs using the TCLP method, and another for the US EPA in 2004 on RCRA toxicity characterization of computer central processing units (CPU) and additional electronic devices.<sup>180, 181</sup>

Lincoln et al. tested cellular telephones for metals and organic compounds using three methods: the US EPA TCLP, the California Department of Toxic Substance Control (DTSC) Waste Extraction Test (WET) and California DTSC's Total Threshold Limit Concentrations (TTLC). Phones were grouped by physical dimensions; phones outside a specific size range were eliminated. Phones were shredded, homogenized, then divided equally by mass. The authors determined that the phones exceeded TCLP limits for lead and failed the standards of the TTLC for five metals (copper, nickel, lead, antimony, and zinc). No limits were exceeded in the WET test.<sup>22</sup>

Keith et al. used four different leachability tests on a selection of computer CRTs, printed circuit boards, computer mice, television (TV) remote controls, and mobile phones. Each of the six devices were crushed and then tested using the US EPA TCLP, the US EPA Synthetic Precipitation Leaching Procedure (SPLP, Method 1312), the Dutch Environmental Agency Availability Test (EA NEN 7371), and method DIN S4 of the Institut für Normung, Germany. The extracts were analyzed for lead, cadmium, chromium, and silver. TCLP extractions were repeated on composite CRT glass (one additional extraction) and filter papers with residual solids (two additional extractions), and then those extracts were filtered and analyzed. Results indicated increased lead leachability with smaller CRT and circuit board particle sizes and extracted lead increased with the repeated TCLP extractions. The SPLP, NEN 7371, and DIN S4 tests yielded very low results for the leachable metals, and the authors did not recommend those tests for assessing potential metal leachability from e-waste.<sup>21</sup>

Researchers are actively seeking ways to detect unwanted chemicals in EEE fabrication, ways to better deconstruct EEE for recovery or reuse of materials, and ways to destroy or isolate



the usable and disposable WEEE. This requires a thorough understanding of the chemical composition of the devices, and most researchers have provided the chemical analysis process details and data in their published work. Lee and Wright developed a standard operating procedure for assessing and confirming compliance with regulations on the use of ozone-depleting chemicals.<sup>23</sup> Cui and Forssberg used a variety of methods to characterize television scrap and determine cost-effective ways to separate the components of a shredded TV mix for potential material recovery.<sup>182</sup> Lee et al. presented a good overview of the recycling process for scrap computers, including tables on compositions of components, separation methods, and CRT coating-removal methods.<sup>183</sup> Méar et al. characterized the waste funnel and panel glass from dismantled CRTs in order to develop reuse applications.<sup>184</sup> Chen et al. tested a pyrovacuum process that removes and recovers lead from the funnel glass of CRTs and makes a porous glass residue.<sup>185</sup> Guo et al. reviewed the recycling methods for the non-metallic fractions of printed circuit boards.<sup>186</sup> Hino et al. developed a method for pulverizing waste printed circuit boards with integrated circuits.<sup>187</sup> Niu and Li studied two methods for making printed wire boards non-hazardous: high-pressure compaction and cement solidification. The low-impact resistance of the compacted samples made them too unstable to be a long-term disposal solution. The results from the TCLP tests on the highly impact-resistant, solidified samples showed lead at concentrations well below the regulatory limits; a good indication that this method could render the boards non-hazardous.<sup>188</sup>

