United States
Environmental Protection
Agency

Office of Water
Program Operations (WH-547)
Washington, DC 20460

November 1976
430/9-76-013

Water

EPA

# Application of Sewage Sludge to Cropland:

## Appraisal of Potential Hazards of the Heavy Metals to Plants and Animals



MCD-33

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EPA—430/9-76-013
November 1976

# APPLICATION OF SEWAGE SLUDGE TO CROPLAND:

## Appraisal of Potential Hazards of the Heavy Metals to Plants and Animals

By
Council for
Agricultural Science and Technology

Report No. 64
November 15, 1976

Prepared at request of

Office of Water Program Operations
U.S. Environmental Protection Agency
Washington, D.C. 20460

MCD—33

## EPA Comment

This report is one of a series planned for publication by the U.S. EPA Office of Water Program Operations to supply detailed information for use in selecting, developing, designing, and operating municipal sewage sludge management systems. The series will provide in-depth presentations of available information on topics of major interest and concern related to municipal sewage sludge management. An effort will be made to provide the most current state-of-the-art information available concerning sewage sludge processing and disposal/utilization alternatives, as well as costs, transport, and environmental and health impacts.

These reports are being prepared to assist EPA Regional Administrators in evaluating grant applications for construction of publicly owned treatment works under Section 203(a) of the Federal Water Pollution Control Act as amended. They also will provide designers, municipal engineers, environmentalists and others with detailed information on municipal sewage sludge management options.

Harold P. Cahill, Jr., Director
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ABSTRACT

This report examines the present state of knowledge regarding the potential effects on agricultural crops and animals by heavy metals in sewage sludges applied to cropland, as well as some consideration of possible groundwater and surface water contamination. Other potential effects associated with land application of sewage sludge are not addressed in detail.

The potential effects of metals in sewage sludges applied to cropland are discussed within a national perspective based upon the volume of sludge being produced (present and projected), the concentrations of heavy metals in sludges, the total acreage of cropland potentially affected and resulting impacts on the crops grown. The problem is also addressed in terms of its localized variability, emphasizing the differences in observed effects as dependent upon the characteristics of the sludge applied, the application site characteristics, the method, rate and duration of application and other agricultural management practices (such as pH control), the particular metal of concern, and the crops and animals involved. Various approaches that could be used singly or in combination to reduce the potential impacts are discussed.

As a current state-of-the-art report, recent research results are summarized and the many gaps in data and understanding are identified. Where possible, the importance of missing data is indicated. The report addresses some of the problems associated with monitoring metals concentrations in sludge, soil and plant tissues as well as with determining toxicity under widely varying circumstances.

The report concludes that the overall impact of sewage sludge use on agricultural practices is very small. Even if all the sewage sludge currently produced were applied to cropland, the actual acreage affected would still be very small and would continue to be so even with the anticipated increase in sludge production after full implementation of P.L. 92-500. Nevertheless, in certain localities (e.g., the Northeast) the percentage of cropland affected could be quite significant. The report further concludes that many metals are probably not a significant potential hazard, either because they are generally present in low concentrations, are not readily taken up by plants under normal conditions, or are not very toxic to plants and/or animals.

Two unanswered questions are identified as crucial in determining the potential hazards of applying sewage sludges to croplands. First, and possibly most important to determining hazards to humans, what percentage of an individual's diet is composed of foods affected by heavy metals from sewage sludge? Second, for determining the relationship between plant uptake and transfer to farm animals as well as to humans (about which relatively little is known) what are the cumulative effects of repeated applications of metals in sewage sludges over time?

The committee-prepared report indicates that most heavy metals are susceptible to control through choice of appropriate application sites, limiting the sludge application rate to that required to meet crop nutrient demands, and applying the sludge to well-aerated soils with pH controlled by sound management practices. Several metals (particularly Cd, Zn, Mo, Ni, Cu) are labeled as posing a potential serious hazard under certain circumstances, however, with cadmium presently being the metal of most concern.

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# APPLICATION OF SEWAGE SLUDGE TO CROPLAND: APPRAISAL OF POTENTIAL HAZARDS OF THE HEAVY METALS TO PLANTS AND ANIMALS

Council for

Agricultural Science and Technology

Report No. 64

November 22, 1976

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## FOREWORD

On June 3, 1976, a proposed EPA technical bulletin entitled "Municipal Sludge Management: Environmental Factors" was published in the "Federal Register" for public comment. During the development of this document by an interagency workgroup, considerable concern and conflicting opinions were expressed regarding the merits and potential hazards of applying sewage sludge to agricultural lands. The fate of heavy metals and other potentially toxic elements in sludge in terms of soil contamination and the uptake of these elements by crops has been expressed as one of the most serious potential problems.

As a possible aid in addressing questions concerning heavy metals, EPA requested that CAST constitute a task force to review the most recent research, especially field research, on the application of sludge to cropland and to prepare a consensus statement on the current understanding of the relationships among the metals applied in the sludge, the chemical and physical properties of the soils, the soil and crop management practices, and the plant growth and uptake of these metals by plants. The report emphasizes these subjects. Implications concerning animal health are dealt with to some extent but not in depth. Possible problems connected with the presence of industrial organic compounds, pathogens, and viruses are not considered.

The report that follows was prepared by a group of 30 scientists, most of whom have been actively engaged in research on the application of sewage sludge to agricultural land. Members of the task force met in St. Louis from September 15 to September 17, 1976, and prepared a preliminary draft of the report. This draft was revised by the chairman of the task force, with special assistance from Dr. Malcolm Sumner, a member of the task force. The revised draft was reviewed by each task force member and was then revised again, edited, and reproduced for transmittal.

CONTENTS

SUMMARY

Application of sewage sludge to cropland usually benefits agriculture because of the value of sludge as a soil conditioner and as a source of many essential plant nutrients. However, there is also the possibility that the heavy metals applied in the sludge might be toxic to crops and might increase the heavy-metal concentrations in edible crops sufficiently to have deleterious effects on animals and humans. This report summarizes current knowledge on plant uptake of heavy metals from sludge-treated soils and the implications for the food supply.

At present, only 25% of the sludge produced is applied to land, and not all of this land is used for production of edible crops. Economic and environmental considerations, however, may increase substantially the percentage of the sludge applied to cropland in the future. At the same time, increases in sewered population and upgrading of treatment plants will result in increases in the total amount of sludge available.

If all the sludge produced in the United States were to be applied to cropland at a rate suitable for purposes of nitrogen fertilization, the estimated proportion of the total 1970 cropland required to accept the sludge would be less than 1%. The proportion of the cropland required could increase to 2% by 1985. This land requirement is relatively small, and the nationwide impact would be even smaller because some sludge will always be disposed of by other means. However, where the population is concentrated (e.g., the Northeast), the ratio of sludge produced to available land exceeds the national average. In New Jersey, for example, about 27% of the cropland would be required in 1970, and 55% would be required in 1985. Thus, in localized areas the amount of land available for sludge application could be a limiting factor.

Semiquantitative estimates indicate that application of all available sludge to cropland would not appreciably increase the total amount of heavy metals in crops harvested in the United States. However, significant amounts of some heavy metals might enter the food supply over a period of several decades. This eventuality could be largely circumvented by designating certain lands as "sludge farms" on which repeated applications would be made. The limited area of land on the sludge farms could be properly managed more easily than could the much larger area needed if the sludge were applied in small quantities on many farms. In general, the increase in metal content of plants is greater from the initial sludge application than from subsequent applications. The long-term impact of repeated applications of sludge on metals in the food supply could be substantially reduced by growing corn and other selected crops harvested for their edible seeds or fruits in place of forages or leafy vegetables. On the other hand, in areas in which the produce from sludge-treated land constitutes a large part of the diet, accumulations of some metals may pose a hazard. Industrial pretreatment of wastewater from highly industrialized areas could decrease substantially the heavy-metal contents of sludges, and this would considerably reduce the hazards associated with use of the sludges.

Several criteria, including the nitrogen content, phosphorus content, and heavy-metal content, may be used in determining the quantities of sludge to apply. If sludges are not excessively high in heavy metals, the applications might be based initially on the quantities needed to supply the crop with adequate nitrogen or phosphorus and, with time, on permissible metal levels in the soil. In this way, the life of the application sites would be extended, and the food supply would be protected.

Suggestions have been made to limit the application of metals in sewage sludge to land on the basis of (1) a "zinc equivalents" equation which attributes to nickel and copper a certain toxicity to plants relative to that of zinc and which assumes that the several toxicities are additive; (2) a zinc-to-cadmium ratio which presumes that zinc will become toxic to plants before excessive levels of cadmium can accumulate in the plants; and (3) the cumulative amounts of the metals supplied. Each of these methods has its limitations. None of the methods, if used alone, is universally applicable.

The impact of heavy metals in sludges on plants, animals, and humans may be limited by using rational management methods intelligently. When sludge is applied to land, special attention must be paid to good management of the site. For example, if the soil is allowed to become acid, the solubility of a number of the heavy metals increases, and this could result in their toxicity to plants and in unnecessary accumulation of some of these metals in the food supply. Moreover, adequate steps must be taken in site management to ensure the protection of both ground water and surface water from contamination.

The impact of heavy metals in sludges applied to cropland can be reduced considerably by proper selection of the crops. The benefits of growing non-edible (fiber) or subsequently processed crops such as sugar beets and sugarcane are obvious. In addition, however, considerable flexibility can be achieved by proper selection of edible crops. For example, the entry of heavy metals into the seeds of some crops is limited, and the potential hazard can be reduced by harvesting only the grain for consumption. In general, leafy vegetable tissues accumulate higher levels of heavy metals than do grain crops.

The heavy metals and other potentially toxic elements present in sludges can be divided into two categories, based on whether or not they present a potentially serious hazard to plants, animals, or humans. This subdivision assumes that correct management practices are implemented at the application site. (The classification used here applies to sludge-borne metals that enter plants through the roots and not to metals that may be ingested directly by grazing animals from sludge present on plant foliage or on the soil surface.)

Manganese, iron, aluminum, chromium, arsenic, selenium, antimony, lead, and mercury pose relatively little hazard to crop production and plant accumulation when sludge is applied to soil because all either have low solubility in slightly acid or neutral, well-aerated soils or, as with selenium, are present in such small amounts that the concentration is low in soils. As a result, the availability of these elements to plants is relatively low, and little uptake by plants occurs. Even though many sludges, particularly those from tertiary treatment plants, contain considerable quantities of iron and/or aluminum, these elements will not pose a problem provided that the application site is well managed. In addition to having low solubility in soil, chromium and lead are not readily taken up by plants, and this also limits their entry into the food supply. Addition of sludge to soil seldom increases the chromium concentration in plant tissue; however, because there is evidence that chromium may be deficient in the diets of animals and humans, such small increases in concentration of chromium in plants as might result from application of sewage sludge are not to be viewed with alarm. Most sludges are relatively low in mercury, and very little increase in mercury concentration in plants has resulted from sludge application. Considerable quantities of arsenic can be added to soil in the form of sludge; but, because most plants tend to exclude arsenic from their aerial tissues, little hazard arises from this element. In the case of selenium, very few reports are available to indicate the quantities likely to be applied to land in sludge. Data now available indicate that selenium does not present a hazard. In some cases it is deficient in animal diets, and so somewhat elevated levels in plant tissue could be an advantage. Research is lacking on antimony, but on the basis of present evidence antimony is unlikely to be a potential hazard to plants or animals.

The remaining heavy metals -- cadmium, copper, molybdenum, nickel, and zinc -- can accumulate in plants and may pose a hazard to plants, animals, or humans under certain circumstances. Because of the potential problems associated with these elements, they will be dealt with in greater detail than the preceding group which poses less hazard.

Cadmium is a nonessential element which can be a serious hazard to animals and humans if dietary levels are increased substantially. Median concentrations of cadmium in sludge are low, but some sludges contain appreciable quantities of cadmium. The chemistry of cadmium in soil is not well understood, but its lability in soil is reduced by organic matter, clay, hydrous iron oxides, high pH, and reducing conditions. Annual cadmium application rates, soil pH, and crop species and varieties have a major influence on the cadmium concentration in plant tissue. Many crops may contain undesirable concentrations of cadmium in their vegetative tissues without showing symptoms of cadmium toxicity. The literature on cadmium is replete with seemingly contradictory findings which no doubt result from incomplete knowledge of the systems and reactions involved. The following management options are, nevertheless, available to limit cadmium accumulation in the food supply to a relatively low level on sludge-treated land: (1) maintain soil pH at or above 6.5; (2) grow crops which tend to exclude cadmium from the whole plant or from reproductive tissue; (3) apply low annual rates of cadmium, and use sludges which have a low cadmium concentration; and (4) grow nonedible crops. The last option may be useful in instances in which problems have occurred. Because the greatest detrimental impact of applying sludge to agricultural land is likely to be associated with the cadmium content of the sludge, the potential for limiting the entry of cadmium into the sewage system and methods for removal of cadmium from sludge prior to application to the soil are worthy of investigation.

Copper, although essential to plants, can become toxic to them at high concentrations. Sludges often contain appreciable levels of copper, but application of sludge to soil results in only slight to moderate increases in the copper content of plants. In general, animal diets are deficient in copper; hence, slightly elevated concentrations in animal feeding could be advantageous. Under good management practices, copper in sludges will seldom be toxic to plants and should not present a hazard to the food supply. Copper toxicity in animals would be expected to occur only when copper toxicity is severe in the plants used as feed.

Molybdenum is not particularly toxic to plants, even when applied at relatively high levels. As a result, molybdenum may accumulate in plants at concentrations sufficient to cause molybdenosis in ruminant animals without prior warning from plant behavior. The recommended practice of maintaining the soil pH at 6.5 or higher at sludge application sites results in greater solubility and availability of the molybdenum than would occur at lower pH values. However, since sludges are usually very low in molybdenum, it is doubtful that molybdenum in sludge would present a serious hazard to the health of grazing animals except for the unusual circumstances in which forages from sites receiving high-molybdenum sludge form the major part of the animal diet.

Nickel is not essential to plant growth but seems to be required for poultry. Sludges often contain substantial quantities of nickel, which appears to be more readily available from sludges than from inorganic sources. Nevertheless, toxicity of nickel to plants occurs only on acid soils. If the soil pH is maintained at 6.5 or above, nickel should not cause toxicity to plants or pose a threat to the food supply.

Zinc, an essential element for both plants and animals, is often found in sludge at relatively high concentrations. Additions of sludge to soil may cause substantial increases in the zinc content of plants, but toxicity seldom occurs. Many animal diets are deficient in zinc, and a wide margin of safety usually exists between normal dietary intakes of zinc and those that produce toxicity in birds and animals. Slightly elevated levels of zinc in plants may, therefore, be regarded as beneficial. In general, if the pH of sludge-treated soils is maintained at 6.5 or greater, zinc should not be a hazard to plants or to the food supply unless exceptionally high amounts are added in the sludge.

# INTRODUCTION

At present, disposition of most sewage sludge is made in landfills and lagoons, by incineration and dumping in the ocean, and by application to land. Applications to land include those for purposes of disposal, reclamation of marginal or drastically disturbed land, and improvement of cropland. Because of environmental and economic considerations, particularly those associated with incineration, application of sewage sludge to land often appears to be the most feasible method of disposition.

Several beneficial effects may result from application of sludge to land. Since sludge contains considerable quantities of organic matter, it acts as a soil conditioner. When applied at high rates, it is effective in improving the physical properties of marginal or drastically disturbed lands and in supplying the plant nutrients such areas almost invariably need. When applied to cropland, sludge has the same beneficial effects in smaller degree. Even though the nutrient concentrations in sludge are low, sludge can be applied at rates which will supply all the nitrogen and phosphorus needed by most crops. Based on current fertilizer prices, the nutrient value of sewage sludge ranges from 15 to 30 dollars per metric ton on a dry-weight basis.

Since sewage sludge is a low-analysis material, the cost of transportation, handling, and application may place sludge at an economic disadvantage in comparison with high-analysis, commercial fertilizers if the distance of transport is great. On the other hand, projected natural gas shortages and increased production costs will probably keep fertilizer prices relatively high and may maintain sludge as an economically attractive source of nutrients. In addition to supplying significant quantities of most of the essential plant nutrients, sludges may increase the concentration in plants of certain elements which are at or near deficiency levels for animals. For instance, animal diets are often deficient in trace elements such as zinc, copper, nickel, chromium, and selenium. Crops grown on sludge-treated land usually contain slightly elevated contents of some of these nutrients. Application of sludge to land may thus improve the quality of feeds and forages used for animal consumption.

On the other hand, there are also several problems or potential hazards associated with application of sludge to land. These include public acceptance, odor, pathogens, parasites, contamination of surface or ground waters, toxicity to plants, and increased concentration of potentially toxic elements in the food supply.

Long-term soil contamination, toxicity to plants, and accumulation of toxic elements in the food supply are thought to be the most serious potential problems resulting from application of sludge to cropland. Since conflicting opinions are held among scientists and within regulatory agencies regarding these issues, the principal objective of this report is to summarize current research information on the impact of heavy metals in sewage sludge on crop productivity, the metal content of plants, and the consequences for livestock that consume these plants. Principal emphasis throughout is on low rates of application of sludge commensurate with meeting the nitrogen fertilizer requirements of the crops. Additional objectives are to review the factors which influence uptake, translocation, and accumulation of potentially toxic elements by plants. Plant factors include differences in selectivity among plant species and varieties and plant parts. Soil factors include pH, content of clay and sesquioxides, cation-exchange capacity, redox potential, and texture. As long as the tolerance for heavy metal addition to cropland is not set at zero, better management decisions can be made for the application of sludge if the underlying plant and soil factors which affect accumulation of these metals in plants are well understood.

The interpretations presented are, of course, based on the data now available. As more information is obtained, understanding will be improved, and more specific interpretations can be developed for different circumstances and different sewage sludges.

