

Metropolitan Water Reclamation District of Greater Chicago

RESEARCH AND DEVELOPMENT DEPARTMENT

REPORT NO. 08-60

EFFECT OF NU EARTH BIOSOLIDS APPLICATION ON ACCUMULATION OF TRACE METALS IN EDIBLE TISSUE OF GARDEN VEGETABLES

OCTOBER 2008

100 East Erie Street

Chicago, IL 60611-2803

(312) 751-5600

EFFECT OF NU EARTH BIOSOLIDS APPLICATION ON ACCUMULATION OF TRACE METALS IN EDIBLE TISSUE OF GARDEN VEGETABLES

By

Albert E. Cox Soil Scientist III

Thomas Granato Assistant Director of Research and Development Environmental Monitoring and Research Division

> John Gschwind Sanitary Chemist IV, Retired

> > Odona Dennison Sanitary Chemist I

> > > Zainul Abedin Biostatistician

Research and Development Department Louis Kollias, Director

October 2008

TABLE OF CONTENTS

Page

LIST OF TABLES	iv
LIST OF FIGURES	ix
ACKNOWLEDGEMENT	xi
DISCLAIMER	xi
SUMMARY	xii
INTRODUCTION	1
MATERIALS AND METHODS	3
Vegetable Tissue Sampling and Analysis	3
Soil Sampling and Analysis	5
Statistical Analyses	5
RESULTS AND DISCUSSION	6
Effect of Annual Applications of Nu Earth on Concentrations of Trace Metals in Soil	6
Trace Metal Concentrations in Nu Earth and Loading Rates	6
Concentrations of Extractable Trace Metals in Soil	6
Relationship Between Trace Metal Loading Rates and Extract- able Concentrations in Soil	12
Effect of Annual Applications of Nu Earth on Concentrations of Trace Metals in Edible Tissue of Vegetables	14
Cadmium	14
Chromium	22

TABLE OF CONTENTS (Continued)

	Page
Copper	22
Nickel	22
Lead	31
Zinc	31
Residual Effect of Nu Earth on Soil Organic Carbon and Extractable Trace Metals Concentrations in Soil and Vegetables	31
Soil Organic Carbon Content	37
Extractable Trace Metal Concentrations in Soil	37
Concentrations of Trace Metal in Edible Tissue of Vegetables	44
Cadmium	44
Chromium	44
Copper	51
Nickel	51
Lead	51
Zinc	51
Trace Metal Concentrations in Vegetables Compared to USEPA Part 503 Risk Assessment Model Predictions and Cumulative Loading Limits	64
Comparison with Risk Assessment Model Predictions	64
Concentrations in Vegetables Compared to Concentrations Computed Using Part 503 Loading Limits	77
IMPLICATIONS FOR HOME GARDENS AMENDED WITH NU EARTH	80
LITERATURE CITED	81

TABLE OF CONTENTS (Continued)

Page

APPENDICES

Mean Concentration of 0.1 <i>M</i> HCl-Extractable Metals in Soil	AI-1
Mean Concentration of Cd in Edible Tissue of Garden Vegetables	AII-1
Mean Concentration of Cr in Edible Tissue of Garden Vegetables	AIII-1
Mean Concentration of Cu in Edible Tissue of Garden Vegetables	AIV-1
Mean Concentration of Ni in Edible Tissue of Garden Vegetables	AV-1
Mean Concentration of Pb in Edible Tissue of Garden Vegetables	AVI-1
Mean Concentration of Zn in Edible Tissue of Garden Vegetables	AVII-1

LIST OF TABLES

Table No.		Page
1	Analysis of Nu Earth Used in the Nu Earth Garden During 1977 Through 1981	7
2	USEPA Part 503 Biosolids Pollutant Concentration and Loading Limits	8
3	Cumulative Amounts of Trace Metals Applied at Four Nu Earth Rates	9
4	Effect of Nu Earth Application on 0.1 <i>M</i> HCl-Extractable Metals in Soil	10
5	Linear Regression of 0.1 <i>M</i> HCl-Extractable Metals in Soil Biosolids vs. Cumulative Metal Loading During Period of Annual Nu Earth Application	13
6	Effect of Nu Earth Application on Cd Concentration in Edible Tissue of Vegetables	18
7	Range of Trace Metal Concentrations in Samples of Six Vegeta- bles Collected in the District's National Trace Metal Survey	20
8	Effect of Nu Earth Application on Cr Concentration in Edible Tissue of Vegetables	23
9	Effect of Nu Earth Application on Cu Concentration in Edible Tissue of Vegetables	25
10	Effect of Nu Earth Application on Ni Concentration in Edible Tissue of Vegetables	28
11	Effect of Nu Earth Application on Pb Concentration in Edible Tissue of Vegetables	32
12	Effect of Nu Earth Application on Zn Concentration in Edible Tissue of Vegetables	34
13	Variability in Concentration of 0.1 <i>M</i> HCl-Extractable Trace Metals in Soil at Fifteen Locations of Each Replicate of Control Plots in 1990	39

Table <u>No.</u>		Page
14	Variability in Concentration of 0.1 <i>M</i> HCl-Extractable Metals in Soil at Fifteen Locations of Each Replicate of the 500 Mg/ha Cumulative Nu Earth Rate in 1990	40
15	Residual Effect of Nu Earth Application on 0.1 <i>M</i> HCl- Extractable Metals in Soil Receiving Nu Earth Annually During 1977 to 1981	41
16	Residual Effect of Nu Earth Application on Cd Concentrations in Edible Tissue of Vegetables Grown in Soil Receiving Nu Earth Annually During 1977 to 1981	45
17	Residual Effect of Nu Earth Application on Cr Concentrations in Edible Tissue of Vegetables Grown in Soil Receiving Nu Earth Annually During 1977 to 1981	48
18	Residual Effect of Nu Earth Application on Cu Concentrations in Edible Tissue of Vegetables Grown in Soil Receiving Nu Earth Annually During 1977 to 1981	52
19	Residual Effect of Nu Earth Application on Ni Concentrations in Edible Tissue of Vegetables Grown in Soil Receiving Nu Earth Annually During 1977 to 1981	55
20	Residual Effect of Nu Earth Application on Pb Concentrations in Edible Tissue of Vegetables Grown in Soil Receiving Nu Earth Annually During 1977 to 1981	58
21	Residual Effect of Nu Earth Application on Zn Concentrations in Edible Tissue of Vegetables Grown in Soil Receiving Nu Earth Annually During 1977 to 1981	61
22	Frequency of Measured Increases in Vegetable Tissue Metal Concentrations Due to Nu Earth that were Lower than Increases Predicted by the Part 503 Risk Assessment Model During the Period of Nu Earth Application, 1977 to 1981	75
23	Frequency of Measured Increases in Vegetable Tissue Metal Concentrations Due to Nu Earth that were Lower than Increases Predicted by the Part 503 Risk Assessment Model for the Period Following Termination of Nu Earth Application, 1982 to 1998	76

Table <u>No.</u>		Page
24	Safe Lifetime Consumption Concentrations of Cd, Ni, and Zn in Root, Leaf, and Fruit Vegetable Tissues Calculated Based on the Part 503 Biosolids Cumulative Metal Loading Limits and Up- take Coefficients Used in the Risk Assessment Model	78
25	Frequency of Occurrence During the Residual Period Where Metal Concentrations in Edible Tissue of Vegetables were Equal to or Greater than the Computed Part 503 Safe Lifetime Con- sumption Concentrations	79
AI-I	Mean Concentration of 0.1 <i>M</i> HCl-Extractable Cd in Soil	AI-I
AI-II	Mean Concentration of 0.1 <i>M</i> HC1-Extractable Cu in Soil	AI-II
AI-III	Mean Concentration of 0.1 <i>M</i> HC1-Extractable Cr in Soil	AI-III
AI-IV	Mean Concentration of 0.1 <i>M</i> HC1-Extractable Pb in Soil	AI-IV
AI-V	Mean Concentration of 0.1 <i>M</i> HC1-Extractable Ni in Soil	AI-V
AI-VI	Mean Concentration of 0.1 <i>M</i> HC1-Extractable Zn in Soil	AI-VI
AI-VII	Concentration of 0.1 <i>M</i> HC1-Extractable Metals in Soil Samples Collected in 1990 at Fifteen Locations within Replicate 1 of the Control	AI-VII
AI-VIII	Concentration of 0.1 <i>M</i> HC1-Extractable Metals in Soil Samples Collected in 1990 at Fifteen Locations within Replicate 2 of the Control	AI-VIII
AI-IX	Concentration of 0.1 <i>M</i> HC1-Extractable Metals in Soil Samples Collected in 1990 at Fifteen Locations within Replicate 3 of the Control	AI-IX
AI-X	Concentration of 0.1 <i>M</i> HC1-Extractable Metals in Soil Samples Collected in 1990 at Fifteen Locations within Replicate 1 of the 100 Mg/ha Nu Earth Treatment	A1-X
AI-XI	Concentration of 0.1 <i>M</i> HC1-Extractable Metals in Soil Samples Collected in 1990 at Fifteen Locations within Replicate 2 of the 100 Mg/ha Nu Earth Treatment	A1-XI

