A. Robertson

DEPARTMENT OF COMMERCE
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NOAA Technical Memorandum NOS OMA 43

An Historical Reconstruction of Major Pollutant Levels in the Hudson-Raritan Basin: 1880-1980

VOLUME 1
Summary

Rockville, Maryland
October 1988

noaa NATIONAL OCEANIC AND ATMOSPHERIC ADMINISTRATION

National Ocean Service

Office of Oceanography and Marine Assessment
National Ocean Service
National Oceanic and Atmospheric Administration
U.S. Department of Commerce

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Robert U. Ayers (Principal Investigator)
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Rolande C. Widgery (Consultant)

Variflex Corporation
Prepared for Martin-Marietta Environmental Systems (MMES)
Grant #NA 83AA-D-00059
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NOTICE

An Historical Reconstruction of Major Pollutant Levels in the Hudson-Raritan Basin: 1880-1980

VOLUME I
Summary

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Acknowledgments

This summary report is one of 3 volumes. The overall methodology was the responsibility of Robert U. Ayres. The runoff model formulation was due to Mitchell Small; the computer analysis in Chapter 2 was carried out by James McCurley. Chapters 3 through 5 were written by Robert Ayres with computer analysis by McCurley and Leslie Ayres. Historical research on land-use, fuel-use, agriculture and sewage treatment was the responsibility of Joel Tarr, with assistance from McCurley.

The volume *Heavy Metals and Fossil Fuels* (HMFF) was written by Robert Ayres with help from Tarr. The HMFF data base was compiled and computerized by Leslie Ayres. The sections on pesticides and fertilizers in the volume *Chemicals and Other Wastes* (COW) were mainly written by Rolande Widgery. The sections covering PCB's, PAH, Oil and Grease and TOC were written by Robert Ayres, who also edited the rest. Most of the COW data base was tabulated and computerized by Leslie Ayres. The agricultural data were compiled by McCurley under the guidance of Tarr.

Ivars Gutmanis (Sterling-Hobe Corp.) provided background information on several industries, (iron and steel, copper, petroleum refining and town gas) under a separate contract with MMES. We acknowledge his useful contributions.

Most of the word-processing was done by Margery Tamres at Variflex Corporation. Graphs were prepared by Leslie Ayres.

We are also grateful to Joel O'Connor and Gary Mayer of NOAA and Kevin Summers of MMES for helpful criticisms and patience.

Introduction

This report is a summary of the results of a larger effort carried out by Variflex Corp. for Martin-Marietta Environmental Systems, Inc. under a grant from the Ocean Assessment Division of the National Oceanographic and Atmospheric Agency (NOAA).

The background research is described in two independent volumes, on *Heavy Metals and Fossil Fuels* (HMFF) and *Chemicals and Other Wastes* (COW), respectively. The purpose of those volumes is to assemble and document available data on sources, transformation processes, uses and losses of critical pollutants of anthropogenic origin, with particular reference to the Hudson-Raritan basin. The results are presented as historical tables. The two volumes in question are the primary sources of information used herein. We refer to them from time to time, for convenience, as HMFF and COW respectively.

The specific purpose of the present report is to reconstruct historical pollutant loadings in the Hudson-Raritan (H-R) basin from the material production and use data (as compiled in HMFF and COW) consistent with recent empirical evidence.

The larger purpose of NOAA in commissioning this report is to provide input data for a statistical test of various hypotheses concerning the long-term relationships between commercial fish stocks and anthropogenic activities in estuarine water bodies. The analysis of fish stocks is the responsibility of MMES. As a test of the overall methodology, this pilot study has been focussed explicitly on the Hudson-Raritan basin. The choice of the Hudson-Raritan basin as a test case seems appropriate for three reasons:

1. The impact of human activity has been great but not completely overwhelming. The Hudson River has been seriously polluted, but never to the point of being uninhabitable to fish (as the Rhine or Thames were at times).
2. From the perspective of human activity, the Hudson-Raritan basin is as complex as any estuary in the world (perhaps, it would be matched by Tokyo Bay). If the methodology yields reasonable results for the Hudson-Raritan basin it can be expected to do so elsewhere.
3. The Hudson-Raritan basin has been thoroughly studied in recent years in terms of pollutant loadings and sources. Thus there is a reasonable basis for model verification and calibration.

Apart from the specific objectives of the sponsors (NOAA), the research reported here has generated two significant conclusions that deserve some emphasis. One is methodological; the other is substantive.

The major methodological conclusion is that the hybrid 'materials-balance' cum statistical runoff model approach exemplified in our work is the only one that offers any realistic hope of success in an historical reconstruction of the sort undertaken hereafter. Conventional historical methodologies (seeking documentary evidence from archival sources), in the absence of a rigorous framework for the data, cannot be expected to be adequate, although the archives can and do yield many useful clues. We believe future economic and other historians will increasingly have to adopt this approach.

The substantive conclusion -- one for EPA, in particular to consider seriously -- is that dissipative *consumption* rather than production processes are now by far the dominant sources of most waterborne pollutants. This conclusion is particularly strong in connection with the heavy metals. It would seem to follow that EPA should give much greater attention than it has in the past to the 'life cycles' of products destined for end-use embodying toxic elements, or other potential pollutants such as pigments (used in paints and coatings), lubricants (including additives), pharmaceuticals, solvents, cleaning agents, chelating agents, bleaches, etc. Lack of good information about the fate of consumption-related emissions is the major weakness of this study. Even though such studies are necessarily limited to a point in time, they would be of value in future historical reconstructions such as this one.

NOTATION: Metric units are used throughout. It should be noted that

1 metric ton = 1 tonne = 1 Mg = 1000 kg.

We have generally used tonnes as measures of production or consumption, while Mg is generally used in measuring emissions.

1. Methodology

1.1 Material Cycles and Balances

The problem of estimating pollutant loadings in the absence of direct measurements is complicated by the fact that pollutants are, for the most part, secondary and unintended consequences of other activities. These unintended consequences have not been systematically studied for their own sake until relatively recently. There are very few direct data on industrial emissions available for periods before the 1960's.

On the other hand, pollutants (except for heat) are inherently material and hence subject to the fundamental principle of conservation of mass. Many priority pollutants are compounds of one of the heavy metals (Ag, As, Cd, Cr, Cu, Hg, Pb, Zn). Another group consists of chlorinated hydrocarbons, pesticides, herbicides and fertilizers (N, P), mostly used in agriculture. A third miscellaneous group of organics includes polychlorinated biphenyls (PCB's), polynuclear aromatics hydrocarbons (PAH), oil and grease, and total organic carbon (TOC). Materials are 'mobilized' in the environment by natural processes and by anthropogenic production and/or consumption activities. Anthropogenic processes do not necessarily dominate natural processes in material cycles on a global scale. However, in a densely populated and highly industrialized area such as the Hudson-Raritan basin, non-anthropogenic sources can be ignored in most cases. With the possible exceptions of N, P and TOC, anthropogenic processes of materials mobilization tend to be much more important than natural ones. This is an important point to which we will return later.

Another important distinction must be made between materials that were extracted and utilized primarily for their own sake, such as lead and copper lost in processing, and materials mobilized as minor contaminants in other fuels or minerals that are processed on a large scale. Examples of the latter include trace elements found in copper ore, coal, petroleum, iron ore, phosphate rock and limestone. As will be seen, this secondary route is significant on a global scale for several minor metals like Ag, As, Cd, Hg. Combustion of coal and oil is the dominant environmental source of sulfur (not of concern in this study) and PAH, both globally and locally. High temperature combustion processes also generate significant amounts of atmospheric NOx, although this is not locally important as a source of waterborne nitrates in comparison with sewage and commercial fertilizer use.

The derivation of material cycles and materials balances is the most important analytical tool for inferring pollutant loadings from historical data on economic activity. In broad terms, the procedure comprises the following steps:

1. Trace the major **sources** of each pollutant, identifying relevant natural processes (such as leaf fall) and relevant production/consumption activities resulting in emissions.
2. Trace the relevant **material cycle**, taking into account purposeful industrial transformations, natural (e.g. biological) transformations, dissipative uses, and environmental transport processes.
3. Construct a **materials balance** (at least for chemical elements) to permit quantitative inferences of emissions from available economic data.

The materials balance can be constructed in a very formal manner, using a standard accounting format to balance inputs and outputs. This can be done for any bounded system, from a single factory (or even a piece of process equipment such as a furnace or reactor) up to the earth as a whole. In general, it will be done for the sake of convenience at fairly high levels of aggregation. But there are cases where materials balance analysis of characteristic individual processes can be invaluable, e.g. to determine the partitioning of trace metals in coal combustion processes between condensed (on fly ash) and vaporized forms. Similarly, this methodology is essential to account for the partition of pesticides (such as DDT) among various possible environmental fates after initial deposition on the plant or soil surface.

The fact that some key data is available only at the national level, while some consumption patterns tend to be regional, is particularly critical in the case of pollutants associated with agriculture. The calculation of estimated use of any agricultural chemical in the Hudson-Raritan river system is made difficult by the fact that most available statistics on use are either for the U.S. as a whole or for a few states that happen to collect data. The following generalized procedure scheme seems to be the most appropriate to the situation. It may be conceptualized as a tabular spread-sheet with rows corresponding to years and columns as follows:

1. **U.S. use of chemical.** Insecticides, herbicides, fertilizers, etc. by weight.
2. **Use (of chemical) by region.** Depending on source these are NE (northest) or MA (Middle Atlantic, EPA region 11). By dividing Column 2 by Column 1, we calculate
3. **Percentage use (of chemical) by region.** Then, assuming that crop planting patterns, pest infestation and so on are similar throughout each region, we can multiply these percentages by
4. **The reported use (of chemical) by crop.** (These data may only be available for a relatively few years and require interpolation.) This in turn provides the data in Column 5.

5. **Chemical use per crop in region.** By dividing this by
6. **Acres of crop in the region** we obtain
7. **Chemical per acre of that crop in region.** This amount multiplied by
8. **Acres of the crop in the Hudson-Raritan river system** yields the desired
9. **Total of specific chemical used in the Hudson-Raritan system.**

Because of the complexity of these calculations, a separate table will have to be made for each material. Year data can be collected and a summary table showing total application each year and total applied can then be interpolated for years for which no data are available.

Dividing (1) by total land area devoted to agriculture yields an average application intensity for the nation as a whole in lb/acre (kg/ha). Similarly, dividing (9) by agricultural land area in the Hudson-Raritan basin yields an average application intensity for the basin.

A similar but simpler scheme can be applied for pollutants associated with combustion of fuel, viz:

1. **U.S. consumption of the fuel** (gasoline, residual oil, distillate oil, bituminous coal, coke, etc.)
2. **Fuel use by state (e.g. NJ, NY)**
3. **Allocation to the Hudson Raritan basin** in proportion to the fraction of each state's population in the basin (except where special adjustments may be required).
4. **Computation of emissions by multiplying regional fuel consumption by appropriate coefficients.**

Emissions associated with dissipative end-uses of products embodying the pollutant as an essential element, rather than as a contaminant, can be allocated to the region in question by a similar procedure, viz:

1. **U.S. consumption of the material by end-use category** (or fractional allocation of total consumption)
2. **Allocation of end-use by state**
3. **Allocation to Hudson-Raritan basin,** using the proportional fraction method outlined above for fuels
4. **Computation of emissions** by multiplying regional consumption by appropriate coefficients

It must be pointed out that gross emissions coefficients for trace contaminants (such as metals or PAH in coal) are relatively small, ranging from less than one part per million up to one or two percent. Gross or uncontrolled emissions calculations must be further adjusted to take into account the extent -- if any -- to which emissions have been further reduced by electrostatic precipitators, scrubbers, etc. In the case of emissions resulting from dissipative end-uses (e.g. weathering of painted surfaces or tire-wear) the key question is: What fraction of the polluting material is volatilized, incinerated, or reduced to powder by weathering and thus escapes to air or water? This fraction can be described as *environmentally mobilized*. The alternative possibility, of course, is that the material is solid to begin with and/or more or less permanently embodied in a solid waste that is disposed of safely on land. Note that the fraction of materials lost by dissipative end-uses is often much larger than a typical processing emissions coefficient. In some cases virtually all of the material is environmentally mobilized by its 'normal' use. For this reason, as will be seen later, emissions associated with consumptive uses tend to dominate point-source emissions associated with production processes.

Most of the pollutant materials considered in detail hereafter are primarily anthropogenic in origin. The methodology outlined above applied to these materials. It happens that three pollutants (N, P and TOC) are partly attributable to non-anthropogenic processes that are widely distributed. These contributions appear as a component of runoff from land surfaces. Where no production or consumption process can be specifically identified as the origin of the pollution, we are limited to the use of empirical relationships between land-use and runoff. These are considered in the next section.

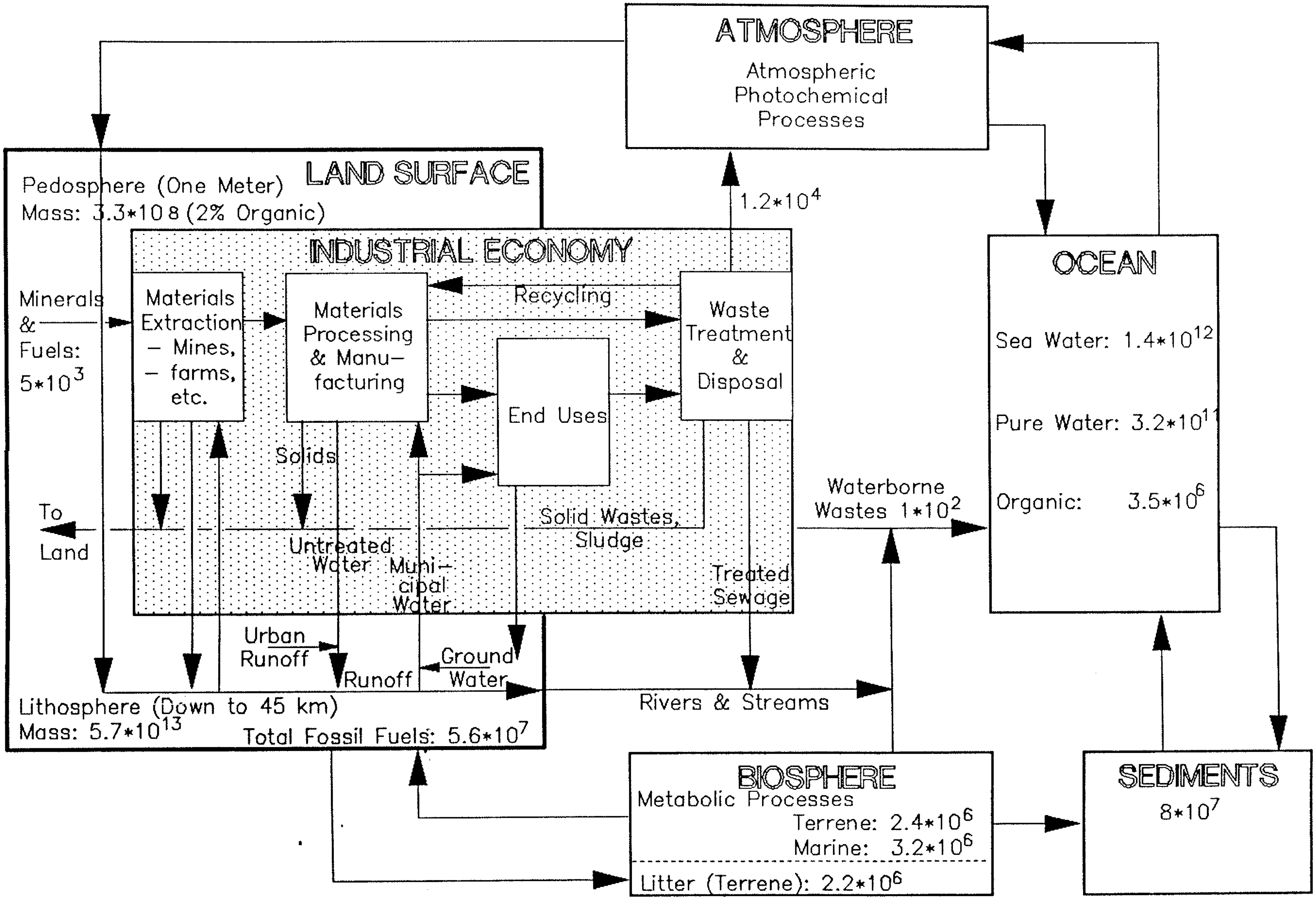
1.2 Environmental Transport and Runoff

Pollutants in rivers are generally being transported *en route* from one environmental reservoir to another, as is the water itself. The transport phase is only one element of a materials cycle that includes products-in-use, various natural reservoirs (atmosphere, soil, groundwater, oceans, biosphere), and various transfer paths (leakage, volatilization, precipitation, incineration, leaching, etc.). A generalized material-cycle is shown in Figure 1-1. In practice, waterborne pollutants are generally divided into two basic groupings based on route of entry, viz

- point-sources
- non-point sources ('runoff')

The first category includes wastes from industrial outfalls, as well as sewage (both treated and untreated)

FIGURE 1-1: Global Materials Cycle (units = 1,000,000 metric tons)



and oil spills in the estuaries themselves. Runoff, on the other hand, accounts for virtually all pollutants of agricultural origin as well as pollutants that are deposited on the land surface from rainfall or dustfall, no matter how they reach the river.

Some of the environmental transfer processes that are theoretically possible occur very slowly under normal conditions and, to a reasonable approximation, can be neglected in comparison to other modes of transport. This is generally true, for example, of transport from the soil surface (or from landfills) to groundwater and thence to rivers and streams, although some of this is included in the runoff category. We can also generally neglect direct deposition from the atmosphere to the water surface. Interestingly, direct emissions to water also tend to be relatively small in the eastern U.S. river basins, in comparison to contributions from sewage and urban runoff. The latter, however, contains major inputs from atmospheric deposition.

1.3 A Runoff Estimation Model (c.1975)

In principle, it should be possible to account for observed concentrations in a water body such as NY harbor by application of materials balance considerations. Measurable or calculable inputs include the flow from upstream, tributary rivers, sewage, urban runoff, and bottom sediments. Outputs include resuspension of deposition to sediments and outflows to the bight. Runoff contribution, in turn can be calculated, in principle, from a similar materials balance matching inputs (emissions within the airshed, advection from upwind) and outputs (deposition on the ground and advection downwind. In effect, one can estimate urban runoff in two different ways:

1. By accounting for observed pollutant inputs to and concentrations in the air, together with estimates of soil permeability and
2. By accounting for observed concentrations in known volumes of water and allowing for known point-source inputs and inflows (from upstream or elsewhere).

A hybrid approach to estimating runoff loads from non-point sources, combining the above methods appears to be the most feasible alternative. Such models have been developed at the national level by Heany [Heany et al 76], Hydroscience Inc [Hydroscience 78] and McElroy [McElroy et al 76]. The approach of Heany et al has been selected as the underlying basis for our loading estimates. The Heany model is modified, however, by reducing the number of land-use categories, j, from 9 to 4, eliminating the rainfall dependence and introducing temporal variability. These adjustments are discussed hereafter. The (modified) Heany model estimates non-point source storm runoff loads F^i (kg of i^{th} pollutant) at the national level for a typical recent period (roughly 1970-1975).

$$F^i(t) = 10^{-3}P\sum_j g_j^i(t)A_j(t) \tag{1}$$

where P is average annual rainfall in cm, $A_j(t)$ is the amount of the j^{th} land-use in hectares in year t and the summation is carried out over the whole drainage basin. The emission coefficient $g_j^i(t)$ in kg/ha is defined as follows:

$$g_j^i(t) = y_j^i + x^i E^i(t) \tag{2}$$

where y_j^i is the annual non-anthropogenic contribution of the i^{th} pollutant by the j^{th} land-use, while x^i is the anthropogenic contribution per tonne of gross annual emissions of the i^{th} pollutant in the region, $E^i(t)$. Note that both land-use and gross emissions change over time, while y_j^i and x^i are constants. Non-anthropogenic terms y_j^i are zero for chemicals, but non-zero for most metals, N, P, hydrocarbons and TOC.

Nine land use categories are used in the pollutant runoff model of Heany et al (1976). These include (1) residential, (2) commercial, (3) industrial, (4) agricultural, and (5) undeveloped land with separate runoff conveyance; and (6) residential, (7) commercial, (8) industrial, and (9) undeveloped land with combined sewer conveyance. Historical data on land-use is not available in this degree of detail however, necessitating the use of a simpler classification. In our modified version of the model residential land-use is lumped together with commercial and industrial land-use because there is insufficient historical data to distinguish them. (Prior to the 1950's these activities were in fact thoroughly mixed up in most of the New York metropolitan area. Residential suburbs developed largely after that time. Also, large-scale industrial activities began to move away from densely populated Manhattan and Brooklyn after WWII). The land use categories we use hereafter are as follows:

1. Developed, Combined Sewerage;
2. Developed, Separate Sewerage;
3. Agricultural Cropland; and
4. Undeveloped Land.

Estimates of historical land use divisions in the Hudson-Raritan planning area are summarized in Table I-1 in Appendix I, together with an explanation of the estimating procedure that was used.

1.4 Initial Parametrization of the Model

The loading factors for the nine land-use categories in the Heany model are given in Appendix I (Tables I-2, I-4, I-6). All load factors are given in terms of kg/ha per cm of rainfall. Average rainfall in the Hudson-Raritan basin is 109 cm. We use this value throughout. These factors have been combined and revised to represent the new grouping. The Heany loading factors for commercial and industrial land -- independent of population density -- were simply averaged together. The resulting load factors are equivalent to what would be expected for residential areas having a density of 20,000-25,000 persons per square mile, which happens to be about right for most of the urbanized area. Thus, the use of a single category for 'developed' land seems reasonable. Other modifications of the Heany model for this study include the removal of a street sweeping factor, based on more recent findings that street sweeping actually does little to improve runoff quality [Athayde et al 83]. We assumed load-factors for undeveloped rural land equal to Heany's category for undeveloped land with separate sewer conveyance.

The values of emission factors g_j^i (Appendix I) are determined from runoff quality data and loading function summaries from Betson [Betson 76], Heany [Heany et al 76], Hydroscience [Hydroscience 78], McElroy [McElroy et al 76], Lager [Lager et al 77], Athayde [op. cit.], and Cole [Cole et al 84].

The suspended solids, TOC ($\sim$=BOD_5)[1] and nitrogen factors are taken directly from Heany et al [op. cit.] with the exception of the agricultural estimates which are based on Lager et al [op. cit.] and Hauck and Tanji [Hauck & Tanji 82]. Phosphorus loading factors are estimated as one sixth and one fifth of those for nitrogen for separate and combined sewer areas, respectively. These factors are consistent with national runoff quality summaries. Oil and grease is estimated from the suspended solids factors by assuming 0.01 gOG/gSS in developed areas and 0.001 gOG/gSS for nonurbanized land uses [Lager et al 77]. These emission factors are given in Table I-2, Appendix I.

Heavy metals loading factors were derived from various sources, mainly by using metal-to-suspended solid (SS) ratios. Athayde et al [op. cit.] provided data on all eight metals; Lager et al [op. cit.] covered Cd, Cr, Cu, Pb and Zn: McElroy et al [op. cit.] dealt with As, Cu, Pb and Zn, Betson [op. cit.] provided data on Pb. The loading ratios are shown in Table I-3 and the resulting loading factors indicated in Table

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[1] On the average, TOC is numerically the same as BOD_5, even though the definitions are technically different [Metcalfe & Eddy 72]

I-4 in Appendix I. The highest values are indicated for lead and zinc while the lowest values are shown for mercury and silver. Note that older studies indicate lead values higher than those for zinc, while more recent studies suggest zinc equal to or greater than lead, probably reflecting the effects of recent lead limitations in gasoline.

Environmental transport of pesticides and herbicides is a complex and recent area of study [McEwen & Stephenson 79]. In a well defined agricultural plot, it is possible to estimate the fraction of applied material mobilized or leached to surface waters. Estimated values for pesticides are generally 0.5-1.0 percent of the applied material, and 1-5 percent for herbicides [Haith & Loehr 79; Kuhner 80]. For a large area, however, tight mass balances are not possible, and we again settle for empirical loading functions based on currently observed (though more limited) runoff data. The key sources for all of the toxic organics are Athayde et al [op. cit.] and Cole et al [op. cit.]. Data on PAH also come from Hoffman [Hoffman et al 84].

Nutrients (N,P), total organic carbon, PAH and metals are mainly transported by the suspended and dissolved solids loads of streams, even in the absence of (direct or indirect) anthropogenic influence. To estimate the "preindustrialization" background levels, average natural global loading rates were taken from Garrels [Garrels et al 75]. Combined with the global suspended sediment load of 180 x 10^8tonne/yr [Garrels et al 75] these yield the background loading factors indicated in Table I-7 in Appendix I. Note that when compared to present day factors determined independently, the preindustrialization values range from 5% to 100% of current values. Constituents indicated to be currently 5 to 20 times above background levels include lead, cadmium, mercury, arsenic, and zinc. However, silver, copper and chromium are indicated to be nearer to background levels. The values in Table I-7 provide a baseline for historical scaling of loading factors.

Chapters 3, 4, 5 and 6 are concerned with estimating values of the $E^{(i)}(t)$ variables. Having done so, it will be possible to make explicit "predictions" of past runoff pollution loadings in the Hudson-Raritan basin. These loadings (added to point-source data) can, in turn, be used to predict ambient concentrations, given a suitable model of pollutant transfer and degradation in the river. Several empirical studies have been carried out in the past two decades in the Hudson-Raritan basin, [Mueller et al 82; Breteler 83; Greig & McGrath ??], that can be compared explicitly with predicted loadings. Comparisons of this sort can be used as a basis for recalibrating both the water models and some of the assumptions used in constructing the materials cycles/balances. Samples of sediments taken from the bottom of rivers and bays at various times [e.g. Bopp et al 82] can also be used to some extent for verification and calibration purposes. Results are discussed in the next chapter.