## ASSESSING THE IMPACTS

It is important to maintain a reasonable perspective of the potential impact of heavy metals and other potentially toxic elements in sludges applied to cropland. The analysis presented in this report overestimates the impacts. The analysis is based on the assumptions that (1) all sludge is applied to cropland, (2) the area of land required to accept the sludge is relatively high because the rate of application of the sludge is not in excess of the amount needed to meet the nitrogen fertilizer requirements of the crop, and (3) all sludge-treated land is used to produce crops for food or feed. Qualifying explanations are then presented as an aid to developing qualitatively realistic and reasonable interpretations of the numerical estimates.

In this portion of the report, we develop first some estimates of sludge production under certain assumptions. Then we estimate the amounts of cropland required to accept the sludge. Then we estimate the background levels of certain metals in crops and the increases in the metal content of these crops due to application of sludge. Selected for special consideration are cadmium, a metal for which an increase in crops is undesirable, and zinc, a metal for which an increase in crops could be desirable.

### Sludge Production

Table 1 gives the human population and sludge production for a densely populated state (New Jersey), for an agricultural state (Illinois), and for the United States. Included also are estimates of the cropland that would be needed if all sludges were applied at annual rates to supply the nitrogen requirements of the crops. Reference to these figures on cropland requirements will be made in the next section. Analogous data by states are given in Appendix Tables 1 and 2.

Sludge production in the United States is influenced by many factors. Basic to any estimation are the present and projected numbers of people served by municipal sewers. The portion of the population served by sewers is estimated to be 67% in 1970 and 75% by 1985. On the basis of population estimates of 204 million people in 1970 and 235 million in 1985 (Water Resources Council, 1972), the sewered population will increase from 135 million in 1970 to 176 million in 1985.

Table 1. Human population, sludge production, cropland, and cropland required annually for the application of sewage sludge in Illinois, New Jersey, and the United States in 1970, and projected values for 1985

| | | | | Cropland required for indicated sludge | | | |
|---|---|---|---|---|---|---|---|
| | | | | Sludge containing 1% avail. N | | Sludge containing 4% avail. N | |
| Area | Population, millions | Sludge produced, thousands of metric tons | Cropland[1/], thousands of hectares | Thousands of hectares | Percent of total cropland | Thousands of hectares | Percent of total cropland |
| | | | 1970 | | | | |
| Illinois | 11.14 | 197 | 9,242 | 17.4 | 0.19 | 70.5 | 0.76 |
| New Jersey | 7.20 | 127 | 170 | 11.3 | 6.68 | 45.4 | 26.71 |
| USA | 203.90 | 3,602 | 131,548 | 322.0 | 0.24 | 1,287.7 | 0.98 |
| | | | 1985 | | | | |
| Illinois | 12.56 | 390 | 8,586 | 34.8 | 0.38 | 139.3 | 1.51 |
| New Jersey | 8.49 | 264 | 166 | 23.5 | 13.83 | 94.0 | 55.34 |
| USA | 234.50 | 7,278 | 126,526 | 650.0 | 0.49 | 2,560.0 | 1.98 |

1/ Harvested cropland in 1975 (Appendix Tables 1 and 2). The total surface areas in Illinois, New Jersey, and the United States are 14.62, 2.03, and 937.05 million hectares, respectively.

Production of sludge on the dry-weight basis per person per day by the sewered population is estimated at 0.055 kg for primary sludge, 0.091 kg for combined primary and secondary sludges (Farrell, 1974), and 0.11 kg for combined primary, secondary, and tertiary sludges. The corresponding figures on an annual basis are 20, 33, and 40 kg. Implementation of advanced wastewater treatment increases sludge production by additional removal of suspended and dissolved solids. The type of sewage treatment employed may thus have greater impact on sludge production than will increases in population. Production of municipal sludge in the United States in 1970 is estimated at approximately 3.6 million metric tons on the dry-weight basis (Appendix Table 1). With projected improvement of wastewater treatment, sludge production is estimated to increase to 7.3 million metric tons per year by 1985 (Appendix Table 2). Production of sludge would be increased still further by pretreating combined sewer overflows to remove 50% of the biological oxygen demand.

The heavy-metal content of sewage sludges, which is of principal concern in this report, is a consequence of the affinity of the organic material in the sludge for the metals and the presence of the metals in the wastewater from which the sludge is developed. The most obvious way to reduce the level of metals that might enter the food supply through application of sludge to cropland is to reduce the amount of metals entering the sewage by pretreating industrial wastes. Although metals will always be present in sludge as a result of their natural content in food and human wastes, leaching from plumbing systems, surface runoff, etc., an effective industrial pretreatment program could significantly reduce the concentrations in many sludges. Principal emphasis could reasonably be placed on metals which could present a potential hazard to animals and humans.

A specific case in point is the experience of the Metropolitan Sanitary District of Greater Chicago, where an industrial pretreatment ordinance has been in effect since 1969. To evaluate the effects of this ordinance, the metal content of anaerobically digested sludges from the Calumet Plant which were placed in storage lagoons before 1969 were compared with sludges from this same plant through the summer of 1974 (Table 2). It is clear from the data that the metal content of the sludge was lower after passage of the ordinance than before. The concentrations of metals of greatest environmental concern (nickel, copper, lead, and cadmium) were reduced in the digested sludges by 92, 81, 73, and 72%, respectively, during the period from 1969 to 1974. Data for 1976 are similar to those for 1974, which indicates that this level of industrial pretreatment has reached its limits of effectiveness for this plant.

Although an industrial pretreatment program can effect substantial reductions in the metal content of sludges in industrial areas, it must be emphasized that each sewage treatment plant or the area served by one has an indigenous concentration for all metals which may not be further reduced by more stringent industrial pretreatment. For example, Chicago's industrial waste control ordinance has had no appreciable effect on the metal content of sludge from the Hanover Park Plant, which serves a nonindustrial area. Digested sludges from this plant had a cadmium content of 60 ppm in 1969 and 56 ppm in 1975. The cadmium content of the Hanover Park sludge before and after the 1969 ordinance is similar to that of the Calumet sludge in 1974.

An industrial pretreatment program implies the existence of effective monitoring and enforcement procedures. In view of the political ramifications

Table 2. Metal content of sewage sludge from the Calumet Sewage Treatment Plant of the Metropolitan Sanitary District of Greater Chicago produced before and after passage of an ordinance in 1969 limiting the content of certain metals in industrial wastewater, and the limits established for the metals in the wastewater (McCalla et al., 1977)

| Metal | Maximum concentration per liter of wastewater by ordinance, mg | Content of metal per gram of dry sludge from indicated source, μg | | | Reduction in metal content of sludge | |
|---|---|---|---|---|---|---|
| | | Samples from lagoons filled before 1969[1/] (A) | Samples from anaerobic digesters 1972[2/] | Samples from anaerobic digesters 1974[3/] (B) | A/B | Percent |
| Cadmium | 2.0 | 190 | 100 | 54 | 3.51 | 72 |
| Chromium (total) | 25.0 | 2,100 | 1,100 | 790 | 2.66 | 62 |
| Copper | 3.0 | 1,500 | 900 | 282 | 5.32 | 81 |
| Iron | 50.0 | 53,700 | 36,800 | 24,200 | 2.22 | 55 |
| Mercury | 0.0005 | 3.3 | 3.0 | 2.15 | 1.53 | 34 |
| Nickel | 10.0 | 1,000 | 200 | 77 | 12.99 | 92 |
| Lead | 0.5 | 1,800 | 1,800 | 486 | 3.70 | 73 |
| Zinc | 15.0 | 5,500 | 3,500 | 2,800 | 1.96 | 49 |

1/ Mean of 10 samples.

2/ Mean of 22 samples from June to October, 1972 (Peterson et al., 1973).

3/ Mean of 6 samples from June to October, 1974.

of these procedures, the effectiveness of an industrial pretreatment program cannot be generalized from one area to another unless the monitoring and enforcement are federally mandated and implemented.

Tertiary treatment of wastewater may entail chemical precipitation of phosphate and coagulation of additional solids with lime, ferric chloride, or alum. Tertiary treatment could remove additional metals, but only a slight increase would be expected since the secondary sludge already contains most of the metals found in the influent. Since tertiary treatment generates additional sludge, the metal concentration in the combined primary, secondary, and tertiary sludges probably will not change appreciably from the concentration in the combined primary and secondary sludge before advanced wastewater treatment. Care is needed to ensure that the sources of ferric chloride and alum used in tertiary treatment do not contribute significantly to the content of metals in the sludge. Some sources of these chemicals contain appreciable levels of heavy metals.

Finally, because in this report the nitrogen content of sludge will be used as a basis for calculating the appropriate quantity of sludge to apply to cropland, comment on the nitrogen content is appropriate. The processing to which the sludge is subjected has a marked influence on the nitrogen content, particularly the available nitrogen, which represents the portion of the total nitrogen that is available to plants. Available nitrogen is the inorganic nitrogen plus the mineralizable portion of the organic nitrogen. The total nitrogen content of a liquid sludge on a dry-matter basis may decrease from 6% to less than 3% after composting, with little change in metal content. To provide equal amounts of total nitrogen, therefore, the amount of composted sludge required would be more than twice that of liquid sludge, on a dry-matter basis, resulting in the addition of more than twice as much of the metals per unit area and affecting less than half as much cropland.

## Cropland Requirements

The disposition of municipal sewage sludge in the United States in 1975 has been estimated to be 15% in the ocean, 25% in landfills, 35% by incineration, and 25% by application to land (Bastian, 1976). Environmental regulations and economic forces are placing restrictions on all disposal methods. Elimination of ocean dumping and pipe discharges in 1981 will make available an increasing amount of sludge for disposition in other ways. The greatest relative increase is expected to occur in application to land.

Increasing attention is being given to the application of sewage sludge to cropland for use in agricultural production because the sludge is indeed a valuable resource for this purpose. This section of the report contains estimates of cropland required to accept all the sludge at rates of application sufficient to meet the nitrogen requirements of the crops.

Gross acreage requirements for agricultural utilization of municipal sludges can be estimated on the basis of sludge production, generalized application rates, and assumed values for the average nitrogen percentage in the sludge. If the sludge contains 1% nitrogen, the proportion of the total U.S. cropland required to accept all U.S. sludge at rates low enough to provide for efficient use of the nitrogen by the crops may be estimated at 0.24% in 1970 and 0.49% in 1985. Cropland requirements on this basis thus seem negligible. Even so, the estimates of cropland requirements are inflated because the figure used for the nitrogen percentage in the sludge is low. Moreover, the assumption is made that the nitrogen percentage in the sludge will be the same in 1985 as in 1970. This assumption also tends to overestimate the cropland requirement in 1985. As municipalities increasingly adopt tertiary treatment of wastewater, the amount of sludge produced will increase, but the nitrogen percentage in the sludge will decrease because the tertiary sludge contains a lower percentage content of nitrogen than do the primary and secondary sludges.

Estimates of the type given in the preceding paragraph do not tell the whole story, however, because the centers of greatest sludge production are the major metropolitan areas. Population centers in the Northeast, for example, are not in close proximity to extensive agricultural land. If the sludge produced in such areas is to be applied close to the source, therefore, the proportion of the total cropland needed for sewage sludge application will be far above the national average.

Population, sludge production, and cropland estimates are given in Table 1 for a densely populated state (New Jersey), an agricultural state (Illinois), and the United States as a whole, along with estimates of cropland needed if all sludges were applied at annual rates to supply the nitrogen requirements of the crops. The table indicates the variability of cropland needs among states and the importance of the ratio of sludge production to cropland availability. Figures by states are given in Appendix Tables 1 and 2.

Another factor of importance in application of sewage sludge to cropland is the transportation cost. The limited availability of cropland for application of sludge near urban centers increases the distance the sludge must be transported and the cost of the transportation. Although these costs may be relatively high, they may nevertheless be considered acceptable in light of the economic and environmental costs of the next best alternative. It seems likely, however, that, to save on transportation costs, an appreciable proportion of the sludges in such areas will be applied to nonagricultural land near the municipality.

## Metal Content of Crops

As a basis for assessing the potential impact of sludge on metals in crops, average background levels of cadmium and zinc (from such sources as native levels of the metals in soil, additions of phosphate fertilizer, and air pollution) were estimated for most agronomic crops in Illinois, New Jersey, and the United States. These values are found in Table 3.

The estimates of the increases in cadmium and zinc content of the crops due to application of sludge were based on the assumption that the recovery of these metals applied in the sludges would not exceed 1 and 3%, respectively. This approach, based on field data from Wisconsin (Appendix Table 3), was used to calculate the values in Tables 4 and 5. Crop recovery of metals varies with many factors, including the pH and cation-exchange capacity of the soil, crop species, plant part harvested, and number of years of cropping over which

Table 3. Background levels of cadmium and zinc in crops grown in Illinois, New Jersey, and the United States

| Crop | Total yield,[1] millions of metric tons | Cadmium[2] Mg per kg of crop | Cadmium[2] Total kg in crop | Zinc[2] Mg per kg of crop | Zinc[2] Total metric tons in crop |
|---|---|---|---|---|---|
| | | | Illinois | | |
| Corn grain | 31.56 | 0.05 | 1578 | 25 | 789 |
| Small grain | 2.23 | 0.1 | 223 | 40 | 89 |
| Soybean grain | 7.94 | 0.1 | 794 | 25 | 199 |
| Forages | 3.22 | 0.5 | 1613 | 25 | 81 |
| TOTAL: | | | 4208 | | 1158 |
| | | | New Jersey | | |
| Corn grain | 0.17 | 0.05 | 8.6 | 25 | 4.3 |
| Small grain | 0.08 | 0.1 | 7.7 | 40 | 3.0 |
| Soybean grain | 0.06 | 0.1 | 5.5 | 25 | 1.4 |
| Forages | 0.28 | 0.5 | 136.4 | 25 | 6.9 |
| TOTAL: | | | 158.2 | | 15.6 |
| | | | United States | | |
| Corn grain | 146.50 | 0.05 | 7325 | 25 | 3662 |
| Small grain | 75.88 | 0.1 | 7588 | 40 | 3035 |
| Soybean grain | 41.43 | 0.1 | 4143 | 25 | 1036 |
| Forages | 120.70 | 0.5 | 60350 | 25 | 3017 |
| TOTAL: | | | 79406 | | 10750 |

[1] Total yields are for 1975 crops (Crop Reporting Board, SRS, USDA); small grains include oats, barley, and all wheats.

[2] Based on average cadmium and zinc contents of crops derived from currently available data. Sources of the metals include natural contents in soils and additions from fertilizers and other sources.

Table 4. Potential impact of the application of sewage sludge on the cadmium content of crops grown in Illinois, New Jersey, and the United States on the basis of recovery of the applied cadmium in crops and on the assumption that all the sludge produced is applied to cropland

| Area | Sludge produced, thousands of metric tons | Total Cd in sludge,1/ kg | Background Cd in crops presently grown2/, kg | Increase in Cd content in all crops, assuming 1% recovery of Cd applied in sludge3/ | |
|---|---|---|---|---|---|
| | | | | Kg | Percent |
| Illinois | 197 | 3,940 | 4,220 | 39.4 | 1.0 |
| New Jersey | 127 | 2,540 | 160 | 25.4 | 15.9 |
| USA | 3,602 | 72,040 | 79,406 | 720.4 | 0.9 |

1/ Based on an average concentration of 20 mg of Cd per kg of sludge (dry-weight basis).

2/ Based on 1975 crop yields and levels of 0.05, 0.1, 0.1, and 0.5 mg of Cd per kg in corn, soybeans, small grains, and forages, respectively. Values are for the grain of the grain crops and for the total above-ground parts of the forages.

3/ See Appendix Table 3.

Table 5. Potential impact of the application of sewage sludge on the zinc content of crops grown in Illinois, New Jersey, and the United States on the basis of recovery of the applied zinc in crops and on the assumption that all the sludge produced is applied to cropland

| Area | Sludge produced, thousands of metric tons | Total Zn in sludge,1/ metric tons | Background Zn in crops presently grown2/, metric tons | Increase in Zn content in all crops, assuming 3% recovery of Zn applied in sludge 3/ | |
|---|---|---|---|---|---|
| | | | | Metric tons | Percent |
| Illinois | 197 | 394 | 1,160 | 11.8 | 1.0 |
| New Jersey | 127 | 254 | 16 | 7.6 | 47.5 |
| USA | 3,602 | 7,204 | 10,750 | 216.1 | 2.0 |

1/ Based on an average concentration of 2,000 mg of Zn per kg of sludge (dry-weight basis).

2/ Based on 1975 crop yields and levels of 25 mg of Zn per kg in corn, soybeans, and forages, and 40 mg of Zn per kg in small grains. Values are for the grain of the grain crops and for the total above-ground parts of the forages.

3/ See Appendix Table 3.

the recovery is determined. Nevertheless, the 1 and 3% maximum recovery levels for cadmium and zinc were not exceeded in the Wisconsin work, in which the soil pH averaged about 5.5 (Appendix Table 11), even with cropping for several years after application of the sludge.

The values in Tables 4 and 5 indicate a considerably greater impact of application of sludge on the metal content of crops in New Jersey than in Illinois. The reason is that, although the production of sludge in Illinois in 1970 is estimated to be 56% greater than that in New Jersey (Table 1), there is far more cropland in Illinois than in New Jersey. A 16% increase in cadmium content is estimated for the crops in New Jersey and a 1% increase in cadmium content for those in Illinois. A 48% increase in zinc content is estimated for the crops in New Jersey and a 1% increase for those in Illinois. Values for the United States as a whole are similar to those for Illinois.

The values for cadmium and zinc in Tables 4 and 5 are based on the assumption that all sludges applied have concentrations of cadmium and zinc slightly above the 16 and 1,890 ppm median values found in over 200 different U.S. sludges by Sommers (1976). If the cadmium and zinc values were twice as high in the sludges, the estimated increases in metal content of the crops would be twice as great as those given in Tables 4 and 5.

Soil reaction has an important influence on the absorption of heavy metals by crops from sludge-treated soils. Almost a ten-fold reduction in zinc, cadmium, and manganese content may be achieved by liming acid soils (pH 4.5 to 6) to a nearly neutral condition (pH 6 to 7). Because of this fact, pH control is recognized as a fundamental requirement for proper management of sludge-treated soils, and the estimates of heavy-metal uptake by crops in this section apply to soils that are in a nearly neutral condition.