Table <u>No.</u>		Page
AI-XII	Concentration of 0.1 <i>M</i> HC1-Extractable Metals in Soil Samples Collected in 1990 at Fifteen Locations within Replicate 3 of the 100 Mg/ha Nu Earth Treatment	AI-XII
AII-I	Mean Concentration of Cd in Beet	AII-I
AII-II	Mean Concentration of Cd in Carrot	AII-II
AII-III	Mean Concentration of Cd in Spinach	AII-III
AII-IV	Mean Concentration of Cd in Swiss Chard	AII-IV
AII-V	Mean Concentration of Cd in Tomato	AII-V
AII-VI	Mean Concentration of Cd in Green Beans	AII-VI
AIII-I	Mean Concentration of Cr in Beet	AIII-I
AIII-II	Mean Concentration of Cr in Carrot	AIII-II
AIII-III	Mean Concentration of Cr in Spinach	AIII-III
AIII-IV	Mean Concentration of Cr in Swiss Chard	AIII-IV
AIII-V	Mean Concentration of Cr in Tomato	AIII-V
AIII-VI	Mean Concentration of Cr in Green Bean	AIII-VI
AIV-I	Mean Concentration of Cu in Beet	AIV-I
AIV-II	Mean Concentration of Cu in Carrot	AIV-II
AIV-III	Mean Concentration of Cu in Swiss Chard	AIV-III
AIV-IV	Mean Concentration of Cu in Spinach	AIV-IV
AIV-V	Mean Concentration of Cu in Tomato	AIV-V
AIV-VI	Mean Concentration of Cu in Green Beans	AIV-VI
AV-I	Mean Concentration of Ni in Beet	AV-I

Table		
<u>No.</u>		Page
AV-II	Mean Concentration of Ni in Carrot	AV-II
AV-III	Mean Concentration of Ni in Swiss Chard	AV-III
AV-IV	Mean Concentration of Ni in Spinach	AV-IV
AV-V	Mean Concentration of Ni in Tomato	AV-V
AV-VI	Mean Concentration of Ni in Green Beans	AV-VI
AVI-I	Mean Concentration of Pb in Beet	AVI-I
AVI-II	Mean Concentration of Pb in Carrot	AVI-II
AVI-III	Mean Concentration of Pb in Swiss Chard	AVI-III
AVI-IV	Mean Concentration of Pb in Spinach	AVI-IV
AVI-V	Mean Concentration of Pb in Tomato	AVI-V
AVI-VI	Mean Concentration of Pb in Green Beans	AVI-VI
AVII-I	Mean Concentration of Zn in Beet	AVII-I
AVII-II	Mean Concentration of Zn in Carrot	AVII-II
AVII-III	Mean Concentration of Zn in Swiss Chard	AVII-III
AVII-IV	Mean Concentration of Zn in Spinach	AVII-IV
AVII-V	Mean Concentration of Zn in Tomato	AVII-V
AVII-VI	Mean Concentration of Zn in Green Beans	AVII-VI

LIST OF FIGURES

Figure <u>No.</u>		Page
1	Nu Earth Garden Plot Layout	4
2	Effect of Cumulative Metal Loading on $0.1 M$ HCl-Extractable Metals in Soil	15
3	Effect of Cumulative Cd Loading on Concentration of Cd in Edible Tissue of Vegetables	21
4	Effect of Cumulative Cu Loading on Concentration of Cu in Edible Tissue of Vegetables	27
5	Effect of Cumulative Ni Loading on Concentration of Ni in Edible Tissue of Vegetables	30
6	Effect of Cumulative Zn Loading on Concentration of Zn in Edible Tissue of Vegetables	36
7	Residual Effect of Nu Earth Application on Soil Organic Carbon Content Following Annual Nu Earth Treatments Applied Annually During 1977 through 1981	38
8	Measured and Predicted (Part 503 Risk Assessment) Increases in Cd Concentrations in Edible Tissue of Root Vegetables Grown in Soil Receiving 40 Mg/ha (a) and 100 Mg/ha (b) Nu Earth Annually from 1977 to 1981	65
9	Measured and Predicted (Part 503 Risk Assessment) Increases in Cd Concentrations in Edible Tissue of Leaf Vegetables Grown in Soil Receiving 40 Mg/ha (a) and 100 Mg/ha (b) Nu Earth Annually from 1977 to 1981	66
10	Measured and Predicted (Part 503 Risk Assessment) Increases in Cd Concentrations in Edible Tissue of Fruit Vegetables Grown in Soil Receiving 40 Mg/ha (a) and 100 Mg/ha (b) Nu Earth Annually from 1977 to 1981	67
11	Measured and Predicted (Part 503 Risk Assessment) Increases in Ni Concentrations in Edible Tissue of Root Vegetables Grown in Soil Receiving 40 Mg/ha (a) and 100 Mg/ha (b) Nu Earth Annually from 1977 to 1981	69

LIST OF FIGURES (Continued)

Figure No. Page 12 Measured and Predicted (Part 503 Risk Assessment) Increases in 70 Ni Concentrations in Edible Tissue of Leaf Vegetables Grown in Soil Receiving 40 Mg/ha (a) and 100 Mg/ha (b) Nu Earth Annually from 1977 to 1981 13 Measured and Predicted (Part 503 Risk Assessment) Increases in 71 Ni Concentrations in Edible Tissue of Fruit Vegetables Grown in Soil Receiving 40 Mg/ha (a) and 100 Mg/ha (b) Nu Earth Annually from 1977 to 1981 14 Measured and Predicted (Part 503 Risk Assessment) Increases in 72 Zn Concentrations in Edible Tissue of Root Vegetables Grown in Soil Receiving 40 Mg/ha (a) and 100 Mg/ha (b) Nu Earth Annually from 1977 to 1981 73 15 Measured and Predicted (Part 503 Risk Assessment) Increases in Zn Concentrations in Edible Tissue of Leaf Vegetables Grown in Soil Receiving 40 Mg/ha (a) and 100 Mg/ha (b) Nu Earth Annually from 1977 to 1981 16 Measured and Predicted (Part 503 Risk Assessment) Increases in 74 Zn Concentrations in Edible Tissue of Fruit Vegetables Grown in Soil Receiving 40 Mg/ha (a) and 100 Mg/ha (b) Nu Earth Annually from 1977 to 1981

ACKNOWLEDGEMENT

The authors wish to acknowledge the field and laboratory staff of the Biosolids Utilization and Soil Science Section of the Metropolitan Water Reclamation District of Greater Chicago (District's) Research and Development Department. In particular Robert Hermann, Laboratory Technician II (retired); Alberta Johnson, Laboratory Technician II (retired); Joseph Calvano, Sanitary Chemist III; and Susan Feeney, Sanitary Chemist III (retired) made major contributions to the conduct and management of the study. Special thanks are extended to the staff of the Analytical Laboratories Division of the District for their support on analysis of extracts and digests of soil and plant tissue. The assistance of Ms. Sabina Yarn and Ms. Joan Scrima in formatting this report is also greatly appreciated.

DISCLAIMER

Mention of propriety equipment and chemical in this report does not constitute endorsement by the Metropolitan Water Reclamation District of Greater Chicago.

SUMMARY

Air-dried Imhoff biosolids (product name, Nu Earth) were produced by the Metropolitan Water Reclamation District of Greater Chicago (District) from the early 1930s to 1974. Nu Earth was distributed to the public within Cook County, Illinois to be used as a soil amendment in home gardens through 1979. In response to the United States Environmental Protection Agency (USEPA) concerns of high metal concentration in biosolids and imminent regulatory controls, the District began studies to evaluate the impact on Nu Earth application on metal accumulation in vegetables.

This Nu Earth Garden Study began at the Stickney Water Reclamation Plant (SWRP) in 1977. Nu Earth biosolids were applied to small plots at five rates of 0 (Control), 20, 40, 80, and 100 Mg/ha annually. The control plots received inorganic fertilizer. In 1980, the plots were split into two sets of subplots. Biosolids application continued in one set of subplots until 1981 (subplots treated for 5 years). The other subplots (subplots treated for 3 years) received only inorganic fertilizer thereafter. Every year from 1977 to 1998 (except 1995 and 1997), six species of common garden vegetables representing three groups of edible tissue, root (beet and carrot), leaf (spinach and Swiss chard), and fruit (tomato and green beans) were grown. At the end of each growing season, a sample of the vegetables was analyzed for metal content and soil was sampled and analyzed for 0.1 M HCl-extractable metals, pH, and organic carbon.

Concentrations of Cd, Cr, Cu, Ni, Pb, and Zn in the Nu Earth were higher than the current limits for exceptional quality biosolids and Cd concentrations were higher than the current ceiling concentration limits for land applied biosolids according to the USEPA Part 503 Rule. Annual loading rates of Cd and cumulative loading rates of Cd and Zn in some treatments were higher than the current Part 503 limits for annual and cumulative loading rates.

During the period of Nu Earth application, concentrations of extractable soil metals increased with metal loading rates. Relatively high concentrations of extractable metals observed in the Control indicate that the imported soil used as the upper 10 cm of the Nu Earth garden probably contained relatively high metal concentrations, which is typical of urban soils. The extractability of applied Cu, Ni, Pb, and Zn expressed as the percentage of total amounts applied which were found to be extractable, tended to decrease with cumulative loading rate. Mean extractability was lowest for Cr (6 percent) and ranged from 31 to 41 percent for the other metals.

During the period of Nu Earth application, concentrations of Cd, Ni, and Zn in most of the vegetables tended to increase with annual and cumulative loading rates. In most of the vegetables, concentrations of those metals increased linearly with cumulative loading rates. The effect of Nu Earth rate on concentrations of Cu, Cr, and Pb in vegetables was either slight or non-existent. Trends in the concentrations of metals in the subplots receiving Nu Earth for five years were similar to that of the subplots receiving Nu Earth for three years, but concentrations were usually slightly higher.

Seventeen years after termination of annual Nu Earth applications, OC contents in the amended plots were reduced by up to 34 percent of levels in the final year (1981) of Nu Earth application. During the 17-year residual period, the levels of extractable metals in soil also

decreased by as much as 57 percent of the concentrations attained in the final years of Nu Earth application. The increase in concentration of Cd, Ni, and Zn in the vegetable tissues due to Nu Earth observed during the period of Nu Earth application was also evident after application was terminated, but concentrations tended to decrease with time. This tendency for reduction in metal concentration in the vegetables following termination of Nu Earth application is opposite to predictions of the "Time Bomb Hypothesis."

