GLOBAL AND SITE SPECIFIC

MULTIMEDIA (FIELD) STUDIES

by

.Norman H. Cutshall and Michael R. Guerin
Environmental Sciences Division
Analytical Chemistry Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee 37831

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INTRODUCTION

Environmental pollution requires control to protect: 1)
human health and safety, 2) the safety of non-human populations,
3) other resource uses of the environment. Although pollutants
are typically released into only one medium--soil, air or water--
natural processes act upon all materials to disperse and
distribute them among multiple media.

Evidence of cross-media pollutant transfer and of multimedia
exposure is often found in field studies. Examples of current
note include the release of airborne dioxins and dibenzofurans
upon solid waste incineration (Karasek and Hutzinger, 1986;
Ozvacic et al., 1985), groundwater contamination from landfills,
and air contamination from sewage treatment plants (Haemisegger
et al., 1985). Evidence of the importance of multimedia
processes in the regional and global distribution of pollutants
frequently results from basic research efforts. For example,
Czuczwa and Hites (1986) found that dioxins and dibenzofurans in
Great Lakes sediments are, in part, products of solid waste
incineration.

To what degree must protection from pollution consider
multimedia factors? Is it sufficient to control releases into
each medium separately or is it necessary to adopt a holistic
approach that includes environmental transport, transformation
and transfer?

In consideration of these issues, we will review recent test
programs in multimedia exposure and in cross-media transfers. We
will also discuss experiences from both global and small-scale
field data that indicate the role of multimedia factors in
pollutant exposures.

RECENT MULTIMEDIA PROGRAMS

Integrated Environmental Management Program (IEMP)

The USEPA Integrated Environmental Management Division has
completed a study (Hinman et al., 1986; Haemisegger et al., 1985)
of exposure and health risk for the Philadelphia metropolitan

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area. Similar studies are underway considering the Santa Clara
Valley in California and the Baltimore, Maryland area. The IEMP,
which was developed to test alternative approaches to
environmental management, emphasized the importance of cross-
media transfers and of multimedia effects.

The IEMP was unique in several respects. First, it was
based on examining a geographic area as a unit rather than on
examining a specific pollutant or a specific source of
pollutants. Second, it sought to consider exposures from all
media simultaneously rather than focusing on a single route of
exposure. The Philadelphia study, (Haemisegger et al., 1986) the
first to be initiated, considered primarily surface water and
ambient air. The Santa Clara Valley study (Hinman et al., 1986)
included a thorough examination of groundwater in addition to
ambient air and surface water. Finally, the IEMP was unique in
that it employed human health risk as the common denominator for
ranking sources and pollutants and identifying those most in need
of control technology implementation.

The general approach involved definition of a unit
geographic area, collection of data on the nature and quantities
of pollutants in each media considered, estimation of exposure
levels, and combination of exposure data with pollutant
biological potency data to estimate health risk. Exposure
estimates were derived using relatively simple EPA transport and
fate models and potency estimates were based largely on those of
EPA's Carcinogen Assessment Group. Data on the quantities of
toxic compounds of interest, primarily noncriteria volatile
organic chemicals (e.g., Table 1), were gathered from existing
databases. Supplementary field measurements were carried out in
some cases to fill data gaps.

IEMP results suggest that cross-media transfers are
important contributors to health risks. The Philadelphia study,
for example, found that risk to airborne volatile organic
chemicals was greater from a sewage treatment plant than to any
of seven other industrial point sources considered. Releases of
volatile organics into the air from publicly owned treatment
works and from waste disposal (e.g., Pellizzari, 1982) sites is a
well-documented concern. The principal cross-media issue
identified in the Santa Clara Valley study was the observation of
groundwater contamination by industrial chemicals. The most
likely sources are thought to be underground storage tanks and
surface spills.

The IEMP experience suggests that a major limitation to
multimedia assessments is the general lack of sufficient
monitoring data. The investigations relied heavily on existing
data gathered for other purposes and in some cases required the
use of source apportionment models to predict the composition of
materials measured simply as VOC (Volatile Organic Chemicals).
The inability to acquire the data needed is presumably due to the
cost or the absence of suitable monitoring methods.