**Table 4. Publications from the Townsend Research Group, University of Florida, Gainesville**

Title, Author (s), Year	Description of Research and Results
<p><b>Characterization of Lead Leachability from Cathode Ray Tubes Using the Toxicity Characteristic Leaching Procedure</b><sup>189</sup> SE Musson, Y-C Jang, TG Townsend, and I-H Chung 2000</p>	<p>Used TCLP procedure on 36 CRTs from television and computer monitors; found that most color monitors (21 of 30) exceeded regulatory limits for lead, but no monochrome monitors exceeded lead limits; the frit seal in color CRTs was a major source of lead</p>
<p><b>Leaching of Lead from Computer Printed Wire Boards and Cathode Ray Tubes by Municipal Solid Waste Landfill Leachates</b><sup>190</sup> Y-C Jang and TG Townsend 2003</p>	<p>Varied the leachates used to test printed wire boards and CRTs from television and computer monitors; test methods and leachates used were the standard TCLP method; leachates that were sampled from 11 Florida landfills, California’s Waste Extraction Test, and the US EPA Synthetic Precipitation Leaching Procedure; leaching test results indicated that lead concentrations were highest with the TCLP test, followed by WET, then leaching with the landfill leachates, and, lastly, the SPLP test</p>
<p><b>RCRA Toxicity Characterization of Discarded Electronic Devices</b><sup>191</sup> SE Musson, KN Vann, Y-C Jang, S Mutha, A Jordan, B Pearson, and TG Townsend 2006</p>	<p>Thirteen electronic devices (printers, cell phones, laptops, flat-panel displays, computers peripherals, etc.) were tested using a standard TCLP test and/or a modified TCLP test; tests were modified to be either large-scale or small-scale; all toxicity characteristic metals were below regulatory limits except lead; a correlation was observed between the amounts of ferrous metal in the device and lead concentrations in the leachate</p>
<p><b>Factors Affecting TCLP Lead Leachability from Computer CPUs</b><sup>192</sup> KN Vann, SE Musson, and TG Townsend 2006</p>	<p>Performed TCLP particle reduction on computer central processing units, then leached different combinations of the CPU components; high lead concentrations were observed when leaching printed wire boards; again observed the correlation between decreased lead concentrations when ferrous metal was included and discussed the connection of iron and zinc concentrations and the hydrous ferric oxide interaction as previously reported by Kendall;<sup>193</sup> also noted increased lead concentration with increased head space and that additional particle size reduction below the size required for TCLP did not increase the lead concentration</p>
<p><b>Evaluation of a Modified TCLP Methodology for RCRA Toxicity Characterization of Computer CPUs</b><sup>194</sup> K Vann, S Musson, and T Townsend 2006</p>	<p>Large-size leaching modification to TCLP test to allow disassembled CPUs to be leached almost whole in a large vessel in order to be more representative of the device and decrease sample preparation time; observed higher lead concentrations for the large-scale leaching when compared to standard TCLP conditions; reported that the large-scale leaching did reduce sample processing time (sample particle reduction), but increased the cost of analysis</p>
<p><b>Impact of Electronic Waste Disposal on Lead Concentrations in Landfill Leachate</b><sup>195</sup> E Spalvins, B Dubey, and T Townsend 2008</p>	<p>Created simulated landfills in large columns using a manufactured municipal solid waste (MSW) in five columns and excavated waste and soil in the sixth column; identical sets of e-waste were added to three of the manufactured MSW columns, and dechlorinated tap water was added to all columns on nearly 30 occasions; analysis of extracts from the columns indicated lead concentrations between columns with e-waste were not statistically significant when compared with lead concentrations in the control columns</p>

# BIOLOGICAL WASTE

## CONCENTRATED ANIMAL FEEDING OPERATIONS (CAFOS)

Animal Feeding Operations (AFOs) are defined as agricultural operations where animals are kept and raised in a confined area.<sup>196</sup> Animals live and grow in a production setting that contains feed, excrement, and sometimes dead animals, all in a limited space. The US EPA has defined AFOs by animal confinement times and the lack of natural vegetation in the confined area. Concentrated Animal Feeding Operations (CAFOs) must meet the definition of an AFO, and, in addition, CAFOs must meet one of the specific regulatory definitions for small, medium, or large CAFOs. CAFOs may be regulated by the US EPA under the National Pollutant Discharge Elimination System (NPDES) which is a part of the Clean Water Act.<sup>196-198</sup>

### Environmental Impacts

CAFOs are a concern in the environment due to the large amounts of waste produced from the confined animal productions. Potential problems include leaking, leaching, or runoff from manure stockpiles or storage pits, which can contaminate ground water and fresh water sources.<sup>199,200</sup> Water contamination is not the sole concern; emissions of gases and particulate matter from the production facilities may cause harm to human health.