The measured increases in Cd, Ni, and Zn concentrations in vegetables that were due to Nu Earth varied in comparison to the increases predicted by the USEPA in the Part 503 risk assessment model. Over the range of metal loading rates established during the study, Cd concentrations in the fruit vegetables and Zn concentration in fruit vegetables and carrot were much lower than the concentrations predicted by the Part 503 risk assessment model. The model does not account for decreases in bioavailability of biosolids-applied metal with time following termination of biosolids application. Therefore, following termination of Nu Earth application, the tendency of measured increases in concentrations to be lower than predicted by Part 503 increased with time. Since this relationship between measured and predicted increases in tissue concentrations existed under conditions where metal concentrations in the Nu Earth were higher than the Part 503 EQ limits for all of the metals evaluated in this study and ceiling limits for most of the metals, these results show that the Part 503 Rule provides very high levels of protection to humans by ensuring that metal transfer from biosolids to edible crops grown in biosolids amended soil does not pose a risk to human health.

Among the trace metals regulated in the Part 503 Rule, the concentration of Cd in Nu Earth appears to pose the highest potential for risk to human health and the environment. However, the data on concentrations of trace metals in the vegetables found in this study and the conservative assumptions the USEPA used to develop the cumulative loading limits of biosolidsborne trace metals in the Part 503 Rule indicate that there is a very low risk of health effects to consumers of vegetables grown in Nu Earth amended home gardens in the Chicago area. Two of the Part 503 conservative assumptions that are noteworthy to the use of Nu Earth in home gardens in the Chicago area are: (1) the assumption that 59 percent of the home gardener's vegetable consumption is grown in biosolids amended soil is unrealistic because of the short growing season in the Chicago area, and (2) it has been well demonstrated that the bioavailability of trace metals in biosolids amended soils decreases with time; an important factor that was not considered in the Part 503 risk assessment. In addition, the soil Pb level resulting from Nu Earth application in home gardens is much lower than the limit of 300 mg/kg stipulated in the Part 503 rule to protect the pica child. Owners of home gardens that have been amended with Nu Earth who might still be concerned about the risk of consuming vegetables grown in their gardens in the long term could further reduce the risk of elevated concentrations of trace metals in their vegetables by maintaining the home garden soil pH above 6.5. This can be done by obtaining a soil pH test and applying agricultural lime as recommended by a soil test laboratory.

INTRODUCTION

From the early 1930s to 1974 air-dried Imhoff digested biosolids (Nu Earth) produced by the Metropolitan Water Reclamation District of Greater Chicago (District) was distributed to the public within Cook County, Illinois. Nu Earth was used as a soil builder because of its high content of organic matter and plant nutrients (mainly nitrogen and phosphorus). Nu Earth also contained relatively high amounts of trace metals that can be potentially toxic to plants and animals. Increasing concerns of trace metal accumulation in the food chain and subsequent promulgation of regulatory standards for land application of municipal biosolids prompted the District to begin studies designed to evaluate the impact of Nu Earth on metal uptake in crops. These studies included a national survey of metals content in garden vegetables and a small plot experiment to study the impact of biosolids amendment on metal uptake by vegetables.

The national survey of metals content in garden vegetables (National Survey) was conducted during 1975 to 1978. Samples of seven common garden vegetables were collected from supermarkets and vegetable farms in seven U.S. states and analyzed for metal content. The results of the study were reported by Feeney et al. (1984).

The Nu Earth garden study was started in 1977 to evaluate the effect of Nu Earth application on metal uptake in edible portions of garden vegetables. Some results from this experiment have been presented in interim reports (Feeney et al., 1984; Granato et al., 1990). Granato et al. (1990) evaluated the results obtained from the study through 1989 and recommended that the study be terminated because it had generated adequate information to assess both short-term and long-term effects of Nu Earth on metal accumulation by garden vegetables. The experiment was terminated in 1998. In this report, the results of the Nu Earth garden experiment are presented primarily as they relate to uptake of Cd, Cr, Cu, Ni, Pb, and Zn and to current hypotheses on the bioavailability of these metals in biosolids amended soils. The results of the study are also used to evaluate the risk assessment methodology used by the USEPA to develop the 40 CFR Part 503 land application standards (503 Rule). Conversely, the Part 503 risk assessment is applied to the results of this study to evaluate long-term risk from gardening in Nu Earth amended soil.

The primary concern associated with land application of biosolids is the addition of trace metals that can potentially accumulate to phytotoxic levels in soils and high levels can be translocated in the food chain. Accumulation of trace metals by crops grown on soils receiving high rates or continuous application of biosolids is well documented (McGrath et al., 2000; Logan et al., 1997; Bell et al., 1991; Jackson and Alloway, 1991, Page et al., 1987). Trace metals vary in their potential to accumulate in plants and uptake rates depend on metal loading rates, biosolids characteristics, soil properties, and crop type (McGrath et al., 2000; Basta and Sloan, 1999; Corey et al., 1987; Sommers et al., 1987; Chaney et al., 1987). Corey et al. (1987) concluded that a plateau response curve of metal availability vs. loading rates is expected as metal concentration in the soil solution approaches equilibrium at high loading rates.

In evaluating the impacts of biosolids application on metal accumulation in crops, shortterm (during and immediately following cessation of applications) and long-term (beyond five years after cessation of applications) effects are important. Data from short-term studies that were conducted to determine the relationship between metal uptake by plants and metal loading in biosolids amended soil were utilized in the risk assessment used to develop USEPA's Part 503 Rule (USEPA, 1992). The USEPA acknowledged that the data used in the risk assessment were conservative because of the limited amount of data available from long-term field experiments at the time the risk assessment was done. This risk assessment assumed that bioavailability of metals applied in biosolids does not change over time. This approach tends to over-estimate the long-term bioavailability of metals in biosolids amended soils. Many researchers have shown that with time, biosolids constituents such as oxides of Al, Fe, and Mn provide surfaces that bind metals in less bioavailable forms (Bell et al., 1991; Corey et al., 1987). More information on long-term bioavailability of metals in biosolids amended soils is required to evaluate the adequacy of the risk assessment used to develop the 503 Rule and to improve the quality of future risk assessment.

The specific objectives of this report are to:

- 1. Evaluate short-term and long-term (residual) effects of annual application of Nu Earth biosolids on extractable metals in soil and metal concentration in edible portions of garden vegetables.
- 2. Compare the measured increases in trace-metal concentrations in edible tissue of vegetable crops to the concentration increases predicted by the Part 503 risk assessment model.
- 3. Evaluate likely risk from long-term consumption of vegetables produced on Nu Earth amended soils.

The report focuses on bioavailability of Cd, Cr, Cu, Pb, Ni, and Zn. Soil pH and OC content are also discussed, but only in the context of the effects of these parameters on bioavailability of trace metals.

MATERIALS AND METHODS

The Nu Earth garden experiment began in 1977 at the Stickney Water Reclamation Plant (SWRP). The grass sod on the experimental site, which was located in the northeast corner of the plant, was stripped and a 10-cm depth of purchased topsoil was spread and rototilled. Five treatments consisting of 0, 20, 40, 80, and 100 Mg/ha annual applications of Nu Earth were established. The 40 Mg/ha treatment is equivalent to the two bushels per 100 ft² Nu Earth application rate recommended by the District during the period that Nu Earth was distributed to the public. The control plots (0 Mg/ha Nu Earth rate) received 100 kg/ha N, P, and K annually. The plot layout of the experiment is presented in <u>Figure 1</u>. The experimental design was a completely randomized block design of five treatments and three blocks established on plots that were 24 feet long and 15 feet wide. A five foot wide grass buffer strip separated each block.

In 1980 each plot was split into subplots. In each plot, the annual Nu Earth application rates were continued in one set of subplots and the other subplots received inorganic fertilizer based on soil tests. After 1981 biosolids application was terminated and all subplots received inorganic fertilizer based on the results of soil tests. Therefore, after 1979, two sets of treatments were established based on the number of annual Nu Earth biosolids application. Cumulative application for subplots that received Nu Earth for three years (1977 to 1979) were 0, 60, 120, 240, and 300 Mg/ha. Cumulative application rates for subplots that received Nu Earth for five years (1977 to 1981) were 0, 100, 200, 400, and 500 Mg/ha. Six species of garden vegetables were planted in May in each experimental plot annually (1977 to 1998), except in 1995 and 1997. The vegetables represented three categories: root vegetables (beet- *Beta vulgaris* var. Early Wonder and carrot-*Daucus carrota* var. Royal Chantenay), leafy vegetables (spinach–*Spinacia oleracea* var. Bloomsdale Long-standing and Swiss chard-*Beta vulgaris* var. Fordhook Giant), and fruit vegetables (tomato–*Lycopersicon lycopersicum* var. Rutgers and green beans-*Phaseolus vulgaris* var. Tendergreen).

Vegetable Tissue Sampling and Analysis

A sample of the edible tissue (root-beet and carrot, fruit-green beans and tomato, leavesspinach and Swiss chard) of the vegetables from two rows of each plot (1977 to 1979) or subplots (1980 to 1998) was harvested at maturity once per growing season. Whole pods of green beans were sampled. Biomass produced and crop yields were not measured.

The beet and carrot samples were scrubbed but not peeled. All vegetable samples were rinsed in deionized water, dried at 65 °C, and ground. Samples were digested with nitric and perchloric acids and analyzed for Cd, Cr, Cu, Pb, Ni, Zn, K, Ca, and Mg by atomic absorption or graphite furnace atomic absorption spectrophotometry.