The data in Tables 3, 4, and 5, showing the impact of sludge on metal uptake by crops, are based on estimated levels of metals present in the grain of grain crops and the total above-ground parts of forages. The total use of grains for food and industrial purposes as a percentage of total production has been estimated at 7.5% for corn, 35% for wheat, 28% for barley, and 5% for oats. The remainder is used for livestock feed, exports, and seed (U.S. Department of Agriculture, 1971). Therefore, only a minor part of the grain and none of the forages are used for direct consumption by humans. If humans consume meat from animals raised on crops grown on sludge-treated soils, the heavy-metal content will be lower in the muscle meat than in the crops used as animal feed. In the liver and kidneys, however, the heavy-metal levels may be above those in the feed.

If the total diet were derived from sludge-treated cropland, the increase in intake of heavy metals over the background level would thus be greater if the persons in question were vegetarians than if they consumed some animal products. In metropolitan areas, where the ratio of sludge production to available cropland is greatest, there might be a few agricultural producers whose diet would come mostly from sludge-treated land. Because of the modern food system, however, the major part of the diet of most of the population would be derived from areas that have not been treated with sludge. The excess of metals from sludge-treated soils would thus be diluted with food from soils that have not been treated.

## CONTROLLING THE IMPACTS

### Limiting the Rate of Application

The quantities of sludge applied to agricultural land will be limited in many respects by regional agronomic practices. Guidelines proposed by USDA and various states are based upon fertilizer recommendations for nitrogen. Nitrogen is the fertilizer element applied in greatest amount to soils, and it is found in sludge in substantial amounts. Therefore, there is good reason to base the application of sludge on its nitrogen content.

In addition to application rates based upon the nitrogen fertilizer value of sludge, constraints have been placed on other constituents in the sludge which may adversely affect crop productivity or may result in concentrations in the edible part of the plant which may adversely affect the health of animals or humans. In Pennsylvania, for example, applications of sludge are based on the nitrogen requirement of the crop, the nitrogen and heavy-metal content of the sludge, and the toxicity of the sludge to plants as estimated by chemical analysis and biological assay (Baker and Chesnin, 1975). The constituents in sludge generally considered to be toxic to plants when they occur in soils at elevated levels are boron, cadmium, copper, molybdenum, nickel, and zinc. The tolerance of plants to levels of these elements in soils varies widely with plant species as well as soil chemical and physical properties. Consequently, it is not possible to select a single level of an element or combination of elements which would be suitable for all crops and soils.

Limiting the concentration of potentially toxic metals in the sludge was one of the first methods suggested for limiting the application of sludge-borne metals to land. Such a limitation would have the salutary effect of encouraging industrial pretreatment and other programs which would lower the concentration of heavy metals in the sludge. Although lowering the heavy-metal content would improve the acceptability of many sludges for application to cropland, the concentration of sludge-borne metals in plants is related more closely to the total amount of the metals applied than to the concentration of the metals in the sludge, fertilizer, or other sources. Thus, a limit on the concentration of metals in sludge would not necessarily protect plants and animals from the hazards that might result from applying excessive amounts of metals to land.

Recently, guidelines for maximum permissible metal application have been developed by several state agencies. These have included recommendations based upon maximum amounts of single metals which can be applied (Sommers and Nelson, 1976) and on the maximum amount of zinc, copper, and nickel which can be applied together. The latter is the so-called zinc equivalent ($Zn + 2Cu + 4Ni$), where 2 and 4 are coefficients to express the toxicity of copper and nickel relative to that of zinc. The various alternatives for limiting sludge application to land based on recent research information and our best judgment will now be evaluated.

Nitrogen Basis

Sludges typically contain from 1 to 6% nitrogen (Sommers, 1976; Keeney et al., 1975), which is partly in inorganic form and partly in organic form. Generally 30 to 60% of the total nitrogen in anaerobically digested fluid sludges is present in the ammoniacal ($NH_4^+$) form, and the remainder is present in the organic form (King, 1976). If fluid sludges are applied directly to land, a certain percentage of the ammoniacal nitrogen is lost by volatilization as ammonia as the sludge dries. The actual amounts of loss vary, depending upon soil and sludge properties and environmental conditions, but they will range from about 30% to essentially complete loss. Amounts of this form of sludge-borne nitrogen which will be available for utilization by plants must be determined for the conditions under which the sludges are applied. Incorporation of fluid sludges below the soil surface by injection or by immediate incorporation following application minimizes the amount of nitrogen lost by volatilization.

Nitrogen in organic forms is unavailable to plants and is not lost following surface application. In soils, organic nitrogen must undergo mineralization (i.e., conversion from the organic to inorganic state) before it can be utilized by plants. The rate of this microbially related conversion depends upon a variety of soil environmental factors such as water content, aeration, pH, temperature, and level of nitrogen in the inorganic state. Although precise rates for the various climatic regions are not completely worked out, the available data indicate that from 15 to 40% of the organic nitrogen is mineralized in the year of application (Pratt et al., 1973; Keeney et al., 1975). Lesser percentages of the remaining nitrogen are mineralized in succeeding years. So-called decay series have been worked out for a few regions, and these can be utilized to approximate the proportion of the nitrogen which will become available each succeeding year. For example, Keeney et al. (1975) suggest 15% availability in the first year, 6% of the remaining nitrogen in the second year, 4% in the third year, and 2% in the fourth year.

Generally, the quantities of sludge required to satisfy nitrogen fertilizer requirements of crops will range from 5 to 40 metric tons per hectare. Techniques to compute the quantities of a particular sludge needed to supply specific amounts of available nitrogen have been worked out (Keeney et al., 1975; Sommers and Nelson, 1976).

It is well known that nitrogen which leaches below the root zone will be in the nitrate form, and eventually this nitrate may contaminate ground-water supplies. For this reason, it is advisable to gear sludge applications to supply sufficient but not excessive nitrogen for crop needs.

Basing sludge application rates on the needs of the crop will often be a safeguard in that use of this criterion will limit the application of potentially toxic heavy metals to levels that are of no concern except for sludges that have an excessively high content of metals. However, when applications are made continually to the same tract of land over a long period of time, metals could eventually accumulate to toxic levels.

Metal Basis

Metal concentrations in sludges vary over wide limits. Recent information on the composition of more than 200 sludges from the North Central Region of the United States has been compiled by Sommers (1976). The literature contains numerous references which show that, when concentrations of certain metals build up in soils from application of inorganic salts of these metals, growth of a wide variety of crops is affected (Baker and Chesnin, 1975; Chaney and Giordano, 1977; Page, 1974). For this reason, some guidelines for the application of sewage sludge to agricultural land have suggested limits on the quantities of metals, particularly copper, nickel, zinc, and cadmium, which can be safely applied.

Early attempts to limit metals applied to soils in the form of sludges were based upon the zinc-equivalents concept. Briefly, this concept assumes that toxicities of copper and nickel can be expressed in terms of some multiple of zinc and that the toxicities of these three elements to plants are additive. The zinc-equivalents equation was based upon a limited amount of data. Information developed since its introduction shows that the toxicity of these elements generally is not additive and that use of the equation greatly underestimates the amounts of sludge-borne metals which can be safely applied to near neutral, neutral, and calcareous soils. Furthermore, the equation does not apply uniformly over a broad spectrum of plant species.

During the past two years, the NC-118 and W-124 Cooperative State Research Service Technical Committees have been working on the development of suggested maximum rates of metal application to land (Sommers and Nelson, 1976). They recognize that the maximum safe applications of individual metals may differ among soils as a consequence of differences in cation-exchange capacity of the soils and differences in relative toxicity of the various metals. They have eliminated the concept of additivity of the toxic effects of the metals that appeared in the zinc-equivalents equation and instead have made suggestions based on the total limits of addition of the heavy metals on an individual basis. It should be pointed out, however, that the rates of application suggested by these committees cannot be considered definitive and that the rates are likely to be changed as additional research information becomes available.

To judge from studies of the effects of adding inorganic salts of metals to soils and studies of soils contaminated with metals from mining and smelting activities, it is certain that there is a limit to the extent to which soils can be enriched in heavy metals and still maintain their normal crop productivity. Data currently available are not sufficient to determine the maximum amounts of heavy metals that can be tolerated in additions of sludge. The information at hand strongly suggests that the maximum additions tolerated will depend upon interactions with other constituents in the sludge and soil. These interactions will depend upon soil chemical and physical properties and regional factors such as climate and water quality. The question of maximum rates of application is complex and cannot be answered with certainty without additional research.

It has also been suggested that rates of application of sludge should be limited by the ratio of zinc to cadmium in the sludge. This concept has as its basis two premises. First, the ratio of zinc to cadmium averages 500 for parent rocks and 100 for soils, which means that, during weathering, cadmium has not been lost as rapidly as has zinc. Second, regulation based on the ratio of zinc to cadmium would result in high enough zinc concentrations in soil to kill plants before cadmium could accumulate to levels in foods considered hazardous to animals and humans. More recent research results (Chaney et al., 1976b; Giordano and Mays, 1976b; Page, 1976) show that this premise often is not correct and that many plants grown on nearly neutral to calcareous soils will tolerate high levels of zinc in the soil and will still show an increase in the concentration of cadmium. Furthermore, at a given ratio of zinc to cadmium, the concentration of cadmium in plants increases with increasing cadmium applications (Appendix Tables 19, 20, and 21). Hence, it seems advisable to abandon the ratio of zinc to cadmium in sludges as a criterion for limiting or regulating applications of sludge to soil, especially if the pH is above 6.5. In these soils, cadmium limits can be based solely on annual and total or "lifetime" rates of application. In acid soils, use of a combination of the ratio of zinc to cadmium in the sludge and annual and total rates of application may be advisable.

If sludge applications are based initially on supplying the crop with adequate nitrogen and safe annual applications of toxic metals and, subsequently, as metals accumulate with time, on permissible metal levels in soil, the life of the site will be extended, and the food supply will be protected. The impact of heavy metals in sludges on plants, animals, and humans may be limited by using rational management methods intelligently, making adjustments with time as may be appropriate.

## Phosphorus Basis

Sludges generally contain from 1 to 3% phosphorus. If sludge is applied in amounts sufficient to satisfy the nitrogen requirements of crops, more than adequate amounts of phosphorus will nearly always be added. The nutrients in sludge could thus be used more efficiently if the sludge were applied in quantities to meet the phosphorus needs of the crops instead of the nitrogen needs. This practice, however, would result in a low rate of sludge application, perhaps 1 to 2 metric tons per hectare, and the cost of application would be increased accordingly.

If sludge applications are based on the phosphorus needs of the crop, the additions of heavy metals will be even less than those applied if nitrogen is used as the base. This management option will nearly always require the application of supplemental nitrogenous fertilizer to nonleguminous crops.

## Boron Basis

Boron may limit the amounts of sludge which can be applied in irrigated regions. Concentrations of boron in sludges typically range from 100 to 1,000 ppm. If application of sludge causes the concentration of boron in the soil solution to reach 1 ppm, damage to boron-sensitive crops may occur. In humid regions and in irrigated regions where the irrigation water is low in boron, the boron added in the sludge will be diluted below the threshold toxic concentration and will present little hazard.

### Soluble Salt Basis

Fluid sludges contain a variety of soluble salts. In irrigated arid and semi-arid regions where the soils and/or irrigation water contain relatively high concentrations of salts, practices to maintain salinity levels in the root zone at acceptable levels are required for successful sludge utilization in agriculture.

## Accessory Criteria

### Soil Properties

Many soil properties are important in considering the use of sewage sludge on land. Among these are pH, texture of the surface soil, cation-exchange capacity, organic matter, and profile characteristics. Of these, the pH is perhaps the most important. Most heavy metals are more soluble under acid conditions than under neutral to alkaline conditions, molybdenum being the main exception. It is important that the soil pH be maintained at 6.5 or above following sludge application.

The cation-exchange capacity of a soil is a reflection of the content of organic matter and of the nature and content of clay and sesquioxides. The cation-exchange capacity thus gives an integrated value that relates to the chemical behavior of a soil. In general, the higher is the cation-exchange capacity, the more sludge a soil can accept without potential hazards.

Although little information is available, application of sludges to Histosols (organic soils) does not seem desirable. Organic soils often have low pH values, high nitrification rates, and high water tables. Application of sludge may result in increased nitrification and loss of nitrates. Even though organic soils have high capacities for metal sorption, excessive amounts of these metals may become available in acid soils.

Care should be exercised not to apply sludges on wet soils with heavy application equipment such as tank trucks or tank wagons. The heavy equipment may compact the soil, causing later problems in tillage and in plant growth. Compaction may also enhance runoff. Equipment with large flotation-type tires has been developed which may reduce this problem.

### Drastically Disturbed Lands

Sewage sludge is an excellent soil amendment for renovating drastically disturbed lands. For plant establishment, it is often desirable to apply larger amounts of sludge than would be recommended on productive soils. The sludge will usually raise the pH of acid soil material, improve its physical properties, and supply nutrients long enough to establish plant growth. If large amounts of sludge are applied, transport of metals to surface waters may present a hazard. Because of the wide range of soil materials and landscapes, recommendations for application of sludges on drastically disturbed lands must be considered on a site-by-site basis.

## Ground-Water Protection

Although long-term studies have not been completed, those working on the subject agree that soils readily remove heavy metals from the soil solution and prevent them from reaching the ground water. Contamination of the ground water with metals, therefore, is not likely to result from application of sewage sludge to soils.

Sludge application at rates supplying more nitrogen than the crop requires on very permeable soils or on soils with water tables or bedrock within a few feet of the soil surface can result in ground-water contamination with nitrate. When selecting sites for sludge application, the permeability and drainage of the soil and the depth of the water table and bedrock should be considered.

## Surface-Water Protection

Because heavy metals can be transported in surface runoff waters, good engineering and soil management practices to limit runoff and sediment transport are appropriate. Factors affecting runoff include land slope, distance from receiving waters, rate of sludge application, water content of the sludge, existing vegetation, soil permeability, and weather conditions. The steeper is the slope of land receiving sludge and the shorter is the distance to receiving waters, the greater is the potential for surface-water contamination. Use of conservation tillage practices and engineering designs available from the Soil Conservation Service for sloping land will help reduce transport of metals by erosion and runoff.

Fluid sludges are more prone to loss in runoff shortly after application than are dried sludges. If surface-applied to cultivated land, fluid sludges should be worked into the soil as soon as practicable after application. Transport in runoff is a distinct possibility where sludges are applied to frozen and snow-covered soils.

With fluid sludges low in available nitrogen, which would otherwise be applied in relatively large quantities on the basis of their nitrogen content, the amount of any single surface application should preferably be limited to surface layers no more than 0.8 to 1.2 cm (1/3 to 1/2 inch) in thickness to avoid undue runoff.

# Crop Selection

Crops differ in their ability to absorb heavy metals such as cadmium from the soil. They differ, moreover, in their tendency to exclude certain metals from particular organs. In many crops, the seeds contain lower concentrations of most heavy metals than do the vegetative tissues. Hence, the potential hazard from heavy metals is reduced if only the grain is harvested. The foliage of pastures sprayed or surface-treated with sludge may have surface coatings of sludge (Chaney et al., 1976a). Ingestion of sufficient quantities of such forage by grazing animals could result in animal-health problems due to heavy metals or other constituents. Thus, in general terms, grain crops present a lesser heavy-metal hazard to the food supply than do forages, pastures, and leafy vegetables.

Little or no hazard to the food supply from heavy metals applied in sewage sludge would be presented by crops that are processed to supply refined substances such as sugar and distilled alcohol. If absolutely no hazard is to be permitted from occurrence of heavy metals above the background level, sewage sludge could be applied only to nonagricultural land or to land used only for production of crops that are grown for their nonedible products.

## Monitoring

When sewage sludge is applied to soil, the heavy metals it contains may be retained by the soil or removed by erosion. Losses by transport in water moving downward through the soil are negligible. Uptake by crops is small. The major part of the metals added normally remains in the upper part of the soil with which the sludge is mixed. With repeated applications of sludge, therefore, the content of heavy metals in the soil gradually increases.

The strong retention of the heavy metals by soil is a consequence of their low solubility. In almost all instances of soils and heavy metals, the availability of the metal to plants is expected to decrease with time after application because of reactions the metal undergoes in the soil.

The capacity of soil to react with substances that are added to it is of great importance where the potentially toxic elements applied in sewage sludge are concerned. Two general situations may be discerned. With elements such as aluminum, which may be inactivated merely by pH control, the capacity of any soil for inactivating the element is infinite as long as the pH is maintained in the proper range by additions of limestone or otherwise. Aluminum may react with specific soil constituents as well, but such interactions are not essential to inactivation as long as proper pH control is practiced. With elements such as arsenic, which are inactivated by interaction with soil constituents such as hydrous iron oxides and not by pH control as such, the capacity of the soil to react decreases with each succeeding application of the element until, in the limit, no reaction capacity remains. At the limit, the soil will have accumulated considerable arsenic and will be highly toxic to plants. The amount of arsenic accumulated will depend on the nature of the soil.

Now under investigation is the extent to which the hazard of the metals to crops and to the food supply increases with cumulative additions of sludge under different conditions. Further research is needed to clarify the trends in availability of the various potentially toxic elements that are added to soils in sewage sludges and to provide some explanation for the trends and for the differences among soils.

If sludges are applied to cropland at rates sufficient to meet the nitrogen fertilizer requirement of the crop, the initial addition of heavy metals to the soil will be small. No measurable detrimental effect of the metals on the crop is expected, and the heavy metal content of the plants will be increased only a little above the background level normally present in the plants without sludge addition. In the view of most members of the task force, there is no hazard at this stage, and monitoring is unnecessary. However, a few members consider that some monitoring is needed even when low rates of sludge are applied.