2. Estimates of Historical Pollutant Loads from the Hudson-Raritan Basin

2.1 Validation and Recalibration of the Model

A general, nonpoint source pollutant (runoff) loading model, applicable to urban areas of the United States, was described in the previous chapter. The model is further adapted to estimate historical runoff loads in the Hudson-Raritan basin in this chapter. We first apply the model in 'raw' form to estimate pollutant loadings for the basin. These results are then compared to the results of detailed studies of the estuary carried out by Mueller [Mueller et al 82]. This comparison can then be used to calibrate the modified Heany model. Historical estimates of gross anthropogenic emissions levels $E^{(i)}(t)$ for the Hudson-Raritan area, derived in the next four chapters are, the basis of our final calculations of pollutant loads for earlier periods.

To recapitulate: the aggregated national runoff loading factors (Appendix I, Tables I-2, I-4 and I-6) are used with the current New York area land use data (Appendix I, Table I-1) to estimate pollutant loads for the period 1970 - 1980, assuming an average of 109 cm of rainfall (Central Park, NYC). These loads were compared to those reported by Mueller et al [op. cit.]. Table 2-1 shows the ratios of the "actual" loads estimated by Mueller et al to loads "predicted" by our model for 1980, before calibration. For many of the constituents there is reasonably good agreement (within a factor of 2). In three cases (Cu, PAH and DDT) the differences are greater. That is to say the "actual" numbers for the Hudson-Raritan appear to exceed the predictions based on national runoff data. It should be noted that both estimates are based on surveys of *available* information and data for urban areas in the U.S. and New York, respectively. A portion of the differences in these two compilations may be due to noise and uncertainty. In the case of DDT, which has not been used much in the U.S. since the 1960's, Mueller's estimates themselves are suspect,[2] because they were based on data from the west coast where DDT use was heavier. However, it is possible that some of the difference is real, arising from a peculiar historical circumstance. Most DDT, nationally, was used in agriculture or for public health purposes (e.g. to kill mosquito larvae). New York State was unique in having used DDT in very large amounts in a *forested* area (to control the gypsy moth). While this occurred almost 25 years before the 1980 calibration date, it is known that DDT degrades much more slowly in shaded, undisturbed forest soil than in cropland or other locations exposed

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[2] We are grateful to Joel O'Connor of NOAA for calling this to our attention.

to sunshine and oxygen (COW, Chapter 2). Thus, the forested areas of the upper Hudson River may constitute one of the last significant reservoirs of soil-bound DDT in the U.S. However this is speculative.

Table 2-1: Ratio of Present Day Runoff Loads Reported by Mueller et al (1982) to Those Predicted for Hudson-Raritan Basin by Uncalibrated Heany Model

Constituent	SS	N	P	Cd	Cr	Pb	Hg	Zn	Cu	PCB	PAH	DDT
Mueller et al Heany (uncal.)	0.55	1.99	0.92	1.08	1.09	1.10	0.78	2.28	9.03	1.09	10.0	11.5

For Cu and PAH, it is even more likely that real differences in Hudson-Raritan vs. U.S. runoff factors still exist. In the case of copper, the high levels in the Hudson-Raritan are certainly attributable to leachate from an accumulation of copper-bearing slag from the smelters and refineries in the Hudson-Raritan basin, much of which was used as land-fill in past years. The locations of highest copper concentration within the Hudson-Raritan basin correspond very closely to the locations of the smelters. Such urban accumulations of copper smelter slag are unique to New York, Baltimore harbors. It is known that slag granules leach (see HMFF, Chapter 5). Thus, it is not surprising that actual loadings of copper in Hudson-Raritan basin are as much as 10-fold larger than "average" loadings from other urbanized areas.

In the case of PAH, the Hudson-Raritan excess over national averages in runoff is harder to account for (the contribution from crude oil spills and bilge-washing operations is presumably not included in this category). The most important source of PAH in runoff appears to be waste motor oil. It is probable that a much higher percentage of waste oil finds its way into storm sewers in the NY metropolitan area than elsewhere, due to the scarcity of alternative means of disposal on land. However part of the big discrepancy between national estimates and Mueller's figures is probably due to unreliable data at the national level.

Given the above explanations, the comparisons shown in Table 2-1 can be regarded as a reasonable basis for recalibrating the national model to adjust for Hudson-Raritan conditions. To do so, loading factors for constituents listed in Table 2-1 (for all four land use categories) were uniformly multiplied by the indicated factor, except in the case of N and P. For other pollutants, the national values initially estimated in Appendix I were maintained.

In the case of nitrogen (N) and phosphorus (P) the detailed analysis in COW (Chapters 10, 11) clearly indicated that average agricultural fertilizer use in the Hudson-Raritan basin is much lower than the national average. This is primarily due to the specialized crop mix (dairy farms and orchards). As a consequence, we altered the agricultural runoff load factors for N and P (Table I-2) reducing them by approximately a factor of 4 to equal the load factors for developed land with separate sewer conveyance. Historical runoff loading becomes a straightforward function of changing land use. The final adjusted runoff load factors are exhibited in Tables 2-2, 2-3, 2-4.

Table 2-2: Adjusted Hudson-Raritan Basin Load Factors for Major Pollutants $g^i(1975)$ (units = kg/ha - cm)

	Developed Land		Agri-cultural Land	Unde-veloped Land
	Separate Sewers	Combined Sewers		
Suspended Solids (SS)	6.25	25.76	5.50	0.550
BOD_5 or TOC	0.97	4.00	0.20	0.050
Nitrogen (N)	0.30	1.25	0.30	0.065
Phosphorus (P)	0.02	0.12	0.02	0.005
Oil & Grease (OG)	0.11	0.47	0.01	0.001

Table 2-3: Adjusted Hudson-Raritan Basin Loading Factors for Heavy Metals $g^i(1975)$ (units = g/ha - cm)

	Developed Land		Agri-cultural Land	Unde-veloped Land
	Separate Sewers	Combined Sewers		
Ag	0.02	0.10	0.01	0.001
As	2.30	9.55	1.00	0.100
Cd	0.61	2.55	0.27	0.027
Cr	3.71	15.30	1.64	0.164
Cu	10.30	42.50	4.52	0.452
Hg	0.04	0.18	0.02	0.002
Pb	19.30	79.62	8.39	0.839
Zn	26.30	108.00	11.40	1.140

2.2 Historical Runoff Estimates

The next to last step in our chain of reasoning is to 'backcast' runoff estimates, based on present-day values. This can be done using equation 2, rewritten as follows:

Let $R^i(t) = E^i(t)/E^i(1975)$, then

$$g_j^i(t) = y_j^i + g_j^i(1975) - y_j^i R^i(t) \tag{3}$$

Table 2-4: Adjusted Hudson-Raritan Basin Loading Factors for Toxic Organics $g^i(1975)$ (units = mg/ha - cm)

	Developed Land		Agricultural	Undeveloped
	Separate Sewers	Combined Sewers	Land	Land
DDT	13.0	53.9	11.4	1.15
Chlordane	34.0	140.0	30.0	3.00
Dieldrin	0.1	0.5	0.1	0.01
Lindane	1.3	5.4	1.1	0.11
Heptachlor	0.1	0.5	0.1	0.01
H.epoxide	0.1	0.5	0.1	0.01
PCB	12.4	51.2	10.9	1.09
PAH	1140.0	4700.0	1000.0	100.00

Plugging 3 back into 1 (Chapter 1) yields

$$\begin{aligned} F^i(t) &= 10^{-3}P\sum_j y^i_j A_j(t) \\ &+ R^i(t)\sum_j dg^i_j(1975) - y^i_j A_j(t) \end{aligned} \tag{4}$$

In most cases $y^i_j << g^i_j$ (i.e. the non-anthropogenic or 'prehistoric' component is negligible), so by setting $y^i_j = 0$ in 4 we can express the average runoff as

$$F^i(t) = 10^{-3}PR^i(t)\sum_j g^i_j(1975)A_j(t) \tag{5}$$

There are two time-dependent factors, $E^i(t)$ and $A_j(t)$.

In some cases -- notably the metals -- runoff is by far the major source of waterborne pollutants in the Hudson-Raritan basin and the backcast is quite straightforward. Using $g^i(1975)$ values from Table 2-3, $R^i(t)$ from Table 5-9 in Chapter 5 and areas $A_j(t)$ by land-usage and decade from Table I-1 in Appendix I and assuming P=109 cm/yr rainfall, we obtain the results shown in Table 2-5 . Since point sources are either negligible or impossible to estimate, these are also our final estimate of total pollutant loading for the metals.

In the case of the pesticides and PCB's it is also true that runoff is the only significant contributory route but the situation is complicated by the fact that (except for chlordane) domestic production and use of most chlorinated hydrocarbons ceased anywhere from 10 to 20 years ago. Pesticide in runoff was therefore rapidly declining thoughout the 1970's, whence the exact temporal distribution of the

Table 2-5: Hudson-Raritan Basin; Historical Runoff Estimates; Metals (tonnes)

	Silver	Arsenic		Cadmium		Chromium	Copper		Mercury	Lead		Zinc	
		low	high	low	high	avg (a)	low	high	avg (a)	low	high	low	high
1980	4.0	282	283	60	60	431	1547	1549	5.3	1413	1415	3613	3614
1970	3.3	439	455	135	135	746	1711	1723	8.8	2583	2622	4682	4692
1960	2.5	349	408	114	114	771	1311	1337	4.5	1879	1976	3815	3838
1950	2.3	355	478	92	93	659	1469	1521	5.0	1283	1657	3783	3835
1940	1.1	548	660	82	83	683	1670	1711	4.8	786	1284	3419	3484
1930	0.7	377	470	42	51	211	724	758	3.9	236	534	2685	2731
1920	0.5	199	256	37	59	218	811	837	2.9	232	516	1740	1801
1900	0.2	61	66	17	19	209	606	616	1.6	244	281	474	512
1880	0.05	15	15	6	6	52	212	213	1.6	8	9	152	168

Calculated from Equation 5 with $Y^i=0$ and rainfall assumed = 109 cm/yr

(a) No significant difference between high and low cases

obervations that provided the data for determining runoff load factors becomes critical. In fact, for these materials, a steady-state runoff model is clearly inappropriate. Accordingly, we use the (adjusted) runoff model results *only* for the single year 1975.

It is important to realize that the current pesticide runoff load at any time t has two components. The first is proportional to the *current use rate*, and represents losses at the time of first application of the pesticide. As noted earlier [McEwen & Stephenson 79] the loss rates for pesticides to runoff tend to range from 0.5% to 1%. As an area of comparatively high rainfall, the higher figure can probably be assumed to prevail in the Hudson-Raritan basin. Thus we have chosen the following to represent the concurrent use portion of runoff:

$$F_{CUR}(t) = Pc^i\overline{X}^i(t) \tag{6}$$

where P is average annual rainfall and $\overline{X}^i(t)$ is the average pesticide consumption in year t. The $\overline{X}^i(t)$ are given in Table II-2, Appendix II and summarized in Table 6-13, Chapter 6. The c^i constants (in general 1%) are shown in Table 2-7. Obviously this contribution ceased in the last year of the pesticide's use in the area (c.1965-1975).The second contribution is roughly proportional to the total mobile environmental reservoir (MER) of the pesticide. As pointed out in COW, pesticides tend to be volatilized or converted to other forms in the environment (much like radionuclides). Thus a 'half-life' approximation is reasonable.

For DDT -- the longest-lived pesticide -- the half-life seems to be about 12 years (COW, Chapter 2). For other chlorinated pesticides, a figure like 5 years is probably more realistic.

We approximate the MER for DDT in year t by the following 12 year half-life formula:

$$MER(t) = \sum_{n=0}^{\infty} (.5)^{\frac{n}{12}} \overline{X}^{i}(t-n) \tag{7}$$

For DDT a single year extra contribution of 400 tonnes was assumed for 1957, the year of the massive spraying campaign against the gypsy moth in NY state forests.

For other pesticides we use the 5 year half-life formula

$$MER(t) = \sum_{n=0}^{\infty} (.5)^{\frac{n}{5}} \overline{X}^{i}(t-n) \tag{8}$$

The results of these calculations are given in Table II-3, Appendix II and summarized in Table 2-6.

The runoff load from MER will be roughly proportional to the size of MER at all times. Given a reasonably uniform pattern of use, the MER peaks in the last year of production and use, whence the runoff load also peaks in that year. During the year from introduction to withdrawal, there will be a gradual increase in the MER-contribution. Given the necessary crudeness of the model and the unreliability of year-to-year data on regional usage, a simple linear function will suffice for approximating the runoff contribution from MER during the period prior to the end of production and use. That is;

$$F^{i}_{MER}(t) = Pk^{i}MER^{i}(t) \tag{9}$$

and

$$F^{i}(t) = F^{i}_{CUR}(t) + F^{i}_{MER}(t) \tag{10}$$

Based on the assumed use and degradation rates noted above, we can derive runoff/MER ratios, k^i. These are shown in Table 2-7.

The "observed" runoff (column 2) includes a loss from concurrent surface uses, transport, or disposal of containers, normally assumed to be 1%. The very low indicated loss for heptachlor is almost certainly

Table 2-6: Estimated MER of Chlorinated Pesticides in the Hudson-Raritan Basin 1945-1980 (tonnes)

	DDT	BHC/ Lindane	Aldrin	Chlordane	Dieldrin	Endrin	Heptachlor	Toxaphene
Half-Life	12	5	5	5	5	5	5	5
Constant	.944	.871	.871	.871	.871	.871	.871	.871
1980	1255	93	2.7	813	11.2	2.8	93.0	29.5
1976	1581	151	4.8	1416	19.5	4.2	91.3	35.8
1971	2049	235	5.2	1632	28.6	5.1	89.1	37.8
1966	2275	390	4.6	1220	38.3	5.7	78.5	36.6
1964	2263	472	3.4	1124	39.4	5.7	73.0	35.4
1960	2103	651	1.8	1032	33.7	5.1	59.0	31.0
1955	1204	651	0.7	832	13.4	3.1	38.4	20.9
1950	619	214	0.1	431	0.4	0.6	10.5	5.2
1945	167	14		16				

Table 2-7: Pesticide & PCB Runoff from MER & Concurrent Uses in the Hudson-Raritan Basin in 1975 (tonnes)

Pesticide	(1) Mueller's Estimated H-R Runoff (1975)	(2) Contribution from Concurrent Use c^i	Amt	(3) MER (c1975)	(4) Contribution from MER (1) - (2)	Ratio Col 4 to Col 3 k^i
DDT	2.5(a)	(1%)	0	1676	2.5	.0015
BHC/Lindane	0.22	(1%)	.09	164	0.13	.0008
Chlordane	6.5	(1%)	1.3	1536	5.2	.0034
Dieldrin	.022	(1%)	.012	21	0.01	.0005
Heptachlor	.022	(.2%)	.022	91	0(b)	0
PCB	2.23		0	16,700(c)	2.23	.00013

(a) We did not use Mueller's (higher) figure for DDT runoff. Instead we use the data in Tables I-5 and I-6 in Appendix I.

(b) Used almost exclusively for control of underground termites, not contributing to runoff after application.

(c) Assuming a national MER of 223,000 tonnes (COW, Chapter 6) and assuming the Hudson-Raritan "share" is proportional to its population (~7.5%)

due to the fact that heptachlor is almost exclusively used for underground termite control. The annual loss rate for this use is negligible (See COW, Chapter 4), and runoff can presumably be related to handling, application and disposal of containers.

Lacking other data, we assume that aldrin and endrin contribute to runoff in the same proportion to MER as dieldrin. In the case of toxaphene, which has been used at a relatively constant level for the past

40 years, declining modestly since the mid-1970's, the simple 1% rule seems adequate, since any contribution from MER would still be proportional to current use. Our composite estimates, based on combining equations (8) and (9), are given in Table 2-8, as follows (year by year detail is shown in Appendix II, Table II-4).

Table 2-8: Pesticides in the Hudson-Raritan Estuary (tonnes/yr)

	DDT	BHC/ Lindane	Aldrin	Chlordane	Dieldrin	Endrin	Heptachlor	Toxaphene
1980	1.9	0.08	0.001	2.8	0.005	0.001	0.025	0.020
1976	2.4	0.21	0.006	5.6	0.021	0.005	0.023	0.041
1971	3.5	0.28	0.010	8.5	0.030	0.007	0.026	0.050
1966	4.6	0.50	0.014	6.2	0.064	0.010	0.025	0.052
1964	4.7	0.64	0.010	5.5	0.066	0.010	0.023	0.054
1960	5.0*	1.20	0.006	5.1	0.078	0.012	0.022	0.053
1955	3.4	2.04	0.003	4.4	0.067	0.011	0.019	0.053
1950	2.5	1.65	0.001	3.1	0.003	0.004	0.014	0.037
1945	1.1	0.10		0.21				

* There was a large single-year peak in 1957 due to Gypsy Moth spraying

In the case of PCB's, point sources, especially the upper basin have been quite important. They are summarized in Chapter 6 and COW, Chapter 6. The point source contribution peaked in 1973 at about 15 tonnes from the upper basin and 1.0 tonnes via sewage. The rate of buildup of MER has not been estimated in aggregate terms, since its chemical composition and the special uses of the materials have more influence on the contribution to runoff. Our summary estimate of historical loadings is shown in Table 2-9 . We combine sewage and runoff, since both are presumably proportional to MER.

Table 2-9: Estimated PCB Loading in the Hudson-Raritan Estuary (tonnes/yr)

Year	Upper Basin	Sewage & Runoff	Total
1980	1.0	1.5	2.5
1975	7.4	2.9	10.3
1973*	15.0	4.0	19.0
1970	1.8	5.0	6.8
1960	1.8	2.0	3.8
1950	1.8	1.0	2.8
1940	0	0.5	0.5

Source: Table 6-14 and text

*Peak year

PAH loadings in the estuary also arise both from point sources (mainly oil spills and bilge-washing in the harbor) and runoff. Currently, the major contributor to PAH in runoff is almost certainly waste motor oil. Unfortunately, historical estimation is complicated by the fact that prior to the 1950's coal (and coke) were widely used for space heating purposes. It is known that low temperature combustion and coking produce significant quantities of airborne PAH, some of which later appears in runoff. Unfortunately, we have no empirical data on which to base any particular assumption. We therefore guess that 25% of the airborne PAH will appear in runoff. Thus we have calculated runoff as the average (17.5%) estimate of PAH to sewers from Table 6-19 plus 25% airborne PAH from Table 6-17. The results are shown in Table 2-10.

Oil and grease loading in the estuary prior to 1970 is primarily attributable to point sources (oil spills and bilge washing), but there is now a significant contribution from waste motor oil in runoff. On land-use runoff factors derived from national average data (Appendix I, Table I-2), one would have expected about 15,000 tonnes of oil and grease in runoff (1975) as compared to our estimate of 58,000 tonnes of waste lube oil for the same year (Table 6-18). The latter should roughly coincide with the runoff figure. This comparison suggests that waste lube oil is about 4 times more likely to appear in runoff in the NY metropolitan area than in the nation as a whole. Such an inference seems quite reasonable, in view of the lack of open land in the area suitable for waste lube oil disposal purposes (either sanctioned or unsanctioned). All oil and grease data shown in Table 2-10 are summarized from Table 6-18.

Nitrogen, phosphorus and total organic carbon are similarly attributable to both point-sources (mainly sewage) and runoff. Historical estimates of point source contributions have been made in Tables 6-20 and 6-21. Runoff estimates are problematical. Using national runoff load factors (Appendix I) for agricultural land yields N and P estimates that are far too large to be credible. Accordingly, we have treated agricultural land in the Hudson-Raritan basin as being equivalent to developed land with separate sewage, for purposes of estimating N and P runoff, as shown in Table 2-2. Taking into account historical land use changes (Table I-1, Appendix I) and rainfall of 109 cm/yr, the results are as shown in Table 2-11.

The contribution from agriculture is the most uncertain and really depends strongly on the type of agriculture. The range of uncertainty is probably plus or minus 50% in a given year. Other runoff figures are probably within about 20%. Decade-to-decade comparisons are certainly much more accurate than

Table 2-10: Historical PAH and OG Loading in the Hudson-Raritan Estuary

	--- Total PAH (tonnes) ----			Oil and Grease (1000 tonnes)		
Year	Point Source (a)	Runoff (b)	Total	Point Source (c)	Runoff (d)	Total
1980	459	434	893	45	59	104
1970	4522	344	4866	507	57	564
1960	4387	273	4659	325	61	386
1950	3937	208	4145	234	53	287
1940	3397	202	3599	156	45	201
1930	2790	157	2947	128	45	173
1920	1192	126	1318	55	28.5	83
1900	405	53	458	18.5	4.5	23
1880	135	5	140	6.0	2.0	8

(a) Average of "low" and "high" figures in Table 6-19.

(b) Runoff includes average of low and high PAH to Sewers from Table 6-19 + 25% of airborne emissions from Table 6-17

(c) Sum of all categories other than lube oil from Table 6-18

(d) Lube oil from Table 6-18

Table 2-11: Historical Nitrogen & Phosphorus Loadings in the Hudson-Raritan Estuary (1000 tonnes)

	--------- Nitrogen --------			-------- Phosphorus ---------		
	Runoff	Point Sources	Total	Runoff	Point Sources	Total
1980	66.7	46.0	112.7	5.36	26.45	31.81
1970	67.8	50.1	117.9	5.56	33.86	39.42
1960	68.7	48.0	116.7	5.65	24.01	29.66
1950	69.0	42.0	111.0	5.68	14.87	20.55
1940	70.0	36.5	106.5	5.73	9.11	14.84
1930	67.2	33.5	100.7	5.48	8.37	13.85
1920	66.0	28.3	94.3	5.23	7.07	12.30
1900	68.0	15.8	83.8	5.28	3.96	9.24
1880	75.0	5.4	80.4	5.78	1.34	7.12

Table 2-12: Total Organic Carbon Loadings in the Hudson-Raritan Estuary (1000 tonnes)

	---------Runoff by Land Use Category--------				Sewage	
	Separate Sewers	Combined Sewers	Agri-cultural	Unde-veloped	(Point Source)	Total
1980	77.9	46.5	9.6	10.6	263.0	407.7
1970	53.8	68.9	9.8	11.7	551.8	696.0
1960	33.9	74.4	13.2	11.8	668.0	801.3
1950	16.9	75.5	16.7	11.8	607.7	728.6
1940	12.8	69.5	20.0	11.2	544.7	658.2
1930	7.7	61.6	20.7	11.4	501.7	603.0
1920	4.0	30.1	28.3	10.1	502.5	575.0
1900	0.8	8.5	36.1	8.5	335.0	388.9
1880	0	0	44.3	6.6	174.5	225.5

the numbers themselves. The estimates of TOC in sewage come from table 6-21. Runoff accounts for perhaps 1/3 of the total.

The production of waste heat is summarized in Table 6-23. There is no contribution from runoff. Table 6-23 is reproduced here for convenience as Table 2-13. An index number is used because we do not have good current information on the quantity of waste heat in the Hudson-Raritan basin *per se*, although relative changes from one decade to another are probably quite well approximated.

Table 2-13: Waste Heat Index

	Total U.S. Electricity Generated (million kwh)	Electric Power Generated in H-R basin (million kwh)	e: Thermal Efficiency	Waste Heat Index (normalized)
1980	2286.5	164.6	0.34	100.0
1970	1284.2	106.6	0.32	69.3
1960	607.7	53.5	0.31	35.7
1950	232.8	21.6	0.24	21.1
1940	94.0	9.1	0.20	10.9
1930	59.6	5.8	0.17	8.7
1920	23.5	2.1	0.09	6.6
1902	3(E)	0.3	0.07	1.2

3. Hudson-Raritan Regional Allocation of Production-Related Heavy Metal Emissions

3.1 Approach

The purpose of this chapter is to estimate anthropogenic heavy metal (HM) emissions in the Hudson-Raritan basin, over time.

The computation of emissions from production activities (i.e. metal smelters), (in principle,) is inherently region-specific. Indeed, the most straightforward approach is to identify major point sources in the region and directly compute emissions (subject to any abatement procedures or controls that may have been applied) as a function of output level. This is preferable if (and only if) one has emissions data specific to individual plants. (We do not have such data for the copper and lead smelters located in the Hudson-Raritan Basin). Alternatively, however, one can estimate the regional share of national emissions from these metallurgical processes as a proportion of national production. The emission computation proceeds in the same way as outlined above, using industry-average emission coefficients. As noted earlier, the source of all data used in this chapter is the separate volume entitled *Heavy Metals and Fossil Fuels* or HMFF.

The eight metals are considered hereafter as a single group because of the complex inter-relationships in their production and uses. All except chromium are obtained from sulfide ores. Arsenic is a by-product of copper ores (and is also found in iron ores and phosphate rocks); cadmium is a by-product of zinc ore; silver is a by-product of copper, zinc and lead ores; and copper, zinc and lead are all contaminants of each other's ores. On the use side, arsenic, copper, chromium, lead and mercury have major overlapping and competing pesticidal, fungicidal, and bactericidal uses; lead, cadmium, chromium and zinc have major overlapping uses as pigments; cadmium, chromium and zinc have overlapping and competing uses in metal plating; cadmium, mercury, zinc and silver are all used in electric batteries, and so on.

Major sources for several of the metals in the Hudson-Raritan basin in the past have been primary non-ferrous (Cu, Pb) smelters and refineries. All of the large metal refineries in the Hudson-Raritan basin are now closed. There may still be a few small secondary refiners. In the late 19th and early 20th century there was also a number of small iron/steel mills and foundries which presumably emitted significant quantities of trace metals, but these were mostly gone by WWII.

In addition, trace metals are emitted in significant quantities via the combustion of coal, oil (especially residual oil) and possibly wood. Though fuels are utilized for residential heating as well as for utility and industrial pruposes, we consider all fuel-related emissions in this chapter. In this category we include lead additives to gasoline and zinc additives to lubricating oil.