Water Environment Research publishes several annual reviews on the types of pollutants that can contaminate surface or ground water. Several review papers address the contaminants that can originate from agricultural waste and the detection of chemicals or pathogens in the environment.<sup>201-209</sup>

Lee et al. prepared an in-depth review of the effect of agricultural antimicrobials and hormones on soil and surface and ground waters. The authors covered the use and occurrence, sorption, degradation, transport processes, and ecological and health effects of the chemicals. Analytical methods and suggestions for future research were also included. The authors stated that more studies were needed to make conclusive connections between animal-derived antibiotic-resistant pathogens and human health impacts.<sup>210</sup>

Dolliver and Gupta addressed the potential for manure runoff to be a source of emerging contaminants to the environment. The authors quantified three commonly used beef antibiotics, chlortetracycline, monensin, and tylosin, in runoff from beef manure stockpiles. Due to concerns from the spread of antibiotic resistance in the environment, they suggested covered facilities to decrease runoff. The authors also compared the antibiotic runoff concentrations to the antibiotic concentrations reported in studies about composting and manure-applied fields; the latter concentrations were generally found to be much less than those found in runoff losses.<sup>199</sup>

Sapkota et al. sampled surface and ground water up-gradient and down-gradient of a swine facility to determine if antibiotic-resistant enterococci and other fecal indicators were present. The results indicated higher concentrations of fecal bacteria in the down gradient surface and ground water samples. The down-gradient surface water concentrations were consistently in excess of the US EPA bacterial water quality for recreational standards.<sup>200</sup>

Seepage of animal wastes stored and treated in anaerobic lagoons can lead to an increase of nitrate concentrations in the ground water. Excess nitrates in rivers can lead to areas of hypoxia, or oxygen deficiency, which can dramatically affect the plant and fish life. Mariappan et al. used nitrogen isotope ratios in their study to identify sources of lagoon seepage. <sup>15</sup>N isotope enrichment was dependent on the ambient temperatures and the rates of mixing or addition of fresh wastes.<sup>211</sup>

Burkholder et al. reviewed the possible contaminants in CAFO waste and the potential impacts to water and wildlife.<sup>212</sup> The authors asserted that current livestock waste management practices are inadequate to protect water resources. This review was prepared as part of the mini-monograph, “Environmental Health Impacts of Concentrated Animal Feeding Operations: Anticipating Hazards—Searching for Solutions,” which resulted from a 2004 scientific conference and workshop addressing CAFO environmental health issues.<sup>213</sup>

Emissions from CAFOs can be more than just malodorous and can include ammonia, hydrogen sulfide, and particulate matter contaminated with endotoxins. Endotoxins are inflammatory agents produced by bacteria that can either aid in the development of immunity or cause harm by inducing asthmatic reactions. Heederik et al. reviewed the human health effects from CAFO emissions as part of the CAFO mini-monograph mentioned above.<sup>214</sup>

Wash and runoff water from CAFOs can be used beneficially as a fertilizer and soil amendment when an acceptable Nutrient Management Plan (NMP) is followed.<sup>215, 216</sup> An additional advantage is the reduction in the need for fresh water irrigation in fields when the CAFO water is applied.

Bradford et al. provided a thorough review on the use of CAFO wastewater on agricultural lands. The review was divided into three sections: environmental contaminants, land application, and treatments. Six classes of potential waste lagoon contaminants were addressed in the environmental contaminant section: nutrients and organics, heavy metals, salts, pathogens, antibiotics, and hormones. For each of these classes, the authors described the potential environmental problems. For example, the authors stated that some waste lagoon water exceeded the recommended copper and zinc concentrations for irrigation water. The potential negative effects from an excess of these metals could be phytotoxicity and surface and ground water contamination. The land application section of the review discussed the US EPA requirement for NMPs that are designed to consider all nutrient input sources, nutrient losses, and nutrient uptake rates for the plants.<sup>215</sup> The authors explained the basic assumption of the NMP, that all CAFO contaminants would be absorbed or degraded in the root zone and no contaminants would enter the surface or ground water. Possible weaknesses to NMP designs and implementation also were discussed in the paper.<sup>216</sup>