If additions of sludge at these low rates were to be repeated indefinitely, however, all would agree that there would be some time at which monitoring of soil and plant tissue would be desirable to ensure that the accumulation is not producing toxic effects on the crop and that the increases in metal content of the crop are not sufficient to represent a hazard to the use of the crop as food for animals or humans. Exactly when this time might be cannot be foretold because too little is known at this stage. There is no question, however, that periodic monitoring would signal an undesirable build-up of heavy metals before irreparable damage results. The warning provided by timely monitoring would enable the operator to introduce corrective measures or, if necessary, to discontinue sludge application.

It is to be expected that, as research proceeds, the kind of guidance needed to develop effective but reasonable monitoring programs will gradually unfold. The prognosis at present is that little monitoring of soils and crops would be needed if sludges were monitored, heavy-metal levels were controlled, and good soil and crop management practices were followed.

If and when agricultural monitoring is necessary, plants are to be regarded as the most appropriate evaluator of the system. Analyses of the harvested portions of plants at intervals will, in most instances, indicate the rate of increase of availability of the metal to the plants and will signal the approach of harmful levels well in advance of permanent damage to either soil or crop. Interpretations of such analyses must recognize normal seasonal differences in plant composition and possible sampling errors. As further toxicological information is obtained, the plant analyses can be interpreted with increasing precision in terms of potential toxicity of the plants to animals and humans.

Plant analyses have three main limitations where monitoring of heavy metals and other toxic elements is concerned. First, although the analyses indicate the approach of a hazardous level for the crop analyzed, the level attained may already be toxic to another more sensitive crop. Second, both tops and roots must be analyzed to diagnose all toxicities. Third, plant analyses usually cannot be used to indicate the level of soil nitrogen that may lead to ground-water contamination.

Soil analyses can be used to monitor fields being treated with sludge. When properly correlated over a wide range of soils and crops, soil tests can indicate a potential metal hazard for any crop. Such extensive testing and correlation has not, as yet, been done. Regardless of the rate and frequency of sludge application, soils should always be routinely tested for pH and available phosphorus and potassium. Proper site management requires that recommended amounts of limestone and nutrients such as potassium be applied. Soil analysis can also be used as an index of potential ground-water pollution.

Because the properties of the soil in the rooting zone of crops vary with depth, there is always some question about where the samples should be collected and, if samples from different depths are collected and analyzed, how the results should be interpreted. For agricultural purposes, soil samples are usually collected from the plowed layer only. Use of the same convention for monitoring the effects of applications of sewage sludge would have the advantage that analyses of this layer should indicate the build-up of heavy metals well in advance of permanent damage.

## Analytical Problems

Soils, plants, and sewage sludges are extremely diverse and complex, and methods for sampling, sample preparation, and analysis have been inadequate in many instances. Sensitive, accurate, analytical methods have not been available for some of the metals until recently. Therefore, data from some of the earlier investigations are questionable. In some instances, high levels of certain elements can interfere in the determination of other elements, resulting in values which are either too high or too low if the problems are not taken into proper account. Difficulties of this kind often result in a lack of confidence in reported work. Use of a common set of standard samples by analysts in different laboratories can be a valuable aid to accuracy (Ellis et al., 1975).

### Sampling

Sludge materials are extremely variable in composition (Sommers, 1976). In addition, sludge from a given source varies in composition with time. A coefficient of variation as great as 50% may occur over time for some of the metals (Baker and Chesnin, 1975). This variability emphasizes the need for obtaining samples over a fairly long period of time when characterizing the composition of sludge from a given source.

The concentration of metals in plants varies with the plant part sampled and the stage of growth. The concentration of metals in sludge-treated soils may vary considerably from sample to sample due to variation in the sludge and poor mixing of the sludge with the soil. Obviously, great care must be taken in sampling plants and soils if the analyses are to be used to identify or predict heavy-metal problems. Sampling plants and soil for quantitative analysis is discussed in detail by Ellis et al. (1975).

### Sample Preservation

Preservation of soil and plant samples is not a major problem in most cases, but preservation of sludge samples is a major problem. Methods for preservation are outlined in a publication by the Environmental Protection Agency (1974).

### Analysis

Plant materials and sludges are normally digested and analyzed for total content of metals. Dry ashing ($<500^oC$) and wet digestion methods have both been used in preparing plant and sludge samples for analysis. Dry ashing is not satisfactory for mercury, selenium, iron, and copper.

For soils, a total analysis for the metals seldom correlates with uptake by plants or leachability of the element in soils. Better correlations with uptake of elements by plants in the deficiency range are obtained by use of appropriate equilibrium extraction solutions that remove a labile fraction of the element from the soil, but with most extractants the quantities extracted have not been correlated with uptake by plants in the toxic range. Further research is needed to establish more precisely the relationships in the toxic range.

Once metals are in solution, a competent analyst can obtain reliable results for many of the metals by any one of several analytical procedures. Atomic absorption is used in most laboratories, but some of the new techniques, such as neutron activation and plasma-source emission spectroscopy, offer good precision and provide lower limits for detection for some of the metals.

Analyses of sludge, plant material, and soil extracts by atomic absorption and flame emission methods are subject to problems because of high salt content and interference by similar ionic species in the matrix. The deuterium background corrector seems to be the best method for correcting "background" problems. Background correction is very critical for the accurate determination of cadmium.

## HAZARD OF HEAVY METALS AND OTHER ELEMENTS TO PLANTS AND ANIMALS

When the chemistry of the heavy metals and other potentially toxic elements in sewage sludges is considered in conjunction with their uptake by plants, it is possible to subdivide the elements into two categories depending on whether or not they represent a potentially serious hazard to plants or animals. In allocating the various elements to the low-hazard category, certain management constraints were taken for granted. These are mentioned in the relevant sections to follow. It should be clearly understood that if poor management is practiced at a sludge-treated site, some of the elements rated here as relatively innocuous could constitute a hazard.

### Elements Posing Relatively Little Hazard

#### Manganese, Iron, and Aluminum

Manganese, iron, and aluminum form sparingly soluble oxides and hydroxides in soils. Those of iron are the least soluble. Most soils contain large quantities of iron and aluminum, and so the addition of sludge containing high amounts of iron or aluminum will not appreciably increase the concentration of these elements in the soil. Under good soil management practices (i.e., soil pH greater than 5.5), little of either element remains in solution.

Aluminum and iron added to soil in sludge should rapidly precipitate as the hydroxides. Later the ferric hydroxide may be expected to revert to less soluble oxide-hydroxide mixtures. A small quantity of each element may remain in the soil as iron or aluminum organic complexes.

Under well-aerated soil conditions, added manganese should rapidly revert to one of the insoluble tetravalent manganese oxides. Manganese may also form stable organic complexes.

The major soil property to affect the solubility (and availability to plants) of aluminum, iron, and manganese is soil pH. But both manganese and iron may be rendered more soluble by reducing conditions in soils. Good management would include applying sludge only to well-aerated soils because this would prevent solubilization of manganese.

At soil pH values above 5.5, uptake of manganese, aluminum, and iron is restricted, and plants will not accumulate these elements to toxic levels. But below pH 5.0, aluminum toxicity is common, and excessive manganese may accumulate in plants if the soil contains large quantities of manganese. Although iron does not accumulate in plant tissue to a great extent, it has been reported that relatively high availability of iron in the soil will induce manganese deficiency.

The levels of iron, manganese, and aluminum in sludge usually will not be of any environmental concern. Even though many tertiary sludges may be high in iron, aluminum, or manganese, these elements will not be a limiting factor in determining the quantity of sludge that may be applied to agronomic crops if the soil pH is maintained above 5.5 and the soil receiving the sludge is well aerated.

Chromium

Little soluble chromium is found in soils. If soluble trivalent chromium is added to soils, it rapidly disappears from solution and is transformed into a form that is not extracted by ammonium acetate or complexing agents. However, it is largely extractable by strong acids, indicating the formation of insoluble hydroxides or oxides. Hexavalent chromium remains as such in a soluble form in soil for a short time but is eventually reduced to trivalent chromium and then changed to forms of low solubility. Hexavalent chromium is toxic to plants, but sludges contain little, if any, hexavalent chromium because it is reduced to the trivalent state during the sewage digestion process.

There have been few studies of the chemistry of chromium added in sludge to soils. The concentration of chromium in sludges ranges from very low values to well over 20,000 ppm. Decomposition of sludge in soils is likely to progress at a sufficiently slow rate so that the released chromium will change to insoluble compounds without build-up of appreciable levels of soluble chromium in the soil.

Most crops absorb relatively little chromium, but some species can contain levels up to 10 ppm. Schueneman (1974) found chromium concentrations less than 0.5 ppm in leaf tissue of corn grown on sandy soils treated with inorganic chromium up to the rate at which growth was reduced (200 ppm on a dry-soil basis). However, a few crops can take up appreciably higher levels of chromium. Field bean tissue reached a chromium concentration of 30 ppm when chromium was applied to the soil at 200 ppm. This addition produced a 25% reduction in yield. Chromium accumulated in tomato tissue to a concentration of 35 ppm, but yield reductions were associated with tissue concentrations as low as 5 ppm.

Clapp et al. (1976) found a chromium concentration of 4 ppm in corn tissue with no increase due to application of 135, 270, or 530 kg of chromium in sludge per hectare (Appendix Table 4). The concentration of chromium in the grain was less than 0.1 ppm, indicating little translocation of chromium from the leaf tissue into the grain. Other researchers (Appendix Table 5) found low concentrations of chromium in leaf and stover tissue of corn grown on soil treated with sludge supplying 833 kg of chromium per hectare.

Although it is generally considered that chromium is not essential for plant growth, there have been several reports of slight yield increases due to chromium additions. These increases may be associated with release of other ions (for example, manganese) that plants require as nutrients.

Chromium is not expected to be a limiting factor in determining the quantity of sludge that may be applied to soil producing agronomic crops because (i) plants can tolerate relatively high levels of chromium applied in sludge, (ii) plants do not accumulate chromium even when it is present in the soil at high levels, and (iii) there is evidence that chromium is required by humans and animals and that diets are deficient in chromium in certain areas (Underwood, 1971).

Arsenic

Inorganic arsenicals have been used as agricultural pesticides and defoliants for many years, and they have seriously polluted soils in certain areas. Since the banning of inorganic arsenicals in 1967, the application of arsenic to soil has decreased very markedly.

Sludge is known to contain arsenic in concentrations ranging from 10 to 1,000 ppm. The form of combination of arsenic in sludge is unknown. Research studies with inorganic sources of arsenic have shown that rates in excess of 90 kg of arsenic per hectare must be applied before toxicity to plants is observed, even on sandy soils (Jacobs et al., 1970). To judge from these results, approximately 90 metric tons of a high-arsenic sludge would have to be applied to cause toxicity to plants.

Once incorporated into the soil, arsenic reverts to the chemical form of arsenate, which is strongly held by the clay fraction of most soils. As a consequence, arsenic has a low availability to plants on soils which have an appreciable clay content. Because of the low clay content of sandy soils, arsenic toxicity to plants could develop if high-arsenic sludge were applied at high rates.

Although plants take up arsenic, they tend to accumulate it in the roots, and the arsenic content of most of the edible portions of plants is well below the critical concentration of 2.6 ppm (U.S. Department of Agriculture, 1968) considered safe for animal or human consumption. Hence, arsenic is not readily passed on to animals and humans. Since sewage sludge generally contains low levels of arsenic, application of sludge usually will not be limited by its arsenic content.

Selenium

Little evidence exists to suggest that selenium is an essential element for plants, but it is definitely required by certain animals. Despite its seeming nonessentiality, selenium is taken up by plants, which serve as carriers of selenium from soil to animal. The range between deficiency and toxicity in animals is fairly narrow. At levels of 0.05 ppm in the diet, degeneration of muscle tissue results; when the diet contains more than 4 ppm, selenium toxicity may occur.

Furr et al. (1976b) found the selenium concentration in sludge from 16 U.S. cities to range from 1.7 to 8.7 ppm. Bates et al. (1976) analyzed nine Ontario sludges and found a selenium concentration of 0.86 ppm in one, 0.73 ppm in another, and less than 0.05 ppm in the remaining seven. Data from greenhouse trials showed that application of up to 8.8 kg of selenium in sludge per hectare to a sandy loam soil did not increase the uptake of selenium by ryegrass. Furr et al. (1976a), on the other hand, found that selenium uptake by a variety of plants was greater from sludge-treated than from control soils but that, in all instances, the selenium content of the tissue was well below hazardous levels.

Selenium in soil is least soluble under acid conditions, which would be in conflict with the general requirement that the soil be near neutral in reaction for appliction of sludge. Under neutral to alkaline conditions, selenium occurs as the selenate ion, which forms neither highly insoluble salts nor stable sorption complexes with the clay fraction of soil. It is under such conditions that selenium poses the greatest hazard to animals. Selenium toxicity to livestock is known to occur under certain conditions in dry regions of the United States where the soils have developed on seleniferous parent materials. Before the potential hazard of selenium applied in sewage sludges can be properly evaluated, more needs to be known about the selenium content of sludges and the rate of loss of selenium from soils in humid regions.

Antimony

Antimony is not known to be essential to the growth of plants but has been reported to be moderately toxic. Significant amounts of antimony can be taken up by plants from contaminated soil, and plant leaves tend to contain more antimony than do stems. Soil investigations have shown that antimony is sorbed very strongly by kaolinite and sesquioxides under acid conditions, but under neutral to alkaline conditions it appears to move readily. Antimony generally occurs in sewage sludge in very low concentrations; one value of 900 ppm has been reported. Although antimony could be a potential hazard to plants and animals if applied in large amounts, no evidence of hazard is currently available.

Lead

Lead is a nonessential element that exhibits a low degree of potential toxicity to plants and a high degree of potential toxicity to animals. Sewage sludge contains lead in significant amounts. Sommers (1976) gives the median concentrations as 540 ppm for anaerobically-digested sludges and 290 ppm for aerobically digested sludges. Additions of 20 metric tons of these sludges per hectare would supply 10.8 and 5.8 kg of lead per hectare or approximately 4.4 and 2.6 ppm in the soil.

Soluble lead added to soil reacts with clays, phosphates, carbonates, hydroxides, sesquioxides, and organic matter, and these reactions greatly reduce the solubility. Jurinak and Santillan-Medrano (1974) concluded that lead is retained as the hydroxide or hydroxy-phosphate in acid soils and as the carbonate in calcareous soils. Hassett (1974) found that the lead sorption capacity of Illinois soils could reach several thousand kg per hectare and was related to the cation-exchange capacity, pH, and extractable phosphorus content of the soil.

Plants take up lead in the ionic form from soils. The amount of lead taken up from soil decreases as the pH, cation-exchange capacity, and available phosphorus of the soil increase. Miller et al. (1975a,b) observed an inverse relationship between the lead uptake by corn and soybeans and the lead sorption capacity of soils. Lead taken up by five plant species from lead-contaminated soil was reduced by liming (Cox and Rains, 1972).

Studies on the effect of lead on plant growth are limited; however, the effect appears to be minor. Baumhardt and Welch (1972) applied up to 3,200 kg of lead per hectare to a soil having a pH of 5.9 with no effects on corn growth or grain yield. In general, the lead content of roots is higher than that of plant tops, with fruits and seeds showing the lowest content. It appears that the lead content of soils would have to approach 1 percent and that the soil pH would have to be below 5.5 before significant effects on plant growth would be encountered.

Investigations by Sabey and Hart (1975), Haze et al. (1975), and Clapp et al. (1976) (Appendix Tables 4 and 6) indicate that, when sewage sludge is incorporated in the soil, the lead content of the above-ground portion of the plants or seeds is not significantly changed. If the sludge is applied as a surface dressing when a crop is growing (Boswell, 1975; Chaney et al., 1976a), there may be an increase in the lead content.

Most soils treated with sewage sludge have a pH above 5.5 and a high labile phosphorus content. Under these conditions, movement of lead from treated soil into plant tops and seeds is not found. Accumulation of lead in crops thus would seldom be a cause for limiting the application of sludge to cropland. One may ensure that lead does not become a problem, however, by maintaining the pH of sludge-treated soils above 5.5, by avoiding the growth of root crops, and by avoiding repeated application of sludges with very high lead content.

Mercury

The mercury contents of soils not receiving additions of sewage sludge lie in the range from 0.01 to 0.5 ppm. In soils receiving sludge for protracted periods, the concentration could approach 1.0 ppm. Once in the soil, mercury enters into reactions with the exchange complex of the clay and organic fractions, forming both ionic and convalent bonds.

Many mercurial compounds, both organic and inorganic, decompose to yield elemental mercury, which may volatilize or be converted to HgS, $HgCl_3^-$, or $HgCl_4^{2-}$. The last of these forms may be adsorbed by sesquioxide surfaces or may move freely in soils (Newton et al., 1976). Organic mercurial compounds can be sorbed on clay minerals by molecular forces, and there is evidence that mercury can be chelated by organic matter. Chemical and microbiological degradation of mercurials can take place side by side in the soil, and the products -- ionic or molecular -- are retained by organic matter and clay or may be volatilized if gaseous. Because of the high affinity between mercury and the solid soil surfaces, mercury persists in the upper layer of soil and is not a threat to ground water.

Mercury can enter plants through the roots, it can be translocated, and it has been reported to cause injury to plants (Smart, 1964; Stewart and Ross, 1967). Once absorbed, it appears to be readily translocated throughout the plant. In many plants, mercury concentrations range from 0.01 to 0.20 ppm, but when plants are supplied with high levels of mercury, these concentrations can exceed 0.50 ppm. Mercury ingested in food undergoes "biological magnification" in animals, indicating a lower rate of excretion than of intake.

In sludges the mercury contents may be high if industrial sources of mercury contamination are present. Little is known about the form in which mercury occurs in sludge. Mercury may undergo biological methylation in sediments (Jernelov, 1972), but no methylation has been observed in soils, mud, or sewage sludge (Rissanen et al., 1970).

Most sludges are relatively low in mercury, and very little increase in concentration of mercury in plants has resulted from application of sludge. However, Van Loon (1974) found mercury concentrations up to 12.2 ppm in tomato fruit after application of a high-mercury sludge to an alkaline soil. Nevertheless, sludge application would seldom be limited because of concern over mercury.