It must be pointed out that incineration of refuse and sewage sludge also results in heavy metal emissions. But this is an environmental transfer, not a true source of metallic pollutants. All of the metals emitted by incinerators must have been originally embodied in items of consumption discharged as wastes. Consumption related wastes are considered in the next chapter. Data on incinerator emissions is only relevant to the extent that it provides evidence of final disposal routes.

3.2 Emissions Coefficients for Production

Emissions coefficients and estimates abound in the literature [Nriagu 80; Nriagu 80a; Nriagu 80c; Nriagu 80z; Nriagu & Davidson 82; Watson & Brooks 79; NRC 77a; NRC 77c; NRC 81; GCA 73; GCA 81; URS 75; WEDavis 72; WEDavis 80; MRI 80; PEDCO 80, chapter 27; PEDCO 80, chapter 29; EPA 84; Nriagu 78; AD Little 76-xiv; Battelle 77; Lowenbach & Schlesinger 79; Ottinger et al 73; APCC 56; Hofman & Hayward 24]. Published numbers disagree spectacularly in some instances. Some of the EPA sponsored studies [such as NRC 77a; GCA 73; WEDavis 72 & 80] were admittedly crude, but -- being readily available -- have been widely quoted, especially in surveys such as those carried out by the National Academy of Sciences /National Research Council in the mid-1970's [NAS 80; NRC 77; NRC 77c; NRC 81] and also in the more recent series of symposium volumes edited by Nriagu [Nriagu 78, 80, 80a & 80z]. Reasons for disagreement among authors include use of questionable data in some early estimates, aggregation from non-representative samples, failure to distinguish between fundamentally different process stages (for example in the primary copper industry), failure to distinguish carefully between land-destined and other wastes, and failure to distinguish carefully between gross (uncontrolled) emissions and net emissions after the implementation of controls.

Unfortunately, some of the industry studies sponsored by EPA are almost useless for our purposes because they present a great deal of detailed but disparate and inconsistent data from which it is virtually impossible to generalize. This is particularly true of the EPA's "Industrial Process Profile" series. The only truly satisfactory approach for a complex industry like primary copper or lead is a combination of materials-balance and plant specific emissions data, as exemplified by *Survey of Cadmium Emission Sources* [GCA 81] in the case of cadmium. In the case of copper, the best source we have uncovered was published in 1924; it is the second edition of *Metallurgy of Copper* [Hofman & Hayward 24]. This book lacks only a discussion of more recent developments, most of which are irrelevant as far as the NJ-NY refineries are concerned. It is regrettable that the few such studies that have been done recently generally focussed on only one pollutant and neglected to estimate the overall effectiveness of controls in effect at the time of the study. This has made it very difficult to establish meaningful benchmarks. We have made our own rough estimates (discussed later).

Taking into account these factors, the sources we consider to be reasonably up-to-date and authorative are as follows: arsenic [Lowenbach & Schlesinger 79], cadmium [GCA 81], mercury [URS 75], and lead [EPA 84]. In the cases of silver, chromium, copper and zinc no single source suffices and a range of estimates will be used.

Table 3-1 and 3-2 summarize our assumed emissions coefficients for metallurgical activities and fossil fuel combustion, respectively. Table 3-3 shows our assumed particulate emission control efficiencies since 1880. The data for coal-fired utility boilers is derived from HMFF (See HMFF, Chapter 10 for fuller discussion). The dust and smoke control technologies first applied to utility boilers around 1915 were first applied to some non-ferrous smelters and refineries at an earlier date, partly because the inherent value of the flue dust (e.g. to recover arsenic and precious metals). Cottrell precipitators were used in addition to dust chambers in the copper industry prior to 1914 [Hofman & Hayward 24, pp 229-241]. The Balbach lead refinery in Newark achieved 90% particulate recovery using a Cottrell precipitator as early as 1915 [Hofman 18]; by 1933 virtually all copper smelters included Cottrell treaters obtaining better than 90% dust collection efficiency. Efficiencies cited in [Newton & Wilson 42, Table 5, p 245] included Garfield, Utah, (90%), Anaconda, Mont., (90%), Noranda, Quebec, (95%), and Cerro de Pasco, Peru, (97%). However, dust recovery efficiency from ferrous melting furnaces was much lower because of the low value of the recovered materials. We have very little data on this, however. The ranges in Table 3-3 are themselves estimates.

Table 3-1: Uncontrolled Emissions From Metallurgical Operations (ppm)

	Steel & Foundries	Smelt/ Convert Copper	Smelt/ Refine Lead	Secondary Copper	Secondary Lead	Secondary Zinc
Arsenic [NRC 77; L&S 79]	15.2	8000 (Refinery 800-900)				
Cadmium [GCA 81]	3.5-4	350-650	1750-2100	-		
Chromium [GCA 73]	6.5-7	-	-	-	-	-
Copper [Nriagu 80; WEDavis 72; PEDCO 80]	17.5-22.5	2500-5000	-	500-1000		
Mercury [URS 75]	-	26 Air 1 Water	9 Air 0.5 Water	-	-	-
Lead [Nriagu 78]	200-300	2000-3000 (U Refinery 25)	20000-23000	500-1000	20000-23000	-
Zinc [Nriagu 80z; WEDavis 80]	27-370	9000-11000	500-1000	500-1000	300	9000-11000

Table 3-2: Uncontrolled Emissions From Fossil Fuel Combustion (ppm)

	Coal	Residual Oil	Distillate Fuel Oil
Arsenic [NRC 77a, Lowenbach & Schlesinger 79]	0.10 (coal)	0.3	-
Cadmium [GCA 81]	0.88 (coal)	2.3	0.15
Chromium [GCA 73]	17 (coal) 260 (ash)	2.9 (oil) 1300 (ash)	-
Copper [WEDavis 72] [PEDCo 80]	15.6 (coal) 240 (ash) [Nriagu 80]	0.7 (oil) [Nriagu 80]	
Mercury [URS 75]	0.16	0.13	0.066
Lead [Nriagu 78]	4.5		
Zinc [Nriagu 80z, WEDavis 80]	4.8-8.5	.025 [Nriagu 80z]	

Table 3-3: Particulate Emission Control Efficiencies Over Time % of Particulates Removed, by Weight

	Copper & Lead Smelters	Other Smelters/ Melting Furnaces	Coal-Fired Utility Boilers
1980	99	97	99
1970	95 - 97	80 - 90	98
1960	94 - 96	50 - 75	97
1950	93 - 95	0 - 70	95
1940	92 - 94	0 - 65	90
1930	90 - 93	0 - 60	85
1920	80 - 90	0 - 50	60
1900	30 - 60	0	0
1880	0	0	0

Table 3-4 gives average national production by decade for seven categories of metals: steel, blister copper, primary refined copper, secondary refined copper (from old scrap), lead bullion, secondary refined lead, and secondary slab zinc. Table 3-5 gives national production capacity data (and estimates) for steel, primary copper smelting, primary copper refining and primary lead smelting. Table 3-6 gives comparable capacity data for the Hudson-Raritan basin, taken from unpublished Bureau of Mines files. Table 3-7 gives the derived or estimated Hudson-Raritan fraction of national capacity and emissions for each category of production, as estimated by authors.

A similar procedure is involved in estimating the Hudson-Raritan fraction of national emissions from combustion of coal and oil. Table 3-8 gives the average national consumption of bituminous coal (by use category), anthracite coal, residual oil and distillate fuel oil by decade. Data for 1880 is estimated on the basis of back extrapolation from the 1900 breakdown of coal uses. Table 3-9 gives our estimated Hudson-Raritan shares. The fossil fuel consumption data, by state, has been compiled (mostly from Bureau of Mines) by G. Gschwandtner et al [Gschwandtner 83]. We allocated fuel consumption in NJ and NY to the Hudson-Raritan basin in proportion to the region's share of each state's population, except for certain adjustments noted in the table. State-by-state or categorical breakdowns do not exist prior to 1920 in general (and 1930 for some categories). Earlier figures are estimated by backward trend extrapolation and judgment. (See footnotes to the table).

The Hudson-Raritan share of nationwide gasoline consumption is not nowadays in proportion to its population due to the congestion of the New York metropolitan area, the high cost of insurance and parking and the availability of public transportation [Jones et al 74]. The share figures in Table 3-9 are computed from gasoline consumption for NJ and NY by assuming that the part of the population of each state *not* living in the Hudson-Raritan basin consumes gasoline at the national average per capita rate. When this amount is subtracted from the total consumed by each state, respectively, the remainder is the (assumed) amount of gasoline consumed within the basin. It is noteworthy that, while residents of the area consumed their *per capita* share of gasoline in 1930 (and earlier), the current Hudson-Raritan level is barely 62% of the national average.

The computation of emissions from coal and fuel oil combustion now proceeds in a straightforward fashion by using the gross emissions coefficients (Tables 3-1, 3-2), assumed control efficiencies (Table 3-3), national average annual metals production data (Table 3-4), national average fossil fuel consumption data (Table 3-8), and The Hudson-Raritan fractions of each (Tables 3-7, 3-9).

In the case of tetra-ethyl lead (TEL) emissions from gasoline, we do not compute emissions from an emission coefficient. Instead, data on lead use as a gasoline additive is taken directly from HMFF (Vol II, Table 8-3, col q) and multiplied by a factor of .75 to reflect the fact that at least 25% of the lead is trapped in the oil, oil filters or exhaust system of the cars and not emitted directly to the atmosphere [Hirschler et al 57; Hirschler & Gilbert 64].

3.3 Metallurgical Emissions in the Hudson-Raritan Basin

It is reasonable to assume that emissions related to metallurgical activities are and have been regionally distributed in proportion to the regional distribution of the activities themselves. As it happens, the Hudson-Raritan Basin has had a very minor share of the U.S. iron/steel industry (except for a few years after 1880 when Albany was briefly a steel producer). However, both the copper and lead refining industries were highly concentrated in the Hudson-Raritan basin from the 1890's until recently. In the case of copper this was due in part to the early dominance of NY as an electro-metallurgical center [Trescott 81] and to the convenience of NY Harbor as a site for processing imported copper ores from Chile for re-export as finished products [Navin 78]. Most of the important copper-using industries were also centered in the area, including copper wire manufacturers such as General Cable, and brass mills in nearby Connecticut. These industries also provided a major market for lead (as cable sheathing). NY also provided a big market for lead pigments. Later the electric battery manufacturers such as Electric Storage Battery Co. (ESB) located plants nearby.

Table 3-4: Average Annual U.S. Production of Metals (1000 tonnes)

	Total Raw Steel Production	Total Copper Smelter Output	Primary Copper Refinery Output	Copper Recovered From Old Scrap	Total Lead Smelter Output	Secondary Lead, inc. Refined From For. Bullion	Slab Zinc From Scrap
Table Ref.	HMFF	6-1 c2	6-1 c3	6-1 c4	8-1 c2	8-1 c5	9-1 c3
1980	100250	1162.6	1271.2	567.1	539.2	691.7	48.4
1970	119371	1362.1	1525.9	454.2	489.8	538.1	69.3
1960	86060	900.8	1293.9	388.0	366.3	403.5	52.1
1950	74295	792.9	1026.9	407.7	414.2	435.2	52.3
1940	58133	768.9	1075.1	313.5	460.6	284.0	44.2
1930	37500	617.8	867.9	292.9	530.4	277.7	30.4
1920	35176	531.6	714.6	151.1	477.8	159.7	
1900	11619	268.8	241.9	26.9	309.7	225.0	
1880	1268	27.4	24.7	2.7	102.8		
	(b)		(d)	(e)		(g)	

All data are five year averages centered on year shown, except as indicated below

(b) source is Historical Statistics, Business Statistics (5 yr avgs)

(d) 1880,1900 estimated at 90% smelter output (remainder=old scrap)

(e) Assumes secondary production is 10% of smelter output in 1880 and 1900 (probably too high)

(g) 1900 estimated from Census data for 1899 (lead refined from imported bullion)

Following the approach described previously, we calculate emissions in the Hudson-Raritan basin as regional fractions of national totals for the metallurgical industries. This, in turn, requires data on time-averaged national production levels by decade, and data on regional production share (calculated on the basis of regional capacity shares). The results are shown in Tables 3-10 and 3-11. Note that high and low estimates in Table 3-10 are based on high and low estimates for emission coefficients in Table 3-1 combined with low and high estimates for particulate controls in Table 3-3. High and low estimates for zinc in Table 3-11 correspond to the range of emission coefficients for zinc in Table 3-2.

Table 3-5: U.S. Metallurgical Production Capacity: 1880-1980 (1000 tonnes)

	Raw Steel	Primary Copper Smelting (Blast or Reverberatory Furnace)	Primary Copper Refining	Primary Silver-Lead Smelting
1980	139344	1687 (79)	2177 (c)	783(79)
1970	140614 (a)	1778 (74)	2722 (74)	739 (c)
1960	134781	1315 (c)	1715 (c)	626 (c)
1950	90703	939	1412	570
1940	74043	939 (e)	1426	772
1930	64448	1030 (e)	1386	699
1920	54631	984 (e)	1256 (d)	662 (e)
1900	21116 (01)	313 (e)	357 (d)	376 (e)
1880	1985 (b)	35 (e)	35	

(a) Assuming 1970 capacity utilization of 85%

(b) Assuming capacity utilization of 0.5 (based on data for 1887) and 1880 output of 1,110,300 tonnes (See HMFF, Chapter 11 and Appendix E)

(c) Assuming maximum capacity utilization of 0.77 in year of peak production during prior 10 year period. The 0.77 figure was estimated on the basis of typical capacity utilization data for the earliest period of available data. For years before 1906 assumes Cu smelter/refinery capacity ratio of 0.85 (approximately true for later periods).

(d) Assuming capacity utilization of 0.77 in 1907 and 1900 output of 270,000 tonnes, 1880 output of 27,400 tonnes

(e) Assuming maximum capacity utilization of 0.88 in prior ten year period

(01) Year of datum (74) Year of datum (79) Year of datum

Table 3-6: Metallurgical Production Capacity - Hudson-Raritan Basin: 1880-1980 (1000 tonnes)

	Raw Steel Capacity (All Types)	Primary Copper Smelter Capacity (a)	Primary Copper Refining Capacity (Based on Refined Output)	Primary Silver-Lead Refining Capacity	Secondary Lead Recovery Capacity
1980	154	0	352	0	69 (g)
1970	0	163	683	0	54 (g)
1960	69	404	775	0	87
1950	41	363	679	0	131
1940	182	290	662	0	158
1930	138	290	766	150 (d)	143 (f)
1920	249	354	662	154 (d)	98 (f)
1900	122	47 (b)	139 (c)	1.4 (e)	91 (f)
1880	113	5.3 (b)	14 (c)		

Source: Gutmanis from U.S. Bureau of Mines

(a) Capacity based on ore throughput -- assumed 10% Cu

(b) Based on 1899 ratio of NJ-NY smelter output to total U.S. output

(c) Based on 1899 ratio of NJ-NY refinery output to total U.S. output

(d) Based on smelter charge (BuMines data)

(e) Assuming 1899 ratio of NJ smelter output to U.S. total

(f) Assuming lead classed as "imported ores" was actually imported bullion, refined in NJ

(g) Assuming 10% of national secondary recovery is in the Hudson-Raritan Basin

Table 3-7: Hudson-Raritan Fraction of Metallurgical Emissions

	Steel (a)	Total Smelter Copper (a)	Primary Refinery Copper (a)	Secondary Refinery Copper (b)	Smelter Lead (a)	Secondary Lead-incl. imp.Bullion (c)	Secondary Slab Zinc (b)
1980	0.0011	0	0.16	0.10	0	0.10	0
1970		0.01	0.25	0.10	0	0.10	0.10
1960	0.0005	0.03	0.45	0.10	0	0.14	0.10
1950	0.0005	0.04	0.48	0.10	0	0.21	0.10
1940	0.0025	0.03	0.46	0.10	0	0.47	0.10
1930	0.0021	0.03	0.55	0.10	0.21	0.31	0.10
1920	0.0045	0.036	0.53	0.10	0.23	0.63	0.10
1900	0.0061	0.015	0.39	0.10	0.35	0.32	0.10
1880	0.0570	0.015	0.39	0	0	0	

(a) Based on ratio of Hudson-Raritan capacity to U.S. capacity (Bureau of Mines data)

(b) In the absence of useable data, we assume NY-NJ accounts for 10% of secondary copper and secondary zinc. This may be an underestimate for copper and an over-estimate for zinc, but final results are insensitive to these assumptions.

(c) Assuming refineries of lead not smelted (including imported bullion) in this category. Perth Amboy, NJ is still one of the three areas of highest concentration of secondary lead smelters. However, the industry is now decentralized, with over one hundred plants scattered around the U.S. Figures for 1920-1960 based on capacity data on lead refineries, assuming capacity utilization, of 0.7(1920), 0.5(1930) and 0.7(1940,1960). Figures for 1970,1980 are a guess.

Table 3-8: Average Annual Combustion of Fossil Fuels, by Decade (million tonnes)

	Bituminous Coal, Electric Utility	Bituminous Coal, Class I Railroads	Bituminous Coal, Industry (d)	Bituminous Coal, Coke Product. (e)	Bituminous Coal, Resid/ Commer. (f)	Anthracite Coal	Residual Fuel Oil	Distillate Fuel Oil	Direct TEL in gasoline (g)
1980	501.25	0.00	57.84	57.39	5.81	3.41	128.94	160.19	0.118
1970	290.04	0.00	76.77	81.60	11.26	8.01	114.91	137.60	0.186
1960	156.59	1.53	84.14	69.84	27.41	15.63	79.80	99.45	0.111
1950	85.22	57.32	108.27	93.16	72.73	36.50	77.51	57.82	0.079
1940	45.58	81.81	119.15	69.95	75.57	43.87	51.35	23.21	0.030
1930	35.96	85.80	125.31	57.03	83.54	57.78	50.68	30.32	0.003
1920	31.30	110.75	130.68	57.76	87.12	72.46	30.48	25.92	
1900	7.60	57.20	67.20	30.20	28.10	49.29	1.08	2.82	
1880	1.97	14.81	17.39	7.82	7.27	12.76		0.05	

(d) Includes cement, steel, rolling mills and other

(e) Includes gas manufacture

(f) Residential and commercial establishments (retail distributors)

(g) Lead content of tetra-ethyl lead (TEL) in gasoline, not gasoline consumed. See HMFF, Chapter 8.

Table 3-9: Hudson-Raritan Fraction of Emissions from Combustion of Fossil Fuels

	Bituminous Coal, Electric Utilities (a)	Bituminous Coal, Class I Railroads (b)	Bituminous Coal, Industry (c)	Bituminous Coal, Resid/ Commer. (c)	Bituminous Coal, Coke Production (d)	Anthracite Coal	Residual Fuel Oil	Distillate Fuel Oil (f)	Gasoline
1980	0.006	0.000	0.003	0.000	0.000	0.000	0.106	0.064	0.045
1970	0.014	0.065	0.011	0.008	0.022	0.036	0.176	0.114	0.052
1960	0.058	0.090	0.064	0.030	0.027	0.205	0.121	0.128	0.065
1950	0.086	0.090	0.044	0.020	0.029	0.324	0.088	0.054	0.062
1940	0.097	0.110	0.052	0.051	0.029	0.347	0.070	0.060	0.083
1930	0.124	0.100	0.070	0.069	0.029	0.195	0.044	0.075	0.096
1920	0.107	0.100	0.080(c)	0.080(c)	0.044	0.244	0.090(g)	0.090(g)	
1900	0.120(a)	0.100	0.080(c)	0.080(c)	0.051	0.210	0.080(g)	0.080(g)	
1880	0.150(a)	0.100	0.070(c)	0.070(c)	0.170	0.250(e)	0.072(g)	0.072(g)	

(a) After 1960 utilities in NYC switched from coal to oil to reduce emissions. Prior to 1920 it can be assumed that the urbanized Hudson-Raritan basin consumed at least 20% more electricity per capita than the nation as a whole and that all of it was generated from coal-fired boilers.

(b) We assumed that the Hudson-Raritan basin accounted for 50% of the fuel consumption by railroads in NY and 75% of the fuel consumed by railroads in NJ. (This may be an over-estimate, especially from 1940 on because all trains serving NYC directly were electrified by that time.) Prior to 1920 the same percentage is assumed.

(c) Usage per capita was slightly below the national average in 1930. We extrapolate back to usage at the average per capita level in 1880.

(d) Excluding coke production for the iron and steel industry in western NY (Bethlehem Steel Co's Lackawanna Works).

(e) Author's estimate

(f) Not separately analysed prior to 1930

(g) Hudson-Raritan share assumed proportional to population.

Table 3-10: Average Annual Emissions from Metallurgical Operations
Hudson-Raritan Valley (tonnes)

	Arsenic		Cadmium		Chromium		Copper		Mercury		Lead		Zinc	
	low	high	low	high	low	high	low	high	low	high	low	high	low	high
1980	4.9	5.5	.0	.0	0.02	0.02	0.9	1.8	0.00	0.00	43	51	1.6	3.5
1970	33.8	74.1	0.1	0.4	0.00	0.00	3.3	12.5	0.01	0.02	112	261	13.8	35.1
1960	125.3	275.3	0.4	1.1	0.07	0.15	7.7	28.0	0.03	0.04	295	688	30.8	82.3
1950	131.2	462.0	0.6	1.6	0.07	0.26	10.3	52.7	0.04	0.06	564	2173	43.0	163.8
1940	150.3	462.1	0.7	1.8	0.33	1.02	9.8	43.8	0.04	0.05	957	3163	47.3	194.1
1930	163.6	445.6	14.2	24.9	0.20	0.55	9.7	40.3	0.11	0.16	864	2306	43.5	149.2
1920	168.0	373.9	20.2	49.3	0.51	1.11	9.9	37.8	0.16	0.31	1254	2902	43.7	167.9
1900	89.5	108.6	1.6	3.7	0.46	0.50	6.6	18.4	0.05	0.08	2328	3436	39.6	82.3
1880	12.1	13.1	0.4	0.6	0.47	0.51	2.3	3.7	0.01	0.01	16	23	5.7	31.3

Table 3-11: Metallic Emissions due to Combustion of Fossil Fuels - Hudson-Raritan Basin (tonnes)

	Arsenic	Cadmium	Chromium	Copper	Mercury	Lead	Zinc low	Zinc high
1980	4.1	31.5	41.7	10.1	.0	5312.4	0.5	0.6
1970	6.1	46.6	63.4	16.4	0.1	9684.3	1.2	1.7
1960	3.0	22.8	40.4	16.4	0.2	7203.1	3.2	5.5
1950	2.2	17.2	48.5	30.7	0.3	4884.2	8.1	14.3
1940	1.5	11.9	79.9	66.0	0.7	2504.3	19.6	34.7
1930	1.3	10.5	110.1	96.3	1.0	266.4	29.2	51.7
1920	2.9	24.6	362.4	326.8	3.5	73.9	100.0	177.1
1900	2.6	23.2	444.8	408.0	4.2	91.9	125.5	222.3
1880	0.8	7.1	136.4	125.1	1.3	29.4	38.5	68.2

4. Regional (Hudson-Raritan) Share of Consumption-Related Heavy Metal Emissions

4.1 Approach

The regional-share approach is the only possible one for consumption-related emissions. In most cases consumption must first be subdivided into source subcategories. There are two reasons for this. In the first place different commodities or products (e.g. anthracite coal vs natural gas) may have very different regional distribution patterns.

In the second place, and just as important, the emissions resulting from dissipative end-uses may be radically different from one source category to another. (An obvious illustration: tetraethyl lead from auto exhaust is rapidly dispersed into a highly mobile form and a high proportion of it evidently appears in urban runoff; on the other hand lead used in batteries, bullets, cable covering or solder is scarcely mobilized at all and little or none appears in runoff).

In the case of the heavy metals (Ag, As, Cd, Cr, Cu, Hg, Pb, Zn) we have found that there are ten categories of consumption that are readily distinguishable in terms of their different degrees of dissipation in use and different modes of release to the environment. These are as follows:

1. Metallic uses, e.g. in alloys. Environmental losses occur mainly in the production stage (discussed previously) and as a result of corrosion in use or discharge to landfills.
2. Plating and surface treatment (excluding paints and pigments) generate some losses in the platings or treatment process and some corrosion losses as above.
3. Paints and pigments generate losses at the point of application and from weathering and wear. Some are ultimately disposed of (e.g. in landfills) along with discarded objects or building materials.
4. Batteries and electronic devices have relatively short useful lives I-I0 years. Production losses can be significant. Most are discarded to landfills.
5. Other electrical equipment as above, but may be longer lived.
6. Industrial chemicals and reagents (e.g. catalysts, colvents, etc) not embodied in products have short useful lives, catalysts and solvents are usually recycled, others are lost directly to air or water.
7. Chemical additives to consumer products include fuel additives,[3] rubber vulcanizing agents and pigments, detergents, plasticizers, photographic film, etc. They are disposed of mainly to landfill or incinerators. There is no recycling.

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[3] Actually, we have already considered tetraethyl lead (TEL) in the context of fuel consumption. It is, therefore, omitted from the calculations hereafter.

8. Agricultural pesticides, fungicides and herbicides are used dissipatively, on farms, nurseries, etc. Most are immobilized by soil or biologically degraded and volatilized. There is some uptake into the food chain and a small amount of loss via runoff.
9. Non-agricultural biocides, include the above, as used in homes and gardens, for termite control, etc. These uses are dissipative but most biocides are immobilized by soil, as above.
10. Pharmaceuticals, germicides, etc. are used in the home or in health service facilities and are largely discharged via sewage or to incenerators.

4.2 Emission Coefficients

The term "emission coefficient", as used in this context, means the fraction of the material in question that is released in mobile form within a decade (more or less). We exclude wastes that are recycled or disposed of in landfills or in sludge dumped offshore. In a few cases we also include production-related losses that were not included in the previous chapter (e.g. process wastes in the plating, tanning and chemical industries).