### **Analytical Methods for Emissions**

Bunton et al. reviewed methods for ammonia and hydrogen sulfide monitoring. The review contained information on the type of equipment and analyzers that have been used in various studies. The authors provided cautionary summaries regarding odor measurements and

the monitoring of particulate matter and described potential problems with each. For odor measurements, a chemical characterization can be performed and constituents quantified, but the result may be a complex mixture of compounds and the correlation of the compounds to human scent responses can be poor. Also, taking only air samples in the field may leave a study incomplete because some odorous material may attach to particulate matter that could be filtered out, depending on the sampling method. For particulate matter emitted from CAFOs, instrumentation does exist for sampling and measurement, but the authors pointed out that dust in the rural areas around CAFOs has the potential to interfere with those results. The authors also included sections on plume dispersion models and recommendations for future studies.<sup>217</sup>

Ni and Heber provided an extensive review of ammonia sampling and measurement techniques for animal facilities in volume 98 of the *Advances in Agronomy* series. The review included sampling methods categorized into closed, point, and open-path methods and advice for selecting a method. Measurement techniques and devices were described in brief and were well-referenced. A comparison of ammonia measurement devices and summaries from several comparative studies were provided at the end of the measurement section. An important addition to the review was the detailed table of potential sources of error in the sampling or measurement of ammonia concentrations. The authors categorized the errors (calibration, sampling, measurement, and data processing) and provided suggestions to reduce or eliminate each source of error.<sup>218</sup>

Todd et al. used acid gas washing samplers and a calibrated flow injection analyzer to obtain ammonia concentrations from sampling sites at a beef cattle feedyard. Continuous ammonia measurements were taken during a summer period using a chemiluminescence analyzer. Measurements for wind velocity and direction, air temperature and humidity, and precipitation were also collected to provide further data for modeling ammonia emission rates in spring, summer, and winter months.<sup>219</sup>

Trabue et al. measured volatile sulfur compounds at swine and poultry feeding operations. They employed a canister-based method and used fused silica-lined mini-canisters to collect the air samples at lagoon and building locations. The sampled air was focused into a gas chromatograph system with both mass spectrometer and pulsed flame photometric detectors (PFPD). Target compounds for the analysis were hydrogen sulfide, carbonyl sulfide, methanethiol, dimethyl sulfide, carbon disulfide, dimethyl disulfide, and dimethyl trisulfide. Storage stability for samples was a problem especially when sampled from warm, humid environments.<sup>220</sup>

## **Methods for Tracking Waste Releases**

Several research groups have published their efforts to find one or more optimal methods to trace the source of an overflow or leak from a CAFO.<sup>28, 199, 200, 211</sup> Causes of leaks or overflows can be obvious, for example, from a weather related event (heavy rains filling an uncovered lagoon) or a structural or engineering fault (a lagoon wall failure). Observed lagoon problems like these likely would not require tracking methods to find the source of waste contamination and encourage a CAFO operator to improve the facilities to avoid future contamination events. But, when fecal environmental contamination occurs and several potential

sources of fecal waste exist, using source identification tools may be the solution to track down the cause of contamination.