Total Exposure Assessment Methodology (TEAM) Study

The USEPA Environmental Monitoring Systems Division's TEAM

study (Wallace, 1986) was designed to develop methods to determine total human exposure to toxic organics. The study centered on volatile organic compounds and included their measurement in outdoor air, indoor air, drinking water, and exhaled breath. A total of six hundred individuals from locales in New Jersey, California, North Carolina, and North Dakota participated in the study by collecting personal air samples, drinking water samples, and providing samples of exhaled breath. Outdoor air samples and indoor air samples were collected by the investigators for comparison with the results of personal air monitoring.

An important finding (e.g., Wallace et al., 1984, 1985; Wallace, 1986) of these studies was that overall exposure to volatile organic chemicals is dominated by indoor air. Concentrations of the chemicals are often an order of magnitude or more greater in indoor air than in ambient outdoor air. Further, quantities of the chemicals in exhaled breath were found to correlate very well with indoor air, occupational (e.g., dry cleaning) exposure, and personal habits (e.g., smoking) and not well with outdoor air concentrations. Indoor air is a medium not commonly considered in developing environmental control strategies to reduce health risk.

The TEAM study also identified an unexpected example of cross-media transfer. The most common source of exposure to airborne chloroform was found to result form showering. Chloroform, formed as a result of chlorination to disinfect drinking water, is released into the air when individuals shower.

The TEAM study initially sought to consider volatile organic chemicals, semivolatile pesticides and PCB's, several metals, and several polycyclic hydrocarbons. The investigators also planned to consider food and house dust as sources of exposure. As for the IEMP study, excessive cost or the unavailability of suitable methods for sampling and analysis required a more limited scope.

Other Related and Site Specific Studies

Site surveys and detailed studies of specific issues which are somewhat unique to individual sites or industries frequently involve some degree of multiple media characterization. The US Department of Energy Environmental Survey, for example, includes consideration of surface water, groundwater, soil, and air. The Department of Defense Installation Restoration Program is engaged in environmental monitoring and remedial action involving soil, groundwater, surface water, and air emissions from active and inactive sites. Industry and USEPA site characterization studies generally include analyses of multiple media samples.

The USEPA-sponsored study by Erickson et al. (1977) of polychloronaphthalenes (PCN), a dielectric used in capacitors, involved collection of soil samples, air samples, and water samples from the vicinity of a suspected PCN manufacturer. The distribution of PCN isomers among these media was found to agree well with their physical properties. All the isomers were found in both soil and air but the more volatile, less chlorinated, isomers predominated in the air samples while the less volatile,

more chlorinated, isomers were predominant in the soil samples. Water samples contained only trace levels of PCN's in agreement with their expected low water solubility. These data suggest that, regardless of the initial source, the PCN's distribute between media in accord with their physical properties.

GLOBAL AND LOCAL RADIONUCLIDE HISTORIES

Global Fallout from Atmospheric Weapons Tests

Among the many examples of global-scale, cross-media transfers of pollutants, perhaps none have been so influential on our thinking as the history of radioactive fallout from atmospheric nuclear weapons testing. Tests conducted during the 1950s and 1960s contaminated not only the immediate zones in which they were conducted but also introduced substantial quantities of fission products into the atmosphere. The subsequent distribution and fate of two of these fission products, Cs-137 and Sr-90, in particular, illustrate cross-media phenomena and, in addition, demonstrate the utility of comprehensive field studies of their behavior.

Cs-137 and Sr-90 have many attributes in common (Table 2). With roughly the same half-life and fission yields, these two nuclides followed very similar atmospheric pathways. Both daughters of gaseous precursors, they tend to condense onto particles relatively late in fireball evolution and were, consequently, more readily solubilized into precipitation or surface waters than other fission products. Members of the soluble alkali metal and alkaline earth groups, they would be expected to be quite mobile in aquatic and marine environments and to substitute readily for their biologically important analogs potassium and calcium.

Atmospheric testing rates and fallout deposition on the earth's surface increased from 1950 through 1957, then declined for the remainder of the decade (Figure 1a). Testing resumed during the early 1960s; the highest rate was in 1962 (Joseph et al., 1971). Devices in the megaton or larger TNT equivalent range produced fireballs that carried virtually all their fission products into the lower stratosphere. Debris introduced into the stratosphere, was transported around the earth in periods of 3-4 weeks. Residence times for fallout particles in the stratosphere were approximately 10 months before they were transferred downward into the troposphere. In the troposphere residence times were less than one month, followed by deposition as dry fallout or with precipitation. Deposition on the earth's surface (Figure 1a) reflected the rates of testing but with approximately 1-year delay in the maximum. Deposition allowed the fission products to participate in the transport processes driven by the hydrologic cycle and to enter into food chains. The records for concentrations in drinking water (Figure 1b) are similar to deposition records with two exceptions. First, there is a further delay in the maximum reflecting the time for transfer from precipitation into public water supplies. Second, the decline in concentrations in drinking water following the test