It is unfortunate (and curious) that there is almost no published data on emissions coefficients for consumption activities. Obviously, most analysts have not considered such activities to be "sources" of pollutants.[4]

In the absence of a body of literature (and of time to undertake more exhaustive research on this topic ourselves) we are led to a rather *ad hoc* choice of emissions coefficients. These are displayed in Table 4-1, together with explanatory notes. See HMFF for more extended discussion of consumptive uses and processes.

It must be pointed out that, while the numerical estimates in many cases are very uncertain--sometimes even by a factor of two or three--there are only a few important routes which clearly dominate the rest for each metal. In general, only the consumption categories with "large" coefficients would need to be considered in greater detail in a more refined future version of this study.

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[4] On the other hand, sewage treatment and waste incineration have inconsistently been so treated.

Table 4-1: Consumption-Related Emissions Factors

Metal	Metallic Use (a)	Plating & Coating (b)	Paint & Pigments (c)	Electron Tubes & Batteries (d)	Other Electrical Equipment (e)	Chemical Uses, Not Embodied (f)	Chemical Uses, Embodied (g)	Agricultural Uses (h)	Non Agricultural Uses (i)	Medical, Dental (j)	Misc. (NEC) (k)
Ag Silver	0.001	0.02	0.5	0.01	0.01	1	0.4	na	na	0.5	0.15
As Arsenic	0.001	0	0.5	0.01	na	na	0.05	0.5	0.8	0.8	0.15
Cd Cadmium	0.001	0.15	0.5	0.02	na	1	0.15	na	na	na	0.15
Cr Chromium	0.001	0.02	0.5	na	na	1	0.05	na	1	0.8	0.15
Cu Copper	0.005	0	1.0	na	0.1	1	0.05	0.05	1	na	0.15
Hg Mercury	0.05	0.05	0.8	0.2	na	1	na	0.8	0.9	0.2	0.5
Pb Lead	0.005	0	0.5	0.01	na	1	0.75	0.05	0.1	na	0.15
Zn Zinc	0.001	0.02	0.5	0.01	na	1	0.15	0.05	0.1	0.8	0.15

Notes for Table 4-1

(a) As alloys or amalgams (in the case of Hg) not used in plating, electrical equipment, catalysts or dental work. Losses can be assumed to be due primarily to wear and corrosion, primarily the latter, except for mercury which volatilizes.

(b) Protective surfaces deposited by dip coating (e.g. galvanizing, electroplating vacuum deposition, or chemical bath (e.g. chromic acid). The processes in question generally resulted in significant waterborne wastes until the 1970's. Cadmium plating processes were particularly inefficient until recently (see discussion in HMFF). Losses in use are mainly due to wear and abrasion (e.g. silverplate), or flaking (decorative chrome trim). In the case of mercury-tin "silver" for mirrors, the loss was largely due to volatilization.

(c) Paints and pigments are lost primarily by weathering (e.g. for metal-protecting paints), by wear, or by disposal of painted dyes or pigmented objects, such as magazines. Copper and mercury based paints slowly volatilizes overtime. A factor of 0.5 is rather arbitrarily assumed for all other paints and pigments.

(d) Includes all metals and chemicals (e.g. phosphorus) in tubes and primary and secondary batteries, but excludes copper wire. Losses in manufacturing may be significant. Mercury in mercury vapor lamps can escape to the air when tubes are broken. In all other cases it is assumed that discarded equipment goes mainly to landfills. Minor amounts are volatilized in fires or incinerators or lost by corrosion lead-acid batteries are recycled.

(e) Includes solders, contacts, semiconductors and other special materials (but not copper wire) used in electrical equipment control devices, instruments, etc. Losses to the environment primarily via discard of obsolete equipment to landfills. Mercury used in instruments may be lost via breakage and volatilization or spillage.

(f) Chemical uses not embodied in final products include catalysts, solvents, reagents, bleaches, etc. In some cases a chemical is basically embodied but there are some losses in processing. Losses in chemical manufacturing per se are included here. Major examples include copper and mercury catalysts (especially in chlorine mfg.) copper, zinc and chromium as mordants for dyes; mercury losses in felt manufacturing; chromium losses in tanning; lead in desulfurization of gasoline; zinc in rayon spinning; etc. In some cases annual consumption is actually makeup and virtually all of the material is actually dissipated. We include detonators such as mercury fulminate and lead azide (and explosives) in this category.

(g) Chemical uses embodied in final products other than paints or batteries include fuel additives (e.g. TEL), anti-corrosion agents (e.g. zinc dithiophosphate), initiators and plasticizers for plastics (e.g. zinc oxide), etc. Also includes wood preservatives and chromium salts embodied in leather. Losses to the environment occur when the embodying productivity is utilized, for example gasoline containing TEL is burned and largely (0.75) dispersed into the atmosphere. However copper, chromium and arsenic are used as woood preservatives and dispersed only if the wood is later burned or incinerated. In the case of silver(photographic film) we assume that 60% is later recovered.

(h) Agricultural pesticides, herbicides, and fungicides. Uses are dissipative but heavy metals are largely immobilized by soil. Arsenic and mercury are exceptions because of their volatility.

(i) Non-agricultural biocides are the same compounds, used in industrial, commercial or residential applications. Loss rates are higher in some cases.

(j) Medical/dental uses are primarily pharmaceutical (including cosmetics) germicides, etc. Also dental filling material. Most are dissipated to the environment via waste water. Silver and mercury dental fillings are likely to be buried with cadavers.

4.3 Historical Usage Patterns

The next step is to allocate total domestic usage of each of the 8 metals among the I0 categories, over the past I00 years. The allocation is far from unchanging. Many formerly important uses have disappeared, while others have emerged as recently as the last decade. Consumption data by use is available, in general, only since WWII. For earlier periods one must rely on a scattering of real data supplemented by a variety of other clues. Our composite picture of historical heavy metals usage patterns for the whole United States is summarized in Tables 4-2 through 4-37, at the end of the chapter. A set of five tables is given for each metal, arranged as follows:

1. % of metal use by consumptive category
2. consumption in tonnes (U.S.)
3. emissions due to consumptive use (U.S.)
4. consumption in tonnes (Hudson-Raritan Basin)
5. emissions due to consumptive use (Hudson-Raritan Basin)

4.4 Regional Distribution of Metals Consumption

In principle, regional distributions patterns for intermediate products could vary almost as much as types of usage, as consuming industries grow or decline or shift locations. However, on reflection, it is clear that industrial locational patterns actually tend to be quite stable. Moreover final consumption for most products tends to be (more or less) proportional to population. The Hudson-Raritan region, in particular, is a fairly large microcosm of the whole industrial landscape of the U.S. in many ways; although it has less than it proportional "share" of certain industries (e.g. ferrous metals, petrochemicals, auto manufacturing) and more than it proportional share of others (including copper and lead refining) [See Hall 59]. In a more elaborate study, the regional share of intermediate consumption could be computed with the help of a regionalized I-O model. However, in the present case we assume that consumption and consumption-related emissions are proportional to regional (Hudson-Raritan) population share.

The fourth and fifth tables for a given metal in Tables 4-2 through 4-41 display respectively Hudson-Raritan consumption in tonnes (4th table) and Hudson-Raritan emissions, in tonnes (5th table).

Table 4-2: Silver: Consumptive Uses -% Use by Category

			Electrical		Chemical		Biocidal Poison Uses			
Year	Metallic Uses (Except Electrical) + Protective Cover: Plating & Coating	Protective Cover: Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.
1980	26	0	4.8	22	2.5	40	0	0	1	3.0
1972	42	0	4.0	24	2.2	25	0	0	1	1.6
1967	42	0	4.0	21	2.0	26	0	0	2	3.0
1958	57	0	2.0	11	2.0	22	0	0	2	7.0
1942	70	0	0	7	3.0	15	0	0	2	3.0
1929	75	0	0	3	3.0	15	0	0	4	0
1919	80	0	0	0	1.5	12	0	0	6	0
1900	85	0	0	0	0	5	0	0	5	5.0
1880	90	0	0	0	0	0	0	0	5	5.0

Table 4-3: Silver: Consumptive Uses, United States (tonnes)

			Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Electrical) + Protective Cover: Plating & Coating	Protective Cover: Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	1096	0	202	927.4	105.4	1686.1	0	0	42	126.5	4215.3
1972	1819	0	173	1039.2	95.3	1082.5	0	0	43	69.3	4329.9
1967	1318	0	126	658.9	62.8	815.8	0	0	63	94.1	3137.7
1958	1790	0	63	345.4	62.8	690.8	0	0	63	219.8	3140.1
1942	1233	0	0	123.3	52.8	264.2	0	0	35	52.8	1761.2
1929	817	0	0	32.7	32.7	163.5	0	0	44	0	1089.9
1919	906	0	0	0	17.0	135.9	0	0	68	0	1132.1
1900	1005	0	0	0	0	59.1	0	0	59	59.1	1181.9
1880	420	0	0	0	0	0	0	0	23	23.3	466.6

Table 4-4: Silver: Emissions from Consumptive Uses, United States (Tonnes)

			Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Electrical) + Protective Cover: Plating & Coating	Protective Cover: Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	11.508	0	2.023	9.27	105.4	674.4	0	0	21.08	6.323	830.03
1972	19.095	0	1.732	10.39	95.3	433.0	0	0	21.65	3.464	584.58
1967	13.837	0	1.255	6.59	62.8	326.3	0	0	31.38	4.707	446.84
1958	18.793	0	0.628	3.45	62.8	276.3	0	0	31.40	10.990	404.40
1942	12.945	0	0	1.23	52.8	105.7	0	0	17.61	2.642	192.94
1929	8.583	0	0	0.33	32.7	65.4	0	0	21.80	0	128.80
1919	9.510	0	0	0	17.0	54.3	0	0	33.96	0	114.79
1900	10.548	0	0	0	0	23.6	0	0	29.55	2.955	66.69
1880	4.409	0	0	0	0	0	0	0	11.67	1.166	17.24

Table 4-5: Silver: Consumptive Uses, Hudson-Raritan Basin (Tonnes)

			Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Electrical) + Protective Cover: Plating & Coating	Protective Cover: Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	79	0	14.53	66.59	7.57	121.07	0	0	3.03	9.08	300.55
1972	154	0	14.64	87.86	8.05	91.52	0	0	3.66	5.86	365.36
1967	117	0	11.11	58.34	5.56	72.23	0	0	5.56	8.33	277.81
1958	167	0	5.86	32.21	5.86	64.43	0	0	5.86	20.50	301.64
1942	119	0	0	11.91	5.10	25.52	0	0	3.40	5.10	170.15
1929	79	0	0	3.17	3.17	15.84	0	0	4.22	0	105.61
1919	81	0	0	0	1.52	12.15	0	0	6.08	0	100.76
1900	79	0	0	0	0	4.63	0	0	4.63	4.63	92.56
1880	30	0	0	0	0	0	0	0	1.69	1.69	33.89

Table 4-6: Silver: Emissions from Consumptive Uses, Hudson-Raritan Basin (Tonnes)

			Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Electrical) + Protective Cover: Plating & Coating	Protective Cover: Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	0.826	0	0.145	0.666	7.57	48.43	0	0	1.51	0.454	59.60
1972	1.614	0	0.146	0.879	8.05	36.61	0	0	1.83	0.293	49.43
1967	1.225	0	0.111	0.583	5.56	28.89	0	0	2.78	0.417	39.56
1958	1.753	0	0.059	0.322	5.86	25.77	0	0	2.93	1.025	37.72
1942	1.251	0	0	0.119	5.10	10.21	0	0	1.70	0.255	18.64
1929	0.832	0	0	0.032	3.17	6.34	0	0	2.11	0	12.48
1919	0.851	0	0	0	1.52	4.86	0	0	3.04	0	10.27
1900	0.826	0	0	0	0	1.85	0	0	2.31	0.231	5.22
1880	0.320	0	0	0	0	0	0	0	0.85	0.085	1.25

Table 4-7: Arsenic: Consumptive Uses -% Use by Category

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses			
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.
1980	2	0	0	1	0	5	0	30	60	1	0
1970	2	0	0	1	0	4	0	45	45	1	0
1960	2	0	0	1	0	5	0	80	10	2	0
1950	2	0	0	1	0	5	0	80	10	2	0
1940	2	0	0	1	0	5	0	80	10	2	0
1930	2	0	0	0	0	5	1	80	10	2	0
1920	2	0	2	0	0	5	3	75	10	3	0
1900	0	0	5	0	0	10	10	60	10	5	0
1880	1	0	10	0	0	10	10	40	20	10	0

Table 4-8: Arsenic: Consumptive Uses, United States (tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	0.393	0	0	0.197	0	0.983	0	5.901	11.801	0.197	0	19.67
1970	0.513	0	0	0.256	0	1.025	0	11.532	11.532	0.256	0	25.63
1960	0.358	0	0	0.179	0	0.895	0	14.315	1.789	0.358	0	17.89
1950	0.369	0	0	0.185	0	0.923	0	14.762	1.845	0.369	0	18.45
1940	0.592	0	0	0.296	0	1.479	0	23.663	2.958	0.592	0	29.58
1930	0.415	0	0	0	0	1.037	0.207	16.590	2.074	0.415	0	20.74
1920	0.253	0	0.253	0	0	0.633	0.380	9.494	1.266	0.380	0	12.66
1900	0	0	0.190	0	0	0.380	0.380	2.281	0.380	0.190	0	3.800
1880	0.012	0	0.123	0	0	0.123	0.123	0.494	0.247	0.123	0	1.230

Table 4-9: Arsenic: Emissions from Consumptive Uses, United States (Tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	0.039	0	0	1.97	0	492	0	2950	5901	19.7	0	9364
1970	0.051	0	0	2.56	0	513	0	5766	5766	25.6	0	12072
1960	0.036	0	0	1.79	0	447	0	7158	895	35.8	0	8537
1950	0.037	0	0	1.85	0	461	0	7381	923	36.9	0	8804
1940	0.059	0	0	2.96	0	739	0	11832	1479	59.2	0	14112
1930	0.041	0	0	0	0	518	166	8295	1037	41.5	0	10058
1920	0.025	0	127	0	0	316	304	4747	633	38.0	0	6164
1900	0	0	95	0	0	190	304	1140	190	19.0	0	1939
1880	0.001	0	62	0	0	62	99	247	123	12.3	0	605

Table 4-10: Arsenic: Consumptive Uses, Hudson-Raritan Basin (Tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	0.028	0	0	0.014	0	0.071	0	0.424	0.847	0.014	0	1.398
1970	0.043	0	0	0.022	0	0.087	0	0.975	0.975	0.022	0	2.123
1960	0.032	0	0	0.016	0	0.079	0	1.267	0.158	0.032	0	1.584
1950	0.034	0	0	0.017	0	0.086	0	1.377	0.172	0.034	0	1.721
1940	0.057	0	0	0.029	0	0.143	0	2.286	0.286	0.057	0	2.858
1930	0.040	0	0	0	0	0.100	0.020	1.608	0.201	0.040	0	2.010
1920	0.023	0	0.023	0	0	0.057	0.034	0.849	0.113	0.034	0	1.132
1900	0	0	0.015	0	0	0.030	0.030	0.179	0.030	0.015	0	0.298
1880	0.001	0	0.009	0	0	0.009	0.009	0.036	0.018	0.009	0	0.091

Table 4-11: Arsenic: Emissions from Consumptive Uses, Hudson-Raritan Basin (Tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	0.0028	0	0	0.141	0	35.3	0	212	424	1.412	0	672.4
1970	0.0043	0	0	0.217	0	43.3	0	488	488	2.167	0	1020.7
1960	0.0032	0	0	0.158	0	39.6	0	634	79	3.169	0	755.9
1950	0.0034	0	0	0.172	0	43.0	0	688	86	3.442	0	821.1
1940	0.0057	0	0	0.286	0	71.4	0	1143	143	5.715	0	1363.4
1930	0.0040	0	0	0	0	50.2	16.1	804	100	4.019	0	974.6
1920	0.0023	0	11.3	0	0	28.3	27.2	425	57	3.397	0	551.4
1900	0	0	7.4	0	0	14.9	23.8	89	15	1.488	0	151.8
1880	0.0001	0	4.5	0	0	4.5	7.2	18	9	0.896	0	43.9

Table 4-12: Cadmium: Consumptive Uses -% Use by Category

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses			
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.
1979	2	50.9	13.0	21.9	0	0	11.0	0	0	0	1.1
1970	2	49.9	14.4	3.4	0	0	26.3	0	0	0	4.1
1963	2	57.6	15.0	7.4	0	0	17.5	0	0	0	0.5
1950	3	68.0	17.0	3.0	0	0	9.0	0	0	0	0
1940	4	75.0	20.0	1.0	0	0	0	0	0	0	0
1930	4	75.0	20.0	1.0	0	0	0	0	0	0	0
1920	0	0	100.0	0	0	0	0	0	0	0	0
1900	0	0	0	0	0	0	0	0	0	0	0
1880	0	0	0	0	0	0	0	0	0	0	0

Table 4-13: Cadmium: Consumptive Uses, United States (tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1979	35.6	907.0	231.7	390.3	0	0	196.0	0	0	0	19.6	1782.0
1970	89.5	2232.8	644.3	152.1	0	0	1176.8	0	0	0	183.5	4474.6
1963	96.5	2779.6	723.9	357.1	0	0	844.5	0	0	0	24.1	4825.7
1950	114.7	2598.8	649.7	114.7	0	0	344.0	0	0	0	0	3821.8
1940	134.3	2517.8	671.4	33.6	0	0	0	0	0	0	0	3357.0
1930	40.1	752.5	200.7	10.0	0	0	0	0	0	0	0	1003.3
1920	0	0	67.8	0	0	0	0	0	0	0	0	67.8
1900	0	0	0	0	0	0	0	0	0	0	0	6.4
1880	0	0	0	0	0	0	0	0	0	0	0	0

Table 4-14: Cadmium: Emissions from Consumptive Uses, United States (Tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1979	0.036	136.06	116	7.81	0	0	29	0	0	0	0.980	290
1970	0.089	334.92	322	3.04	0	0	177	0	0	0	9.173	846
1963	0.097	416.94	362	7.14	0	0	127	0	0	0	1.206	914
1950	0.115	389.82	325	2.29	0	0	52	0	0	0	0	769
1940	0.134	377.66	336	0.67	0	0	0	0	0	0	0	714
1930	0.040	112.87	100	0.20	0	0	0	0	0	0	0	213
1920	0	0	34	0	0	0	0	0	0	0	0	34
1900	0	0	0	0	0	0	0	0	0	0	0	0
1880	0	0	0	0	0	0	0	0	0	0	0	0

Table 4-15: Cadmium: Consumptive Uses, Hudson-Raritan Basin (Tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1979	2.56	65.13	16.63	28.021	0	0	14.075	0	0	0	1.407	127.82
1970	7.57	188.79	54.48	12.863	0	0	99.501	0	0	0	15.512	378.71
1963	8.55	246.11	64.09	31.618	0	0	74.772	0	0	0	2.136	427.27
1950	10.69	242.37	60.59	10.693	0	0	32.079	0	0	0	0	356.43
1940	12.97	243.24	64.86	3.243	0	0	0	0	0	0	0	324.32
1930	3.89	72.91	19.44	0.972	0	0	0	0	0	0	0	97.22
1920	0	0	6.06	0	0	0	0	0	0	0	0	6.06
1900	0	0	0	0	0	0	0	0	0	0	0	0
1880	0	0	0	0	0	0	0	0	0	0	0	0

Table 4-16: Cadmium: Emissions from Consumptive Uses, Hudson-Raritan Basin (Tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1979	0.0026	9.769	8.317	0.560	0	0	2.111	0	0	0	0.0704	20.83
1970	0.0076	28.318	27.240	0.257	0	0	14.925	0	0	0	0.7756	71.52
1963	0.0085	36.916	32.045	0.632	0	0	11.216	0	0	0	0.1068	80.92
1950	0.0107	36.356	30.297	0.214	0	0	4.812	0	0	0	0	71.69
1940	0.0130	36.486	32.432	0.065	0	0	0	0	0	0	0	68.99
1930	0.0039	10.937	9.722	0.019	0	0	0	0	0	0	0	20.68
1920	0	0	3.032	0	0	0	0	0	0	0	0	3.03
1900	0	0	0	0	0	0	0	0	0	0	0	0
1880	0	0	0	0	0	0	0	0	0	0	0	0

Table 4-17: Chromium: Consumptive Uses -% Use by Category

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses			
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.
1980	58.5	3.0	5.0	0	0	0.5	2.7	0	0.4	0	16.5
1968	62.5	4.2	5.0	0	0	0.5	3.0	0	0.3	0	25.0
1960	54	4.0	6.0	0	0	0.5	3.0	0	0.2	0	32.0
1950	50	4.0	6.5	0	0	0.5	3.0	0	0.2	0	35.0
1940	50	3.5	14.5	0	0	0.5	4.0	0	0.2	0	27.0
1930	60	2.5	7.7	0	0	0.5	4.0	0	0.2	0	30.0
1920	50	0	15.0	0	0	0.5	10.0	0	0.1	0	25.0
1900	0	0	100.0	0	0	0	0	0	0	0	0
1880	0	0	100.0	0	0	0	0	0	0	0	0

Table 4-18: Chromium: Consumptive Uses, United States (tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	151.8	7.78	12.97	0	0	1.297	7.78	0	1.038	0	42.8	259.46
1968	223.8	15.04	17.91	0	0	1.791	9.67	0	1.074	0	89.5	358.16
1960	176.1	13.04	19.56	0	0	1.630	9.78	0	0.652	0	104.3	326.02
1950	129.9	10.39	16.88	0	0	1.299	7.79	0	0.519	0	90.9	259.73
1940	81.4	5.70	23.61	0	0	0.814	6.51	0	0.326	0	44.0	162.83
1930	41.7	1.74	5.35	0	0	0.347	2.78	0	0.139	0	20.8	69.42
1920	16.9	0	5.08	0	0	0.169	3.38	0	0.034	0	8.5	33.85
1900	0	0	6.49	0	0	0	0	0	0	0	0	6.49
1880	0	0	0.67	0	0	0	0	0	0	0	0	0.67

Table 4-19: Chromium: Consumptive Uses, United States (tonnes)

Year	Metallic Uses (Except Coatings & Electrical)	Protective Cover		Electrical		Chemical		Biocidal Poison Uses		Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
		Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical			
1980	0.1518	0.1557	6.49	0	0	1.297	0.389	0	1.038	0	2.141	11.659
1968	0.2238	0.3009	8.95	0	0	1.791	0.484	0	1.074	0	4.477	17.304
1960	0.1761	0.2608	9.78	0	0	1.630	0.489	0	0.652	0	5.216	18.205
1950	0.1299	0.2078	8.44	0	0	1.299	0.390	0	0.519	0	4.545	15.532
1940	0.0814	0.1140	11.81	0	0	0.814	0.326	0	0.326	0	2.198	15.665
1930	0.0417	0.0347	2.67	0	0	0.347	0.139	0	0.139	0	1.041	4.415
1920	0.0169	0	2.54	0	0	0.169	0.169	0	0.034	0	0.423	3.351
1900	0	0	3.25	0	0	0	0	0	0	0	0	3.246
1880	0	0	0.34	0	0	0	0	0	0	0	0	0.337

Table 4-20: Chromium: Consumptive Uses, Hudson-Raritan Basin (Tonnes)

Year	Metallic Uses (Except Coatings & Electrical)	Protective Cover		Electrical		Chemical		Biocidal Poison Uses		Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
		Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical			
1980	10.90	0.559	0.931	0	0	0.093	0.559	0	0.075	0	3.07	16.189
1968	18.93	1.272	1.514	0	0	0.151	0.818	0	0.091	0	7.57	30.343
1960	15.59	1.155	1.732	0	0	0.144	0.866	0	0.058	0	9.24	28.779
1950	12.11	0.969	1.575	0	0	0.121	0.727	0	0.048	0	8.48	24.029
1940	7.87	0.551	2.281	0	0	0.079	0.629	0	0.031	0	4.25	15.684
1930	4.04	0.168	0.518	0	0	0.034	0.269	0	0.013	0	2.02	7.057
1920	1.51	0	0.454	0	0	0.015	0.303	0	0.003	0	0.76	3.046
1900	0	0	0.508	0	0	0	0	0	0	0	0	0.508
1880	0	0	0.049	0	0	0	0	0	0	0	0	0.049

Table 4-21: Chromium: Emissions from Consumptive Uses, Hudson-Raritan Basin (Tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	0.0109	0.0112	0.466	0	0	0.093	0.028	0	0.075	0	0.154	0.837
1968	0.0189	0.0254	0.757	0	0	0.151	0.041	0	0.091	0	0.379	1.463
1960	0.0156	0.0231	0.866	0	0	0.144	0.043	0	0.058	0	0.462	1.612
1950	0.0121	0.0194	0.787	0	0	0.121	0.036	0	0.048	0	0.424	1.449
1940	0.0079	0.0110	1.141	0	0	0.079	0.031	0	0.031	0	0.212	1.513
1930	0.0040	0.0034	0.259	0	0	0.034	0.013	0	0.013	0	0.101	0.428
1920	0.0015	0	0.227	0	0	0.015	0.015	0	0.003	0	0.038	0.300
1900	0	0	0.254	0	0	0	0	0	0	0	0	0.254
1880	0	0	0.024	0	0	0	0	0	0	0	0	0.024

Table 4-22: Copper: Consumptive Uses -% Use by Category

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses			
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.
1980	99.670	0	0	0	0	0.190	0	0.14	0	0	0
1970	99.610	0	0	0	0	0.230	0	0.10	0	0	0
1960	99.440	0	0	0	0	0.260	0	0.30	0	0	0
1950	99.420	0	0	0	0	0.270	0	0.31	0	0	0
1945	99.355	0	0	0	0	0.265	0	0.70	0	0	0
1940	99.290	0	0	0	0	0.260	0	0.45	0	0	0
1930	99.460	0	0	0	0	0.270	0	0.27	0	0	0
1920	99.040	0	0	0	0	0.290	0	0.67	0	0	0
1900	99.220	0	0	0	0	0.280	0	1.12	0	0	0
1880	93.200	0	0	0	0	0.300	0	3.10	0	0	0