Environmental contamination from fecal waste is not limited to AFOs or CAFOs. When urban areas are involved, fecal material entering the environment may come from humans (leaks in sanitary sewers or sewer connections to storm drains), domestic animals (including pets), and urban wildlife (birds, raccoons, rats, etc.). Mallin et al. listed these contributors to waste from urban areas and studied the impacts from stormwater runoff in urban, suburban, and rural areas. The authors noted that, in some rural areas, unsuitable septic systems, pastureland, and farmland receiving applications of manure as fertilizer can contribute to fecal material found in waterways.<sup>26</sup>

Chemicals such as artificial sweeteners, pharmaceuticals, detergents, fragrances, and caffeine can be used to trace discharges into a waste stream.<sup>30, 221</sup> Buerge et al. in Switzerland sampled untreated and treated wastewater, ground water, and tap water and tested for the presence of four sweeteners, acesulfame, cyclamate, saccharin, and sucralose. The authors observed that acesulfame was prevalent in all waters and was not removed through wastewater treatment, but cyclamate and saccharin were removed. Buerge's team concluded that assumptions could be made about the source of a leak from those sweeteners. Assuming that some contamination has occurred, ground water samples without saccharin and cyclamate could point to a treated wastewater leak and ground water samples with those two sweeteners could indicate a leak from untreated wastewater.<sup>221</sup>

Using specific chemicals to locate the source of a release of fecal waste into water resources is one of the tracking methods discussed in the literature and, in the paper by Cimenti et al., it is listed as “chemical microbial source tracking” or chemical MST. Microbial source tracking encompasses many different methods that can be used alone for a single-source investigation or in combination for scenarios in which multiple sources of contamination exist. The large volume of research and developments in MST is complex, but some excellent papers exist to help the investigator decide the most efficient and informative tracking approach.<sup>30</sup>

## **MICROBIAL SOURCE TRACKING**

The goal of microbial source tracking is to determine the source of a fecal contamination event. To trace back to the origin, it is necessary to determine what species produced the fecal material. Is the fecal material of human or animal origin? If animal, can indicators point to the type of animal (agricultural animals, domestic animals, urban wildlife)?

Contamination can also be from multiple sources. More than one method of tracking may be needed to locate those sources and the pathways used to spread the fecal waste in the environment. Considering the ecology of an area can be an essential tool in understanding the spread of contamination.<sup>25</sup>

Cimenti et al. provided an excellent overview of microbial source tracking in their concise, informative review paper. The authors acknowledged several fundamental papers in

microbial source tracking in their text.<sup>222-225</sup> In the paper, MST methods were sorted into five classifications: direct monitoring of pathogens, culturing methods, phenotypic methods, genetic methods, and chemical methods. The classifications and several examples of each were listed in a summary table and later described in detail with methods and references to published work. The authors suggested that direct monitoring of pathogens, while effective for determining health risks, may not be the most efficient or affordable. More than one pathogen may exist in water affected by fecal contamination, and detection of pathogens can be complex and costly.<sup>30</sup>

The other four method classes (culturing methods, phenotypic methods, genetic methods, and chemical methods) track fecal indicators, which, if chosen well, should only be present whenever fecal contamination is present; the indicators should not be a normal part of the environment. An ideal indicator should also be effective for tracking in all types of waters (surface, ground, and marine waters). These indicator characteristics and a few others (fast analysis, cost-effective, etc.) were listed in the Cimenti paper and credited to a book by Maier et al.<sup>226</sup> Gerba contributed the chapter on indicator microorganisms, and in it he listed the criteria for an ideal indicator organism.<sup>227</sup>

Detection of enteroviruses falls into the category of direct monitoring of pathogens. Enteroviruses are very host-specific, and detection of these confirms fecal contamination. But, the authors pointed out, the lack of enteroviruses in a sample does not exclude fecal contamination because water polluted by feces does not always have enteroviruses present.<sup>30</sup> Rajtar et al. detailed the danger of enteroviruses in the aquatic environment and listed some methods of detection.<sup>228</sup>

Culturing methods involve isolating a microorganism that is specific to a potential source of the fecal pollution (humans, agricultural animals, domestic animals, urban wildlife). These microorganisms can include fecal coliforms, fecal streptococci, protozoa, or viruses. The authors cautioned that culturing media is rarely selective and that few organisms are host-specific and have “ideal indicator” criteria as presented in Maier et al.<sup>30, 226, 227</sup>