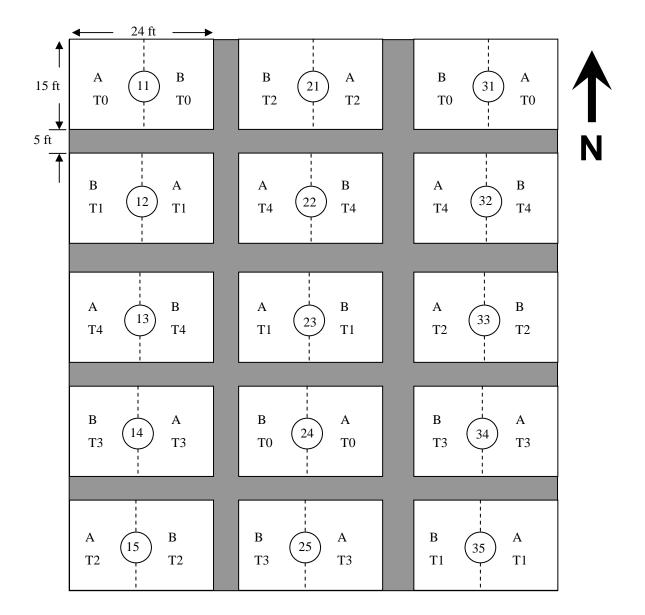


FIGURE 1: NU EARTH GARDEN PLOT LAYOUT

ANNUAL NU EARTH TREATMENTS

T0 = Control T1 = 20 Mg/ha Nu Earth T2 = 40 Mg/ha Nu EarthT3 = 80 Mg/ha Nu Earth

T4 = 100 Mg/ha Nu Earth

SUB-PLOT DESIGNATION

- A = Received Nu Earth 1977 to 1981
- B = Received Nu Earth 1977 to 1979
- \bigcirc = Plot number
- = Buffer

Soil Sampling and Analysis

Each spring after biosolids application, soil samples were taken from the 0-15 cm depth of each plot (1977 to 1979) or subplots (1980 to 1998). The soil samples were air-dried and crushed to pass a 2-mm sieve. Samples were extracted with 0.1 M HCl (Baker and Amacher, 1982) and analyzed for Cd, Cr, Cu, Pb, Ni, and Zn. Samples were also analyzed for extractable P (Bray P1), organic carbon (OC) content (wet oxidation), and pH (1:2 soil to water ratio). Soil samples collected prior to 1981 were not analyzed for OC content.

Statistical Analyses

The data was analyzed using SAS statistical software (SAS, 1994). For analysis of variance, the Shapiro-Wilk test for normality was done (SAS, 1995). If the test was significant, Kruskal-Wallis test was used. Otherwise the F test was used. For mean comparisons, the nonparametric Wilcoxon rank-sum test was used if the Shapiro-Wilk test for normality was significant. Otherwise the F test was used. Student's t test was used to compare Cd, Ni, and Zn concentrations in edible tissues to concentrations computed based on the Part503 risk assessment model and cumulative metal loading limits.

RESULTS AND DISCUSSION

Effect of Annual Applications of Nu Earth on Concentrations of Trace Metals in Soil

Trace Metal Concentrations in Nu Earth and Loading Rates. Selected chemical analyses of Nu Earth used in the study from 1977 to 1981 are presented in <u>Table 1</u>. Except for Cr and Pb, there was little variation (CV < 20 percent) in concentration of metals in the Nu Earth used during the five-year period (<u>Table 1</u>). Concentration of metals were in the order Zn > Cr > Cu > Pb > Ni > Cd. Concentrations of all metals in Nu Earth were much higher than concentrations in biosolids currently produced at the District. Except for Cu in 1977 and 1981 and Ni in 1978 and 1980, the concentration of trace metals in Nu Earth used in the study (<u>Table 1</u>) were higher than the current concentration limits for exceptional quality biosolids in the Part 503 Rule (<u>Table 2</u>). Concentrations of Cd, Ni (except 1978), and Pb (except 1977 and 1980) were higher than the current ceiling concentration limits for biosolids that can be land applied under the Part 503 Rule.

Cumulative metal loading rates for the five treatments are presented in <u>Table 3</u>. Cumulative loading rate in a given year was calculated as the sum of loading from previous years and the amount applied in that year. The annual loading in any year can be deduced as the difference between the cumulative loading in that year and the previous year.

Metal loadings in each treatment (<u>Table 3</u>) were in the same order as metal concentrations (<u>Table 1</u>). In some years, annual loading rates of some metals were higher than the limits currently in the Part 503 Rule. In all Nu Earth treatments, annual loading rates of Cd were always higher than the current Part 503 annual pollutant loading rates (APLR). In all years, loading rates of Cu (80 and 100 Mg/ha Nu Earth), Ni (40 and 100 Mg/ha Nu Earth), Pb (40 to 100 Mg/ha biosolids rates), and Zn (40 to 100 Mg/ha Nu Earth) were higher than the current Part 503 APLR.

Cumulative Cd loading (<u>Table 3</u>) was higher than the current Part 503 cumulative pollutant loading limits (CPLR) at Nu Earth rates of 40 Mg/ha (1981), 80 Mg/ha (1979 to 1981), and 100 Mg/ha (1978 to 1981). At the end of the Nu Earth application period, cumulative loading rates of Cd, Cu, Pb, and Zn at the 100 Mg/ha rates (<u>Table 3</u>) were at 300, 50, 89, and 74 percent of the CPLR. The relatively high cumulative metal loading rates established by the end of the application period provide a wide range of loading rates to evaluate the protection established in the Part 503 Rule.

Concentrations of Extractable Trace Metals in Soil. Extractable metals were determined by the 0.1 *M* HCl method. This method is used as an index of plant availability of trace metals and it extracts water-soluble metals as well as a portion of the metals held on the inorganic and organic fractions of the soil (Baker and Amacher, 1982). Nu Earth application significantly (P<0.001) increased extractable concentrations of all metals in soil during all years of biosolids application (Table 4). Annual mean extractable metal concentrations generally were in the

Constituent	1977	1978	1979	1980	1981
	Percent DW				
Total Solids	60.4	45.5	56.6	58.7	56.8
Total Volatile Solids	35.1	35.2	34.4	38.0	33.4
Total P	1.64	0.70	1.80	0.76	1.50
Total Kjeldahl-N	1.35	1.90	1.18	1.29	1.26
NH ₃ -N	0.06	0.10	0.11	0.10	0.12
Κ	0.07	0.13	0.22	0.13	0.06
Na	0.05	0.06	0.06	0.06	0.02
Ca	4.48	3.99	4.72	4.23	4.95
Mg	1.26	0.835	1.33	1.32	1.45
Al	1.06	0.942	1.54	0.839	1.41
Fe	2.86	2.70	2.96	2.06	2.44
	mg/kg DW				
Cd	200	220	300	230	210
Cr	2,800	2,290	3,340	1,590	2,580
Cu	1,290	1,680	1,750	1,530	1,270
Hg	4.0	10.0	4.0	NA	NA
Ni	430	360	470	340	510
Pb	720	1,450	1,310	1,260	870
Mn	280	270	290	300	410
Zn	3,310	4,660	4,860	4,010	3,890

TABLE 1: ANALYSIS OF NU EARTH USED IN THE NU EARTH
GARDEN DURING 1977 THROUGH 19811

¹Mean of three replications. NA = no analysis.

Pollutant	Ceiling Concentration ² (mg/kg)	Pollutant Concentration for EQ and PC Biosolids ³ (mg/kg)	Cumulative Pollutant Loading Rate ⁴ (kg/ha)	Annual Pollutant Loadir Rate ⁵ (kg/ha)
Cd	85	39	39	1.9
Cr ⁶	3,000	1,200	3,000	150
Cu	4,300	1,500	1,500	75
Pb	840	300	300	15
Ni	420	420	420	21
Zn	7,500	2,800	2,800	140

TABLE 2: USEPA PART 503 BIOSOLIDS POLLUTANT CONCENTRATION¹ AND LOADING LIMITS

¹Concentrations on dry weight basis. ²All biosolids that are land applied. ³Bulk or bagged biosolids. ⁴Bulk biosolids. ⁵Bagged biosolids. ⁶Limits for Cr were deleted from the final Part 503 Rule (USEPA, 1995).

Annual Nu Earth Rate (Mg/ha)	1977	1978	1979	1980	1981
			Cd		
20	4	8	14	19	$23(59)^1$
40	9	16	29	38	46 (118)
80	18	33	58	76	93 (238)
100	22	41	72	95	116 (297)
			Cr		
20	62	101	171	202	254
40	123	201	341	405	508
80	246	402	683	810	1,016
100	308	503		1,012	1,270
-					
20	28	57	94	124	150 (10)
40	57	114	187	249	299 (20)
80	114	228	375	497	599 (40)
100	142	285	468	621	748 (50)
-			Ni		
20	9	16	25	32	42 (10)
40	19	31	51	65	85 (20)
80	38	62	102	129	170 (40)
100	47	78	127	161	212 (50)
-			Pb		
20	16	40	50	57	75 (25)
40	32	81	101	114	149 (50)
80	63	162	201	229	298 (100)
100	79	202	252	286	373 (124)
-			Zn		
20	73	152	254	334	412 (15)
40	146	304	508	669	824 (29)
80	291	608	1,016		1,648 (59)
100	364	760	1,271	1,672	2,061 (74)

TABLE 3: CUMULATIVE AMOUNTS OF TRACE METALS APPLIED (kg/ha)AT FOUR NU EARTH RATES

¹Numbers in parentheses represent cumulative loading rates established in 1981 as a percentage of Part 503 cumulative pollutant loading limits. There are no Part 503 limits for Cr.