ban treaty is much less abrupt than the decline in deposition rates reflecting storage of the isotope in soils and plants. Concentrations in milk followed a similar time trend (Figure 1b). In contrast, although the Cs-137:Sr-90 concentration ratio in deposition followed the production ratio of 1.5 very closely, the ratio in tap water was markedly lower, averaging 0.11 for the ten years 1965-1974 (Health and Safety Laboratory, 1975). Fallout Cs-137 was more effectively retained by soils than was Sr-90.

Local-scale Radionuclide Cycling

Binding of fallout Cs-137 to soil particles was often irreversible and that subsequent transport was by erosion and movement of the soils. This tenacity of binding is sufficient that sedimentologists now use Cs-137 as an age marker in their work. Profiles of Cs-137 concentrations in undisturbed sediment cores (Figure 2) faithfully record the history of fallout deposition and show the effects of soil erosion. When the deposition history is scaled to the sediment accumulation record as dated by Pb-210, the 1963 maximum is clearly evident in the core but, like the Sr-90 in tap water, concentrations in sediments deposited after 1965 did not decline as rapidly as the deposition record. The mid-1970s rise in Cs-137 concentrations in the core shown is the result of releases from the Surry nuclear power plant (Cutshall, et al. 1981). Because these releases entered the brackish waters of the James River Estuary, the trapping efficiency was lower than previously seen for fallout which was distributed over the watershed (Zucker et al, 1984).

Environmental monitoring in the vicinity of Oak Ridge National Laboratory (ORNL) has yielded similar observations. Although Cs-137 has been disposed of by shallow land burial at ORNL since the mid 1940s, offsite escape at the present time is restricted to erosion of sediments that were contaminated through direct surface water discharges carried out in the 1950s. Leaching and transport of Sr-90 from wastes in shallow land burial sites, on the other hand, provides the greatest source of this nuclide to surface waters (Stueber, et al., 1981).

Studies of the binding mechanisms on soils have revealed the reason for the contrasting behavior. While Sr-90 attaches to soil particles principally by reversible mechanisms, such as ion exchange, Cs-137 can be trapped by clay minerals having 10-$\overset{\circ}{A}$ interlayer spacings such as illite (Jacobs, 1963). Once trapped, Cs-137 is not readily released; Lomenick and Gardiner (1965) found that treatment with nitric acid to destroy the clay minerals was necessary to achieve Cs-137 solubilization.

Non-radioactive Pollutants

The environmental transport and transfer pathways that have been traced by Cs-137 and Sr-90 are followed by many other pollutants. A large class of pollutants, many with seemingly very different chemistry, are particle-seekers and follow the pathways typified by Cs-137 (Olsen, et al. 1982). Turner, et al.

(1985) used the discharge records for Cs-137 from ORNL and for mercury from the Y-12 plant in Oak Ridge to correlate the deposition record at several locations downstream in the Clinch and Tennessee Rivers. By Pb-210 dating, they showed that deposition in sediments accurately reflected the historical release patterns for these two particle-seeking contaminants. In the James River, cores such as shown in Figure 2 contained the chlorinated hydrocarbon kepone in proportion to the deposition rate (Cutshall, et al. 1981). Polychlorinated biphenyl concentrations in Hudson River sediments are highly correlated with Cs-137 concentrations (Olsen, 1979).

Although generalized transport pathways can be identified for many pollutants, it may also be critical to consider quantitatively minor factors in evaluating exposure or risk. For example, as indicated above mercury is a particle-seeking contaminant and its general distribution is controlled by soil and sediment particle movements. Nonetheless, the importance of a minor pathway in the environmental toxicology of mercury is well known. Following the disaster at Minimata Bay, where a number of deaths were caused through consumption of fish that contained methylmercury which had been discharged, it was shown that the methylation process also occurred in nature. No matter what the form of mercury, it could be converted to methylmercury and, once converted, biological uptake soon followed. The principal mechanism of methylation is microbially controlled and is therefore dependent on the microbial ecology of the medium in which it occurs. In turn, chemical factors which affect the microbial ecology, oxidation potentials, pH, and sulfate/sulfide concentrations have all been shown to be important to methylation rates.