## Elements Posing a Potentially Serious Hazard

### Cadmium

During the past decade, there has been increased concern over cadmium in the environment because cadmium has been linked to certain health problems. Because of the cumulative characteristics of cadmium in animals and humans from low-level exposure, there is much greater concern about the possible hazard to humans from elevated concentrations of cadmium in plants than there is for possible toxicity to the plants.

Sewage sludge may contain cadmium concentrations from 3 to over 3,000 ppm, with a mean value of 106 ppm and a median value of 16 ppm (Sommers, 1976), whereas soil contains from 0.01 to 7 ppm with 0.06 being common (Allaway, 1968). Therefore, the addition of sewage sludge to soil usually results in an increase in concentration of total cadmium in the soil. To maintain perspective, it is important to note that superphosphate can also add cadmium to soil. For example, Lee and Keeney (1975) have shown that cadmium added to land in Wisconsin as a contaminant in fertilizer is equivalent to that produced in sludges from all Wisconsin treatment plants. However, because of a wide difference in the rate of application, cadmium addition to a given tract of land will be much less from fertilizer than from sludge. In either case the amount of cadmium added is low (about 2000 kg for Wisconsin).

The chemistry of cadmium in soil is not well understood, but cadmium appears to be influenced by soil organic matter, clay content and type, hydrous oxide content, soil pH, and redox potential. The assumption has been that the total amount of cadmium added to the soil would ultimately control the amount of soluble cadmium present and thus the uptake of cadmium by plants. Soils must have some "saturation" limit for cadmium at which the addition of a quantity of soluble cadmium results in a nearly equivalent increase in soluble cadmium in the soil. However, data in Appendix Table 7 on repeated annual applications of sludge to soil cropped to corn show that the amounts applied in a given year influenced the cadmium content in the leaves to a greater extent than did the total cumulative amounts of cadmium applied. The implication of these results is that, at the rates used, most of the applied cadmium was being converted to forms of relatively low availability to plants.

The observations just made point to the importance of an understanding of the chemistry of cadmium in soil-water systems. Soil properties and their importance to the removal of cadmium from the soil solution need further study and quantification.

In investigations in which cadmium addition was discontinued, there was a decrease in cadmium uptake by corn in the first year with a smaller decrease in the second year (Hinesly et al., 1976b). The number of years after termination of sludge application required for plant uptake to approach background levels has not been established.

Field plots have been established on several farms in the northeastern United States on which sewage sludge containing cadmium concentrations of 5 to 800 ppm had been applied over a period of 7 to 25 years. The period after cessation of sludge application ranged from 1 to 12 years. Although the total amount of sludge or cadmium applied could not be definitely established, the cadmium content of crops grown on these plots reflected the influence of pH on the availability of the cadmium to plants as well as the importance of agronomic practices, including crop selection. Many farms were managed without the benefit of pH control during sludge applications. Subsequent additions of lime to these acid areas showed that cadmium was thereby rendered less available to crops (Appendix Table 8). This decrease in cadmium uptake with increasing pH was noted for both low and high cadmium application levels. In one set of observations (Appendix Table 9), the effect of application of cadmium-bearing sludge on the cadmium content of plants was evident 5 years after application of sludge had ceased.

Annual cadmium application rates, soil pH, and crop species and varieties have a major influence on the cadmium concentration in plant tissue. To a lesser extent, soil temperature, nitrogen and phosphorus fertilization, and addition of metals such as zinc and copper may also influence the cadmium content of crop tissues.

In experiments in which soil pH was varied (Anderson and Nilsson, 1974; Chaney et al., 1976b; Giordano and Mays, 1976b), an increase in soil pH usually markedly reduced the cadmium content of crop tissues and grains (Appendix Tables 8 and 9). However, raising the pH of sludge-treated soils probably will not reduce the content of cadmium in plants to the same level as that on untreated soils. Moreover, differences in cadmium uptake by plants from different soils at a given pH may conceivably exceed the differences in uptake from a given soil at different pH values.

Research studies (Bingham et al., 1975; Giordano and Mays, 1976a; Dowdy and Larson, 1975) have shown that different plant species, varieties, and plant tissues contain different cadmium concentrations from similar rates of application (Appendix Tables 4, 6, 8, 12, 13). Cadmium concentrations in corn grain are usually only 3 to 15% of those in the leaf, whereas in the grain of soybeans, wheat, oats, and sorghum, cadmium reaches 30 to 100% of the foliar levels.

Interactions of other metals with cadmium have been observed in soybeans and corn. Application of copper increased the cadmium content of corn and rye shoots (Cunningham et al., 1975a). As shown in Appendix Table 21, application of zinc or zinc plus copper plus selenium plus molybdenum reduced the cadmium content of soybean grain (Chaney et al., 1976b; Baker et al., 1975) even though foliar cadmium content increased (Haghiri, 1974). On phosphorus-deficient soils, phosphorus application appears to decrease the cadmium content of corn leaves. Ammonium fertilizer applications may increase cadmium uptake.

Bingham et al. (1975, 1976) found that cadmium concentrations in the dry matter of edible tissues of various crops ranged from 1.7 to 80 ppm at cadmium additions to soils which caused 25% yield reduction (Appendix Table 12). It appears that crops may contain undesirable concentrations of cadmium in their tissues without showing visible symptoms of toxicity of cadmium to the plants. Leafy vegetables such as lettuce, chard, spinach, and turnip greens may contain cadmium concentrations exceeding 100 ppm without showing any toxicity symptoms.

Crops grown on different soils were found by John et al. (1972) and Miller et al. (1976) to differ widely in cadmium uptake from equal cadmium additions as inorganic cadmium salts. Cadmium uptake was related to the ratio of cadmium added to the cadmium sorbing capacity of the soil. Where cadmium salts were added in sludge, some work showed no influence of the clay content of the soil on cadmium absorption by plants (Kelling et al., 1976), and some showed a significant influence of soil clay content (Singh et al., 1976).

An increase in soil temperature leads to an increase in uptake of cadmium by crops whether the added cadmium is inorganic (Haghiri, 1974) or sludge-borne (Shaeffer et al., 1975; Giordano and Mays, 1976a). Sludge processing may affect subsequent availability of the cadmium to the crop. For example, application of high-lime sludge cake may cause soils to become alkaline, thus reducing cadmium availability, and anaerobic digestion may be necessary to produce the lack of additivity of cadmium uptake by plants grown on soils that have received repeated applications of cadmium in sludge. Clapp et al. (1976), however, have not observed this phenomenon with an aerobically stabilized sludge. As shown in Appendix Table 10, composting of digested or raw sludge appears to reduce the crop uptake of cadmium in field studies during the first and subsequent years (Chaney et al., 1975).

Field experiments involving use of anaerobically digested sewage sludge (single application) for corn production have led to conflicting results. Kelling et al. (1976) found that the application of 4.3 kg of cadmium per hectare did not change the cadmium concentration in corn grain, Decker et al. (1976) found that application of 2.0 kg of cadmium per hectare increased the cadmium concentration five-fold, and Baker et al. (1975) found that application of 24.8 kg of cadmium per hectare increased the concentration eight-fold (Appendix Table 11). The same treatments, however, gave a twenty-fold increase in cadmium concentration in sorghum grain. Findings with sweet corn (Giordano et al., 1975; Singh et al., 1976) also show conflicting results (Appendix Table 11). The inconsistency in results obtained by the various investigators cannot be explained on the basis of existing knowledge of the systems involved.

In summary, several management options are available to keep the concentration of cadmium in food and feed crops at a low level on sludge-treated land: (1) Maintain the soil pH at or above 6.5. (2) Grow crops which accumulate relatively low concentrations of cadmium or, for crops harvested for grain, those which have a low concentration of cadmium in the grain. (3) Make only small annual applications of cadmium when food or feed crops are to be grown. (4) Use sludges low in cadmium on cropland. (5) Grow nonedible crops.

Inadequate information is available to set forth either specific soil properties other than pH or specific sludge processing methods which may lead to lower availability of cadmium to crops. Because cadmium is the heavy metal which is likely to have the greatest impact on the food produced on land treated with sewage sludge, the potential for limiting the entry of cadmium into the sewage system and for removing cadmium from sludge prior to application is worthy of investigation.

A specific metal-binding protein called metallothionein, which has been characterized in several animal species and microorga isms, is responsible for the accumulation of cadmium in animals. The content of this protein increases following continued exposure to cadmium.

Williams et al. (1976) found that the liver, kidney, and muscle of meadow vole contained lower concentrations of cadmium than were present in the diet supplied by corn or sorghum grown with or without application of sewage sludge as a source of cadmium (Appendix Table 16). Concentrations of cadmium in the liver and kidney considerably exceeded those in the muscle. The maximum concentration of cadmium in the diet in this work was 2.76 ppm.

In research by Baker et al. (1975) on chickens, shown in Appendix Table 15, the cadmium content of eggs and muscle was lower than that in the diet at all levels of cadmium feeding, but the cadmium content of liver and kidney exceeded that in the diet. In this work, the lowest concentration of cadmium fed was 3 ppm and the highest was 48 ppm.

Doyle et al. (1974) found that, with lambs, the muscle and the fat had much lower concentrations of cadmium than did the diet, and the liver and kidney had much higher concentrations than did the diet (Appendix Table 17). Dietary concentrations of cadmium supplied in this work ranged from 5 to 60 ppm. Up to 5.3% of the cadmium supplied in the diet was absorbed by the lambs.

In all the investigations mentioned, the cadmium content of all tissues analyzed increased with the cadmium content of the diet. The poorest correlation observed was between cadmium in the diet and in the wool of the lambs fed by Doyle et al. (1974).

Preventive effects of zinc in cadmium toxicity suggest that a simultaneous increase of zinc intake may prevent cadmium absorption or accumulation (Parizek, 1957). Interactions with other elements are not well known.

As the foregoing discussion has indicated, there are many gaps in our knowledge of the behavior of cadmium in soils, plants, animals, and humans, and much more research is needed to clarify the relationships which control the fate of cadmium in the environment. Under some circumstances, cadmium could represent a very real hazard in the food supply, and so caution should be exercised whenever cadmium is applied to land used for producing food crops. The hazard is greatest when sludges high in cadmium are applied at high rates to limited tracts of land and where the produce from such land constitutes a considerable portion of the diet.

## Copper

Copper is found in all soils, and its concentration is usually from 10 to 80 ppm. In soils, copper occurs in association with hydrous oxides of manganese and iron and also as soluble and insoluble complexes with organic matter. Keeney and Walsh (1975) found that the extractable copper content of sludge-treated soil decreased with time, which suggests that a reversion of the copper to less soluble forms was occurring.

Copper is essential to the growth of plants, and the normal range of concentrations in plant tissue is from 5 to 20 ppm. Copper concentrations in plants normally do not build up to high levels when toxicity occurs. For example, Walsh et al. (1972) found that the concentrations of copper in snapbean leaves and pods were less then 50 and 20 ppm, respectively, under conditions of severe copper toxicity. Even under conditions of copper toxicity, most of the excess copper accumulates in the roots; very little is translocated to the aerial portion of the plant.

Copper toxicity may develop in plants from application of sewage sludge if the concentration of copper in the sludge is relatively high. Copper toxicity to plants from sludge applications was reported by Cunningham et al. (1975a), who suggested that copper is about twice as toxic as zinc. Corn and rye took up more copper from soil treated with an inorganic salt of copper than from soil treated with sewage sludge supplying an equal amount of copper (Cunningham et al., 1975c).

Sheep are most susceptible to copper toxicity, followed by cattle, swine, and poultry, in that order. Swine, sheep, and cattle are capable of accumulating high concentrations of copper in the liver under some conditions. Accumulation of copper in animal livers with resultant copper toxicity may occur with rations having normal levels of copper if the molybdenum intake is extremely low. This toxicity can be prevented by controlling the molybdenum content of the ration. Relatively high concentrations of copper can be tolerated in ruminants when an adequate amount of molybdenum is available. Copper poisoning of animals associated with consumption of certain plants such as Heliotropium species is considered to be due to liver damage caused by hepatotoxic agents of the plant in which the injured animal hepatic cells also accumulate copper.

High levels of copper in the diet are beneficial for chickens and pigs and have been widely used in feeding these animals. Modern research with pigs followed from the observation that pigs have a craving for copper. The optimum level for pigs seems to be a copper concentration of 250 ppm in the diet. The dietary copper is supplemented with extra zinc and iron. Pigs fed the high-copper rations gain weight more rapidly and use less feed per pound of gain than do control pigs. The physiological effect of the copper overlaps that of antibiotics and often seems to be a substitute for antibiotics. Although feeding copper at a concentration of 250 ppm increases the concentration of copper in the liver of the pigs, the accumulations do not seem harmful if the copper is properly supplemented with zinc and iron. Literature on this subject was reviewed by Braude (1965) and Wallace (1967). Another review by Braude is in publication. The principal issue surrounding the feeding of high levels of copper has not been the possible detrimental effects on the animals but the possibility that toxicity of copper to plants may develop over a period of years if the animal wastes are applied in large quantities to limited areas of land.

## Molybdenum

Molybdenum is required in very small amounts by plants. It does not appear to be very toxic to plants, even at levels of a few hundred ppm in plant tissues.

Sorption of molybdenum by acid soils depends on the iron oxide and phosphorus status of the soil, iron oxide having a great affinity for molybdenum. If phosphorus is present in large quantities, some molybdenum bonded to iron oxide will be replaced. Maximum molybdenum sorption in soils occurs at pH values near 4.2. The availability of molybdenum increases as the soil pH increases, a behavior the reverse of that observed with copper, nickel, and zinc. The practice of keeping soils at pH values near neutrality to limit the availability to plants of heavy metals applied in sludge thus is ineffective in limiting molybdenum availability.

The molybdenum content of sewage sludges has been found to vary from 5 to 39 ppm with a mean of 28 ppm (Sommers, 1976). If sludge with a molybdenum concentration of 39 ppm were applied at the rate of 60 metric tons per hectare, the molybdenum application would be about 2.3 kg of molybdenum per hectare, an addition that would not pose a threat to the health of grazing animals. However, repeated applications of high-molybdenum sludge over a long period of time might cause animal health problems, especially on soils of high pH which are not subject to leaching (Hornick et al., 1976). It is doubtful that molybdenum in sludge would present a serious hazard to the health of grazing animals except where forages from sites treated with sludge high in molybdenum form the major part of the animal diet.

The tolerance of animals to molybdenum varies with species and age, and is dependent upon the copper status and copper intake of the animal, the inorganic sulfate and organic sulfur oxidizable to sulfate in the diet, and the intake of other metals such as zinc and iron. Cattle are considered the most susceptible of the farm animals to molybdenum toxicity. Sheep are less susceptible, and horses and pigs are least susceptible. Forages containing molybdenum concentrations exceeding 10 to 20 ppm may produce molybdenosis in ruminants. The symptoms of molybdenosis are essentially those of copper deficiency. Cattle provide the most overt clinical signs. Excessive molybdenum in the diet causes copper deficiency accompanied by phosphorus deficiency. The condition is correctable by supplementing the diet with copper sulfate and phosphorus.

Nickel

Nickel is found in nearly all soils, plants, and waters. The content of total nickel in soils is commonly in the range from 10 to 100 ppm, with higher values in soils derived from serpentine. The concentration of extractable nickel in soils seems to be governed by the surfaces of iron and manganese hydrous oxides, which act as a "sink" for nickel, as well as by organic chelates, which complex nickel less strongly than copper.

Sludges vary greatly in their nickel content -- from 2 to over 3,500 ppm -- with a mean of 300 to 400 ppm (Sommers, 1976). Considerable quantities of nickel can be added to soil in sewage sludges, but whether the nickel becomes toxic to plants depends on several soil factors, the amount of nickel applied, and the contents of other metals in the sludge. Unlike copper and zinc, which are more available to plants from inorganic sources than from sludge, nickel uptake by plants seems to be promoted by the presence of the sludge organic matter (Cunningham et al., 1975c).

Nickel has no known essential function in plants. It is toxic to plants at concentrations above 50 ppm in plant tissues. No significant plant toxicity due to nickel was observed by Cunningham et al. (1975b), however, when nickel in quantities of 81 ppm in sludge and 100 ppm as nickel sulfate was added to sandy loam and silt loam soils, respectively. Thus far, toxicity of nickel to plants has been observed only on acid soils.

The daily ingestion of nickel by grazing sheep is approximately 2 mg per day. Nickel ingested as the sulfate or acetate has a very low toxicity to rats, mice, monkeys, and chickens (nickel may be required by chickens). Nickel salts in quantities to supply nickel at 700 ppm in the diet of chicks suppressed growth and nitrogen retention. This effect was not observed at lower levels. Soluble nickel salts in quantities to supply nickel at 1,000 ppm in the diet did not affect the growth rate or reproduction in rats.

Soil treatments such as liming that reduce the solubility of nickel reduce the toxicity. Thus far, toxicity of nickel to plants has been observed only on acid soils. If sludge-treated sites are well managed, nickel is not likely to be taken up by plants in quantities sufficient to cause toxicity to the plants or to pose a threat to the food supply.

Zinc

Zinc is essential for both plants and animals, being an essential component of a number of enzyme systems. Under acid conditions, zinc occurs in solution as the $Zn^{2+}$ ion. The most important mechanisms for zinc retention in soils are sorption on clay and hydrous iron oxide surfaces and chelation by organic matter.

Zinc is taken up by plants as $Zn^{2+}$ and, in excessive quantities, can be toxic. However, few records of toxic effects of zinc are available in the literature. Heavy dressings of zinc (up to 1,390 kg of zinc per hectare) had little or no effect on the growth of a number of crops (Murphy and Walsh, 1972; Walsh et al., 1972). When zinc toxicity does occur, the tissue of most crops will contain zinc at concentrations of several hundred ppm.