2	$\frac{\text{Annual Nu Earth Rate (Mg/ha)}^{1}}{0^{3}} \frac{20}{40} \frac{40}{80} \frac{80}{100}$									
Year ²	0^{3}	20	40			Mean				
			(Cd						
1977	0.5	4.8	5.4		17.3	7.9				
1978	3.6	11.3	12.6	23.1	19.2	14.0				
1979	2.7	7.4	13.0	17.2	25.8	13.2				
1980	2.7	44.8	96.7	122.6	37.4	60.8				
1981	3.2	12.0	19.9	38.1	42.3	23.1				
Mean	2.6	16.1	29.5	42.5	28.4	23.8				
P-value ⁴	0.1071	16.1 0.0043	0.0005	0.0003	0.0001					
			(Cr						
1977	5.9	6.0	6.7	17.2	17.3	10.6				
1978	2.7	8.7	15.9	35.7	33.8	19.3				
1979	5.0	10.1	20.4	27.5	42.5	21.1				
1980	3.7*	11.8	29.1	42.4	72.3	38.9				
1981	4.7	22.9	40.4	81.3	91.6	48.2				
Mean	4.6	11.9	22.5	40.8	51.5	26.3				
P-value ⁴	0.0259	0.0003	0.0001	0.0003						
			(Cu						
1977	11.7	28.8	29.4	51.4	83.7	41.0				
1978	16.1	41.0			111.7					
1979	16.4	32.8	61.3	84.3	123.8	63.7				
1980	19.8*	45.2	92.4	146.2	215.8	124.9				
1981	8.1*	54.7	116.3	162.7	126.5	115.1				
Mean	14.7	40.5	72.9	114.6	132.3	83.4				
P-value ⁴	0.0007	0.0667	0.0002	0.0206	0.0297					
			Ì	Ni						
1977					28.7					
1978	6.3	12.0	18.2	29.0	39.0	20.9				
1979	7.4	10.8	16.0	20.2	28.0	16.5				
1980	10.5*	15.4	31.8	32.2	53.7	33.3				
1981	7.5*	19.4	30.2	53.7	60.2	40.9				
Mean	6.6	13.7	21.8	31.4	41.9	23.1				
\mathbf{P}^4	0.0016	0.0026	0.0001	0.0003	0.0007					

TABLE 4: EFFECT OF NU EARTH APPLICATIONON 0.1 *M* HCI-EXTRACTABLE METALS (mg/kg) IN SOIL

Annual Nu Earth Rate (Mg/ha) ¹											
Year ²	0^{3}	20	40	80	100	Mean					
]	Pb							
1977	17.7	30.9	33.4	53.9	72.9	41.8					
1978	19.6	42.9	57.6	106.5	91.9	63.7					
1979	24.5	37.9	58.0	79.4	105.2	61.0					
1980	29.7	37.6	88.8	120.4	181.8	91.7					
1981	22.9	69.4	106.0	180.0	190.2	113.7					
Mean	22.9	43.7	68.7	108.1	128.4	74.4					
\mathbf{P}^4	0.5146	0.0009	0.0002	0.0002	0.0001						
			2	Zn							
1977	27.4	80.7	90.8	186.7	266.8	130.5					
1978	36.2	107.9	197.2	387.3	347.0	215.1					
1979	75.5	119.2	223.3	298.0	398.1	222.8					
1980	45.2	120.6	258.2	383.7	611.7	283.9					
1981	53.4*	202.0	454.7	698.3	766.7	530.4					
Mean	46.1	126.1	244.8	390.8	478.0	257.2					
\mathbf{P}^4	0.0044	0.0020	0.0045	0.0011	0.0002						

TABLE 4 (Continued): EFFECT OF NU EARTH APPLICATIONON 0.1 M HCI-EXTRACTABLE METALS (mg/kg) IN SOIL

¹ Treatment effect highly significant (P<0.01) in all years.

² Values for 1977 to 1979 are for each plot before they were split into subplots, and 1980 and 1981 values are for subplots receiving Nu Earth from 1977 to 1981.

³ Asterisk (*) denotes significant (P<0.05) difference compared to 1978. Comparisons were made only for the 0 Mg/ha treatment.

⁴ P = Significance probability of year effect on concentrations according to *F* test. Data for 1977 was not included because only mean data for each treatment was available for 1977.

order Zn >> Pb = Cu > Cr = Ni > Cd. Extractable levels of metals in biosolids amended soils are a function of loading rates and intrinsic solubilities of the metals (Logan et al., 1997; Sommers et al., 1987). The difference in the order of extractable levels compared to loading rates (<u>Table 2</u>) is most likely due to differences in metal compound solubility and affinity for sorption on soil solids.

Except for Cu at the 20 Mg/ha Nu Earth rate, there were some year-to-year fluctuations in the concentrations of extractable metals. As generally expected, concentrations of extractable metals tended to increase with time in response to annual metal loadings. In 1980, extractable Cd in the 20, 40, and 80 Mg/ha treatments were high compared to levels in 1979 and 1981. In 1978, extractable Cu in the 20, 40, and 80 Mg/ha treatments were high compared to levels in 1977 and 1979. This variability may be partially due to sampling or analytical error. Variations in the concentrations of extractable metals throughout the period 1977 to 1981 are not attributable to soil pH because there was very little variation in soil pH (pH = 7.0 ± 0.18 ; data not shown) during the period.

Fluctuations in metal concentrations in the Control plots (0 Mg/ha) over time can be used to assess the possibility of cross contamination between treatment plots during the experiment. In the Control plots, there were a few instances where metal concentrations were significantly higher or lower compared to 1977 values (<u>Table 4</u>). The differences observed may be due mostly to year-to-year variability in how representative the samples are and the accuracy of analyses because there was no consistent trend in the changes in concentration over time. Therefore, cross contamination between the plots during the period of Nu Earth application appears minimal.

Relationship Between Trace Metal Loading Rates and Extractable Concentrations in Soil. The relationship between 0.1 *M* extractable metal concentration (mg/kg) in the surface 15 cm of the soil and metal loading rate (kg/ha) can be used as an index of solubility and extractability in biosolids amended soils. This relationship assumes that movement of applied metals below the depth of incorporation (15 cm) is minimal. Leaching of metals below the depth of incorporation in biosolids amended soils is usually very slow (Yingming and Corey, 1993; Sukkariyan et. Al, 2005a.

The relationship between trace metal cumulative loading rates and extractable concentration were linear in most years (<u>Table 5</u>). The magnitude of the slope is directly related to extractability. In the first year of Nu Earth application (1977) metal extractability was in the order Zn = Pb > Ni = Cu > Cd > Cr (<u>Table 5</u>). In all years, Cr was least extractable. The relatively low extractability of Cr may be due to the ability of Cr to complex into insoluble forms in the soil. Extractability of Cu, Ni, Pb, and Zn tended to decrease with time (<u>Table 5</u>). Extractability of Cr tended to increase with time and Cd extractability fluctuated with no obvious trend (<u>Table 5</u>). Some of the annual variation in metal extractability may be due to changes in lability of metals in years following application. Logan et al. (1997) found that in soil receiving a one-time application of biosolids, EDTA-extractability of Cd and Cu tended to increase with time, but found no consistent trend for Ni, Pb, and Zn.

TABLE 5: LINEAR REGRESSION OF 0.1 M HCI-EXTRACTABLE METALS IN SOIL BIOSOLIDS (mg/kg) VS. CUMULATIVE METAL LOADING (kg/ha) DURING PERIOD OF ANNUAL NU EARTH APPLICATION

Cd^2			Cr		Cu		Ni			Pb		Zn						
Year ¹	I^3	S^3	r ³	Ι	S	r	Ι	S	r	Ι	S	r	Ι	S	r	Ι	S	r
1977	1.8	0.27	0.86	4.0	0.044	0.88	10	0.45	0.91	5.7	0.46	0.98	17	0.65	0.96	22	0.62	0.96
1978	4.5	0.43	0.71	2.6	0.069	0.75	20	0.38	0.76	5.4	0.41	0.70	25	0.40	0.74	47	0.46	0.78
1979	2.8	0.31	0.99	2.7	0.045	0.88	12	0.22	0.79	6.5	0.17	0.87	23	0.23	0.76	56	0.27	0.85
1980	35	0.57	0.01	1.3	0.063	0.85	14	0.30	0.88	9.9	0.24	0.82	21	0.32	0.85	29	0.32	0.87
1981	3.8	0.35	0.92	5.1	0.071	0.89	18	0.26	0.95	8.3	0.25	0.93	32	0.31	0.91	84	0.36	0.83
Mean	3.2	0.34	0.87	3.1	0.058	0.85	15	0.32	0.86	7.2	0.31	0.86	24	0.38	0.84	48	0.41	0.86

 ¹ Regressions for 1980 and 1981 are for subplots receiving Nu Earth from 1977 to 1981.
² In computing the mean regression parameter values for Cd, values for 1980 are excluded because of the low regression coefficient.

³ I = Intercept, S = Slope, and r = regression coefficient.

<u>Figure 2</u> shows the relationship between cumulative metal loading and extractable concentrations during the five years of Nu Earth application. The amount of applied metals recovered in the extractable form in the 15-cm soil layer (kg/ha) was calculated by multiplying the soil concentration (mg/kg) by 2, with the assumption that the 15-cm soil layer weighs approximately 2 x 10^6 kg/ha. The 1:1 line represents the relationship in which all applied metals are recovered. For Cd, the data for the 20, 40, and 80 Mg/ha loading rates in 1980 (<u>Table 4</u>) were excluded as outliers in the relationship (see discussion above).