Table 4-23: Copper: Consumptive Uses, United States (tonnes)

Year	Metallic Uses (Except Coatings & Electrical)	Protective Cover		Electrical		Chemical		Biocidal Poison Uses		Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
		Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical			
1980	2214.9	0	0	0	0	4.222	0	3.111	0	0	0	2222.2
1970	1964.7	0	0	0	0	4.537	0	1.972	0	0	0	1972.4
1960	1414.5	0	0	0	0	3.698	0	4.267	0	0	0	1422.5
1950	1559.2	0	0	0	0	4.235	0	4.862	0	0	0	1568.3
1945	1945.1	0	0	0	0	5.188	0	13.704	0	0	0	1957.7
1940	1727.2	0	0	0	0	4.523	0	7.828	0	0	0	1739.5
1930	723.5	0	0	0	0	1.964	0	1.964	0	0	0	727.4
1920	732.2	0	0	0	0	2.144	0	4.953	0	0	0	739.3
1900	469.8	0	0	0	0	1.326	0	5.303	0	0	0	473.5
1880	151.3	0	0	0	0	0.487	0	5.033	0	0	0	162.4

Table 4-24: Copper: Emissions from Consumptive Uses, United States (Tonnes)

Year	Metallic Uses (Except Coatings & Electrical)	Protective Cover		Electrical		Chemical		Biocidal Poison Uses		Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
		Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical			
1980	11.074	0	0	0	0	4.222	0	0.156	0	0	0	15.452
1970	9.824	0	0	0	0	4.537	0	0.099	0	0	0	14.459
1960	7.072	0	0	0	0	3.698	0	0.213	0	0	0	10.984
1950	7.796	0	0	0	0	4.235	0	0.243	0	0	0	12.274
1945	9.725	0	0	0	0	5.188	0	0.685	0	0	0	15.598
1940	8.636	0	0	0	0	4.523	0	0.391	0	0	0	13.550
1930	3.617	0	0	0	0	1.964	0	0.098	0	0	0	5.680
1920	3.661	0	0	0	0	2.144	0	0.248	0	0	0	6.052
1900	2.349	0	0	0	0	1.326	0	0.265	0	0	0	3.940
1880	0.757	0	0	0	0	0.487	0	0.252	0	0	0	1.495

Table 4-25: Copper: Consumptive Uses, Hudson-Raritan Basin (Tonnes)

Year	Metallic Uses (Except Coatings & Electrical)	Protective Cover		Electrical		Chemical		Biocidal Poison Uses		Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
		Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical			
1980	159.0	0	0	0	0	0.303	0	0.223	0	0	0	159.56
1970	166.1	0	0	0	0	0.384	0	0.167	0	0	0	166.67
1960	125.2	0	0	0	0	0.327	0	0.378	0	0	0	125.95
1950	145.4	0	0	0	0	0.395	0	0.453	0	0	0	146.27
1945	184.7	0	0	0	0	0.493	0	1.301	0	0	0	186.45
1940	166.9	0	0	0	0	0.437	0	0.756	0	0	0	168.05
1930	70.1	0	0	0	0	0.190	0	0.190	0	0	0	70.49
1920	65.5	0	0	0	0	0.192	0	0.443	0	0	0	66.13
1900	36.8	0	0	0	0	0.104	0	0.415	0	0	0	37.31
1880	11.0	0	0	0	0	0.035	0	0.366	0	0	0	11.39

Table 4-26: Copper: Emissions from Consumptive Uses, Hudson-Raritan Basin (Tonnes)

Year	Metallic Uses (Except Coatings & Electrical)	Protective Cover		Electrical		Chemical		Biocidal Poison Uses		Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
		Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical			
1980	0.795	0	0	0	0	0.303	0	0.0112	0	0	0	1.109
1970	0.831	0	0	0	0	0.384	0	0.0083	0	0	0	1.222
1960	0.626	0	0	0	0	0.327	0	0.0189	0	0	0	0.973
1950	0.727	0	0	0	0	0.395	0	0.0227	0	0	0	1.145
1945	0.923	0	0	0	0	0.493	0	0.0650	0	0	0	1.481
1940	0.834	0	0	0	0	0.437	0	0.0378	0	0	0	1.309
1930	0.351	0	0	0	0	0.190	0	0.0095	0	0	0	0.550
1920	0.327	0	0	0	0	0.192	0	0.0222	0	0	0	0.541
1900	0.184	0	0	0	0	0.104	0	0.0208	0	0	0	0.309
1880	0.055	0	0	0	0	0.035	0	0.0183	0	0	0	0.109

Table 4-27: Mercury: Consumptive Uses -% Use by Category

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses			
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.
1977	0	0	0	49.2	8.8	20.7	0	1.0	13.2	2.1	0.7
1970	0	0	0	25.9	13.5	28.0	0	2.9	19.7	5.2	2.9
1960	0	0	0	18.1	12.7	13.5	0	5.8	8.2	7.3	2.5
1950	0	0	0	33.8	15.0	12.6	0	12.6	8.7	21.5	1.8
1942	0	0	6	9.2	7.1	40.0	0	3.0	5.0	27.7	3.0
1928	1.3	0	7	7.3	8.6	43.7	0	1.1	4.7	16.7	9.9
1920	5.0	0	7	7.0	8.0	42.0	0	0	8.0	15.0	8.0
1900	10.0	0	7	3.0	8.0	42.0	0	0	8.0	15.0	7.0
1880	42.0	0	5	0	5.0	30.0	0	0	5.0	10.0	5.0

Table 4-28: Mercury: Consumptive Uses, United States (tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1977	0	0	0	979.54	175.20	412.12	0	19.91	262.80	41.81	13.94	1990.94
1970	0	0	0	570.46	297.35	616.72	0	63.87	433.90	114.53	63.87	2202.56
1960	0	0	0	349.1	244.95	260.38	0	111.87	158.15	140.80	48.22	1928.72
1950	0	0	0	547.01	242.75	203.91	0	203.91	140.80	347.95	29.13	1618.36
1942	0	0	66.88	102.54	79.14	445.84	0	33.44	55.73	308.75	33.44	1114.61
1928	11.9	0	64.10	66.84	78.75	400.14	0	10.07	43.04	152.91	90.65	915.65
1920	42.5	0	59.54	59.54	68.04	357.22	0	0	68.04	127.58	68.04	850.52
1900	55.9	0	39.11	16.76	44.70	234.65	0	0	44.70	83.80	39.11	558.70
1880	353.3	0	42.06	0	42.06	252.35	0	0	42.06	84.12	42.06	841.18

Table 4-29: Mercury: Emissions from Consumptive Uses, United States (Tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1977	0	0	0	195.909	17.520	412.1	0	15.93	236.52	8.36	6.96	893.3
1970	0	0	0	114.092	29.735	616.7	0	51.10	390.51	22.91	31.93	1257.0
1960	0	0	0	69.820	24.495	260.4	0	89.49	142.34	28.16	24.10	638.8
1950	0	0	0	109.401	24.275	203.9	0	163.13	126.72	69.59	14.56	711.6
1942	0	0	53.50	20.509	7.914	445.8	0	26.75	50.16	61.75	16.71	683.1
1928	0.595	0	51.28	13.368	7.875	400.1	0	8.06	38.73	30.58	45.32	595.9
1920	2.126	0	47.63	11.907	6.804	357.2	0	0	61.24	25.52	34.02	546.5
1900	2.793	0	31.29	3.352	4.470	234.7	0	0	40.23	16.76	19.55	353.1
1880	17.665	0	33.65	0	4.206	252.4	0	0	37.85	16.82	21.03	383.6

Table 4-30: Mercury: Consumptive Uses, Hudson-Raritan Basin (Tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1977	0	0	0	70.33	12.58	29.59	0	1.430	18.87	3.00	1.00	136.81
1970	0	0	0	48.23	25.14	52.14	0	5.401	36.69	9.68	5.40	182.69
1960	0	0	0	30.91	21.69	23.05	0	9.905	14.00	12.47	4.27	116.29
1950	0	0	0	51.02	22.64	19.02	0	19.018	13.13	32.45	2.72	159.99
1942	0	0	6.46	9.91	7.65	43.07	0	3.230	5.38	29.83	3.23	108.76
1928	1.15	0	6.21	6.48	7.63	38.77	0	0.976	4.17	14.82	8.78	88.99
1920	3.80	0	5.33	5.33	6.09	31.95	0	0	6.09	11.41	6.09	76.08
1900	4.38	0	3.06	1.31	3.50	18.38	0	0	3.50	6.56	3.06	43.75
1880	25.66	0	3.05	0	3.05	18.33	0	0	3.05	6.11	3.05	62.31

Table 4-31: Mercury: Emissions from Consumptive Uses, Hudson-Raritan Basin (Tonnes)

						Chemical		Biocidal Poison Uses				
		Protective Cover		Electrical								
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1977	0	0	0	14.067	1.258	29.6	0	1.144	16.98	0.60	0.500	64.14
1970	0	0	0	9.647	2.514	52.1	0	4.320	33.02	1.94	2.700	106.28
1960	0	0	0	6.182	2.169	23.1	0	7.924	12.60	2.49	2.135	56.56
1950	0	0	0	10.203	2.264	19.0	0	15.214	11.82	6.49	1.358	66.37
1942	0	0	5.17	1.981	0.765	43.1	0	2.584	4.85	5.97	1.615	66.00
1928	0.058	0	4.97	1.295	0.763	38.8	0	0.781	3.75	2.96	4.392	57.75
1920	0.190	0	4.26	1.065	0.609	32.0	0	0	5.48	2.28	3.043	48.88
1900	0.219	0	2.45	0.263	0.350	18.4	0	0	3.15	1.31	1.531	27.65
1880	1.283	0	2.44	0	0.305	18.3	0	0	2.75	1.22	1.527	27.86

Table 4-32: Lead: Consumptive Uses -% Use by Category

						Chemical		Biocidal Poison Uses			
		Protective Cover		Electrical							
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.
1980	18.8	0	7.3	64.0	0	0	11.9	0.0	0	0	2.0
1972	22.7	0	6.0	48.9	0	0	18.7	0.3	0	0	3.7
1957	39.4	0	10.1	31.7	0	0	15.5	0.6	0	0	3.0
1949	43.3	0	11.2	33.3	0	0	9.9	3.0	0	0	1.7
1939	41.0	0	19.8	29.7	0	0	6.0	1.0	0	0	0.5
1929	57.9	0	15.4	21.6	0	0	0.3	0.2	0	0	3.8
1923	53.7	0	22.9	18.6	0	0	0	0	0	0	4.6
1900	60.0	0	30.0	5.0	0	0	0	0	0	0	5.0
1880	60.0	0	35.0	0	0	0	0	0	0	0	5.0

Table 4-33: Lead: Consumptive Uses, United States (tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	250	0	97	851	0	0	0	0	0	0	26.58	1224
1972	288	0	76	621	0	0	0	3.81	0	0	46.96	1036
1957	374	0	96	301	0	0	0	5.70	0	0	28.50	806
1949	443	0	115	341	0	0	0	30.73	0	0	17.41	947
1939	310	0	150	225	0	0	0	7.56	0	0	3.78	696
1929	419	0	111	156	0	0	0	1.45	0	0	27.51	716
1923	374	0	160	130	0	0	0	0	0	0	32.05	695
1900	0	0	0	0	0	0	0	0	0	0	0	0
1880	0	0	0	0	0	0	0	0	0	0	0	0

Table 4-34: Lead: Emissions from Consumptive Uses, United States (Tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	1.249	0	48.5	8.51	0	0	0	0	0	0	1.329	56.9
1972	1.441	0	38.1	6.21	0	0	0	0.190	0	0	2.348	48.3
1957	1.871	0	48.0	3.01	0	0	0	0.285	0	0	1.425	54.6
1949	2.217	0	57.4	3.41	0	0	0	1.536	0	0	0.871	65.4
1939	1.550	0	74.8	2.25	0	0	0	0.378	0	0	0.189	79.2
1929	2.096	0	55.7	1.56	0	0	0	0.072	0	0	1.375	60.9
1923	1.871	0	79.8	1.3	0	0	0	0	0	0	1.602	84.5
1900	0	0	0	0	0	0	0	0	0	0	0	0
1880	0	0	0	0	0	0	0	0	0	0	0	0

Table 4-35: Lead: Consumptive Uses, Hudson-Raritan Basin (Tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	17.9	0	6.97	61.1	0	0	0	0	0	0	1.91	87.9
1972	24.4	0	6.44	52.5	0	0	0	0.322	0	0	3.97	87.6
1957	33.1	0	8.49	26.7	0	0	0	0.505	0	0	2.52	71.3
1949	41.4	0	10.70	31.8	0	0	0	2.866	0	0	1.62	88.4
1939	29.9	0	14.46	21.7	0	0	0	0.730	0	0	0.37	67.2
1929	40.6	0	10.80	15.2	0	0	0	0.140	0	0	2.67	69.4
1923	33.5	0	14.27	11.6	0	0	0	0	0	0	2.87	62.2
1900	0	0	0	0	0	0	0	0	0	0	0	0
1880	0	0	0	0	0	0	0	0	0	0	0	0

Table 4-36: Lead: Emissions from Consumptive Uses, Hudson-Raritan Basin (Tonnes)

		Protective Cover		Electrical		Chemical		Biocidal Poison Uses				
Year	Metallic Uses (Except Coatings & Electrical)	Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical	Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
1980	0.090	0	3.48	0.611	0	0	0	0	0	0	0.0954	4.28
1972	0.122	0	3.22	0.525	0	0	0	0.0161	0	0	0.1985	4.08
1957	0.166	0	4.25	0.267	0	0	0	0.0252	0	0	0.1262	4.83
1949	0.207	0	5.35	0.318	0	0	0	0.1433	0	0	0.0812	6.10
1939	0.150	0	7.23	0.217	0	0	0	0.0365	0	0	0.0183	7.65
1929	0.203	0	5.40	0.152	0	0	0	0.0070	0	0	0.1333	5.90
1923	0.167	0	7.14	0.116	0	0	0	0	0	0	0.1433	7.56
1900	0	0	0	0	0	0	0	0	0	0	0	0
1880	0	0	0	0	0	0	0	0	0	0	0	0

Table 4-37: Zinc: Consumptive Uses -% Use by Category

Year	Metallic Uses (Except Coatings & Electrical)	Protective Cover		Electrical		Chemical		Biocidal Poison Uses		Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.
		Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical		
1977	41	35	12.4	0.5	0	0.2	9.9	0.3	0.2	0.1	0
1973	45	31	11.7	1	0	0.3	8.9	0.2	0.2	0.1	0
1968	0	27	0	0	0	0	0	0	0	0.1	0
1958	43	33	12.6	3.3	0	0.7	6.8	0.3	0.2	0.1	0
1949	25	45	16.8	3.7	0	0.7	7.5	0.2	0.2	0.1	0
1940	27	39	19.8	4.1	0	0.5	9.2	0.2	0.2	0.1	0
1930	22	39	25.7	2.7	0	0.5	9.9	0.2	0.2	0.1	0
1920	26	33	26.0	2.7	0	0.6	5.0	0.3	5.5	0.1	0
1900	30	25	25.0	5.0	0	1.0	5.0	0.5	8.0	0.3	0
1880	35	30	30.0	0	0	2.5	2.0	0	0	0.5	0

Table 4-38: Zinc: Consumptive Uses, United States (tonnes)

Year	Metallic Uses (Except Coatings & Electrical)	Protective Cover		Electrical		Chemical		Biocidal Poison Uses		Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
		Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical			
1977	514.1	438.90	155.50	6.27	0	2.51	124.15	3.76	2.51	1.254	0	1254.0
1973	643.5	443.29	167.30	14.30	0	4.29	127.27	2.86	2.86	1.430	0	1430.0
1968	688.6	422.56	190.15	33.65	0	7.83	122.86	3.91	3.13	1.565	0	1565.0
1958	477.5	366.44	139.91	36.64	0	7.77	75.51	3.33	2.22	1.110	0	1110.4
1949	217.5	391.50	146.16	32.19	0	6.09	65.25	1.74	1.74	0.870	0	870.0
1940	186.5	269.38	136.76	28.32	0	3.45	63.55	1.38	1.38	0.691	0	690.7
1930	104.8	185.82	122.45	12.86	0	2.38	47.17	0.95	0.95	0.476	0	476.5
1920	102.9	130.59	102.89	10.68	0	2.37	19.79	1.19	21.76	0.396	0	395.7
1900	37.3	31.07	31.07	6.21	0	1.24	6.21	0.62	9.94	0.373	0	124.3
1880	11.9	10.23	10.23	0	0	0.85	0.68	0	0	0.171	0	34.1

Table 4-39: Zinc: Emissions from Consumptive Uses, United States (Tonnes)

Year	Metallic Uses (Except Coatings & Electrical)	Protective Cover		Electrical		Chemical		Biocidal Poison Uses		Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
		Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical			
1977	0.514	8.778	77.75	0.063	0	2.508	18.622	0.188	0.251	1.003	0	109.67
1973	0.643	8.866	83.65	0.143	0	4.290	19.090	0.143	0.286	1.144	0	118.26
1968	0.689	8.451	95.08	0.336	0	7.825	18.428	0.196	0.313	1.252	0	132.57
1958	0.477	7.329	69.96	0.366	0	7.773	11.326	0.167	0.222	0.888	0	98.51
1949	0.217	7.830	73.08	0.322	0	6.090	9.787	0.087	0.174	0.696	0	98.28
1940	0.186	5.388	68.38	0.283	0	3.454	9.532	0.069	0.138	0.553	0	87.99
1930	0.105	3.716	61.22	0.129	0	2.382	7.075	0.048	0.095	0.381	0	75.16
1920	0.103	2.612	51.44	0.107	0	2.374	2.968	0.059	2.176	0.317	0	62.16
1900	0.037	0.621	15.54	0.062	0	1.243	0.932	0.031	0.994	0.298	0	19.75
1880	0.012	0.205	5.12	0	0	0.853	0.102	0	0	0.136	0	6.42

Table 4-40: Zinc: Consumptive Uses, Hudson-Raritan Basin (Tonnes)

Year	Metallic Uses (Except Coatings & Electrical)	Protective Cover		Electrical		Chemical		Biocidal Poison Uses		Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
		Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical			
1977	36.92	31.51	11.16	0.45	0	0.1801	8.914	0.2701	0.1801	0.0900	0	89.68
1973	50.30	34.65	13.08	1.118	0	0.3354	9.949	0.2236	0.2236	0.1118	0	110.00
1968	58.22	35.73	16.08	2.845	0	0.6616	10.388	0.3308	0.2647	0.1323	0	124.65
1958	42.28	32.45	12.39	3.245	0	0.6882	6.686	0.2950	0.1966	0.0983	0	98.32
1949	20.28	36.51	13.63	3.002	0	0.5680	6.085	0.1623	0.1623	0.0811	0	80.49
1940	18.02	26.02	13.21	2.736	0	0.3337	6.139	0.1335	0.1335	0.0667	0	66.80
1930	10.16	18.01	11.87	1.247	0	0.2308	4.571	0.0923	0.0923	0.0462	0	46.31
1920	9.20	11.68	9.20	0.956	0	0.2124	1.770	0.1062	1.9468	0.0354	0	35.11
1900	2.92	2.43	2.43	0.487	0	0.0973	0.487	0.0487	0.7786	0.0292	0	9.71
1880	0.87	0.74	0.74	0	0	0.0619	0.050	0	0	0.0124	0	2.48

Table 4-41: Zinc: Emissions from Consumptive Uses, Hudson-Raritan Basin (Tonnes)

Year	Metallic Uses (Except Coatings & Electrical)	Protective Cover		Electrical		Chemical		Biocidal Poison Uses		Medical, Dental, Pharmaceutical	Miscellaneous, N.E.C.	Total
		Plating & Coating	Paints & Pigments	Batteries & Equipment	Other Electrical Uses, Instruments, etc.	Industrial Catalysts, Reagents, Explosives, etc.	Consumer Uses; Additives, Extenders, Photography, etc.	Agricultural Pesticides, Herbicides, Fungicides	Non-Agricultural Pesticides, Except Medical			
1977	0.037	0.630	5.582	0.005	0	0.180	1.337	0.014	0.018	0.072	0	7.87
1973	0.050	0.693	6.540	0.011	0	0.335	1.492	0.011	0.022	0.089	0	9.24
1968	0.058	0.715	8.039	0.028	0	0.662	1.558	0.017	0.026	0.106	0	11.21
1958	0.042	0.649	6.194	0.032	0	0.688	1.003	0.015	0.020	0.079	0	8.72
1949	0.020	0.730	6.816	0.030	0	0.568	0.913	0.008	0.016	0.065	0	9.17
1940	0.018	0.520	6.606	0.027	0	0.334	0.921	0.007	0.013	0.053	0	8.50
1930	0.010	0.360	5.933	0.012	0	0.231	0.686	0.005	0.009	0.037	0	7.28
1920	0.009	0.234	4.601	0.010	0	0.212	0.265	0.005	0.195	0.028	0	5.56
1900	0.003	0.049	1.217	0.005	0	0.097	0.073	0.002	0.078	0.023	0	1.55
1880	0.001	0.015	0.372	0	0	0.062	0.007	0	0	0.01	0	0.47

5. Total Anthropogenic Hudson-Raritan Metal Emissions

Tables 5-1, through 5-7 present the total metal emissions for the Hudson-Raritan basin. These have been taken from Chapters 3 and 4 respectively and summed. A major implication of these results is worth emphasizing: in recent years consumption related emissions dominate production related emissions by a large factor. The apparent exception (lead) is not an exception, since the "fossil fuel" contribution is actually tetraethyl lead in gasoline -- clearly a form of consumption.

In earlier periods the situation was more complex. In some cases, production-related emissions were dominant. That was true for cadmium prior to 1940, for chromium prior to 1930, for copper prior to 1920 and for lead prior to 1950. Note that anthropogenic silver, arsenic, mercury and zinc in the environment were never derived mainly from production processes.

It follows that uncertainties with regard to emissions coefficients and control efficiencies from production (and combustion) processes are relatively unimportant as compared to uncertainties in the material cycle of dissipative uses. For instance, the two major contributions to environmental emissions of zinc in 1980 are from pigments (5582 tonnes) and chemicals (1337 tonnes). These numbers totally overwhelm the 2-4 tonnes attributable to metallurgical processes and coal combustion. Yet they are based on emissions factors (Table 4-1) that are admittedly highly uncertain. It would appear that greater efforts should be made to improve on this part of the data base.

Table 5-8 repeats the Total Emissions category from Tables 5-1 through 5-7 in a single table for convenient reference. Table 5-9 gives the 1975 emissions ratios $R^i(t)$.