Giordano and Mays (1976a) found, in studies with string beans and sweet corn, that zinc in zinc sulfate initially was more available to plants than zinc in sludge. However, in residual studies, more zinc was taken up from sludge-treated plots than zinc-sulfate-treated plots. The pH was the same, and the difference in uptake, therefore, was not due to a pH effect. According to research by Giordano and Mays (1976a) and Touchton et al. (1976), the availability of zinc is reduced more by liming than is that of cadmium, copper, and nickel.

Touchton et al. (1976) found a marked decline of heavy metal content of grass with time. Three years after the last addition of sludge to Coastal bermudagrass, copper was at normal levels, and zinc levels were decreased by almost half. In contrast, where zinc sulfate was applied, Giordano et al. (1975) found that the reduction in yield of snapbeans and the increase in zinc content of the plants grown on Sango silt loam of pH 4.9 were almost as marked in the second year following application as in the first. There was little evidence of reversion of the zinc to forms not extractable by 0.5N hydrochloric acid. Walsh et al. (1972) similarly did not find reversion of zinc added as zinc sulfate at high rates. In this work, however, the toxicity to plants was not studied. To what extent and under which conditions reversion of zinc to forms of low availability in soils can decrease the long-term potential for uptake by plants and toxicity to plants remains uncertain. If the soil pH drops, the availability of the zinc will increase.

A wide margin of safety exists between normal dietary intakes of zinc and the higher intakes that may produce toxicity in birds and mammals. Pigs show no ill effects when they receive either zinc sulfate or zinc carbonate in quantities to supply a zinc concentration of 1,000 ppm in the diet. Sheep and cattle are less tolerant than other species. Dietary zinc levels of 500 ppm do not cause signs of toxicity or other detrimental effects, but levels of 900 ppm cause reduced gains and lowered feed efficiency. Excessive zinc intake decreases the copper content of the liver in sheep and cattle.

In general, if the pH value of sludge-treated soils is maintained at the recommended level, zinc should not be a serious hazard to plants or to the food supply unless exceptionally high levels are added in the sludge. In many instances, a moderate increase in the zinc content of the food supply should be beneficial because there is evidence that diets are often deficient in zinc.

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Appendix Table 1. Estimated production of sewage sludge in the continental U.S.A. in 1970 and annual cropland requirements for utilization of the sludge in agriculture[1/]

| State | Population, millions | Sludge produced,[2/] thousands of short tons | Cropland, thousands of acres | Cropland required to accept the sludge having the indicated content of nitrogen[3/] — 1% Available nitrogen: Acres | 1% Available nitrogen: Percent of total cropland | 4% Available nitrogen: Acres | 4% Available nitrogen: Percent of total cropland |
|---|---|---|---|---|---|---|---|
| AL | 3.45 | 67 | 3550 | 13 | 0.38 | 54 | 1.51 |
| AR | 1.79 | 35 | 1210 | 7 | 0.58 | 28 | 2.30 |
| AK | 1.93 | 38 | 8010 | 8 | 0.09 | 30 | 0.38 |
| CA | 19.99 | 389 | 9700 | 78 | 1.21 | 311 | 4.85 |
| CO | 2.22 | 43 | 5700 | 9 | 0.15 | 35 | 0.61 |
| CN | 3.04 | 59 | 150 | 12 | 7.89 | 47 | 31.58 |
| DE | 0.55 | 11 | 500 | 2 | 0.43 | 9 | 1.71 |
| FL | 6.85 | 133 | 1380 | 27 | 1.93 | 107 | 7.73 |
| GA | 4.60 | 90 | 4870 | 18 | 0.37 | 72 | 1.47 |
| ID | 0.72 | 14 | 4290 | 3 | 0.07 | 11 | 0.26 |
| IL | 11.14 | 217 | 22820 | 43 | 0.19 | 174 | 0.76 |
| IN | 5.21 | 101 | 12260 | 20 | 0.17 | 81 | 0.66 |
| IA | 2.83 | 55 | 24160 | 11 | 0.05 | 44 | 0.18 |
| KS | 2.25 | 44 | 21660 | 9 | 0.04 | 35 | 0.16 |
| KY | 3.22 | 63 | 4700 | 13 | 0.27 | 50 | 1.07 |
| LA | 3.64 | 71 | 3660 | 14 | 0.39 | 57 | 1.55 |
| ME | 1.00 | 19 | 420 | 4 | 0.93 | 15 | 3.71 |
| MD | 3.94 | 77 | 1530 | 15 | 1.00 | 61 | 4.01 |
| MA | 5.70 | 111 | 150 | 22 | 14.80 | 89 | 59.21 |
| MI | 8.90 | 173 | 6270 | 35 | 0.55 | 139 | 2.21 |
| MN | 3.82 | 74 | 20030 | 15 | 0.07 | 60 | 0.30 |
| MS | 2.22 | 43 | 5560 | 9 | 0.16 | 35 | 0.62 |
| MO | 4.69 | 91 | 13240 | 18 | 0.14 | 73 | 0.55 |
| MT | 0.70 | 14 | 9090 | 3 | 0.03 | 11 | 0.12 |
| NB | 1.49 | 29 | 17710 | 6 | 0.03 | 23 | 0.13 |
| NV | 0.49 | 10 | 500 | 2 | 0.38 | 8 | 1.53 |
| NH | 0.74 | 14 | 110 | 3 | 2.62 | 12 | 10.48 |
| NJ | 7.20 | 140 | 420 | 28 | 6.68 | 112 | 26.71 |
| NM | 1.02 | 20 | 1240 | 4 | 0.32 | 16 | 1.28 |
| NY | 18.26 | 356 | 4080 | 71 | 1.74 | 285 | 6.97 |
| NC | 5.09 | 99 | 4750 | 20 | 0.42 | 79 | 1.67 |
| ND | 0.62 | 12 | 18960 | 2 | 0.01 | 10 | 0.05 |
| OH | 10.69 | 208 | 10730 | 42 | 0.39 | 167 | 1.55 |
| OK | 2.57 | 50 | 10380 | 10 | 0.10 | 40 | 0.39 |
| OR | 2.10 | 41 | 2640 | 8 | 0.31 | 33 | 1.24 |
| PA | 11.88 | 231 | 4480 | 46 | 1.03 | 185 | 4.13 |
| RI | 0.95 | 19 | 20 | 4 | 18.50 | 15 | 74.01 |
| SC | 2.60 | 51 | 2740 | 10 | 0.37 | 41 | 1.48 |
| SD | 0.67 | 13 | 15400 | 3 | 0.02 | 10 | 0.07 |
| TN | 3.93 | 77 | 4610 | 15 | 0.33 | 61 | 1.33 |
| TX | 11.25 | 219 | 23150 | 44 | 0.19 | 175 | 0.76 |
| UT | 1.07 | 21 | 1160 | 4 | 0.36 | 17 | 1.44 |
| VT | 0.45 | 9 | 580 | 2 | 0.30 | 7 | 1.21 |
| VA | 4.65 | 91 | 2850 | 18 | 0.64 | 72 | 2.54 |
| WA | 3.41 | 66 | 4820 | 13 | 0.28 | 53 | 1.10 |
| WV | 1.75 | 34 | 760 | 7 | 0.90 | 27 | 3.59 |
| WI | 4.43 | 86 | 9160 | 17 | 0.19 | 69 | 0.75 |
| WY | 0.33 | 6 | 1810 | 1 | 0.07 | 5 | 0.28 |
| USA | 203.90 | 3971 | 328270 | 794 | 0.24 | 3177 | 0.98 |

1/ 1970 population estimates and harvested crop acreages for 1975.

2/ Sludge production estimates assume that 67% of the population was sewered and produced 0.16 lb of sludge per capita per day.

3/ Sewage sludge containing 1% or 4% available nitrogen (i.e., inorganic nitrogen) plus an additional 15 to 20% of this amount in organic form. Acreages are calculated on the basis of application of the sludge at rates to supply 100 lb of available nitrogen per acre.

Appendix Table 2. Estimated production of sewage sludge in the continental U.S.A. in 1985 and annual cropland requirements for utilization of the sludge in agriculture[1]

| State | Population, millions | Sludge produced,[2] thousands of short tons | Cropland, thousands of acres | Cropland required to accept the sludge having the indicated content of nitrogen[3]: 1% Available nitrogen, Acres | 1% Available nitrogen, Percent of total cropland | 4% Available nitrogen, Acres | 4% Available nitrogen, Percent of total cropland |
|---|---|---|---|---|---|---|---|
| AL | 3.91 | 134 | 2500 | 27 | 0.75 | 107 | 3.02 |
| AR | 2.45 | 84 | 1220 | 17 | 1.39 | 67 | 5.54 |
| AK | 2.18 | 75 | 7280 | 15 | 0.19 | 60 | 0.75 |
| CA | 23.66 | 810 | 8390 | 162 | 2.52 | 648 | 10.09 |
| CO | 2.73 | 93 | 6060 | 19 | 0.33 | 75 | 1.31 |
| CN | 3.53 | 121 | 120 | 24 | 16.11 | 97 | 64.42 |
| DE | 0.66 | 23 | 470 | 5 | 0.90 | 18 | 3.61 |
| FL | 9.90 | 339 | 2400 | 68 | 4.91 | 271 | 19.64 |
| GA | 5.51 | 189 | 3870 | 38 | 0.77 | 151 | 3.10 |
| ID | 0.72 | 25 | 4140 | 5 | 0.11 | 20 | 0.46 |
| IL | 12.56 | 430 | 21200 | 86 | 0.38 | 344 | 1.51 |
| IN | 6.07 | 208 | 10990 | 42 | 0.34 | 166 | 1.36 |
| IA | 2.95 | 101 | 22500 | 20 | 0.08 | 81 | 0.33 |
| KS | 2.25 | 77 | 23320 | 15 | 0.07 | 62 | 0.28 |
| KY | 3.79 | 130 | 3180 | 26 | 0.55 | 104 | 2.21 |
| LA | 3.84 | 131 | 3770 | 26 | 0.72 | 105 | 2.87 |
| ME | 0.98 | 34 | 400 | 7 | 1.60 | 27 | 6.39 |
| MD | 4.86 | 166 | 1370 | 33 | 2.17 | 133 | 8.70 |
| MA | 6.56 | 224 | 150 | 45 | 29.93 | 180 | 119.72 |
| MI | 10.18 | 348 | 6100 | 70 | 1.11 | 279 | 4.44 |
| MN | 4.33 | 148 | 18460 | 30 | 0.15 | 119 | 0.59 |
| MS | 2.39 | 82 | 5100 | 16 | 0.29 | 65 | 1.18 |
| MO | 5.25 | 180 | 11880 | 36 | 0.27 | 144 | 1.09 |
| MT | 0.67 | 23 | 11660 | 5 | 0.05 | 18 | 0.20 |
| NB | 1.53 | 52 | 18640 | 10 | 0.06 | 42 | 0.24 |
| NV | 0.68 | 23 | 450 | 5 | 0.93 | 19 | 3.72 |
| NH | 0.88 | 30 | 90 | 6 | 5.48 | 24 | 21.90 |
| NJ | 8.49 | 291 | 410 | 58 | 13.83 | 232 | 55.34 |
| NM | 1.09 | 37 | 830 | 7 | 0.60 | 30 | 2.41 |
| NY | 20.13 | 689 | 3640 | 138 | 3.38 | 551 | 13.51 |
| NC | 6.09 | 208 | 4110 | 42 | 0.88 | 167 | 3.51 |
| ND | 0.57 | 20 | 20610 | 4 | 0.02 | 16 | 0.08 |
| OH | 12.12 | 415 | 8740 | 83 | 0.77 | 332 | 3.09 |
| OK | 2.88 | 99 | 8640 | 20 | 0.19 | 79 | 0.76 |
| OR | 2.43 | 83 | 3420 | 17 | 0.63 | 67 | 2.52 |
| PA | 13.03 | 446 | 3850 | 89 | 1.99 | 357 | 7.96 |
| RI | 1.07 | 37 | 20 | 7 | 36.61 | 29 | 146.46 |
| SC | 2.97 | 102 | 1330 | 20 | 0.74 | 81 | 2.97 |
| SD | 0.65 | 22 | 14950 | 4 | 0.03 | 18 | 0.12 |
| TN | 4.86 | 166 | 3590 | 33 | 0.72 | 133 | 2.89 |
| TX | 12.85 | 440 | 21680 | 88 | 0.38 | 352 | 1.52 |
| UT | 1.23 | 42 | 740 | 8 | 0.73 | 34 | 2.90 |
| VT | 0.50 | 17 | 400 | 3 | 0.59 | 14 | 2.36 |
| VA | 5.70 | 195 | 2210 | 39 | 1.37 | 156 | 5.48 |
| WA | 3.68 | 126 | 6760 | 25 | 0.52 | 101 | 2.09 |
| WV | 1.84 | 63 | 510 | 13 | 1.66 | 50 | 6.63 |
| WI | 4.87 | 167 | 8810 | 33 | 0.36 | 133 | 1.46 |
| WY | 0.33 | 11 | 1440 | 2 | 0.12 | 9 | 0.50 |
| USA | 234.50 | 8024 | 312410 | 1605 | 0.49 | 6419 | 1.98 |

[1] 1985 population estimates for population and cropland projections (Water Resources Council, 1972).

[2] Sludge production estimates assume that 75% of population is sewered and produces 0.25 lb of sludge per capi per day.

[3] Sewage sludge containing 1% or 4% available nitrogen (i.e., inorganic nitrogen) plus an additional 15 to 20% of this amount in organic form. Acreages are calculated on the basis of application of the sludge at rates to supply 100 lb of available nitrogen per acre.

Appendix Table 3. Recovery of sludge-applied metals by four successive crops grown in field trials at two sites1/ in Wisconsin (Kelling et al., 1976)

| Sludge applied per hectare, metric tons | Metals added per hectare with the sludge, kg | | | | Total percentage of applied metals recovered in four crops2/ | | | |
|---|---|---|---|---|---|---|---|---|
| | Copper | Zinc | Nickel | Cadmium | Copper | Zinc | Nickel | Cadmium |
| 3.75 | 6 | 11 | 3 | 0.3 | 0.34 | 2.78 | 0.26 | 0.65 |
| 7.5 | 11 | 22 | 6 | 0.6 | 0.32 | 2.25 | 0.31 | 0.76 |
| 15 | 22 | 45 | 11 | 1.1 | 0.21 | 1.53 | 0.19 | 0.50 |
| 30 | 43 | 90 | 21 | 2.2 | 0.12 | 0.94 | 0.12 | 0.29 |
| 60 | 86 | 180 | 42 | 4.4 | 0.08 | 0.65 | 0.07 | 0.18 |

1/ Plano silt loam (Arlington, WI) and Warsaw sandy loam (Janesville, WI).

2/ The crops were rye or sorghum-sudan followed by three years of corn. Recovery data include both the grain and stover for each crop of corn.

Appendix Table 4. Metal content of corn grown in the field on sandy soil in Minnesota treated with different quantities of sewage sludge (Clapp, Dowdy, and Larson, 1976)

| Sludge applied[1/] per hectare, metric tons | Yield of grain per hectare, metric tons | Metals Zinc | Copper | Cadmium | Nickel | Lead | Chromium |
|---|---|---|---|---|---|---|---|
| | | Applied to soil, kg per hectare[2/] | | | | | |
| 119 | 14.9 | 555 | 290 | 1.3 | 42 | 325 | 135 |
| 237 | 15.2 | 1110 | 580 | 2.6 | 84 | 650 | 270 |
| 466 | 15.6 | 2175 | 1135 | 5.1 | 165 | 1275 | 530 |
| | | Present in dry leaf tissue, µg per g[3/] | | | | | |
| 0 | 9.1 | 19 | 6 | 0.04 | 0.3 | 1.5 | 0.4 |
| 119 | 14.9 | 132 | 11 | 0.12 | 0.8 | 1.3 | 0.4 |
| 237 | 15.2 | 160 | 13 | 0.19 | 1.4 | 1.0 | 0.4 |
| 466 | 15.6 | 182 | 12 | 0.25 | 3.0 | 0.7 | 0.5 |
| | | Present in dry grain tissue, µg per g[3/] | | | | | |
| 0 | 9.1 | 20 | 1.0 | <0.04 | 0.3 | 0.14 | <0.1 |
| 119 | 14.9 | 38 | 0.7 | <0.04 | 1.5 | 0.14 | <0.1 |
| 237 | 15.2 | 40 | 0.8 | <0.04 | 2.6 | 0.14 | <0.1 |
| 466 | 15.6 | 42 | 2.0 | <0.04 | 4.0 | 0.14 | <0.1 |

[1/] Anaerobically digested sewage sludge applied over a four-year period.

[2/] 105°C weight basis; total metals applied over a four-year period.

[3/] 70°C weight basis; metal concentration of tissue grown after final sludge application.

Appendix Table 5. Content of chromium in corn leaf and stover tissue from field plots on Blount silt loam in Illinois with and without application of sewage sludge (Hinesly, 1976)

| | | Chromium in dry tissue with indicated sludge treatment, ppm | | |
|---|---|---|---|---|
| Year | Tissue | No sludge applied | 1/2 Maximum sludge application | Maximum sludge application[1/] |
| 1971 | Leaf | 1.2 | 1.3 | 1.2 |
| 1973 | Leaf | 1.5 | 1.2 | 1.2 |
| 1973 | Stover | 1.0 | 1.2 | 1.2 |

[1/] Digested sludge was applied annually on replicated plots. By 1971, the maximum treatment had received 700 kg of chromium per hectare in 280.5 metric tons of sludge. By 1973, the maximum treatment had received 833 kg of chromium per hectare in 368.6 metric tons of sludge.