Phenotypic methods look at bacterial species that are present in both humans and animals, but differentiate the bacterium based on how it changed while existing within a specific host. Different phenotypic characteristics develop under different intestinal conditions. The authors stated that biochemical responses to these methods could be too similar to distinguish a difference, but, if multiple phenotypic characteristics are detected, the accuracy of the method could be improved. Antibiotic resistance analysis is one phenotypic method. Bacteria are isolated, cultured, and measured for their responses to several antibiotics.<sup>30</sup>

The preferred MST methods are the genetic methods. These use intestinal bacteria to discern sources of fecal pollution by either making a host-specific match for DNA or identifying a genetic characteristic that indicates an adaptation to a specific host. The drawback to the genetic method is that it requires databases or libraries to match the genetic profiles, and these profiles can vary by location and time. The authors provided brief descriptions of a few genetic methods, but point to other papers for more information.<sup>30</sup>

Cimenti et al. also listed several possible chemical tracers that can be either directly associated with fecal material or can be discharged in the same wastewaters as fecal material. These chemical methods, the authors pointed out, do not easily correlate to pathogens, but can still help in locating a source of pollution. Using information about emerging contaminants in water, the authors provided lists of veterinary and human antibiotics, veterinary medicines, prescription and non-prescription drugs, and hormones as possible chemical tracers.<sup>30</sup>

Field and Samadpour published a review of source tracking methods and included details on the applicability of certain methods, the usefulness of the methods, and the drawbacks of the methods. They expressed concerns about public health and the occurrence of pathogens in water and suggested a combination of targeted pathogen monitoring with targeted fecal source tracking to better regulate water quality.<sup>229</sup> Zhang et al. reviewed antibiotic resistance genes and provided detailed tables, categorized by antibiotic, that list the genes, the biological and environmental sources, and references of papers with studies on each specific gene.<sup>230</sup>

Stoeckel and Hardwood prepared a review of MST that addressed the strategies for developing, validating, and evaluating a microbial source tracking study. The authors compared method performance data, including statistics, and outlined considerations for the field study design.<sup>231</sup> Santo Domingo et al. reviewed MST methods and listed the research gaps and suggested some goals for future studies.<sup>27</sup>

Witty et al. addressed the complexity of determining a single source of pollution from multiple potential and contributing sources. The authors used multiple methods but concluded that DNA sequence analysis for MST in combination with a study of the ecosystem is a better approach than microbiological applications alone.<sup>25</sup> Reischer et al. emphasized the need to consider hydrological catchment dynamics in source tracking study design and gathered data for their study over a 31-month period of time.<sup>232</sup> Stapleton et al. performed a catchment scale study and found that MST techniques needed further development in order to become a useful tool in catchment systems.<sup>233</sup> Vogel et al. studied water and sediment samples in a catchment reach for fecal contamination. The authors found *Escherichia coli* in both water and sediment, while the *Bacteroidales* markers were detected more frequently in the sediment samples.<sup>234</sup>

Wolf et al. created a library-independent “viral toolbox” (VTB) to distinguish between human and animal fecal pollution. Their VTB used real-time reverse transcription (RT)-polymerase chain reaction (PCR) assays for norovirus genogroups, adenovirus species, and F+ RNA bacteriophage genogroups. The authors found their assays to be highly sensitive and specific to the targets.<sup>29</sup>

Microbial source tracking papers are filled with PCR assays. Agudelo et al. used a multiplex real-time PCR assay for fast quantification and obtained higher detection levels than with conventional microbiological techniques.<sup>235</sup> Kortbaoui et al. used dot-blotting membranes containing specific oligonucleotides for human and for four animals (bovine, porcine, ovine, and chicken) to detect mitochondrial DNA in water samples. The dot-blot assays were found to be as specific and sensitive as a nested-PCR approach.<sup>28</sup> Lamendella et al. evaluated two swine-specific PCR assays. The *Prevotella* strain-based PCR assay provided positive results more



often than the methanogen-based PCR assay, and it also had a high number of cross-reactions with non-target fecal DNA samples.<sup>236</sup>

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