For all metals, the relationship, extractable concentration vs. cumulative loading rate, was almost linear and the slopes were less than 1. Recovery was highest and similar for Cd (<u>Figure 2a</u>), Pb (<u>Figure 2e</u>), and Zn (<u>Figure 2f</u>) and was lowest for Cr (<u>Figure 2b</u>).

Effect of Annual Applications of Nu Earth on Concentrations of Trace Metals in Edible Tissue of Vegetables

Cadmium. Annual application of Nu Earth caused significant (P < 0.05) increases in Cd concentration in all vegetables except in carrot (1978) and green beans (1978 and 1979) (<u>Table 6</u>). Except for relatively high concentrations in spinach in 1977, Cd concentrations in all vegetables tended to increase with time. This increase is most likely due to the annual increase in cumulative Cd loading. Some changes in Cd concentrations over time are attributed also to year-to-year variability because there were annual fluctuations in Cd concentrations in the control treatment.

Mean Cd concentrations among vegetables during the period 1977 to 1981 were generally in the order spinach = Swiss chard > beet = carrot = tomato > green beans. Brown et al. (1996) found that Cd concentrations in vegetables grown in Nu Earth amended soil were in the order carrot > tomato > beans. Rates of metal accumulation in crops are dependent on cultivar (Chaney et al., 1987). Cadmium concentrations in the vegetables in most Nu Earth treatments were higher than concentrations observed in results of the District's National Survey of Trace Metals in Vegetables (<u>Table 7</u>). In some cases concentrations in the control treatment were higher than survey results. The concentrations of available metals in the soil used in the study were probably relatively high as typical of topsoil in urban areas.

The relationships between cumulative Cd loading and concentrations in vegetables are presented in Figure 3. The effect of loading rate on tissue concentration tended to be linear for root vegetables (Figure 3a) and leafy vegetables (Figure 3b). In Figure 3b, the 1977 data for spinach was omitted since they did not fit any noticeable trend. In the fruit vegetables, tissue concentrations tended to increase with loading rate up to approximately 60 kg/ha Cd loading for tomato and there was very little effect on green beans. This relationship tends to conform to the crop metal uptake plateau theory proposed by Corey et al. (1987) and the uptake response for Cd in lettuce and radish grown in biosolids amended soil observed by Sukkariyah et al. (2005b).

FIGURE 2: EFFECT OF CUMULATIVE METAL LOADING ON 0.1 M HCL-EXTRACTABLE METALS IN SOIL

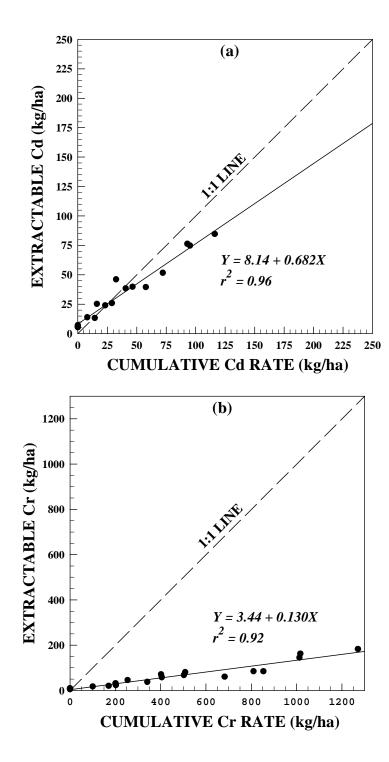


FIGURE 2 (Continued): EFFECT OF CUMULATIVE METAL LOADING ON 0.1 *M* HCI-EXTRACTABLE METALS IN SOIL

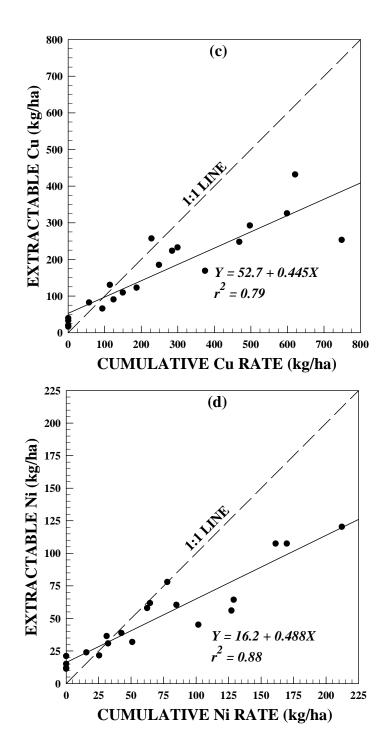
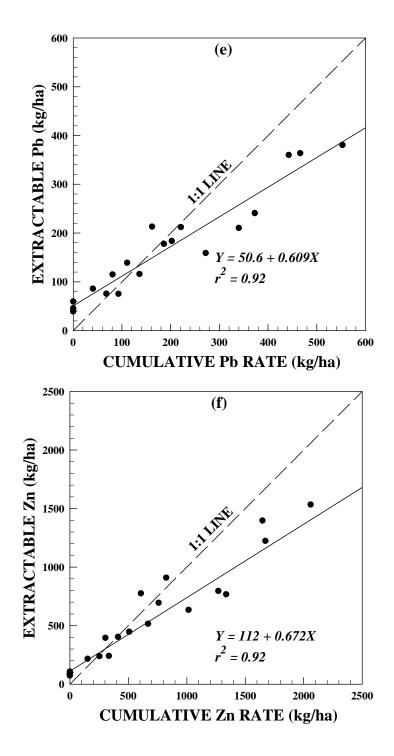


FIGURE 2 (Continued): EFFECT OF CUMULATIVE METAL LOADING ON 0.1 *M* HCI-EXTRACTABLE METALS IN SOIL



1				te (Mg/ha)		_	2
Year ¹	0	20	40	80	100	Mean	\mathbf{P}^2
				Beet -			
1977	0.51	1.04	0.98	2.03	1.95	1.30	0.0001
1978	0.35	0.98	1.11	2.00	1.75	1.24	0.0000
1979	0.22	1.39	1.60	2.69	2.94	1.77	0.0008
1980	0.89	2.31	4.31	7.18	9.10	4.76	0.0000
1981	1.20	2.27	3.52	4.21	4.78	3.20	0.0000
Mean	0.64	1.60	2.30	3.62	4.10	2.45	
\mathbf{P}^2	0.0001	0.0007	0.0001	0.0004	0.0001		
				Carrot -			
1977	1.04	1.25	1.30	1.83	1.73	1.43	0.0312
1978	0.14	0.67	0.60	0.89	0.72	0.60	0.1100
1979	0.67	1.41	1.16	1.90	2.30	1.49	0.0237
1980	1.83	4.57	6.51	2.67	3.98	3.91	0.0105
1981	1.53	2.80	4.09	4.53	4.76	3.54	0.0008
Mean	1.04	2.14	2.73	2.37	2.70	2.20	
\mathbf{P}^2	0.0000	0.0008	0.0002	0.0010	0.0002		
				Swiss Cha	ard		
1977	2.20	4.88	8.43	7.03	10.22	6.55	0.0002
1978	1.31	5.70	6.18	11.46	11.48	7.23	0.0006
1979	1.80	5.01	7.62	10.52	13.84	7.76	0.0011
1980	2.03	4.57	10.62	17.85	26.30	12.27	0.0000
1981	2.65	6.22	11.53	14.90	16.15	10.29	0.0000
Mean	2.00	5.28	8.88	12.35	15.60	8.82	
\mathbf{P}^2	0.0057	0.0090	0.0014	0.0002	0.0016		
				Spinach			
1977	5.38	11.9	18.8	-	14.6	13.5	0.0054
1978	2.94	4.82	5.17	7.3	3.71	4.79	0.0207
1979	3.97	7.88	6.42	9.02	7.55	6.97	0.0302
1980	2.60	3.95	7.45	8.68	8.24	6.18	0.0137
1981	5.70	11.2	14.1	19.2	14.5	12.9	0.0000
Mean	3.80	6.96	8.28	11.04	8.50	7.72	
P^2			0.0017	0.0015	0.0002		

TABLE 6: EFFECT OF NU EARTH APPLICATION ON CdCONCENTRATION (mg/kg) IN EDIBLE TISSUE OF VEGETABLES

		Annual N	u Earth Rat	te (Mg/ha)			
Year ¹	0	20	40	80	100	Mean	\mathbf{P}^2
				- Green Bea	ns		
1977	0.07	0.11	0.21	0.31	0.26	0.19	0.0009
1978	0.31	0.26	0.39	0.50	0.45	0.38	0.0913
1979	0.40	0.27	0.48	0.46	0.56	0.43	0.2128
1980	0.15	0.13	0.23	0.34	0.41	0.25	0.0123
1981	0.07	0.10	0.18	0.37	0.34	0.21	0.0022
Mean	0.20	0.17	0.30	0.40	0.40	0.29	
\mathbf{P}^2	0.0008	0.0018	0.0010	0.0053	0.0903		
				Tomato -			
1977	1.52	1.34	1.50	1.93	2.04	1.67	0.0001
1978	0.85	1.95	1.69	2.76	2.95	2.04	0.0008
1979	1.14	1.78	2.34	2.23	3.37	2.17	0.0002
1980	1.62	1.51	2.98	3.38	3.93	2.68	0.0030
1981	0.86	1.42	1.77	2.85	2.43	1.87	0.0000
Mean	1.20	1.60	2.06	2.63	2.95	2.09	
\mathbf{P}^2	0.0012	0.0442	0.0193	0.0296	0.0000		

TABLE 6 (Continued): EFFECT OF NU EARTH APPLICATION ON Cd CONCENTRATION (mg/kg) IN EDIBLE TISSUE OF VEGETABLES

¹Values for 1980 and 1981 represent data from plots receiving Nu Earth from 1977 to 1981. ²P = Significance probability of Nu Earth rate or Year.