Appendix Table 6. Heavy-metal concentrations in vegetable crops grown in the field on Sango silt loam (pH 6.4) in Alabama with and without application of sewage sludge (Giordano and Mays, 1976a)

| Crop | Treatment[1] | Concentration in dry matter of fruit or root[2], ppm | | | | | Concentration in dry matter of leaves[2], ppm | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Zn | Cu | Ni | Cd | Pb | Zn | Cu | Ni | Cd | Pd |
| Lettuce | C | -- | -- | -- | -- | -- | 47.5 | 5.2 | 2.4 | 0.9 | 2.4 |
| | S | -- | -- | -- | -- | -- | 74.2 | 9.6 | 1.7 | 3.6 | 3.1 |
| Broccoli | C | 86.8 | 7.5 | 3.3 | 0.3 | 2.4 | -- | -- | -- | -- | -- |
| | S | 99.4 | 12.2 | 2.1 | 0.5 | 2.6 | -- | -- | -- | -- | -- |
| Potato | C | 15.7 | 7.8 | 0.8 | 0.1 | 1.3 | 26.7 | 14.9 | 4.1 | 0.8 | 5.1 |
| | S | 19.4 | 8.6 | 0.9 | 0.1 | 1.4 | 33.4 | 31.3 | 2.3 | 0.7 | 4.6 |
| Tomato | C | 25.7 | 5.0 | 1.3 | 0.5 | 1.6 | 37.5 | 20.7 | 2.5 | 1.1 | 7.3 |
| | S | 40.4 | 9.6 | 1.3 | 1.2 | 1.7 | 47.1 | 22.5 | 2.0 | 3.6 | 8.1 |
| Cucumber | C | 40.4 | 7.7 | 3.4 | 0.1 | 2.6 | 39.2 | 17.1 | 6.2 | 7.8 | 0.5 |
| | S | 67.5 | 14.4 | 2.2 | 0.4 | 2.6 | 80.0 | 12.0 | 3.9 | 10.9 | 0.9 |
| Egg Plant | C | 14.8 | 25.1 | 1.1 | 0.5 | 1.2 | 20.9 | 14.4 | 2.3 | 0.8 | 4.7 |
| | S | 22.5 | 26.5 | 1.0 | 1.6 | 1.3 | 22.1 | 17.7 | 1.8 | 2.0 | 4.6 |
| String Beans | C | 45.4 | 8.1 | 7.6 | 0.4 | 2.5 | 32.9 | 8.6 | 4.3 | 0.4 | 5.0 |
| | S | 60.5 | 8.9 | 2.8 | 0.4 | 2.7 | 49.2 | 7.6 | 3.0 | 0.5 | 6.8 |
| Cantaloupe | C | -- | -- | -- | -- | -- | 44.4 | 10.2 | 4.8 | 1.1 | 8.2 |
| | S | -- | -- | -- | -- | -- | 40.0 | 9.2 | 3.1 | 2.3 | 8.4 |

[1] C = control. S = Anaerobically digested sludge from Decatur, Alabama, applied in the fall of 1974 at a rate equivalent to 224 metric tons of dry matter per hectare. The heavy metal concentrations in the sludge were as follows: Zn = 1800 ppm, Cu = 730 ppm, Ni = 20 ppm, Cd = 50 ppm, and Pb = 530 ppm.

[2] The plant samples were taken in the summer of 1976.

Appendix Table 7. Cadmium concentrations in corn leaves in different years and in successive ryegrass cuttings within a single year as influenced by applications of sewage sludge

| Cadmium applied per hectare in sewage sludge, kg | Cadmium in dry matter of corn leaf tissue in indicated years, ppm | | | | | | Cadmium in dry matter of ryegrass in indicated cuttings, ppm | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | 1970 | 1971 | 1972 | 1973 | 1974 | 1975 | 1st | 2nd | 3rd | 4th |
| 0.0 (0.0)[1/] | | | | 0.05 | 0.20 | 0.10 | | | | |
| 2.2 (8.8) | | | | 0.45 | 1.30 | 1.19 | | | | |
| 4.5 (18.0) | | | | 1.07 | 2.25 | 1.62 | | | | |
| 20 (48)[2/] | 17.1 | | | | | | | | | |
| 29 (77) | | 25.4 | | | | | | | | |
| 4 (81) | | | 21.9 | | | | | | | |
| 7 (88) | | | | 22.1 | | | | | | |
| 13 (101) | | | | | 10.09 | | | | | |
| 7 (108) | | | | | | 13.5 | | | | |
| 0.00 (0.00)[3/] | | | | | | | 0.46 | 0.36 | 0.45 | 0.63 |
| 1.08 (4.33) | | | | | | | 1.16 | 0.93 | 0.90 | 1.08 |
| 8.64 (34.2) | | | | | | | 3.30 | 3.68 | 2.70 | 3.12 |

[1/] Research by Baker et al. (1976) in Pennsylvania. The applications of sludge indicated were made in 1973, 1974, and 1975. The values in parentheses are cumulative amounts applied through 1975. The crops were grown on soil in containers in the greenhouse.

[2/] Field research by Hinesly et al. (1976) in Illinois. The first application of sludge was made in 1968, and it supplied 11 kg of cadmium per hectare. The second application in 1969 supplied 17 kg of cadmium per hactare. The third application in 1970 supplied 20 kg of cadmium per hectare, a cumulalative total of 48 kg. The values in parentheses are cumulative total amounts of cadmium applied.

[3/] Field research by Bates et al. (1975) in Ontario. The cadmium applications indicated in the first column were made in applications of sludge in individual years, and the cumulative values in parentheses are the total applications by the time the ryegrass was planted. The four cuttings of ryegrass were all made in a single year.

Appendix Table 8. Cadmium content of swiss chard, soybeans, and oats grown on limed and unlimed soil with and without prior application of sludge from three cities (Chaney, Hornick, and Simon, 1976a)

| Site designation | Soil pH | Sludge treat- ment[2/] | Extractable cadmium in soil[2/], ppm | Cadmium in dry matter of crops, ppm | | | |
|---|---|---|---|---|---|---|---|
| | | | | Swiss chard leaves[3/] | Soybeans[3/] | | Oat grain[4/] |
| | | | | | Leaves | Grain | |
| City 4 | 5.7 | Control | 0.13 | 0.6 | 0.27 | 0.17 | 0.05 |
| | 6.7 | Control | 0.14 | 0.5 | 0.26 | 0.15 | 0.04 |
| | 5.2 | Sludge | 0.53 | 1.9 | -- | -- | 0.23 |
| | 6.2 | Sludge | 0.57 | 0.6 | -- | -- | 0.07 |
| City 9 | 5.3 | Control | 0.13 | 3.6 | 1.04 | 0.36 | 0.22 |
| | 6.7 | Control | 0.10 | 1.2 | 0.55 | 0.28 | 0.04 |
| | 4.8 | Sludge | 1.13 | 73.0 | 10.7 | 3.70 | 2.12 |
| | 6.6 | Sludge | 1.19 | 5.5 | 1.87 | 1.51 | 0.38 |
| City 13 | 5.3 | Control | 0.93 | 0.89 | 0.24 | 0.16 | 0.11 |
| | 6.4 | Control | 0.96 | 0.49 | 0.17 | 0.13 | 0.07 |
| | 5.6 | Sludge | 7.15 | 70.4 | 5.70 | 2.64 | 3.38 |
| | 6.6 | Sludge | 5.45 | 17.7 | 2.38 | 0.65 | 0.54 |

[1/]Applications of sludge were made from 1962 to 1975 at the City 4 site, from 1961 to 1973 at the City 9 site, and from 1967 to 1974 at the City 13 site.

[2/]Extractable by diethylenetriaminepentaacetic acid.

[3/]Grown in 1975.

[4/]Grown in 1976.

Appendix Table 9. Cadmium and zinc content of soils and swiss chard grown in the field (Site City 39) five years after the application of sludge with a cadmium concentration of 100 ppm (Hornick, Chaney, and Simon, 1976a)

| Soil pH | Total content of metals in soil, ppm | | Metals extractable from soil,[1/] ppm | | Metals in dry matter of of swiss chard leaves, ppm | |
|---|---|---|---|---|---|---|
| | Cd | Zn | Cd | Zn | Cd | Zn |
| 5.68 | 13.3 | 458 | 6.38 | 140 | 5.88 | 287 |
| 5.87 | 16.4 | 501 | 8.53 | 156 | 5.43 | 216 |
| 6.37 | 16.5 | 563 | 7.56 | 135 | 3.39 | 103 |
| 6.66 | 14.8 | 583 | 6.41 | 127 | 2.88 | 84 |
| 6.12[2/] | 0.06 | 57 | 0.04 | 1.6 | 0.43 | 28 |

[1/] Extractable by diethylenetriaminepentaacetic acid.

[2/] Control, no sludge applied.

Appendix Table 10. Cadmium content of crops grown on Woodstown silt loam treated with digested sludge or composted digested sludge (Chaney, Simon, et al., 1976)

| Treatment designation | Sludge or compost applied per hectare,[1/] metric tons | Extractable cadmium in soil, ppm | Tests in 1973: Soil pH | Tests in 1973: Cadmium in dry matter, ppm: Corn seedlings | Tests in 1973: Cadmium in dry matter, ppm: Swiss chard[2/] | Tests in 1976: Soil pH | Tests in 1976: Cadmium in dry matter of corn seedlings,[2/] ppm |
|---|---|---|---|---|---|---|---|
| Control | 0 | 0.03 | 5.5 | 2.1 | 2.0de | 5.8 | 0.41h |
| Sludge | 40 | 0.27 | 5.2 | 3.6 | 5.9ab | 4.9 | 3.0cd |
| | 80 | 0.40 | 5.6 | 6.6 | 6.3a | 4.9 | 4.9b |
| | 160 | 0.75 | 6.5 | 5.9 | 3.8cd | 4.4 | 6.2a |
| | 240 | 0.98 | 6.4 | 5.1 | 4.3bc | 4.4 | 6.3a |
| Sludge | 80 | 0.42 | 6.4 | 4.5 | 2.9cde | 6.4 | 1.8fg |
| | 160 | 0.74 | 6.3 | 5.9 | 3.5cde | 6.6 | 2.7cd |
| | 240 | 1.08 | 6.9 | 4.6 | 3.5cde | 6.4 | 3.2c |
| Compost | 40 | 0.10 | 5.8 | 3.2 | 2.9cde | 5.0 | 1.6g |
| | 80 | 0.11 | 6.0 | 2.8 | 2.1de | 4.9 | 2.1ef |
| | 160 | 0.19 | 6.5 | 2.9 | 2.3de | 4.4 | 2.5cdc |
| | 240 | 0.33 | 6.8 | 2.8 | 2.3de | 4.6 | 2.8cd |
| Compost | 80 | 0.14 | 6.8 | 1.2 | 1.5e | 6.6 | 0.5h |
| | 160 | 0.16 | 6.9 | 2.3 | 1.6e | 6.7 | 0.8h |
| | 240 | 0.25 | 7.0 | 2.2 | 1.6e | 6.6 | 1.1gh |

[1/] The sludge or compost were mixed with the soil by tillage in 1973. In 1976, sulfur was added to plots on which the desired low pH had not been achieved due to soil pH alteration by the sludge or compost.

[2/] Values not followed by the same letter or letters differ significantly at the 5% probability level.

Appendix Table 11. Zinc and cadmium content of soil and corn grain with different applications of sewage sludge in field experiments by various investigators

| Sludge applied per hectare, metric tons | Time from sludge application to sampling | Soil pH | Total zinc in soil per hectare kg | Zinc in dry corn grain, ppm | Total cadmium in soil per hectare, kg | Cadmium in dry corn grain, ppm |
|---|---|---|---|---|---|---|
| 0[1/] | 1 year | 5.8 | - | 20.1 | - | 0.09 |
| 3.7 | | 5.5 | 11.2 | 20.3 | 0.26 | 0.08 |
| 7.5 | | 5.4 | 22.5 | 20.6 | 0.53 | 0.10 |
| 15 | | 5.3 | 45 | 22.0 | 1.07 | 0.09 |
| 30 | | 5.3 | 90 | 26.3 | 2.15 | 0.09 |
| 60 | | 5.3 | 180 | 26.9 | 4.30 | 0.10 |
| 0[2/] | 2 years | 5.2 | - | 22 | - | 0.12 |
| 55 | | 5.4 | 64 | 34 | 0.5 | 0.41 |
| 100 | | 5.3 | 129 | 49 | 1.0 | 0.59 |
| 220 | | 5.3 | 258 | 60 | 2.0 | 0.68 |
| 0[3/] | 1 year | 6.6 | 5.6 | 28.5 | 0.2 | 0.015 |
| 5 | | 6.7 | 56 | 30.8 | 6.2 | 0.025 |
| 10 | | 6.9 | 113 | 36.3 | 12.4 | 0.065 |
| 20 | | 6.8 | 221 | 39.9 | 24.8 | 0.120 |
| 0[4/] | 6 months | 4.9 | - | 37 | - | 0.3 |
| 50 | | 5.3 | 90 | 43 | 2.5 | 0.9 |
| 100 | | 5.3 | 180 | 49 | 5 | 1.0 |
| 200 | | 5.6 | 360 | 44 | 10 | 1.2 |
| 0[5/] | 4 months | - | - | 44 | 0 | 0.18 |
| 45 | | - | 79 | 55 | 0.23 | 0.26 |
| 90 | | - | 158 | 46 | 0.46 | 0.31 |
| 145 | | - | 267 | 74 | 0.76 | 0.93 |

[1/] Kelling et al. (1976). Data are for field corn.

[2/] Decker, Chaney, and Wolf (1976). Data are for field corn.

[3/] Baker et al. (1976). Data are for field corn.

[4/] Giordano, Mortvedt, and Mays (1975). Data are for sweet corn.

[5/] Singh et al. (1976). Data are for sweet corn.

Table 12a. Cadmium extracted from calcareous Domino silt loam and cadmium content of various crops associated with a 25% yield depression from addition of cadmium in sewage sludge in greenhouse tests (Bingham et al., 1975)

| Crop | Plant part harvested and analyzed | Cadmium per gram of soil with a 25% reduction in crop yield, µg | | Cadmium per gram of plant dry matter with a 25% yield reduction, µg | |
|---|---|---|---|---|---|
| | | Cadmium added in sludge[1/] | Cadmium extracted by DTPA[2/] | Diagnostic leaf | Edible plant part harvested |
| Spinach | Shoot | 4 | 2.4 | 75.0 | 75.0 |
| Soybean | Dry bean | 5 | 3.0 | 7.0 | 7.0 |
| Curlycress | Shoot | 8 | 4.8 | 70.0 | 80.0 |
| Lettuce | Head | 13 | 7.8 | 48.0 | 70.0 |
| Corn | Grain | 18 | 10.8 | 35.0 | 2.0 |
| Carrot | Tuber | 20 | 12.0 | 32.0 | 19.0 |
| Turnip | Tuber | 28 | 16.8 | 121.0 | 15.0 |
| Field bean | Dry bean | 40 | 24.0 | 15.0 | 1.7 |
| Wheat | Grain | 50 | 30.0 | 33.0 | 11.5 |
| Radish | Tuber | 96 | 57.6 | 75.0 | 21.0 |
| Tomato | Ripe fruit | 160 | 96.0 | 125.0 | 7.0 |
| Zucchini squash | Fruit | 160 | 96.0 | 68.0 | 10.0 |
| Cabbage | Head | 170 | 102.0 | 160.0 | 11.0 |
| Rice | Grain | >640 | >384.0 | 3.0[3/] | 2.0[3/] |

[1/]The sludge used was obtained from a treatment plant that served a residential community and was low in cadmium. This sludge was enriched to different levels with cadmium as cadmium sulfate before addition to the soil. The cadmium-treated sludge was added at the rate of 10 g of dry sludge per kg of soil. An additional quantity of fertilizer containing nitrogen, phosphorus, and potassium was added to all cultures.

[2/]Cadmium extracted by a technique involving shaking 10 g of soil for 2 hours with 20 ml of diethylenetriaminepentaacetate (DTPA)-triethanolamine (TEA) solution of pH 7.3 for 2 hours.

[3/]These are the values measured with the maximum addition of 640 µg of cadmium per gram of soil. No yield depression was noted with this addition of cadmium.

Appendix Table 12b. Cadmium content of crops grown in the greenhouse on calcareous Domino silt loam with and without treatment with sewage sludge (Chaney and Giordano, 1977)

| | Cadmium per gram of dry plant tissue, µg | | | |
|---|---|---|---|---|
| | Control soil | | Sludge-treated soil[1/] | |
| Crop | Diagnostic leaf | Edible tissue | Diagnostic leaf | Edible tissue |
| Paddy rice | <0.1 | <0.1 | <0.1 | 0.2 |
| Upland rice | 0.4 | <0.1 | 0.9 | 0.4 |
| Sudangrass | 0.2 | 0.2 | 5.7 | 5.7 |
| White clover | 0.2 | 0.2 | 6.0 | 6.0 |
| Alfalfa | 0.3 | 0.3 | 8.2 | 8.3 |
| Bermudagrass | 0.3 | 0.3 | 9.4 | 9.4 |
| Field bean | 0.6 | <0.1 | 10.3 | 0.7 |
| Wheat | <0.1 | <0.1 | 11.6 | 5.8 |
| Zuchinni squash | 0.6 | <0.1 | 12.5 | 0.7 |
| Soybean | 0.4 | 0.7 | 15.6 | 10.7 |
| Tall fescue | 1.4 | 1.4 | 17.3 | 17.3 |
| Corn | 3.9 | <0.1 | 27.0 | 1.4 |
| Carrot | 1.4 | 0.9 | 38.0 | 16.0 |
| Cabbage | 0.7 | 0.2 | 39.0 | 1.8 |
| Radish | 4.2 | 0.3 | 40.0 | 4.0 |
| Swiss chard | 1.4 | 1.4 | 42.0 | 42.0 |
| Table beet | 0.8 | 0.2 | 47.0 | 4.5 |
| Romaine lettuce | 0.8 | 0.8 | 62.0 | 62.0 |
| Tomato | 2.6 | <0.1 | 71.0 | 2.4 |
| Curlycress | 2.4 | 2.4 | 89.0 | 89.0 |
| Spinach | 3.6 | 3.6 | 161.0 | 161.0 |
| Turnip | 1.8 | <0.1 | 162.0 | 9.2 |

1/ Sludge treated with cadmium sulfate to supply 10 µg of cadmium per gram of soil. The sludge was added at the rate of 10g per kg of soil.