Table 5-1: Average Annual Silver & Arsenic Emissions, Hudson-Raritan Basin (tonnes)

	Silver (Consumptive Only)	Arsenic Metallurgical Ops		Arsenic Fossil Fuel Combustion	Arsenic Consumptive Uses	Arsenic Total Emissions	
		low	high			low	high
1980	59.6	4.9	5.5	4.1	672.4	681.4	682.0
1970	49.4	33.8	74.1	6.1	1020.7	1060.6	1100.9
1960	39.6	125.3	275.3	3.0	755.9	884.1	1034.2
1950	37.7	131.2	462.0	2.2	821.1	954.5	1285.3
1940	18.6	150.3	462.1	1.5	1363.4	1515.2	1826.9
1930	12.5	163.6	445.6	1.3	974.6	1139.5	1421.5
1920	10.3	168.0	373.9	2.9	551.4	722.3	928.2
1900	5.2	89.5	108.6	2.6	151.8	243.9	263.0
1880	1.3	12.1	13.1	0.8	43.9	56.8	57.8

Table 5-2: Average Annual Cadmium Emissions, Hudson-Raritan Basin (tonnes)

	Metallurgical Operations		Fossil Fuel Combustion	Consumptive Uses	Total Emissions	
	low	high			low	high
1980	.0	.0	31.5	20.8	52.3	52.3
1970	0.1	0.4	46.6	71.5	118.3	118.6
1960	0.4	1.1	22.8	80.9	104.1	104.8
1950	0.6	1.6	17.2	71.7	89.4	90.4
1940	0.7	1.8	11.9	69.0	81.5	82.6
1930	14.2	24.9	10.5	20.7	45.4	56.1
1920	20.2	49.3	24.6	3.0	47.8	76.9
1900	1.6	3.7	23.2	0.0	24.8	26.9
1880	0.4	0.6	7.1	0.0	7.5	7.6

Table 5-3: Average Annual Chromium Emissions, Hudson-Raritan Basin (tonnes)

	Metallurgical Operations		Fossil Fuel Combustion	Consumptive Uses	Total Emissions	
	low	high			low	high
1980	0.02	0.02	41.7	837	878.7	878.7
1970	0.00	0.00	63.4	1463	1526.4	1526.4
1960	0.07	0.15	40.4	1612	1652.4	1652.5
1950	0.07	0.26	48.5	1449	1497.6	1497.8
1940	0.33	1.02	79.9	1513	1593.2	1593.9
1930	0.20	0.55	110.1	428	538.3	538.6
1920	0.51	1.11	362.4	300	662.9	663.5
1900	0.46	0.50	444.8	254	699.3	699.3
1880	0.47	0.51	136.4	24	160.8	160.9

Table 5-4: Average Annual Copper Emissions, Hudson-Raritan Basin (tonnes)

	Metallurgical Operations		Fossil Fuel Combustion	Consumptive Uses	Total Emissions	
	low	high			low	high
1980	0.9	1.8	10.1	1109	1120.0	1120.8
1970	3.3	12.5	16.4	1222	1241.7	1250.9
1960	7.7	28.0	16.4	973	997.1	1017.4
1950	10.3	52.7	30.7	1145	1186.0	1228.4
1940	9.8	43.8	66.0	1309	1384.9	1418.9
1930	9.7	40.3	96.3	550	656.0	686.7
1920	9.9	37.8	326.8	541	877.8	905.7
1900	6.6	18.4	408.0	309	723.6	735.4
1880	2.3	3.7	125.1	109	236.4	237.8

Table 5-5: Average Annual Mercury Emissions, Hudson-Raritan Basin (tonnes)

	Metallurgical Operations		Fossil Fuel Combustion	Consumptive Uses	Total Emissions	
	low	high			low	high
1980	0.00	0.00	0.03	64.1	64.2	64.2
1970	0.01	0.02	0.10	106.3	106.4	106.4
1960	0.03	0.04	0.16	56.6	56.8	56.8
1950	0.04	0.06	0.32	66.4	66.7	66.8
1940	0.04	0.05	0.71	66.0	66.7	66.8
1930	0.11	0.16	1.04	57.8	58.9	58.9
1920	0.16	0.31	3.54	48.9	52.6	52.7
1900	0.05	0.08	4.21	27.7	31.9	31.9
1880	0.01	0.01	1.28	27.9	29.2	29.2

Table 5-6: Average Annual Lead Emissions, Hudson-Raritan Basin (tonnes)

	Metallurgical Operations		Fossil Fuel Combustion	Consumptive Uses	Total Emissions	
	low	high			low	high
1980	43	51	5312	4280	9635	9643
1970	112	261	9684	4080	13875	14024
1960	295	688	7203	4830	12328	12721
1950	564	2173	4884	6100	11548	13157
1940	957	3163	2504	7650	11111	13317
1930	864	2306	266	5900	7560	8473
1920	1254	2902	74	7560	8889	10537
1900	1470	1708	92	0	1562	1800
1880	16	23	29	0	45	53

Table 5-7: Average Annual Zinc Emissions, Hudson-Raritan Basin (tonnes)

	Metallurgical Operations		Fossil Combustion		Consumptive	Total Emissions	
	low	high	low	high	Uses	low	high
1980	1.6	3.5	0.5	0.6	7870	7872	7874
1970	13.8	35.1	1.2	1.7	10225	10240	10262
1960	30.8	82.3	3.2	5.5	8720	8754	8808
1950	43.0	163.8	8.1	14.3	9170	9221	9348
1940	47.3	194.1	19.6	34.7	8500	8567	8729
1930	43.5	149.2	29.2	51.7	7280	7353	7481
1920	43.7	167.9	100.0	177.1	5560	5704	5905
1900	39.6	82.3	125.5	222.3	1550	1715	1855
1880	5.7	31.3	38.5	68.2	470	514	569

Table 5-8: $E^i(t)$; Average Annual Metallic Emissions, Hudson-Raritan Basin (tonnes)

	Silver	Arsenic		Cadmium		Chromium	Copper		Mercury	Lead		Zinc	
From		low	high	low	high	avg	low	high	avg	low	high	low	high
Table	5-1	5-1	5-1	5-2	5-2	5-3	5-4	5-4	5-5	5-6	5-6	5-7	5-7
						(a)			(a)				
1980	60	681	682	52	52	879	1120	1121	64	9635	9643	7872	7874
1975(b)	55	881		85		1203	1183		85	7762		9062	
1970	49	1061	1101	118	119	1526	1242	1251	106	13875	14024	10240	10262
1960	40	884	1034	104	105	1652	997	1017	57	12328	12721	8754	8808
1950	38	954	1285	89	90	1498	1186	1228	67	11548	13157	9221	9348
1940	19	1515	1827	82	83	1594	1385	1419	67	11111	13317	8567	8729
1930	12	1139	1422	45	56	538	656	687	59	7560	8473	7353	7481
1920	10	722	928	48	77	663	878	906	53	8889	10537	5704	5905
1900	5	244	263	25	27	699	724	735	32	1562	1800	1715	1855
1880	1	57	58	7	8	161	236	238	29	45	53	514	569

(a) No significant difference between high and low cases

(b) 1975 = average of 1970,1980, both high and low

Table 5-9: $R^i(t)=E^i(t)/E^i(1975)$
Metallic Emissions Ratios, Hudson-Raritan Basin

	Silver	Arsenic		Cadmium		Chromium	Copper		Mercury	Lead		Zinc	
		low	high	low	high	avg (a)	low	high	avg (a)	low	high	low	high
1980	1.09	0.77	0.77	0.61	0.61	0.73	0.95	0.95	0.75	0.82	0.82	0.87	0.87
1970	0.91	1.20	1.25	1.39	1.39	1.27	1.05	1.06	1.25	1.18	1.19	1.13	1.13
1960	0.73	1.00	1.17	1.22	1.23	1.37	0.84	0.86	0.67	1.05	1.08	0.97	0.97
1950	0.69	1.08	1.46	1.05	1.06	1.25	1.00	1.04	0.78	0.98	1.12	1.02	1.03
1940	0.34	1.72	2.07	0.95	0.97	1.33	1.17	1.20	0.78	0.94	1.13	0.95	0.96
1930	0.23	1.29	1.61	0.53	0.66	0.45	0.55	0.58	0.69	0.60	0.72	0.81	0.83
1920	0.19	0.82	1.05	0.56	0.90	0.55	0.74	0.77	0.62	0.75	0.89	0.63	0.65
1900	0.10	0.28	0.30	0.29	0.32	0.58	0.61	0.62	0.37	0.13	0.15	0.19	0.20
1880	0.02	0.06	0.07	0.09	0.09	0.13	0.20	0.20	0.34	0.004	0.004	0.06	0.06

(a) No significant difference between high and low cases

6. Hudson-Raritan Regional Allocation of Other Critical Pollutants

6.1 Approach

The approach used in this chapter is to derive waste flows ands losses from data on uses. For this reason, conventional source classifications, notably runoff, are not emphasized, although we explicitly consider runoff in a few places. With regard to pesticides, herbicides and fertilizers used primarily in agriculture, it was necessary to estimate pesticide use by crop, and crop distribution by region. These estimates, described in detail in Chemicals and Other Wastes (COW), are further adjusted to take into account regional differences in the severity of pest outbreaks for particular crops.

With regard to other pollutants of concern the approach is necessarily *ad hoc*. Cases are considered individually. Briefly, the key points are summarized hereafter

Dioxins Not enough data is available at this time to estimate emissions in the Hudson-Raritan basin.

PCB's The major problem in the Hudson-Raritan basin arose from a single point source of emissions at Fort Edward in the upper basin of the Hudson River. A large accumulation was discovered about a decade ago. Some has since been removed by dredging, while a considerable amount is still being re-suspended from bottom sediments. The problem is to estimate the rate of emissions to the estuarine zone over the past 50 years.

PAH's in the atmosphere are largely due to low temperature combustion, especially of coal. Thus the atmospheric contribution is historically attributable to coal use, mainly for space heating. The other important contribution to PAH's in the water is spillage of crude oil in the harbor and used motor oil dumped or washed into sewers. Crude oil losses are proportional to the quantities unloaded for refineries in NY harbor, while motor oil losses are proportional to auto use and, therefore, to gasoline consumption.

Oil and Grease

Emissions arise from the same two sources mentioned above, *viz* spillage and losses of lubricants. The non-automotive component of lubricant consumption may be taken to be proportional to the Hudson-Raritan share of heavy machinery usage, which is somewhat lower than the region's share of population (since heavy industry is obviously concentrated in other parts of the country).

Nitrogen and Phosphorus

are environmentally mobilized on the national scale primarily by agricultural usage. However in the Hudson-Raritan basin it is sewage that now accounts for the dominant share of both. Sewage output is essentially proportional to population. In the late 19th and early 20th century, however, horses working on city streets contributed for a significant amount.

Water Withdrawals

are dominated nationally by irrigation, with municipal, utility and industrial uses of lesser importance. In the Hudson-Raritan basin, however, only utility uses (for cooling) are significant. Municipal water supplies for New York City and the industrial counties of northeastern New Jersey are drawn from protected watersheds. Local industries are not large water users. Water withdrawals (except for cooling) have never accounted for more than 4% of the flow of the Hudson-Raritan basin (although withdrawals have occasionally accounted for more than 100% of the flow of the Passaic River, which enters Newark Bay).

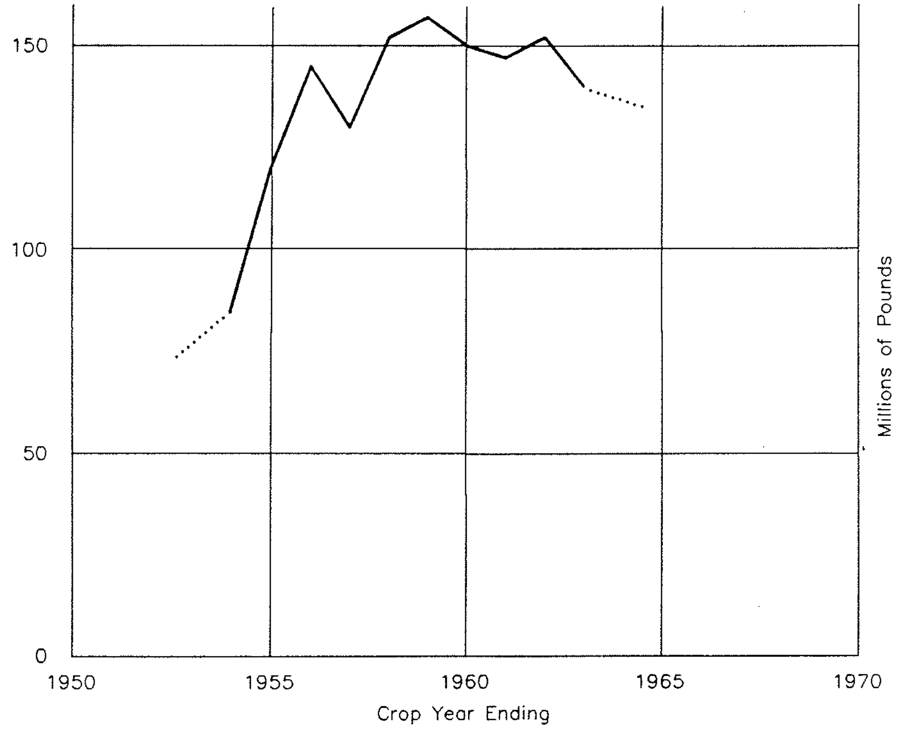
Total Organic Carbon (TOC)

is the only pollutant in the Hudson-Raritan basin that has a significant non-anthropogenic contribution. Since the basin has never been intensively cultivated nor been the site for major paper pulp or food processing industries that generate large quantities of organic wastes, the major anthropogenic source of TOC is sewage. Most of the TOC is from natural sources and may be considered "background".

6.2 Pesticides and Herbicides

In this section we consider all the chlorinated hydrocarbon pesticides and herbicides as a group, due to their obvious similarities. Chlorinated pesticides were in use from 1946 through the early 1970's but restrictions began in the early 1960's, beginning with DDT (Figure 6-1).

Figure 6-1: Trend of Chlorinated Hydrocarbon Insecticide Usage in the U.S (million lbs of technical product)



Source: [*Rosen & Kraybill* 66]

The major farm uses, from the outset, were to control pests of cotton and corn, while lesser amounts were used on soy beans, tobacco, potatoes, apples, citrus fruits and other crops. Pesticide use on farms, by crop are shown in Tables 6-1, 6-2 (1964) and 6-3, 6-4 (1976), taken from USDA stratified sample surveys. Major shifts from 1964 to 1976 are clearly attributable to the fact that many of the chlorinated pesticides that were important in 1964 had been banned by 1976.

Table 6-5 shows the Hudson-Raritan regional share of production for several major crop categories. It is significant that the Hudson-Raritan basin as a whole (with roughly 8% of the nation's population) produces far less than 0.1% of the nation's wheat, much less than .01% of the nation's corn and oats, and around .02% of its potatoes. Deciduous fruit by far is the most important crop in the region (accounting for 4.5% of the national total in 1950 to less than 2% now), with hay being next in regional importance.

Table 6-1: National Pesticide Use by Crop, 1964 (tonnes)

CROP/PEST	DDT	BHC/ Lindane	Al- drin	Di- eldrin	Endrin	Hepta- chlor	Toxa- phene	Other	Total
Corn	48	3	4859	30		498	60	18	5516
Cotton	10699	245	8		846		12208	1294	25300
Soybeans	219		17	13	4		598	4	855
Tobacco	538	34	61	42	68	11	132	1080	1967
Other Field	1022	10	65	129	5	77	1953	181	3442
Potatoes	169			19	28			45	260
Other Veg.	608	60	31	20	9		555	358	1641
Citrus				5				16	22
Apples	744	62		46	14			112	979
Other Fruit	392	20	3	115				236	766
Subtotal	14440	433	5043	420	974	586	15507	3345	40748
Unaccounted	8486	4903	714	510	10	245	2439	29557	46866
Total	22925	5337	5757	931	984	831	17946	32903	87614

The available data do not permit a complete projection of pesticide allocation for all years prior to and later than 1966. However, it is sufficiently clear from available information that agricultural uses in the Hudson-Raritan basin accounted for an insignificant proportion of national usage for all these chemicals over the entire revelant time span. In effect, we are able to conclude that only non-agricultural pesticide uses were at all significant. (The same is true for fertilizers).

The combination of use-by-crop data (Tables 6-1 through 6-4) and crop-distribution by region data (Table 6-5) yields an immediate rough estimate of pesticide usage in the Hudson-Raritan basin. For

Table 6-2: Pesticide Percentage Use Nationally by Crop, 1964

CROP/PEST	DDT	BHC/ Lindane	Al-drin	Di-eldrin	Endrin	Hepta-chlor	Toxa-phene	Other	Crop % Total Pest.
Corn	0.2%	0.1%	84.4%	3.3%		60.0%	0.3%	0.1%	6.30%
Cotton	46.7%	4.6%	0.1%	0.0%	86.0%		68.0%	3.9%	28.88%
Soybeans	1.0%		0.3%	1.4%	0.4%		3.3%	.0%	0.98%
Tobacco	2.3%	0.6%	1.1%	4.5%	6.9%	1.3%	0.7%	3.3%	2.25%
Other Field	4.5%	0.2%	1.1%	13.9%	0.6%	9.2%	10.9%	0.6%	3.93%
Potatoes	0.7%			2.0%	2.8%			0.1%	0.30%
Other Veg.	2.7%	1.1%	0.5%	2.2%	0.9%		3.1%	1.1%	1.87%
Citrus	0.0%			0.6%				.0%	0.02%
Apples	3.2%	1.2%		5.0%	1.4%			0.3%	1.12%
Other Fruit	1.7%	0.4%	.0%	12.4%				0.7%	0.87%
Subtotal	63.0%	8.1%	87.6%	45.2%	99.0%	70.5%	86.4%	10.2%	46.51%
Unaccounted	37.0%	91.9%	12.4%	54.8%	1.0%	29.5%	13.6%	89.8%	53.49%
									100.00%

Table 6-3: National Pesticide Use by Crop, 1976 (tonnes)

CROP/PEST	Al-drin	Chlor-dane	Endrin	Hepta-chlor	Toxa-phene	Other	Total
Corn	386	616		738	43	13	1796
Cotton			141		11924	307	12373
Wheat			89	0.5	252	2	343
Sorghum					454	3	457
Pasture/Range		3.6				1	5
Other Grain					92	0	92
Soybeans			20		1001	0	1021
Tobacco	5.4	6.8	5		6	51	75
Peanuts		0.9			160	0	161
Alfalfa	1.4	18			3	646	668
Subtotal	392	645	254	739	13935	1041	17007
Unaccounted	2308	1605	676	245	4011	17349	26193
Total	2700	2250	931	984	17946	18389	43200

instance, corn accounted for 10.7 million lb of aldrin in 1964, which was 84% of the total domestic sales for that year, while the Hudson-Raritan basin accounted for somewhat less than .01% of total corn production. This simple calculation implies that less than 10,000 lb (5 tons) of Aldrin would have been used on corn in the Hudson-Raritan basin -- assuming uniform distribution of the pesticide over all corn acreage. However, the assumption of uniformity of application is unreliable, due to climatic and soil variations between regions.

Table 6-4: Pesticide Percentage Use Nationally by Crop, 1976

CROP/PEST	Aldrin	Chlordane	Endrin	Heptachlor	Toxaphene	Other	Crop % Total Pest.
Corn	14.28%	27.40%		75.06%	0.24%	0.07%	4.16%
Cotton			15.16%		66.45%	1.67%	28.64%
Soybeans			9.55%	0.05%	1.41%		0.79%
Tobacco					2.53%	0.02%	1.06%
Other Field		0.16%					0.01%
Potatoes					0.51%		0.21%
Other Veg.			2.10%		5.58%		2.36%
Citrus	0.20%	0.30%	0.54%		0.04%	0.28%	0.17%
Apples		0.04%			0.89%		0.37%
Other Fruit	0.05%	0.79%			0.02%	3.51%	1.55%
Subtotal	14.53%	28.69%	27.34%	75.10%	77.65%	5.66%	39.37%
Unaccounted	85.47%	71.31%	72.66%	24.90%	22.35%	94.34%	60.63%
							100.00%

Table 6-5: Hudson - Raritan Fractions of U.S. Crop Totals

	Population	Total Area	Wheat (acres)	Oats (bushels)	Corn	Orchard	Hay	Irish Potatoes	Cropland (in farms)	Wood land (in farms)	Cattle	Horses & Mules
1980	.072	.0034	.0002	-	-	.018	.004	-	.0028	-	.0025	-
1970	.085	.0036	.0004	-	.00004	.024	.004	0	.0029	.0033	.0030	.0060
1960	.089	.0042	.0006	-	-	.030	.006	.00014	.0038	.0027	.0043	.0039
1950	.093	.0042	.001	-	-	.045	.0075	.00026	.0046	.0034	.0054	.0045
1940	.097	.0042	.0009	-	.00008	.060	.008	.0002	.0057	.0050	.0064	.0039
1930	.097	.0042	.0009	<.0001	-	.075	.010	.0002	.0057	.0056	.0064	.0034
1920	.089	.0042	.00184	.00013	-	.075	.013	.00044	.0080	.0066	.0064	.0051
1910	.087	.0042	.00241	.0002	.0001	.075	.0165	.00046	-	-	-	-
1900	.078	.0042	.00282	.00025	.00011	.08	.027	.0007	.0128	.0097	.0081	.0092
1890	.073	.0042	.00325	.0005	.00016	.09	.029	.0010	-	-	-	-
1880	.073	.0042	.0054	.0009	.0002	.10	.048	.0012	.0267	.0076	.0091	.0102

To test this assumption, we have carried out a more detailed allocation by use for each chlorinated pesticide for a single year (1966). The results are shown in Tables 6-6 through 6-11. In five cases (DDT, BHC-lindane, aldrin, chlordane, and dieldrin) the availability of total consumption (domestic sales) data enables us to directly compute the Hudson-Raritan share for all uses (including non-farm) as a fraction of the national total. Shares range from .00014 (aldrin) to .0048 (BHC-lindane), .0052 (dieldrin), .0059 (DDT) to .035 (chlordane). Evidently, the *per capita* use of pesticides in the Hudson-Raritan basin was considerably less than 10% of the national average, for all except chlordane. (The latter was exceptional because 70% of its uses are non-farm).

In the other cases (endrin, heptachlor, toxaphene) there is some doubt of the Hudson-Raritan shares inasmuch as non-farm and public health uses were omitted from the USDA surveys. However,in most cases non-farm uses are known to be minor. In these cases, the Hudson-Raritan shares may be slightly over-estimated. Applying the crop shares from Table 6-5 to the national total sales of the pesticides shown in Table 6-12 gives the Hudson-Raritan use of chlorinated pesticides (Table 6-13).

Table 6-6: 1966 Consumption of Pesticides in the Hudson-Raritan Basin

	DDT			TDE		
	NORTHEAST (tonnes)	H-R (%)	H-R (kg)	NORTHEAST (tonnes)	H-R (%)	H-R (kg)
Corn	1.8	2.9%	52.6	0.9	2.9%	26.3
Other Grain	0.5	6.8%	30.8		6.8%	
Hay (Incl. Alfalfa)		8.3%	0.0		8.3%	
White Potatoes	108.4	4.0%	4336.3	8.2	4.0%	326.6
Other Vegetables	21.8	40.9%	8904.9	51.3	40.9%	20963.7
Apples	139.3	31.1%	43307.6	10.0	31.1%	3103.5
Other Fruits & Nuts	52.2	47.6%	24829.6	9.1	47.6%	4318.2
Other Field Crops	45.4	10.0%	4535.9	6.4	10.0%	635.0
Total Crops	369.2	23.3%	85997.9	85.7	34.3%	29373.3
Total Livestock			1211.1			992.0
TOTAL AGRICULTURAL			87209.0			30365.3
Nursery			102.5			
Turf			54.4			
Home & Garden			300.7			
Pest Control			36854.4			
Forest Use			90.7			
TOTAL NON-AGRICULTURAL			37402.8			
GRAND TOTAL (H-R)			124611.8			30365.3
GRAND TOTAL (U.S.)			21170060			
H-R Share		0.00589				

6.3 Dioxins

The most toxic dioxin (2,3,7,8 TCDD) is apparently a contaminant of chlorophenols, especially 2,4,5-trichlorophenol, mainly used in the manufacture of the herbicides 2,4,5-T and silvex, and the anti-bacterial agent hexachlorophene. It is a known contaminant of these products, and probably of the production wastes. It is also known that the combustion of herbicide treated materials (e.g. in brush fires), or wood

Table 6-7: 1966 Consumption of Pesticides in the Hudson-Raritan Basin

	ALDRIN			BHC-LINDANE		
	NORTHEAST (tonnes)	H-R (%)	H-R (kg)	NORTHEAST (tonnes)	H-R (%)	H-R (kg)
Corn	6.4	2.9%	184.2	6.4	2.9%	184.2
Other Grain	1.8	6.8%	123.4		6.8%	
Hay (Incl. Alfalfa)		8.3%		2.0	8.3%	161.9
White Potatoes		4.0%			4.0%	
Other Vegetables		40.9%		3.2	40.9%	1317.2
Apples	1.8	31.1%	564.3	9.1	31.1%	2821.3
Other Fruits & Nuts	0.5	47.6%	215.9	6.8	47.6%	3238.6
Other Field Crops	0.9	10.0%	90.7		10.0%	
Total Crops	25.0	4.7%	1178.4	27.4	28.2%	7723.2
TOTAL AGRICULTURAL			1178.4			7723.2
Nursery						181.4
Home & Garden			2.7			231.3
Pest Control			43.5			10432.6
TOTAL NON-AGRICULTURAL			46.3			10845.4
GRAND TOTAL (H-R)			1224.7			18568.6
GRAND TOTAL (U.S.)			8766578			3865514
H-R Share		0.00014			0.00480	

treated with the preservative pentachlorophenol, can generate dioxins. In fact, evidence has been presented by Dow Chemical Co. (and challenged by others) that suggests that dioxins may be created to some extent whenever (chlorinated) organic materials are burned at relatively low temperatures. However, the Dow study is widely regarded as methodologically suspect and establishes no firm chain of causation. At present we do not know enough to make any reasonable hindcast of dioxin emissions in the Hudson-Raritan basin.

6.4 PCB Emissions in Hudson-Raritan Basin

The emissions of PCB's in the Hudson-Raritan basin are largely due to a single point source. Two GE capacitor plants at Ft. Edward and Hudson Falls in the upper basin purchased about 35,000 tonnes of PCB's during the 9 years 1966-1974 inclusive. This constituted about 15% of total U.S. consumption during that period of time, and 25% of U.S. consumption for electrical equipment. Assuming a similar percentage of U.S. consumption for electrical equipment manufacturing during the earlier period

Table 6-8: 1966 Consumption of Pesticides in the Hudson-Raritan Basin

	DIELDRIN			ENDRIN		
	NORTHEAST (tonnes)	H-R (%)	H-R (kg)	NORTHEAST (tonnes)	H-R (%)	H-R (kg)
Corn	2.3	2.9%	65.8		2.9%	
Other Grain	0.9	6.8%	61.7		6.8%	
Hay (Incl. Alfalfa)	7.3	8.3%	602.4		8.3%	
White Potatoes		4.0%			4.0%	
Other Vegetables	3.6	40.9%	1484.2	0.5	40.9%	185.5
Apples	4.5	31.1%	1410.7	0.9	31.1%	282.1
Other Fruits & Nuts	0.9	47.6%	431.8	0.5	47.6%	215.9
Other Field Crops		10.0%		0.5	10.0%	45.4
Total Crops	19.5	20.8%	4056.5	2.3	32.1%	728.9
TOTAL AGRICULTURAL			4056.5			728.9
Pest Control			646.4			
TOTAL NON-AGRICULTURAL			646.4			
GRAND TOTAL (H-R)			4702.8			728.9
GRAND TOTAL (U.S.)			906277.4			907184.6
H-R Share		0.00519			0.0008	

1948-1965 for which GE data are lacking (See COW, Chapter 6), the total amount used must have been around 75,000 tonnes. Given the amounts known to have been removed by dredging and the amounts known to remain in sediments, there must have been a total accumulation of the order of 500 tonnes in the upper basin, corresponding to a loss rate from the capacitor plants of 0.67%. Roughly 50-60% of this is now immobilized on dump sites from dredging in 1978-80. Around 64 tonnes remained in the river bed of the upper basin as of 1978.

The rate of escape downstream was 3.6 tonnes/yr in 1976[5] It dropped by 90% in 5 years from 1976 to 1981. The peak year of escape must certainly have been 1974, after the Ft. Edward dam was taken down and a large accumulation was disturbed and subject to scouring by the river. The input to the lower river in 1974 *may* have been as much as 30 or 40 tonnes (although the GE emissions at Fort Edward in that year alone were probably nearer 20-25 tonnes). Even so, given the low observed rates of discharge

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[5] In 1976 there was a major (100-year) flood.