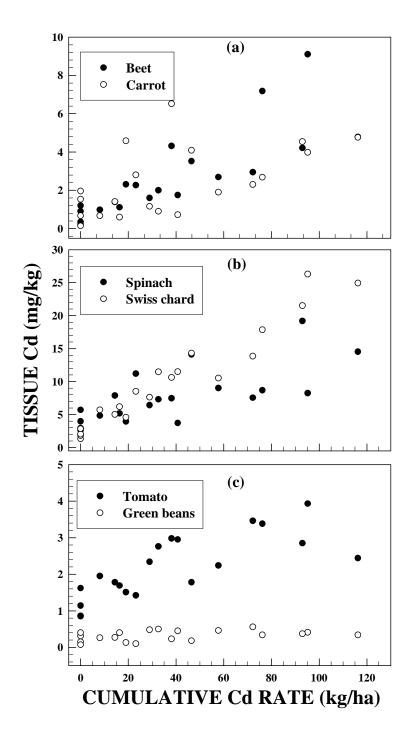
	Beet		Beet Carrot		Sp	Spinach		Swiss Chard		Beans		ato
Metal	Min.	Max.	Min.	Max.	Min.	Max.	Min.	Max.	Min.	Max.	Min.	Max.
Cd	0.08	0.40	0.04	2.79	0.38	3.80	0.07	1.04	0.03	0.22	0.09	4.12
Cr	0.08	0.32	0.04	0.42	0.14	1.57	0.12	0.68	0.08	0.41	0.03	0.40
Cu	5.35	9.82	1.78	12.80	6.80	18.40	9.50	16.80	2.80	17.50	3.80	17.00
Ni	0.04	2.20	0.09	2.10	0.26	6.10	0.55	8.92	0.70	2.44	0.18	1.78
Pb	ND	ND	<0.09	0.24	0.51	2.50	ND	ND	ND	ND	0.04	0.40
Zn	20.2	35.4	5.9	37.4	27.2	148.0	21.5	113.0	26.5	41.7	12.6	33.0

TABLE 7: RANGE OF TRACE METAL CONCENTRATIONS IN SAMPLES OF SIX VEGETABLES COLLECTED IN THE DISTRICT'S NATIONAL TRACE METAL SURVEY $^{\rm 1}$

¹ Data from Feeney et al. (1984).

ND = No data.

FIGURE 3: EFFECT OF CUMULATIVE Cd LOADING ON CONCENTRATION OF Cd IN EDIBLE TISSUE OF VEGETABLES



Chromium. In most years there was no effect (P>0.05) of Nu Earth rate on concentrations of Cr in vegetables (<u>Table 8</u>). In 1977, Cr concentration in Swiss chard increased with Nu Earth rate. Nu Earth rate was also significant in beet (1980), carrot (1980), spinach (1977 to 1979), green beans (1981), and tomato (1980), but the effect showed no consistent trends.

Except in tomato, the apparent effect of cumulative Cr loading on tissue Cr concentrations are attributed mostly to year-to-year fluctuation, because the data showed no trend and Cr in the Control Treatment fluctuated also. Generally, concentrations were lowest in 1978 and 1979 and highest concentrations were in 1980 and 1981. Tomato tissue Cr concentration tended to increase with cumulative Cr loading in all treatments. Since this trend also existed in the Control, the increasing trend in the Nu Earth amended plots is most likely due to factors other than the cumulative Cr additions during the period.

Mean Cr concentrations in the vegetables during the period 1977 to 1981 were generally in the order: spinach > Swiss chard > tomato > carrot > beet = green beans (<u>Table 8</u>). For all treatments (including the control), mean tissue Cr concentrations for all vegetables were higher than maximum concentrations observed in the District's National Survey (<u>Table 7</u>).

Copper. In the first year (1977), Nu Earth application significantly (P<0.05) increased Cu only in the leafy vegetables (<u>Table 9</u>). In all years, the effect of Nu Earth on Cu in carrot, green beans and tomato was slight and mostly insignificant (P>0.05). The relationship between cumulative Cu loading and tissue Cu concentrations tended to be linear for beet, and there was no trend for the other vegetables (<u>Figure 4</u>).

The highest Cu concentrations in the vegetables occurred mostly in 1977 and 1981. There was very little effect of cumulative Cu loading on vegetable tissue Cu concentrations. Changes in Cu concentrations that occurred during this period are attributed mostly to year-to-year variability because there were annual fluctuations in Cu concentration in the control treatment. Except in the 20 to 100 Mg/ha Nu Earth rates for beets and all treatments for Swiss chard, mean Cu concentrations in the vegetables during the period (<u>Table 9</u>) were lower than maximum concentrations found in the District's National Survey of Trace Metals in Vegetables (<u>Table 7</u>).

Nickel. The effect of annual application of Nu Earth on Ni concentration in all vegetables was significant (P < 0.05) (<u>Table 10</u>). Ni concentrations fluctuated with time, but tended to increase with Nu Earth rate. The fluctuations in Ni concentrations over time are attributed mostly to year-to-year variability, because Ni concentrations in the control treatment fluctuated during the period. The effect of cumulative Ni loading on tissue Ni concentrations tended to be linear for beet, carrot, Swiss chard, and tomato (<u>Figure 5</u>) and there was no trend for the other vegetables.

Mean Ni concentrations among vegetables during 1977 to 1981 were generally in the order: green beans > Swiss chard > spinach > tomato > carrot > beet. Except in carrot (100 Mg/ha Nu Earth), green beans (above 20 Mg/ha Nu Earth), and tomato (above 40 Mg/ha Nu Earth),

			u Earth Rat	τ υ ,			2
Year	0	20	40	80	100	Mean	\mathbf{P}^2
				Beet -			
1977	0.14	0.16	0.13	0.16	0.25	0.20	0.3571
1978	0.09	0.07	0.09	0.07	0.11	0.09	0.0660
1979	0.23	0.80	0.51	0.58	0.60	0.54	0.1782
1980	2.77	3.47	2.86	3.06	2.70	2.97	0.0272
1981	0.13	0.15	0.13	0.19	0.25	0.17	0.8889
Mean	0.67	0.96	0.74	0.81	0.78	0.79	
\mathbf{P}^2	0.0001	0.0001	0.0000	0.0001	0.0002		
				Carrot			
1977	0.14	0.14	0.07	0.12	0.14	0.12	0.0686
1978	0.02	0.03	0.01	0.02	0.01	0.02	0.4291
1979	1.74	1.77	1.81	1.49	1.90	1.74	0.6967
1980	3.07	4.78	4.30	2.43	4.20	3.76	0.0428
1981	0.30	0.33	0.51	0.48	0.40	0.40	0.6606
Mean	1.05	1.41	1.34	0.91	1.33	1.21	
\mathbf{P}^2	0.0001	0.0001	0.0000	0.0001	0.0002		
				Swiss Ch	ard		
1977	0.49	0.58	0.59	1.05	1.62	0.87	0.0019
1978	1.75	1.76	1.65	1.79	1.90	1.77	0.6918
1979	0.55	0.79	0.71	0.69	0.66	0.68	0.2440
1980	3.02	2.97	3.16	2.90	3.73	3.16	0.8678
1981	0.63	0.74	0.58	0.57	0.92	0.69	0.1056
Mean	1.29	1.37	1.34	1.40	1.77	1.43	
\mathbf{P}^2	0.0001	0.0001	0.0002	0.0001	0.0005		
				Spinach	۱		
1977	1.04	1.37	1.88	2.99	1.76	1.81	0.0157
1978	0.35	1.13	0.73	0.47	0.63	0.66	0.0051
1979	1.02	2.03	1.84	2.60	2.91	2.08	0.0074
1980	2.54	1.17		1.52	3.63	2.50	0.4711
1981	3.10	3.77	3.35	3.52	3.18	3.38	0.9960
Mean	1.61	1.89		2.22	2.42	2.09	0.7700
P^2	0.0011	0.1016	0.0082	0.0043	0.0018	,	

TABLE 8: EFFECT OF NU EARTH APPLICATIONON ON Cr CONCENTRATION
(mg/kg) IN EDIBLE TISSUE OF VEGETABLES

		Annual N	u Earth Rat	te (Mg/ha)			
Year	0	20	40	80	100	Mean	\mathbf{P}^2
				- Green Bea	ins		
1977	0.78	1.20	0.96	1.58	0.63	1.03	0.4320
1978	0.11	0.22	0.24	0.14	0.10	0.16	0.4795
1979	1.66	0.59	0.94	1.42	0.75	1.07	0.3961
1980	1.07	1.01	1.11	1.19	1.43	1.16	0.4632
1981	0.05	0.07	0.31	0.09	0.08	0.12	0.0206
Mean	0.74	0.62	0.71	0.88	0.60	0.71	
\mathbf{P}^2	0.0002	0.0006	0.0028	0.0003	0.0001		
				Tomato			
1977	0.12	0.09	0.09	0.13	0.05	0.10	0.5400
1978	0.01	0.02	0.02	0.10	0.01	0.03	0.0989
1979	1.69	1.72	1.64	1.70	1.15	1.58	0.1515
1980	2.82	2.12	1.35	2.15	2.46	2.18	0.0345
1981	3.16	2.95	2.89	2.82	2.95	2.96	0.9277
Mean	1.56	1.38	1.20	1.38	1.32	1.37	
\mathbf{P}^2	0.0001	0.0001	0.0000	0.0002	0.0001		

TABLE 8 (Continued): EFFECT OF NU EARTH APPLICATION ON Cr CONCENTRATION (mg/kg) IN EDIBLE TISSUE OF VEGETABLES

¹Values for 1980 and 1981 represent data from plots receiving Nu Earth from 1977 to 1981. ²P = Significance probability of Nu Earth rate or Year.