Appendix Table 13. Cadmium content of vegetable crops grown in 1974 and 1975 on field plots of Sango silt loam with an initial pH value of 6.4 with and without application of sewage sludge from two sources in Alabama (Giordano and Mays (1976a)

| Crop and plant part | Cadmium in dry plant tissue, ppm | | | | | |
|---|---|---|---|---|---|---|
| | Control, no sludge applied | | Treated with Decatur sludge1/ | | Treated with Tuscumbia sludge1/ | |
| | 1974 | 1975 | 1974 | 1975 | 1974 | 1975 |
| Bean leaf | 0.46 | -- | 1.70 | -- | 0.55 | -- |
| Bean pods | 0.04 | -- | 0.23 | -- | 0.07 | -- |
| Okra leaf | 0.59 | 0.67 | 2.00 | 3.10 | 0.59 | 0.44 |
| Okra pods | 0.13 | 0.42 | 0.60 | 1.20 | 0.16 | 0.39 |
| Pepper leaf | 0.71 | 1.04 | 2.70 | 2.92 | 0.76 | 0.78 |
| Pepper fruit | 0.09 | 0.04 | 0.40 | 0.60 | 0.14 | 0.12 |
| Tomato leaf | 0.66 | 1.70 | 2.10 | 6.70 | 0.75 | 1.70 |
| Tomate fruit | 0.12 | 0.33 | 0.39 | 1.12 | 0.20 | 0.40 |
| Squash leaf | 0.34 | 0.70 | 0.63 | 2.15 | 0.36 | 0.87 |
| Squash fruit | 0.03 | 0.27 | 0.20 | 0.72 | 0.15 | 0.19 |
| Turnip leaf | 0.59 | -- | 2.60 | -- | 0.59 | -- |
| Turnip globe | 0.42 | -- | 1.30 | -- | 0.42 | -- |
| Radish leaf | 0.92 | -- | 3.10 | -- | 0.88 | -- |
| Radish globe | 0.29 | -- | 0.92 | -- | 0.33 | -- |
| Kale leaf | 0.63 | -- | 2.30 | -- | 0.63 | -- |
| Lettuce leaf | 1.00 | 1.20 | 8.60 | 7.00 | 3.00 | 2.60 |
| Spinach leaf | 1.00 | -- | 2.80 | -- | 0.84 | -- |

1/ The sludges were applied in the fall of 1973 in quantities to supply 112 metric tons of dry matter per hectare. The Decatur and Tuscumbia sludges supplied 2.45 and 1.75 kg of cadmium per hectare and had pH values of 6.6 and 6.1. The heavy-metal concentrations in the sludges were as follows: Cd = 49ppm, Zn = 1840ppm, and Cu = 740ppm in the Decatur sludge; and Cd = 35ppm, Zn = 3640ppm, and Cu = 516ppm in the Tuscumbia sludge.

Appendix Table 14. Cadmium content of the grain of corn grown in the field in Illinois on three soils receiving different amounts of sewage sludge (Hinesly et al., 1976b)

| Soil type | Cumulative amount of sludge applied per hectare, 1968-1971, metric tons | Cumulative amount of cadmium applied in sludge per hectare, 1968-1971, kg | Cadmium in dry matter of corn grain in 1972, ppm |
|---|---|---|---|
| | 0 | 0 | 0.10 |
| Blount silt loam | 40 | 12.9 | 0.21 |
| | 80 | 25.7 | 0.57 |
| | 160 | 51.4 | 0.96 |
| | 0 | 0 | 0.17 |
| Elliott silt loam | 40 | 12.9 | 0.49 |
| | 80 | 25.7 | 0.53 |
| | 160 | 51.4 | 1.29 |
| | 0 | 0 | 0.22 |
| Plainfield loamy sand | 40 | 12.9 | 0.27 |
| | 80 | 25.7 | 0.87 |
| | 160 | 51.4 | 0.94 |

Appendix Table 15. Content of cadmium in various tissues of chickens at the end of a 12-week feeding trial with various levels of dietary cadmium supplied as an inorganic cadmium salt (Baker et al., 1975)

| Cadmium in the diet, ppm | Cadmium in dry matter of indicated tissues, ppm | | | | | |
|---|---|---|---|---|---|---|
| | Laying hen | | | Broiler chicken | | |
| | Egg | Liver | Kidney | Muscle | Liver | Kidney |
| 0 | 0.047 | 1.6 | 8.5 | 0.07 | 0.2 | 0.4 |
| 3 | 0.075 | 9.0 | 30.7 | 0.15 | 4.7 | 9.3 |
| 12 | 0.071 | 26.5 | 92.6 | 0.26 | 15.1 | 49.7 |
| 48 | 0.12 | 91.8 | 305.9 | 0.75 | 87.2 | 239.0 |

Appendix Table 16. Content of cadmium in meadow vole tissues at the end of a 40-day feeding trial with corn and sorghum forages grown on soil with and without application of sewage sludge (Williams, Shenk, and Baker, 1976)

| Forage | Sludge treatment | Cadmium in dry matter[2/], ppm | | | |
|---|---|---|---|---|---|
| | | Diet | Liver | Kidney | Muscle |
| Corn | Control | 0.10a | 0.08a | 0.09a | 0.02a |
| Corn[1/] | Sludge | 1.09b | 0.43b | 0.42b | 0.05a |
| Sorghum | Control | 0.23a | 0.03a | 0.09a | 0.03a |
| Sorghum[1/] | Sludge | 2.76c | 1.86c | 2.84c | 0.03a |

1/ The material fed was a composite of produce from plots receiving 10 and 20 metric tons of sludge per hectare (Appendix Table 11).

2/ Values not followed by a common letter differ significantly at the 5% level of probability.

Appendix Table 17. Content of cadmium in various tissues of lambs at the end of a 191-day feeding trial with different levels of dietary cadmium supplied as an inorganic cadmium salt (Doyle et al., 1974)

| Cadmium added to the diet, ppm | Cadmium concentration in indicated tissue[1/], ppm | | | | |
|---|---|---|---|---|---|
| | Liver | Kidney | Muscle | Fat | Wool |
| 0 | 1.7a | 4.4a | 0.025a | 0.011a | 0.55a |
| 5 | 14.9ab | 58.9a | 0.047a | 0.010a | 1.20a |
| 15 | 51.7ab | 187.6b | 0.091a | 0.012a | 0.84a |
| 30 | 62.7b | 426.8c | 0.170a | 0.021a | 1.22a |
| 60 | 276.0c | 468.8d | 0.428b | 0.113b | 0.70a |

[1/] Values for fat are on the wet-weight basis. All others are on the dry-weight basis. Values in a given column not followed by the same letter differ significantly at the 5% probability level.

Appendix Table 18. Heavy metal content of sorghum grown in the field in Georgia in two years on soil at different pH values and with different additions of inorganic fertilizer and sewage sludge (Boswell, 1976)

| | Metal concentrations in dry tissue[2/] in indicated year, ppm | | | | | |
|---|---|---|---|---|---|---|
| | 1974 | | | 1975 | | |
| Treatment[1/] | Cu | Mn | Zn | Cu | Mn | Zn |
| Lime level$_0$ | | | | | | |
| Control | 5.7 | 28 | 16.4 | 5.6 | 28 | 20.7 |
| N-P-K (140-25-93)[3/] | 7.1 | 50 | 15.8 | 8.7 | 53 | 27.2 |
| Sewage sludge, 5.6 mt/ha | 5.7 | 53 | 19.6 | 5.4 | 28 | 32.9 |
| Sewage sludge, 11.2 mt/ha | 5.3 | 39 | 20.6 | 6.8 | 29 | 66.0 |
| Lime level$_1$ | | | | | | |
| Control | 5.2 | 30 | 14.1 | 4.8 | 19 | 15.3 |
| N-P-K (140-25-93) | 6.0 | 36 | 13.4 | 8.3 | 34 | 33.1 |
| Sewage sludge, 5.6 mt/ha | 5.7 | 24 | 17.5 | 5.6 | 20 | 31.3 |
| Sewage sludge, 11.2 mt/ha | 6.1 | 22 | 23.1 | 5.6 | 24 | 49.7 |
| Lime level$_2$ | | | | | | |
| Control | 5.9 | 35 | 15.7 | 5.1 | 26 | 17.1 |
| N-P-K (140-25-93) | 5.9 | 32 | 12.9 | 6.8 | 32 | 25.0 |
| Sewage sludge, 5.6 mt/ha | 5.4 | 28 | 14.6 | 5.4 | 22 | 35.4 |
| Sewage sludge, 11.2 mt/ha | 5.2 | 29 | 17.2 | 6.1 | 22 | 38.5 |

[1/]The treatments were applied annually. The numerical values for the inorganic fertilizer are kilograms per hectare. The concentrations of heavy metals in the sewage sludge were as follows: Cu = 656ppm, Mn = 1040ppm, and Zn = 8930ppm. Soil pH values at lime levels 0, 1, and 2 were 6.0, 6.6, and 6.9, respectively, in 1974.

[2/]The tissue analyzed was the third leaf down from the flag leaf.

[3/]The numerical values are kilograms of nitrogen, phosphorus, and potassium per hectare.

Appendix Table 19. Yield and cadmium content of swiss chard on three soils with different applications of sewage sludge having *various ratios* of cadmium to zinc (Page, 1976)

| Sludge applied per hectare, metric tons | Ratio of cadmium to zinc in sludge[1] | Cadmium added per gram of soil, µg | Domino soil | | | Hanford soil | | | Redding soil | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | pH | Yield of dry matter per culture, g | Cadmium per gram of dry matter, µg | pH | Yield of dry matter per culture, g | Cadmium per gram of dry matter, µg | pH | Yield of dry matter per culture, g | Cadmium per gram of dry matter, µg |
| 0 | -- | 0 | 7.4 | 13.6 | 0.8 | 5.2 | 15.0 | 1.8 | 4.5 | 12.3 | 1.6 |
| 20 | 0.005 | 0.10 | 7.2 | 19.8 | 1.3 | 5.1 | 17.2 | 1.9 | 4.6 | 17.3 | 1.8 |
| 20 | 0.0175 | 0.51 | 7.2 | 18.6 | 3.0 | 5.1 | 17.4 | 5.2 | 4.6 | 14.9 | 8.4 |
| 20 | 0.025 | 1.00 | 7.3 | 15.3 | 3.7 | 5.2 | 10.8 | 10.0 | 4.6 | 13.3 | 14.0 |
| 40 | 0.005 | 0.20 | 7.1 | 20.2 | 1.6 | 5.1 | 17.2 | 1.9 | 4.9 | 16.6 | 2.2 |
| 40 | 0.011 | 0.54 | 7.0 | 20.9 | 2.8 | 5.1 | 14.4 | 5.4 | 4.8 | 15.8 | 5.5 |
| 40 | 0.0175 | 1.02 | 7.1 | 15.6 | 3.5 | 5.2 | 16.1 | 7.8 | 4.7 | 15.4 | 10.0 |
| 40 | 0.022 | 1.51 | 7.1 | 17.1 | 4.6 | 5.2 | 3.9 | 6.8 | 4.8 | 7.7 | 18.0 |
| 40 | 0.025 | 2.00 | 7.0 | 15.3 | 5.3 | 5.4 | 1.4 | 7.2 | 4.9 | 2.2 | 16.0 |
| 80 | 0.005 | 0.40 | 6.6 | 20.3 | 1.6 | 5.2 | 13.8 | 1.8 | 5.5 | 12.0 | 2.2 |
| 80 | 0.007 | 0.59 | 6.7 | 20.7 | 1.9 | 5.4 | 15.9 | 3.2 | 5.4 | 12.3 | 3.8 |
| 80 | 0.011 | 1.08 | 6.8 | 20.6 | 3.2 | 5.4 | 15.9 | 5.9 | 5.1 | 12.9 | 7.8 |
| 80 | 0.015 | 1.56 | 6.8 | 17.8 | 3.0 | 5.5 | 11.8 | 12.0 | 5.2 | 13.6 | 11.0 |
| 80 | 0.018 | 2.05 | 6.8 | 16.5 | 4.7 | 5.5 | 4.6 | 15.0 | 5.4 | 7.9 | 13.0 |
| 80 | 0.022 | 3.03 | 6.8 | 18.9 | 5.7 | 5.6 | 0.9 | 11.0 | 5.3 | 1.2 | 16.0 |
| 80 | 0.025 | 4.00 | 6.7 | 16.1 | 10.0 | 5.5 | 0.6 | 5.4 | 5.1 | 0.4 | 6.0 |

1/ The various ratios were obtained by mixing different sludges.

Appendix Table 20. Yield and cadmium content of bermudagrass on three soils with different applications of sewage sludge having various ratios of cadmium to zinc (Page, 1976)

| Sludge applied per hectare, metric tons | Ratio of cadium to zinc in sludge[1/] | Cadmium added per gram of soil, µg | Domino soil | | | Hanford soil | | | Redding soil | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | pH | Yield of dry matter per culture, g | Cadmium per gram of dry matter, µg | pH | Yield of dry matter per culture, g | Cadmium per gram of dry matter, µg | pH | Yield of dry matter per culture, g | Cadmium per gram of dry matter, µg |
| 0 | -- | 0 | 7.4 | 16.75 | 0.21 | 5.2 | 26.0 | 0.35 | 4.5 | 16.9 | 0.40 |
| 20 | 0.005 | 0.10 | 7.2 | 21.3 | 0.27 | 5.1 | 25.6 | 0.52 | 4.6 | 24.1 | 0.43 |
| 20 | 0.0175 | 0.51 | 7.2 | 24.3 | 0.37 | 5.1 | 21.5 | 1.60 | 4.6 | 22.1 | 1.29 |
| 20 | 0.025 | 1.00 | 7.3 | 22.8 | 0.66 | 5.2 | 23.3 | 3.17 | 4.6 | 20.1 | 2.89 |
| 40 | 0.005 | 0.20 | 7.1 | 25.5 | 0.36 | 5.1 | 24.4 | 0.63 | 4.9 | 28.4 | 0.72 |
| 40 | 0.011 | 0.54 | 7.0 | 26.4 | 0.35 | 5.1 | 26.0 | 0.96 | 4.8 | 27.0 | 1.61 |
| 40 | 0.0175 | 1.02 | 7.1 | 30.9 | 0.70 | 5.2 | 24.5 | 1.59 | 4.7 | 26.7 | 2.63 |
| 40 | 0.022 | 1.51 | 7.1 | 25.9 | 1.25 | 5.2 | 22.9 | 3.34 | 4.8 | 22.2 | 4.95 |
| 40 | 0.025 | 2.00 | 7.0 | 27.6 | 1.30 | 5.4 | 23.7 | 3.10 | 4.9 | 23.0 | 5.10 |
| 80 | 0.005 | 0.40 | 6.6 | 36.6 | 0.41 | 5.2 | 28.8 | 0.44 | 5.5 | 29.6 | 1.55 |
| 80 | 0.007 | 0.59 | 6.7 | 25.7 | 0.40 | 5.4 | 26.7 | 0.49 | 5.4 | 25.8 | 2.94 |
| 80 | 0.011 | 1.08 | 6.8 | 30.3 | 0.78 | 5.4 | 27.7 | 1.60 | 5.1 | 27.8 | 5.68 |
| 80 | 0.015 | 1.56 | 6.8 | 27.9 | 0.85 | 5.5 | 22.5 | 1.73 | 5.2 | 25.3 | 4.65 |
| 80 | 0.018 | 2.05 | 6.8 | 29.3 | 1.30 | 5.5 | 24.3 | 2.95 | 5.4 | 24.8 | 4.02 |
| 80 | 0.022 | 3.03 | 6.8 | 24.8 | 2.64 | 5.6 | 25.3 | 4.00 | 5.3 | 29.0 | 6.60 |
| 80 | 0.025 | 4.00 | 6.7 | 26.2 | 3.56 | 5.5 | 25.5 | 3.52 | 5.1 | 40.6 | 8.72 |

[1/] The various ratios were obtained by mixing different sludges.

Appendix Table 21. Cadmium content of various parts of soybean plants grown on Sassafras sandy loam with different additions of cadmium and zinc (Chaney, White, and von Tienhoven, 1976b)

| Metals added to soil, ppm | | Terminal pH of bulk soil | Cadmium in dry plant tissue, ppm | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | | Seedlings (whole tops) | Diag- nostic leaves | Mature plants | | | |
| Zinc | Cad- mium | | | | Old leaves | Pods (bottom) | Beans (bottom) | Beans (top) |
| 0 | 0 | 5.5 d[1/] | 0.38 f | 0.66 e | 0.66 e | 0.14 c | 0.36 d | 0.37 e |
| 100 | 0 | 5.6 cd | 0.62 f | 0.52 e | 0.60 e | 0.12 c | 0.10 d | 0.12 e |
| 200 | 0 | 5.8 bd | 0.47 f | 0.56 e | 0.87 e | 0.16 c | 0.12 d | 0.14 e |
| 0 | 2 | 5.4 d | 1.86 e | 2.98 cd | 7.04 d | 1.37 c | 3.37 b | 5.14 b |
| 100 | 2 | 5.5 d | 3.71 d | 5.70 bc | 11.87 abc | 3.28 b | 2.16 c | 2.27 d |
| 200 | 2 | 6.0 ab | 5.11 c | 7.29 b | 9.18 cd | 3.98 ab | 1.93 c | 1.96 d |
| 0 | 4 | 5.5 d | 2.98 d | 4.73 bcd | 11.5 bc | 4.49 ab | 6.01 a | 8.61 a |
| 100 | 4 | 5.9 ab | 6.76 b | 7.30 b | 15.8 a | 5.27 a | 3.75 b | 4.55 bc |
| 200 | 4 | 6.2 a | 9.83 a | 14.13 a | 13.4 ab | 4.16 ab | 3.42 b | 3.84 c |

[1/] Values in a given column not followed by the same letters differ significantly at the 5% level of probability according to the Duncan multiple range test.

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