Table 6-9: 1966 Consumption of Pesticides in the Hudson-Raritan Basin

	CHLORDANE			HEPTACHLOR		
	NORTHEAST (tonnes)	H-R (%)	H-R (kg)	NORTHEAST (tonnes)	H-R (%)	H-R (kg)
Corn	1.4	2.9%	39.5	2.7	2.9%	78.9
Other Grain		6.8%		0.5	6.8%	30.8
Hay (Incl. Alfalfa)	0.9	8.3%	75.3	2.3	8.3%	188.2
White Potatoes	1.4	4.0%	54.4		4.0%	0.0
Other Vegetables	0.5	40.9%	185.5		40.9%	0.0
Apples	0.5	31.1%	141.1		31.1%	0.0
Other Fruits & Nuts	0.9	47.6%	431.8		47.6%	0.0
Other Field Crops	0.5	10.0%	45.4	1.8	10.0%	181.4
Total Crops	5.9	16.5%	973.0	7.3	6.6%	479.4
TOTAL AGRICULTURAL			973.0			479.4
Nursery			408.2			
Turf			9434.7			
Home & Garden			33905.1			
Pest Control			118024.7			225.4
Forest Use						7370.9
TOTAL NON-AGRICULTURAL			161772.8			7596.3
GRAND TOTAL (H-R)			162745.7			8075.8
GRAND TOTAL (U.S.)			4642971.0 *			589670.0
H-R Share		0.035			0.014	

* Based on data from Velsicol (COW, Chap 4, Table I-3)

into the estuarine zone at Troy, the only way to explain the 75-90 tonnes's of PCB's estimated to be in river sediments in the lower basin (1979-80), together with recent removal rates in excess of 2 tonnes/yr from New York harbor (Bopp et al 81) is to assume that substantial quantities had already escaped to the lower river prior to 1973, cumulatively amounting to not less than 50-60 tonnes.

A simple model consistent with the known facts assumes that pre-1973 discharges to the lower basin were primarily due to diffusion. Since PCB's are almost insoluble, maximum quantity that can be transferred by this mechanism is essentially a function of the amount of water flowing in the river. Once the saturation rate is reached it will tend to remain constant. Thus, from 1950 until 1973 (when the Ft. Edward Dam was removed) the annual PCB input to the lower basin was roughly constant and would not have exceeded recently (1977) observed low-flow loadings in the upper river, *viz.* 4 to 5 kg/day or 1.8 tonnes/yr.

Table 6-10: 1966 Consumption of Pesticides in the Hudson-Raritan Basin

	TOXAPHENE			ENDOSULFAN		
	NORTHEAST (tonnes)	H-R (%)	H-R (kg)	NORTHEAST (tonnes)	H-R (%)	H-R (kg)
Corn		2.9%			2.9%	
Other Grain	0.9	6.8%	61.7		6.8%	
Hay (Incl. Alfalfa)		8.3%			8.3%	
White Potatoes	0.5	4.0%	18.1	11.3	4.0%	453.6
Other Vegetables	0.5	40.9%	185.5	9.1	40.9%	3710.4
Apples		31.1%		8.6	31.1%	2680.3
Other Fruits & Nuts		47.6%		13.2	47.6%	6261.4
Other Field Crops		10.0%		0.5	10.0%	45.4
Total Crops	1.8	14.6%	265.4	42.6	30.8%	13151.0
Beef Cattle			1280.9			
Dairy			1640.2			
Other			809.2			
Total Livestock			3730.3			
TOTAL AGRICULTURAL			3995.7			13151.0
GRAND TOTAL (H-R)			3995.7			13151.0
GRAND TOTAL (U.S.)			15512857	(1966)		
H-R Share		0.0003				

Table 6-11: 1966 Consumption of Pesticides in the Hudson-Raritan Basin

	METHOXYCHLOR		
	NORTHEAST (tonnes)	H-R (%)	H-R (kg)
Corn		2.9%	
Other Grain		6.8%	
Hay (Incl. Alfalfa)	40.8	8.3%	3388.3
White Potatoes		4.0%	
Other Vegetables		40.9%	
Apples	99.8	31.1%	31034.8
Other Fruits & Nuts	27.7	47.6%	13170.5
Other Field Crops	4.5	10.0%	453.6
Total Crops	172.8	27.8%	48047.2
Beef Cattle			113.4
Dairy			1201.6
Other			13.2
Total Livestock			1328.1
TOTAL AGRICULTURAL			49375.3
GRAND TOTAL			49375.3

Table 6-12: Sales of Chlorinated Pesticides (1945-1980) (tonnes)

	DDT	BHC/ Lindane	Aldrin	Chlor- dane	Diel- drin	Endrin	Hepta- chlor	Toxa- phene	NEC + Export	Domestic Sales
H-R	0.0059	0.0048	0.00014	0.035	0.0052	0.0008	0.0140	0.0003		
1980	0	0 RC 78	0 RC 76	0 RC 75	0 RC 76	? RC 79	900(E)	RC 82 6800(E) RP5/77	?	7700(E)
1976	0 RC 1/73	1800(E)	2700(E)	2250(E)	230(E)	450(E)	830 RC 75(b)	13600(E)	13600	29600
1971	8160	1800(E)	5268	8453	320	643	930	16589	13835	51120
1966	21170	3866	8767	5900(E)	906	900(E)	900(E)	17200(E)	23100	58100
1964	22925	5337	5757	4643	931	984	831	17946	16789	47900
1960	31818	13909	3678	4550(E)	1202	1150(E)	770(E)	17700(E)	11800(E)	29000(E)
1955	28032	31521	1983	4550(E)	1113	1150(E)	680(E)	17700(E)	7700(E)	27200(E)
1950	26144	30741	660 PB 48	4550(E)	50(E) PB 48	500(E) PB 47	500(E) PB ?	12200(E) PB ?	0(E)	18100(E)
1945	14331 PB 42	1810(E) PB 42		450(E) PB 44						

PB = Production began RC = Registration cancelled E = Estimated by author
RP = Regulated Production

Table 6-13: Use of Chlorinated Pesticides in the Hudson-Raritan Basin (1945-1980) (tonnes)

	DDT	BHC/ Lindane	Aldrin	Chlor- dane	Diel- drin	Endrin	Hepta- chlor	Toxa- phene
H-R	0.0059	0.0048	0.00014	0.035	0.0052	0.0008	0.014	0.0003
1980	0	0	0	0	0	0	12.6	2.0
1976	0	8.6	0.4	78.8	1.2	0.4	11.6	4.1
1971	48.0	8.6	0.7	295.9	1.7	0.5	13.0	5.0
1966	124.9	18.6	1.2	206.5	4.7	0.7	12.6	5.2
1964	135.3	25.6	0.8	162.5	4.8	0.8	11.6	5.4
1960	187.7	66.8	0.5	159.3	13.8	0.9	10.8	5.3
1955	165.4	333.6	0.6	159.3	13.4	0.9	9.5	5.3
1950	154.2	147.6	0.1	159.3	2.6	0.4	7.0	3.7
1945	84.6	86.9		15.8				

On the other hand, still more recent measurements at Troy, NY, showed only 0.4 tonnes passing over the dam in 1981. Even if this includes no "scouring" contribution, it is hard to explain unless one assumes that the remaining PCB's are no longer in the channel of the river (where flow rates are always higher) but dispersed along banks and in shallows where flow rates are very slow; or it may be the case that older contaminated sediments are now being covered up by newer material.

Assuming the 1.8 tonnes/yr annual transfer rate for years 1950 through 1973 accounts for 40 tonnes accumulated in the lower basin or transported out to sea. The year after the dam was dismantled a large accumulation behind the dam (~560 tonnes) was disturbed and destabilized. This contaminated material was subsequently redeposited along the next 50 miles of the river and a much larger surface area of sediment was exposed to the scouring action of the water. While the diffusion mechanism was limited by the total flow, much larger amounts of PCB's were moved during periods of high (turbulent) flow. The amounts subject to scouring is roughly proportional to the accumulation in the upper basin. This suggests a delta function "pulse" in 1973-1974 followed by an exponential decline. The accumulated PCB's were also rapidly depleted by dredging during the 1975-80 period. Altogether 240 tonnes were removed by dredging, while 30 to 40 tonnes of the residue seems to be "stabilized" (i.e. no longer subject to scouring).

To summarize, we estimate that PCB inputs to the estuary from the upper Hudson river basin averaged 1.8 tonnes per year from 1950 through 1972, peaked at close to 15 tonnes in 1973, and declined thereafter as shown in Table 6-14:

Table 6-14: PCB Inputs from the Upper Hudson Basin (1973-1981) (tonnes)

1973	15.0
1974	10.5
1975	7.4
1976	5.1
1977	3.6
1978	2.5
1979	1.75
1980	1.0
1981	0.4

The cumulative amount entering the lower basin during 1973-1978 would appear to be of the order of 44 tonnes. Added to the 40 tonnes previously accounted for, we estimate a total accumulation of 84 tonnes in the lower basin at the end of 1978, originating from Ft. Edward.

The above analysis is restricted to one point source contribution. In addition, there has been a measurable contribution, especially since the 1950's, from distributed sources, both dissipative uses and losses from the disposal of equipment and products containing PCB's.

As of 1975 223,000 tonnes of PCB's had been disposed of in various ways (COW, Chapter 6) and possibly almost as much again has been disposed of since then, though more carefully. Of the 223,000 tonnes, it can be assumed on the basis of population that 8% or 18,000 tonnes was in the Hudson-Raritan basin. If only 0.01% of this was lost each year to surface runoff (or groundwater leachate), that would still generate a load of 1.8 tonnes per year in the river -- comparable to the annual quantity contributed by the Ft. Edward point source before 1973.

The runoff contribution has been estimated for 1975 (approximately) using the Heany et al model, discussed in Chapter 1. Based on national average runoff loading factors, this model yields an estimate of 2.23 tonnes of PCB's per year. This is close to the amount annually removed by dredging in New York harbor. In addition, about 0.7 tonnes per year from distributed sources reaches the river from sewage [Bopp et al 81].

6.5 Polyaromatic Hydrocarbons (PAH's) in the Hudson-Raritan Basin

Major sources of atmospheric PAH's, nationwide, have been given for recent years in Table 7-1 (COW, Chapter 7). Most are combustion related. The contributions that are revelant to the Hudson-Raritan basin include the following (Table 6-15):

Table 6-15: PAH Emissions by Activity (tonnes)

Activity	PAH Emissions
coal for residential/commercial heating	270 - 380
coke production	0 - 150
open refuse burning	5 - 67
enclosed refuse burning	3 - 31
motor vehicle exhaust and tires	30
wood-burning fireplaces and stoves	23 - 36
petroleum refining	5 - 6
industrial heating (coal)	6 - 9
electric power generation (coal)	4

Data on fuel use are given in HMFF (Chapter 11) and have been summarized already in Chapter 3 of this

Table 6-16: Hudson - Raritan Fractions of U.S. Totals

	--Bituminous Coal Use--					----------Consumption----------							
Year	Electric Utilities	Railroads	Cement, Steel & Other Indust.	Commercial& Residential	Coke Production	Anthracite Coal	Gasoline	Distillate Fuel Oil	Residual Oil	Lube Oil	Petroleum Refining	Vehicle Miles Travelled (VMT)	Tire Wear
1980	.006	-	.003	0	.02	-	.068	.064	.106	.068	.029	.068	.068
1970	.014	.065	.011	.008	.022	.036	.0665	.114	.176	.0665	.033	.0665	.0665
1960	.058	.09	.064	.030	.027	.205	.065	.128	.121	.065	.044	.065	.065
1950	.086	.09	.044	.020	.029	.324	.0634	.054	.088	.0634	.056	.0634	.0634
1940	.097	.11	.052	.051	.029	.347	.083	.060	.070	.083	.079	.083	.083
1930	.124	.10	.070	.069	.029	.195	.096	.075	.044	.096	.095	.096	.096
1920	.107	.10	-	-	.044	.244	.089	-	-	.089	.085	-	-
1910	.12	-	-	-	-	.240	-	-	-	-	-	-	-
1900	.15	-	-	-	.051	.216	-	-	-	-	.24	-	-
1890	-	-	-	-	-	-	-	-	-	-	-	-	-
1880	-	-	-	-	.17	-	-	-	-	-	.22	-	-

report. For a given year (1970) we could compute approximate emissions coefficients, then calculate national PAH emissions for other years, and finally compute the Hudson-Raritan share. The Hudson-Raritan share of each of these activities (except wood burning and refuse burning) is presented in Table 6-16 .

A simplified estimation procedure is justified, however, by the fact that coal combustion for residential heating is by far the dominant source of atmospheric PAH's. (Wood combustion might have been dominant in some locations, but not in the Hudson-Raritan basin.) In 1970 11.3 million tonnes of bituminous coal were used for residential/commercial heating in the U.S. (along with a small amount of anthracite coal), generating on the order of 330 tonnes of PAH's (roughly 30 ppm). The Hudson-Raritan fraction of this was .008, or roughly 1.8 tonnes. By comparison, automobiles generated about 30 tonnes, of which the Hudson-Raritan share was also about .06 or 1.8 tonnes. Contribution from refuse burning and coke ovens are very uncertain: for coke the Hudson-Raritan share is .022, so the range is 0 to 3.3 tonnes (.022X150) while for enclosed refuse burning the Hudson-Raritan share is probably at least 10% of the national total[6], or 0.3 to 3 tonnes. Other contributions in the area are comparatively negligible. Thus for 1970, the range of atmospheric emissions would seem to be 4-10 tonnes. Background data is given in COW, Chapter 7.

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[6] Because of the large number of municipal incinerators and apartment house incinerators in NYC.

To estimate national atmospheric PAH emissions over the past, therefore, we need only calculate the ratio of past to present residential/commercial coal use in the basin (assuming anthracite coal was also used mainly for this purpose. All the relevant data are given elsewhere in this report and need not be repeated. The results are shown in Table 6-17. Curiously, the *range* of uncertainty was probably lower in the past, when coal use was much more prevalent but most other sources were no greater in magnitude than presently.

Table 6-17: Coal Combustion-Related Atmospheric PAH Emissions (tonnes)

	Nationwide (Coal only)	H-R Fraction	H-R Emissions
1980		0	0
1970	330	.028	9.2
1960	737	.093	68.5
1950	1871	.192	228.3
1940	2045	.159	325.2
1930	2420	.121	292.8
1920	2733	.154	420.9
1900	2180	.098	213.6
1880	343	.060	20.6

As noted above, as of 1970 other combustion-related sources may have generated an additional increment of 2-8 tonnes of atmospheric PAH in the Hudson-Raritan basin, but these sources would in all cases have been smaller (or no larger) in the past and would consequently be insignificant in earlier decades.

Atmospheric PAH emissions contribute to river loadings insofar as they are deposited on land and reach the water via runoff. The probability of this happening is quite high if the deposition occurs over a paved or solid surface. It is rather low if the deposition occurs on open land. Of course most of the emissions occur in the most densely populated area, where the paved fraction is highest. Lacking data, we assume that 25% of the airborne PAH ends up in runoff.

Apart from atmospheric PAH emissions there are PAH emissions direct to waterways from crude oil spills or via runoff of used motor oil. These contributions are considered separately in the next section where oil and grease are discussed. It will be seen that the waterborne contributions are dominant.

6.6 Oil and Grease Emissions to the Hudson-Raritan Basin

Major oil and grease losses in the Hudson-Raritan basin arise from three sources: (1) refineries, (2) spillage from tanker operations and bilge washing in NY harbor and (3) waste motor oil. Detailed discussion can be found in COW, Chapter 8.

Refineries in the New York Harbor area are concentrated near the mouth of the Raritan River and Arthur Kill (separating Staten Island from New Jersey). These refineries formerly supplied most of the needs of the New York metropolitan area and the Hudson-Raritan basin, but no longer do so [PANYNJ 82]. In 1920, these refineries refined 8.4% of the crude oil in the U.S., and Hudson-Raritan basin had 8.9% of the nation's population. After World War II, however, more and more refined products were imported from the Gulf Coast, and by 1980 only 2.8% of U.S. crude oil was refined in the New York area (for 7.2% of the U.S. population). Even so the total amount of crude oil refined increased from 37 million bbl (5.27 million tonnes) in 1920 to 143 million bbl (20.4 million tonnes) in 1980 -- almost 400%.

Refinery losses of oil to wastewater have been variously estimated from 0.025 lb/bbl or 75 ppm to 0.096 lb/bbl (300 ppm), after primary treatment (gravity separation). Assuming the *worst* case (.096 lb/bbl) implies a refinery waste oil loss of 5840 tonnes to the lower Hudson-Raritan basin in 1980. The actual number was probably considerably smaller. Note that even in the worst case (above), the contribution from refineries is negligible compared to other sources of oil.

All of the crude petroleum refined in the New York area is transported by tanker. Under the old system (in effect before 1970) up to 1.0% of this amount (1.4 million bbl or 200,000 tonnes) may have been lost by spills and tank washing. By 1980 this loss was reduced by the LOT procedure to ~0.1% of crude oil shipments or 20,400 tonnes in New York Harbor. In addition, there were substantial shipments of residual oil as fuel for electricity generating plants and industrial users in the New York area (19.3 million tonnes in 1970. 11.8 million tonnes in 1980). Of this, ~1.5% or 289,000 tonnes would have been lost to the harbor by spillage in 1970; reduced to .15% or 18,000 tonnes in 1980. Finally, New York Harbor received substantial shipments of lighter refined products (distillate fuels), amounting to approximately 10.9 million tonnes in 1970 and 5.2 million tonnes in 1980. For light refined products the 1970 loss rate was 0.1% (11,000 tonnes), down to .01% (500 tonnes) in 1980. Total estimated losses of oil from shipping operations in New York Harbor were therefore on the order of 501,000 tonnes in 1970 and 39,000 tonnes in 1980.

We assume the Hudson-Raritan basin consumed lubricating oils in proportion to gasoline. It is true that 60% of lubricating oils are used for purposes other than motor vehicles, but it is not unreasonable to assume that these (industrial, agricultural mining and construction) uses are distributed in roughly the same way as motor vehicle usage. The national total for 1980 was 8.8 million tonnes (Average) of which the Hudson-Raritan share would be 5.5% (based on its share of gasoline use). We have estimated that roughly 17% (plus or minus 3%) of waste oils ends up in surface waters (See COW, Chapter 8). This estimate is fairly uncertain, but the lubricant contribution years prior to 1980 is swamped by tanker spillage. A summary of the estimated oil and grease emissions to the Hudson-Raritan basin is given in Table 6-18 as follows:

Table 6-18: Oil & Grease Losses, Hudson-Raritan Basin (1000 tonnes)

	Crude Oil Tonnage Shipped (a)	Residual Oil Tonnage Shipped (a)	LRP Tonnage Shipped (a)	Refinery Losses (b)	Spills of Crude Oil (c)	Spills of Residual Oil (d)	Spills of LRP (e)	Waste Lube Oil (f)	Total Oil & Grease Losses
1980	20370	11800	5190	5.84	20.4	18	0.5	59	104
1970	20090	19290	10860	5.76	201	289	11	57	564
1960	19520	7750	8520	5.60	195	116	8.5	61	386
1950	17520	3610	-140	5.02	175	54	0.1	53	287
1940	15100	0	-530	4.33	151	0	0.5	45	201
1930	12400	0	-310	3.55	124	0	0.3	45	173
1920	5270	0	0	1.51	53	0	0	28.5	83
1900	1850	0	0	0.53	18	0	0	4.5	23
1880	570	0	0	0.16	6	0	0	2.0	8

(a) Imports computed as consumption less quantity refined (See Table 11-8, HMFF)
(b) Worst case calculated as .096 lb/bbl = 300ppm (See text)
(c) Based on 1% loss for 1970 & earlier; .1% in 1980
(d) Based on 1.5% loss for 1970 & earlier; .15% in 1980
(e) Based on .1% loss for 1970 & earlier, .01% in 1980
(f) Based on 15% of lubricants consumed (See Table 11-8, HMFF)
LRP=Light refined products

It should be noted that waste motor oil reaches the river mainly by way of urban runoff (through separate storm drains) although some may pass through combined sewers. There are also other miscellaneous sources of oil and grease, mainly from food processing, that arrive via sewage.

The runoff contribution can be estimated separately, using the Heany et al runoff model discussed in Chapter 1. Based on national average load factors, this yields an estimate of only 15,000 tonnes/yr (for 1979-80) as compared to our independent estimate of about 58,000 tonnes of waste motor oil. The discrepancy may be partly explained by the high proportion of paved land in the New York area, as compared to the national average for built-up areas, and the fact that other means of disposal of waste oils (e.g. in landfill) are virtually unavailable.

Table 6-19: Waterborne PAH, Hudson-Raritan Basin

	Vehicle Miles Travelled (Billions) (a)	PAH in Motor Oil (tonnes) (b)	PAH to Sewers (15%) (tonnes) (c)	PAH to Sewers (20%) (tonnes) (c)	PAH in Crude Oil (1%) (tonnes) (d)	PAH in Crude Oil (3.5%) (tonnes) (d)	Total PAH Low (tonnes)	Total PAH High (tonnes)
1980	99.1	2480	372	496	204	714	576	1210
1970	77.9	1950	293	390	2010	7035	2303	7425
1960	58.5	1460	219	292	1950	6825	2169	7117
1950	34.5	860	129	172	1750	6125	1879	6297
1940	27.7	690	104	138	1510	5285	1614	5423
1930	19.2	480	72	96	1240	4340	1312	4436
1920	4.9	120	18	24	530	1855	548	1879
1900	0	0	0	0	180	630	180	630
1880	0	0	0	0	60	210	60	210

(a) Based on Hudson-Raritan share of national gasoline use (Table 3-9) times total vehicle miles travelled (VMT), from U.S. Department of Transportation *Highway Statistics* [USDOT 77 & annual].

(b) Assumes average vehicle holds 5 kg of oil and generates 5 mg of PAH per gm of oil per 1000 vehicle miles = 25 kg per million VMT.

(c) Low estimate assumes 15% of waste motor oil lost to sewers, high assumes 20%.

(d) Low estimate assumes 1% of crude oil spilled, high assumes 3.5% (See Table 6-18).

As noted in the foregoing discussion of (atmospheric) PAH's, associated with coal, it is appropriate to consider oil-related PAH's separately here. There are two major sources, viz. waste motor oil and oil spills due to harbor operations. The quantities involved in any given year are quite uncertain, so we present "low" and "high" estimates in Table 6-19 . It is likely that the truth lies between. The relative changes from one decade to another should be about right, however.

Note that, a separate estimate has been made (Chapter 2) of the PAH contribution attributable to runoff, using the Heany et al model, based on average national data. For 1975 (approximately) that estimate was 222 tonnes. This would include PAH contributions from atmospheric deposition on land surface -- presumably less than 10 tonnes (See Table 6-17) -- plus PAH in waste motor oil. Our best estimate of the latter for 1975 is about 380 tonnes. The two estimates are within a factor of 2 and it is not surprising that the Hudson-Raritan estimates are on the high side, due to the relatively high proportion of paved area as compared to national averages. Again, it should be pointed out that the point-source contribution from crude oil spillage was unquestionably dominant by far until 1970, although waste motor oil may now be the largest contributor of waterborne PAH.

6.7 Nitrogen (N) and Phosphorus (P) Emissions to the Hudson-Raritan Basin

As pointed out in COW, Chapters 10 and 11, the major sources of anthropogenic nitrogen and phosphorus emissions to the Hudson-Raritan basin are somewhat different from the major sources of nationwide emissions. In both cases agricultural fertilizer uses are nationally predominant, accounting for over 70% of all fixed nitrogen and nearly 80% of phosphorus. However, agriculture is *not* quantitatively important in the Hudson-Raritan basin and relatively little fertilizer is used. See Table 5-3 in this chapter for Hudson-Raritan fractions of major crops. Roughly 0.1% of national fertilizer use is in the Hudson-Raritan basin (1980), down from 0.2% in the mid 1950's. Nutrients are imported into the region mainly as food products, the major sources of N.P are food processing wastes and sewage. Detergents -- which also end in sewage -- also contribute significantly to phosphates in wastewater. The computational procedure is straightforward: from the urbanized human population in the area one can compute the nitrogen and phosphorus content of wastes, using standard coefficients (10 gm of N and 2.5 gm of P per capita per day). In the late 19th and early 20th century, horses were also used as prime movers (in place of internal combustion engines) and their contribution of waste excrement should also.be included. We assume a horse is the equivalent of four humans in body mass, for purposes of calculating N and P losses. The fraction of urban wastes captured by sewers and the degree of sewage treatment must also be taken into account explicitly. Data on the former is available from surveys of sewage treatment requirements carried out at intervals by various public authorities [See Tarr & McCurley 84, pp. 135-139; ISC annual]. Data on treatment efficiency is found in standard engineering texts. Results are given in Table 6-20 .

In addition to the point-sources identified above, there is certainly a contribution from runoff. In general (i.e. nationally), the sources of runoff are as follows:

Table 6-20: Point-Source Loadings of N & P in the Hudson-Raritan Basin

	Urban Population		Organic Nitrogen from (Urban) Human Waste tonnes		Organic Nitrogen from Horse Waste (Urban) tonnes						Discharge	
	Human (millions)	Horse (Thousands)	Nitrogen	Phosphorus	Nitrogen	Phosphorus	Phosphorus from Detergents tonnes	Urban Pop. with Sewers %	Urban Horse Wastes Drained %	N & P Removed by Sewage Treatment %	N tonnes	P tonnes
1980	15.2	0	55438	13859	0	0	18000	1.03	NA	0.194	46024	26449
1970	16.1	0	58943	14736	0	0	25100	1.00		0.150	50102	33861
1960	14.9	0	54361	13590	0	0	13600	0.99		0.108	48005	24011
1950	13.2	0	48141	12035	0	0	5000	0.97		0.100	42027	14872
1940	11.6	0	42351	10588	0	0	0	0.95		0.094	36452	9113
1930	10.9	0	39926	9981	0	0	0	0.92		0.088	33500	8374
1920	8.6	115.9	31382	7845	1692	423	0	0.90	0.90	0.050	28278	7069
1900	5.1	178.8	18692	4673	2610	653	0	0.75	0.75	0.008	15849	3963
1880	3	250	10769	2692	3650	913	0	0.33	0.5	0	5379	1345

1. Excess fertilizer use
2. Animal feeding operations & slaughterhouses
3. Nitrate deposition from acid rain
4. Special uses of nitrogenous chemicals such as urea (used as a de-icer at major airports)
5. Decaying organic matter

In the Hudson-Raritan basin *total* animal agricultural use of N-fertilizer is no more than 9000 tonnes. Animal feeding operations in the area are mainly dairy farms which do not produce unusual amounts of runoff. Acid rain may now account for an annual input of around 16,000-17,000 tonnes (+or-3000 tonnes), mostly over land. It is unreasonable to suppose that all of these rural sources contribute more than 5000 tonnes *directly* to runoff. Any remainder must be due to the decay of dead organic matter. If the remainder is significant, one would have to conclude that the soil of the Hudson-Raritan basin is becoming severely depleted in nitrogen, which seems rather unlikely.