CONCLUSIONS

Experience with radioactive fallout, with organic contaminants and with heavy metals has amply demonstrated that cross-media transfers are common and that understanding the transport, cycling and fate of these contaminants requires a multimedia approach. Nonetheless, pollutants with similar physical and chemical attributes may follow markedly different pathways. The frequency of exceptions to predictions based on simplistic models is also sufficient to show that direct investigation of environmental contamination is essential to confirm validity of models used for conceptualizing a problem or for control. Modeling based on multimedia premises and regulatory controls that encompass multimedia considerations are challenged by a dilemma, however. First, the development of multimedia models or regulatory frameworks represents simplification and generalization. This is true for several reasons: 1) inadequate understanding of physical and environmental factors which control specific cross-media transfer, 2) the absence of specific data on certain multimedia pollutant concentrations, 3) even the most powerful computers do not have sufficient speed and capacity to deal with the known complexities of natural systems. On the other hand, for

contaminants such as mercury, it may be necessary to include
great detail; the overall distribution in the environment may be
less important than the rate of some minor process. With
sufficient experience and good judgement of what can be ignored,
the simplifications and generalizations can be made. For the
present, and for the forseeable future, however, they absolutely
must be accompanied by thorough field validation and monitoring.

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TABLE 1   Selected Organic Chemicals Considered by IEMP and TEAM Studies

| Chemical | TEAM[1] | | IEMP[2] | |
	Air[a]	Drinking Water	Air	Water
Chloroform	++[a]	+++		
1,1,1 Trichloroethane	+++	++	X[b]	X
Tetrachloroethylene	+++	+	X	X
m,p-Dichlorobenzene	+++	+		
o-Xylene	+++		X	X
m,p-Xylene	+++			
Benzene	+++	–		
Trichloroethylene	++	–	X	X
Styrene	++	–		
1,2-Dichloromethane	+	+	X	
Carbon Tetrachloride	+	+		
Bromodichloromethane	+	+++		X
Chlorobenzene	+	+		
o-Dichlorobenzene	+		X	
Bromoform	+	+		
Dibromochloromethane		++		
Vinylidinechloride		+		
Toluene		+	X	X
1,1 Dichloroethylene			X	X
1,2 Dichloroethylene				X
Ethylene Dibromide			X	X
Methylene Chloride			X	X

[a]Frequency of detection: ubiquitous/often/occasionally/"never"
(+++,++,+,-) for North Carolina and North Dakota TEAM studies.
Blank indicates not considered or rarely detected.

[b]"X" indicates the chemical was considered in the IEMP study.

[1]Wallace, 1986. Air samples include personal and fixed site samples,
outdoor and indoor air, and/or exhaled breath. Drinking water samples
taken from taps.

[2]Hinman et al., 1986. Air samples taken outdoors only. Water samples
include surface and/or groundwater Santa Clara Valley study.

TABLE 2   Selected properties of Cs-137 and Sr-90.

Property	Cs-137	Sr-90
Half-Life	30 y	28 y
Chemical analog	K	Ca
Thermal fission yield*	6%	6%
Precursor	Xe-137	Kr-90
Periodic chart	Gp IA (Alkali)	Gp IIA (Alkaline earth)

*Yields from nuclear detonations are significantly lower.

FIGURE CAPTIONS

FIGURE 1   Sr-90 records from 1954-1974.   1a) Deposition by year measured in
New York City; 1b) In New York City tap water; 1c) in milk, normalized
to calcium.   Maxima in tap water and in milk occurred later than in
deposition and the peaks for tap water and milk were broader than for
deposition.   (Data from Health and Safety Laboratory Quarterly Report,
HASL-291, Appendix)

FIGURE 2   Concentrations of Cs-137 in a sediment core from the James River
estuary, Virginia, compared to fallout deposition and local reactor
releases (after Cutshall, et al. 1981).   Scaling of the two profiles is
based on Pb-210 dating of the core.   The correspondence of maxima in
the Cs-137 input to the estuary with concentrations in the core is
excellent.   Sediment retention of Cs-137 from atmospheric fallout (1963
peak) is higher than retention of Cs-137 from the reactor (1975 peak)
because the latter has a shorter contact time with the particles and is
more susceptible to desorption in brackish estuarine waters (Zucker, et
al. 1984).