Using a typical runoff factor for undeveloped rural (i.e. forested) land, the whole Hudson-Raritan basin 3,230,000 hectares would generate about 8700 tonnes of N in runoff. By contrast, using a typical runoff factor for agricultural land, the 441,000 hectares in the Hudson-Raritan basin would account for 22,000 tonnes, which is certainly much too high. Finally, the 840,000 hectares of developed land would generate 10,500 tonnes (assuming separate sewers), which is not unreasonable.

To reconcile these inconsistencies, we assume a modified runoff factor for agricultural land equal to the factor for all developed land with separate sewers; runoff the factor for developed land with combined sewers is assumed to be slightly higher. Detailed estimates are given in Chapter 2.

6.8 Total Organic Carbon in the Hudson-Raritan Basin

The anthropogenic contribution can be computed in a manner very similar to the N and P computations and is shown in Table 6-21 . Note that the figures in the last column refer to (dry) weight of organic material, not carbon content *per se.*

Table 6-21: Organic Carbon Loadings

	Urban Population	Urban Horse Population	Organic Carbon From Human Waste (tonnes) (a)	Organic Carbon From Horse Waste (tonnes)	Fraction of Human Waste Drained to Sewers (b)	Fraction of Horse Waste Drained to Sewers	Fraction of TOC Removedby Sewage Treatmnt (c)	Total Organic Material (tonnes)
1980	15188490	0	803851	0	1.03	1.00	0.6824	262966
1970	16148898	0	854680	0	1.00	1.00	0.3544	551803
1960	14893389	0	788233	0	0.99	0.99	0.1439	668050
1950	13189415	0	698050	0	0.97	0.97	0.1025	607705
1940	11602963	0	614087	0	0.95	0.95	0.0663	544722
1930	10938588	0	578925	0	0.92	0.92	0.0581	501677
1920	8597704	115884	455033	113964	0.90	0.90	0.0188	502495
1900	5121033	178786	271031	175823	0.75	0.75	0.0005	334980
1880	2950314	250014	156145	245871	0.33	0.50	0	174463

(a) Based on urban population

(b) Note that some non-urban human waste reaches sewers from outer suburbs. This accounts for the fraction being slightly greater than unity in recent years.

(c) Estimated by Tarr and McCurley

This calcuation excludes non-anthropogenic sources which contribute to runoff. These can be estimated by the Heany et al runoff model and the results are given in Chapter 2. It is noteworthy that there have been significant changes over time due to shifting land-use categories.

6.9 Water Withdrawals

Water withdrawals cannot be meaningfully discussed on a national basis (although national estimates are available, as shown in COW, Chapter 12, Table 12-1). In the northeastern United States irrigation is not a factor and consumptive uses are minor. In the case of the Hudson-Raritan basin, less than 1% of the water of the upper basin was used consumptively for all purposes in 1975 even in periods of low flow (e.g. flow which is exceeded 90% of the time). In the lower basin withdrawals from the Hudson never exceed 4% on the same basis. The Passaic River, entering Newark Bay is an exception, however [Anderson & Faust 73]. (One reason is that municipal supplies for most population centers in the region are obtained from reservoirs in reserved watersheds.) The only other significant use of water is for purposes of cooling the condensers of steam-electric power plants located in the basin, as shown in Table 6-22. It may be noted that usage for this purpose is of the same magnitude as the river flow (in millions of gallons per day or mgd).

Table 6-22: Water Withdrawals by Electric Utilities (mgd)

	Fresh	Saline	H-R(low flow)
New York (entire state)	4400	8500	11886
New Jersey (entire state)	1000	3200	

It may be assumed that most of the fresh water withdrawals in NY State are taken from Lake Erie, Lake Huron and the upper basin of the Hudson, since the lower Hudson is tidal (i.e. saline) as far north as Troy. In the case of NJ the fresh water withdrawals are taken largely from the Delaware north of Philadelphia. Saline withdrawals are only partly from the Hudson-Raritan estuary. For both NY and NJ many power plants use water from Long Island Sound, the Delaware Estuary or the Atlantic Ocean. Nevertheless, in low flow conditions up to half of the water in the lower Hudson-Raritan basin -- 6000 mgd -- may have been withdrawn for power plant cooling (and subsequently returned) as of 1975. The quantities withdrawn in earlier years can be assumed to be nearly proportional to electricity generation in the basin, adjusted for the somewhat lower generating efficiencies (i.e. greater cooling required) in earlier years.

The derivation of a waste heat index is shown in Table 6-23 . Specifically, we can assume that the water withdrawn by electric utilities is proportional to the amount of waste heat to be dissipated. This, in turn, is proportional to the heat value of the electricity generated (Column 2) times a factor (1-e)/e and normalized to 100 in 1980. In this expression, e is the average thermal efficiency of the steam-electric generating station.[7] Thus the waste heat index is *proportional* to water withdrawals for cooling purposes. It can be converted to mgd in various years by determining the actual withdrawals in any year.

Table 6-23: Waste Heat Index

	Total U.S. Electricity Generated (million kwh)	Electric Power Generated in H-R basin (million kwh)	e: Thermal Efficiency	Waste Heat Index (normalized)
1980	2286.5	164.6	0.34	100.0
1970	1284.2	106.6	0.32	69.3
1960	607.7	53.5	0.31	35.7
1950	232.8	21.6	0.24	21.1
1940	94.0	9.1	0.20	10.9
1930	59.6	5.8	0.17	8.7
1920	23.5	2.1	0.09	6.6
1902	3(E)	0.3	0.07	1.2

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[7] A "heat rate" of 10,000 BTU/kwh is equivalent to a thermal efficiency of 34.12%, since a kwh(e) has a heat equivalent of 3412 BTU.

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I. Landuse & Loading Tables

Table I-1: Landuse - Hudson Raritan Basin (thousand hectares)

	Total	Separate Sewers	Combined Sewers	Agri-cultural	Unde-veloped
1980	3230	737.1	106.7	441	1945
1970	3250	509.1	158.1	449	2134
1960	3260	320.6	170.7	605	2164
1950	3260	159.8	173.2	766	2161
1940	3250	121.0	159.4	916	2054
1930	3250	72.9	141.2	948	2088
1920	3250	37.6	69.1	1299	1844
1900	3250	7.7	19.5	1655	1568
1880	3250			2034	1216

I.1 Notes to Landuse

Total land area and cropland figures were taken from the Census of Agriculture and from *Major Uses of Land and Water in the U.S.: 1974*, U.S. Dept. of Agriculture; Economics, Statistics and Cooperatives Service (1979). Undeveloped land is the remaining unclassified land after accounting for the above variables (see below).

The method of estimation for separate and combined sewer areas for the U.S. is based on Census Bureau estimates of urbanized and specialized land use classifications as presented in the series *Major Uses of Land and Water in the U.S. - 1954, 1959, 1969, 1974*, the 1945 *Inventory of Major Land Uses, the Statistical Abstract of the U.S.* (various issues); and, *Metropolitan Growth in the U.S. - 1920 to 1940* (1954). These figures spanned 1919-1974 at uneven intervals and were then adjusted to obtain estimates for each decade. Data for earlier years was estimated by fitting a curve for 1920-1950 for area based on population (R^2=.97) and extending it to 1880. Figures for the Hudson-Raritan basin urban area were similarly calculated using Census data from the *Social Statistics of Cities* (1880), the series *City and County Data Book* (1947-1972), with supplemental figures reported by the Interstate Sanitation Commission.

Estimates for the U.S. population sewered and treated were previously calculated by the author (see: J.A. Tarr, F.C. McMichael, J. McCurley, et al, *Retrospective Assessment of Wastewater Technology in the U.S., 1800-1972* (NSF, 1977) and indices of the urban population sewered and treated were constructed based on population. Combined versus separate sewerage use indices were then constructed by interpolating from data provided by the Census Bureau in the *Social Statistics of Cities* (1880), the *Statistics of Cities: Special Report* (1909), data from the Public Health Service reported in the series *Inventory of Municipal Waste Facilities* (1941, 1957, 1962, 1968), the National Resources Committee *Report of the Special Advisory Committee on Water Pollution*(1935, 1937, 1939); Athayde, D.N., et al: *Results of the Nationwide Urban Runoff Program* vol. 1 (1983), U.S.EPA, NTIS: PB 84-185552; Heany, J.P., et al: *Assessment of Combined Sewer Overflows*, vol. 1, (1976), EPA-600/2-76-275; McElroy, A.D., et al: *Loading Functions for Assessment of Water Pollution from Non-Point Sources* (1976), EPA-600/2-76-151. Figures to construct comparable indices for the Hudson-Raritan basin utilized the above sources plus data provided in the 1925 Corps of Engineers *Pollution Survey*, several reports from the Interstate Sanitation Commission (1937, 1954, 1972, 1981, 1983); and, Hydroscience, Inc.: *208 Areawide Assessment Procedures Manual*, vol. 1, (1976), EPA-600/9-76-014.

The population based sewerage indices were then used with the land area statistics to derive estimates of the land areas that were encompassed by separate and combined sewers. The runoff concentration factors supplied by M. Small were used with this data to obtain the runoff loading estimates for the Hudson-Raritan basin.

Table I-2: Load Factors for Major Pollutants (kg/ha - cm)

	Developed Land		Agricultural Land	Undeveloped Land
	Separate Sewers	Combined Sewers		
Suspended Solids (SS)	11.35	46.80	10.00	1.000
BOD$_5$ or TOC	0.97	4.00	0.20	0.050
Nitrogen (N)	0.13	0.52	0.50	0.027
Phosphorus (P)	0.02	0.11	0.08	0.005
Oil & Grease (OG)	0.11	0.47	0.01	0.001

Table I-3: Estimated Loading Ratios for Heavy Metals (mg/gSS)

Metal	Developed Land	Undeveloped Land
Ag	0.002	0.001
As	0.2	0.1
Cd	0.05	0.025
Cr	0.3	0.15
Cu	0.1	0.05
Hg	0.005	0.0025
Pb	1	0.5
Zn	1	0.5

Table I-4: Loading Factors for Heavy Metals (g/ha - cm)

	-- Commercial --		-- Industrial --		Developed Land		Agri-	Unde-
	Separate Sewers	Combined Sewers	Separate Sewers	Combined Sewers	Separate Sewers	Combined Sewers	cultural Land	veloped Land
Ag	0.02	0.081	0.026	0.12	0.02	0.10	0.01	0.001
As	2	8.1	2.6	11	2.30	9.55	1	0.1
Cd	0.49	2	0.64	2.7	0.57	2.35	0.25	0.025
Cr	2.9	12	3.9	16	3.40	14.00	1.5	0.15
Cu	0.98	4.1	1.3	5.3	1.14	4.70	0.5	0.05
Hg	0.049	0.2	0.065	0.27	0.06	0.24	0.025	0.0025
Pb	10	40	13	55	12	48	5	0.5
Zn	10	40	13	55	12	48	5	0.5

Table I-5: Loading Ratios for Toxic Organics (micrograms/gSS)

Constituent	Loading Ratio
PCB	1
PAH	10
DDT	0.1
Chlordane	3
Endrin	-
Dieldrin	0.01
Lindane	0.1
Heptachlor	0.01
H. epoxide	0.01
Hexachlorobenzene	-

Table I-6: Loading Factors for Toxic Organics (mg/ha - cm)

	-- Commercial --		-- Industrial --		Developed Land		Agricultural Land	Undeveloped Land
	Separate Sewers	Combined Sewers	Separate Sewers	Combined Sewers	Separate Sewers	Combined Sewers		
DDT	0.98	4.1	1.3	5.3	1.14	4.7	1	0.1
Chlordane	29	120	39	160	34	140	30	3
Dieldrin	0.10	0.41	0.13	0.53	0.11	0.47	0.1	0.01
Lindane	0.98	4.1	1.3	5.3	1.14	4.7	1	0.1
Heptachlor	0.10	0.41	0.13	0.53	0.11	0.47	0.1	0.01
H.epoxide	2	0.41	0.13	0.53	0.11	0.47	0.1	0.01
PCB	10	41	13	53	11	47	10	1
PAH	98	410	130	530	114	470	100	10

Table I-7: Estimated Metals Loading Ratios & Loading Rates for Preindustrial Undeveloped Land

Element	Loading Ratio mg/gSS	Loading Factor g/ha - cm	As Fraction of Current Factor for Undeveloped Land
Ag	0.001	0.001	1
As	0.017	0.017	0.17
Cd	0.0022	0.0022	0.09
Cr	0.094	0.094	0.63
Cu	0.046	0.046	0.92
Hg	0.00028	0.00028	0.11
Pb	0.023	0.023	0.05
Zn	0.1	0.1	0.2

II. Data-Base for Chlorinated Pesticides

Table II-1: Data-Base for Table 6-12
Domestic Sales of Chlorinated Pesticides(1945-1980) (tonnes)

Table	DDT	BHC/ Lindane	Aldrin	Chlor- dane	Diel- drin	Endrin	Hepta- chlor	Toxa- phene
Vol I	6-12	6-12	6-12	6-12	6-12	6-12	6-12	6-12
Vol III	2-2	3-1	4-1	4-3	4-1	4-6	4-8	4-9
1980	0	0	0	0	0	0	900	6800
1979	0	0	0	0	0	100	890	8500
1978	0	600	0	0	0	250	870	10200
1977	0	1200	0	0	0	400	850	11900
1976	0	1800	2700	2250	230	450	830	13600
1975	0	1800	3600	3600	240	490	850	14198
1974	0	1800	4441	4800	250	530	870	14796
1973	0	1800	4536	6000	256	570	890	15393
1972	9979	1800	5383	7200	336	610	910	15991
1971	8165	1800	5268	8453	320	643	930	16589
1970	11638	2000	4041	7900	340	700	925	16711
1969	13724	2250	4491	7400	574	750	920	16833
1968	14857	2500	6210	6900	600	800	915	16956
1967	18260	2741	8206	6400	668	850	910	17078
1966	21170	3866	8767	5900	906	900	900	17200
1965	24034	3152	6476	5272	823	942	865	17573
1964	22925	5337	5757	4643	931	984	831	17946
1963	27744	4454	5512	4625	1218	1020	810	17885
1962	30511	5625	4938	4600	1356	1060	895	17823
1961	29061	10533	4502	4575	1254	1100	880	17762
1960	31818	13909	3678	4550	1202	1150	770	17700
1959	35690	13624	2525	4550	1364	1150	750	17700
1958	30255	14788	2255	4550	1394	1150	730	17700
1957	32205	18741	1103	4550	1212	1150	710	17700
1956	34019	32200	2946	4550	1649	1150	690	17700
1955	28032	31521	1983	4550	1173	1150	680	17700
1954	20465	26009	1358	4550	806	1000	640	16600
1953	28304	27639	560	4550	515	850	600	15500
1952	31785	29504	369	4550	343	700	560	14400
1951	32970	32306	1491	4550	84	600	530	13300
1950	26144	30741	660	4550	50	500	500	12200
1949	16959	9367	300	3700	20	200	200	5000
1948	13608	2287		2900		100	100	1000
1947	16362	2100		2100				
1946	19731	1950		1250				
1945	14331	1810		450				
1944	10000	900						
1943	5000	400						
Created from Vol III references by interpolation and estimation								
-- Estimated Years --								
	43-44	43-47 68-77	49 73-76	All but 64 & 71	49-50 73-76	All but 64 & 71	All but 64 & 71	All but 64 & 71

Table II-2: Data-Base for Table 6-13
Use of Chlorinated Pesticides in the Hudson-Raritan Basin (1945-1980) (tonnes)

	DDT	BHC/ Lindane	Aldrin	Chlor- dane H-R	Diel- drin	Endrin	Hepta- chlor	Toxa- phene
frac	0.0059	0.0048	0.00014	0.035	0.0052	0.0008	0.014	0.0003
Ref	6-6	6-7	6-7	6-9	6-8	6-8	6-9	6-10
1980	0	0	0	0	0	0	12.6	2.0
1979	0	0	0	0	0	0.1	12.5	2.6
1978	0	2.9	0	0	0	0.2	12.2	3.1
1977	0	5.8	0	0	0	0.3	11.9	3.6
1976	0	8.6	0.4	78.9	1.2	0.4	11.6	4.1
1975	0	8.6	0.5	126.2	1.2	0.4	11.9	4.3
1974	0	8.6	0.6	168.2	1.3	0.4	12.2	4.4
1973	0	8.6	0.6	210.3	1.3	0.5	12.5	4.6
1972	58.9	8.6	0.8	252.4	1.7	0.5	12.7	4.8
1971	48.2	8.6	0.7	296.3	1.7	0.5	13.0	5.0
1970	68.7	9.6	0.6	276.9	1.8	0.6	13.0	5.0
1969	81.0	10.8	0.6	259.4	3.0	0.6	12.9	5.1
1968	87.7	12.0	0.9	241.9	3.1	0.6	12.8	5.1
1967	107.7	13.2	1.1	224.3	3.5	0.7	12.7	5.1
1966	124.9	18.6	1.2	206.8	4.7	0.7	12.6	5.2
1965	141.8	15.1	0.9	184.8	4.3	0.8	12.1	5.3
1964	135.3	25.6	0.8	162.7	4.8	0.8	11.6	5.4
1963	163.7	21.4	0.8	162.1	6.3	0.8	11.3	5.4
1962	180.0	27.0	0.7	161.2	7.1	0.9	12.5	5.3
1961	171.5	50.6	0.6	160.4	6.5	0.9	12.3	5.3
1960	187.7	66.8	0.5	159.5	6.3	0.9	10.8	5.3
1959	210.6	65.4	0.4	159.5	7.1	0.9	10.5	5.3
1958	178.5	71.0	0.3	159.5	7.3	0.9	10.2	5.3
1957	590.0	90.0	0.2	159.5	6.3	0.9	9.9	5.3
1956	200.7	154.7	0.4	159.5	8.6	0.9	9.7	5.3
1955	165.4	151.4	0.3	159.5	6.1	0.9	9.5	5.3
1954	120.7	124.9	0.2	159.5	4.2	0.8	9.0	5.0
1953	167.0	132.8	0.1	159.5	2.7	0.7	8.4	4.7
1952	187.5	141.7	0.1	159.5	1.8	0.6	7.8	4.3
1951	194.5	155.2	0.2	159.5	0.4	0.5	7.4	4.0
1950	154.3	147.7	0.1	159.5	0.3	0.4	7.0	3.7
1949	100.1	45.0	0.04	129.7	0.1	0.2	2.8	1.5
1948	80.3	11.0		101.7		.1	1.4	0.3
1947	96.5	10.1		73.6				
1946	116.4	9.4		43.8				
1945	84.6	8.7		15.8				
1944	59.0	4.3						
1943	29.5	1.9						

Calculated by multiplying Table II-1 by appropriate H-R fraction above

Table II-3: Data-Base for Table 2-6; Estimated MER of Chlorinated Pesticides in the Hudson-Raritan Basin (1945-1980) (tonnes)

	DDT	BHC/ Lindane	Aldrin	Chlor- dane	Diel- drin	Endrin	Hepta- chlor	Toxa- phene
Half-Life	12	5	5	5	5	5	5	5
k(i)	0.943874	0.870551	0.870551	0.870551	0.870551	0.870551	0.870551	0.870551
1980	1255	93	2.7	813	11.2	2.8	93.0	29.5
1979	1330	107	3.1	934	12.8	3.3	92.3	31.6
1978	1409	122	3.6	1073	14.8	3.6	91.7	33.3
1977	1493	137	4.1	1233	17.0	4.0	91.4	34.8
1976	1581	151	4.8	1416	19.5	4.2	91.3	35.8
1975	1676	164	5.0	1536	21.0	4.4	91.5	36.5
1974	1775	178	5.2	1620	22.7	4.6	91.4	37.0
1973	1881	195	5.3	1667	24.6	4.8	91.0	37.4
1972	1993	214	5.3	1673	26.7	4.9	90.3	37.7
1971	2049	235	5.2	1632	28.6	5.1	89.1	37.8
1970	2119	261	5.2	1535	31.0	5.3	87.3	37.7
1969	2173	288	5.3	1445	33.6	5.4	85.5	37.5
1968	2216	319	5.4	1362	35.1	5.5	83.4	37.3
1967	2255	352	5.2	1286	36.8	5.6	81.0	37.0
1966	2275	390	4.6	1220	38.3	5.7	78.5	36.6
1965	2278	426	3.9	1164	38.5	5.7	75.7	36.1
1964	2263	472	3.4	1124	39.4	5.7	73.0	35.4
1963	2254	513	3.0	1105	39.6	5.6	70.5	34.5
1962	2215	565	2.6	1083	38.3	5.5	67.9	33.5
1961	2156	617	2.2	1059	35.9	5.3	63.6	32.3
1960	2103	651	1.8	1032	33.7	5.1	59.0	31.0
1959	2029	671	1.4	1002	31.5	4.8	55.3	29.5
1958	1926	696	1.2	968	28.1	4.5	51.5	27.8
1957	1852	718	1.1	929	23.9	4.1	47.4	25.8
1956	1337	721	1.0	883	20.2	3.6	43.1	23.5
1955	1204	651	0.7	832	13.4	3.1	38.4	20.9
1954	1100	573	0.5	772	8.4	2.5	33.1	17.9
1953	1037	515	0.4	704	4.8	1.9	27.8	14.9
1952	922	439	0.3	625	2.4	1.4	22.3	11.7
1951	778	342	0.3	535	0.7	1.0	16.6	8.5
1950	619	214	0.1	431	0.4	0.6	10.5	5.2
1949	492	76	0.04	312	0.10	0.23	4.0	1.8
1948	415	36		209		0.08	1.4	0.3
1947	355	29		124				
1946	274	21		58				
1945	167	14		16				
1944	87	6						
1943	30	2						

Calculated as

$$MER(t) = \sum_{n} (k^{i})^{n} X(t-n)$$

where k(i) are given above, and X(t) are from Table II-2

Table II-4: Data-Base for Table 2-8
Pesticides in the Hudson Raritan Estuary (tonnes/yr)

	DDT	BHC/ Lindane	Aldrin	Chlor- dane	Diel- drin	Endrin	Hepta- chlor	Toxa- phene
1980	1.87	0.08	0.001	2.8	0.005	0.001	0.025	0.020
1979	1.98	0.09	0.001	3.2	0.006	0.002	0.025	0.026
1978	2.10	0.13	0.002	3.7	0.007	0.004	0.024	0.031
1977	2.23	0.17	0.002	4.2	0.008	0.005	0.024	0.036
1976	2.36	0.21	0.006	5.6	0.021	0.005	0.023	0.041
1975	2.50	0.22	0.007	6.5	0.022	0.006	0.024	0.043
1974	2.65	0.23	0.009	7.2	0.023	0.006	0.024	0.044
1973	2.81	0.25	0.009	7.8	0.024	0.007	0.025	0.046
1972	3.56	0.26	0.010	8.2	0.030	0.007	0.025	0.048
1971	3.54	0.28	0.010	8.5	0.030	0.007	0.026	0.050
1970	3.85	0.31	0.008	8.0	0.032	0.008	0.026	0.050
1969	4.05	0.34	0.009	7.5	0.045	0.008	0.026	0.051
1968	4.18	0.38	0.011	7.1	0.047	0.009	0.026	0.051
1967	4.44	0.42	0.014	6.6	0.051	0.009	0.025	0.051
1966	4.64	0.50	0.014	6.2	0.064	0.010	0.025	0.052
1965	4.82	0.50	0.011	5.8	0.060	0.010	0.024	0.053
1964	4.73	0.64	0.010	5.5	0.066	0.010	0.023	0.054
1963	5.00	0.63	0.009	5.4	0.081	0.011	0.023	0.054
1962	5.11	0.73	0.008	5.3	0.088	0.011	0.025	0.053
1961	4.93	1.01	0.007	5.2	0.081	0.011	0.025	0.053
1960	5.01	1.20	0.006	5.1	0.078	0.012	0.022	0.053
1959	5.13	1.20	0.004	5.0	0.085	0.011	0.021	0.053
1958	4.66	1.28	0.004	4.9	0.085	0.011	0.020	0.053
1957	8.66	1.49	0.002	4.8	0.074	0.011	0.020	0.053
1956	4.00	2.14	0.005	4.6	0.095	0.011	0.019	0.053
1955	3.45	2.04	0.003	4.4	0.067	0.011	0.019	0.053
1954	2.85	1.72	0.002	4.2	0.046	0.009	0.018	0.050
1953	3.22	1.75	0.001	4.0	0.029	0.008	0.017	0.046
1952	3.25	1.78	0.001	3.7	0.019	0.006	0.016	0.043
1951	3.11	1.83	0.002	3.4	0.005	0.005	0.015	0.040
1950	2.47	1.65	0.001	3.1	0.003	0.004	0.014	0.037
1949	1.73	0.51	0.0004	2.4	0.001	0.002	0.006	0.015
1948	1.42	0.14		1.7		0.001	0.003	0.003
1947	1.49	0.12		1.2				
1946	1.57	0.11		0.6				
1945	1.09	0.10		0.2				
1944	0.72	0.05						
1943	0.34	0.02						

Calculated as Table 2-7, col 2a times Table II-2 plus
Table 2-7, col 5 times Table II-3