		Annual N	Vu Earth Ra	te (Mg/ha)			
Year	0	20	40	80	100	Mean	\mathbf{P}^2
				Beet -			
1977	8.91	9.31	9.91	10.4	12.0	10.1	0.0676
1978	8.61	9.64	9.75	10.8	11.1	9.97	0.0039
1979	8.78	10.5	9.82	11.1	12.8	10.6	0.0031
1980	8.44	10.4	10.4	14.9	16.5	12.1	0.0001
1981	10.4	11.5	10.5	12.1	12.1	11.3	0.0020
Mean	9.02		10.1	11.9	12.9	10.8	
\mathbf{P}^2	0.0125	0.0718	0.4823	0.0002	0.0000		
				Carrot			
1977	8.61	5.93	7.53	7.41	7.82	7.46	0.0697
1978	8.54	8.48	8.94	10.6	10.2	9.36	0.0000
1979	5.37	5.96	5.83	5.91	6.51	5.92	0.3901
1980	6.33	7.53	9.02	5.12	6.64	6.93	0.0438
1981	7.61	9.70			9.99	9.35	0.0026
Mean	7.29	7.52	8.10	7.88	8.23	7.80	
\mathbf{P}^2	0.0035			0.0003			
				- Swiss Cha	rd		
1977	21.1	28.8	35.6	26.8	32.4	28.9	0.0000
1978	18.2	19.3	19.6	22.8	24.3	20.9	0.0000
1979	14.8	13.0	16.0	19.3	18.2	16.9	0.0447
1980	17.1	17.4	19.9	20.4	29.3	20.8	0.0032
1981	22.3	25.4	27.7	31.3	33.0	28.0	0.0000
Mean	16.9	18.5	21.6	21.8	24.7	20.7	
\mathbf{P}^2	0.0019	0.0000	0.0001	0.0000	0.0003		
				Spinach			
1977					18.2		
1978	13.4	15.7	15.3	14.6	14.8	14.8	0.0193
	8.37						0.0010
1980		11.1					
1981		15.2					0.1498
Mean			14.8		14.7	14.1	
P^2	0.0002	0.0007	0.0000	0.0000	0.0001		

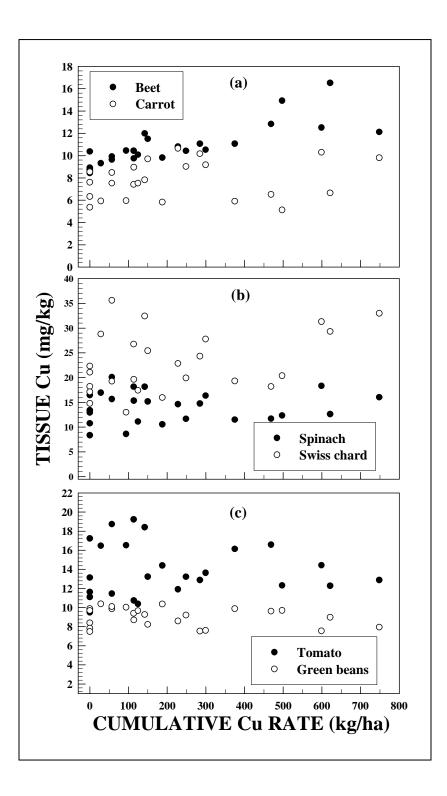
TABLE 9: EFFECT OF NU EARTH APPLICATION ON Cu CONCENTRATION (mg/kg) IN EDIBLE TISSUE OF VEGETABLES

Year	0	20	40	80	100	Mean	P^2
1 Cui	0	20	-10	00	100	wiedh	1
				Green Bear	ns		
1977	9.88	10.4	9.87	9.40	9.27	9.76	0.4390
1978	8.40	10.1	8.70	8.59	7.53	8.66	0.0138
1979	9.69	10.0	10.4	9.88	9.61	9.91	0.7720
1980	7.83	9.67	9.21	9.70	8.98	9.08	0.0022
1981	7.49	8.23	7.61	10.4	7.94	8.33	0.1525
Mean	8.66	9.68	9.15	9.59	8.67	9.15	
\mathbf{P}^2	0.0078	0.0060	0.0012	0.0017	0.0011		
				Tomato			
1977	16.8	16.5	18.7	25.6	18.4	19.2	0.2258
1978	9.50	11.5	10.7	11.9	12.9	11.3	0.0004
1979	13.1	16.5	14.4	16.1	16.6	15.4	0.0022
1980	11.1	10.4	13.2	12.3	11.4	11.7	0.1495
1981	12.4	13.2	13.6	14.4	12.9	13.3	0.0120
Mean	12.6	13.6	14.1	16.1	14.4	14.2	
\mathbf{P}^2	0.0000	0.0001	0.0001	0.0000	0.0002		

TABLE 9 (Continued): EFFECT OF NU EARTH APPLICATION ON Cu CONCENTRATION (mg/kg) IN EDIBLE TISSUE OF VEGETABLES

¹Values for 1980 and 1981 represent data from plots receiving Nu Earth from 1977 to 1981. ²P = Significance probability of Nu Earth rate or Year.

FIGURE 4: EFFECT OF CUMULATIVE Cu LOADING ON CONCENTRATION OF Cu IN EDIBLE TISSUE OF VEGETABLES



			u Earth Rat				2
Year	0	20	40	80	100	Mean	\mathbb{P}^2
				Beet -			
1977	0.38	0.58	0.57	1.08	1.56	0.84	0.0001
1978	0.34	0.38	0.51	0.76	0.85	0.57	0.0003
1979	0.46	0.54	0.90	1.37	1.52	0.96	0.0005
1980	0.53	0.47	0.78	1.28	2.84	1.18	0.0000
1981	0.32	0.50	1.66	1.51	1.47	1.09	0.0001
Mean	0.41	0.49	0.88	1.20	1.65	0.93	
\mathbf{P}^2	0.2885	0.0651	0.0075	0.0092	0.0002		
				Carrot -			
1977	1.10	0.92	1.46	1.69	2.71	1.58	0.0027
1978	0.64	0.64	1.47	1.63	1.96	1.27	0.0004
1979	0.03	0.36	0.52	1.43	2.05	0.88	0.0002
1980	0.81	0.99	0.98	0.61	3.07	1.29	0.0053
1981	0.12	0.41	0.92	1.92	2.88	1.25	0.000
Mean	0.54	0.66	1.07	1.46	2.54	1.25	
\mathbf{P}^2	0.0009	0.0000	0.0008	0.0030	0.2082		
				- Swiss Cha	rd		
1977	- 1.89	1.34	1.69	2.79	3.99	2.34	0.0018
1978	2.28	2.45	2.99	3.60	4.97	3.26	0.0010
1979	0.80	0.86	1.56	2.20	2.70	1.63	0.0002
1980	3.31	2.07	3.59	4.22	4.90	3.62	0.001
1981	7.31	6.99	7.35	7.62	8.13	7.48	0.000
Mean	3.12	2.74	3.44	4.09	4.94	3.66	0.000
P^2	0.0018	0.0002	0.0001	0.0005	0.03	5.00	
-							
1977	5.59	5.91		9.33			0.0038
	1.48						
1979	0.90	0.96	1.04	1.71	1.65	1.25	0.002
1980	1.17	0.55	2.77	2.13	7.77	2.88	0.0150
1981	0.89	1.40	2.18	3.04	3.86	2.27	0.0002
Mean	2.00	2.05	3.54	3.62	4.86	3.21	
P^2	0.0010	0.0030	0.0032	0.0004	0.0012		

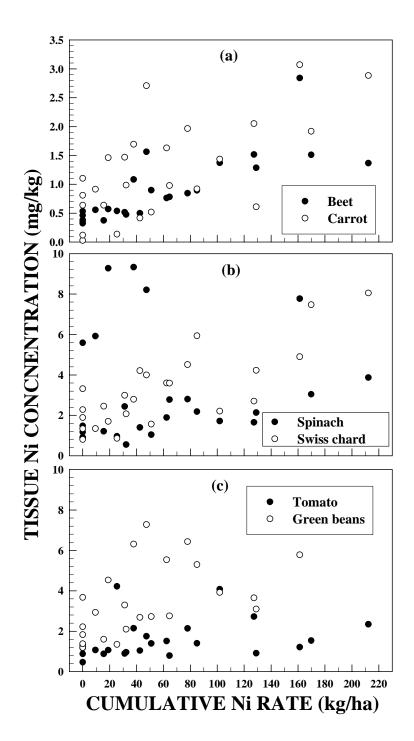
TABLE 10: EFFECT OF NU EARTH APPLICATIONON Ni CONCENTRATION (mg/kg) IN EDIBLE TISSUE OF VEGETABLES

		Annual N	Ju Earth Ra	te (Mg/ha)			
Year	0	20	40	80	100	Mean	\mathbf{P}^2
				Green Bea	ans		
1977	2.22	2.93	4.53	6.31	7.27	4.65	0.0001
1978	1.18	1.60	3.29	5.53	6.43	3.61	0.0000
1979	3.67	1.34	2.72	3.92	3.65	3.06	0.0363
1980	1.83	2.10	2.75	3.09	5.78	3.11	0.0001
1981	1.38	2.68	5.30	11.1	12.2	6.53	0.0000
Mean	2.06	2.13	3.72	5.99	7.07	4.19	
\mathbf{P}^2	0.0062	0.0001	0.0000	0.0002	0.0001		
				Tomato)		
1977	1.71	1.31	1.06	2.15	1.75	1.60	0.0003
1978	0.46	0.88	1.02	1.50	2.14	1.20	0.0002
1979	1.17	4.22	4.09	4.08	2.72	3.25	0.0054
1980	0.88	0.96	0.79	0.90	1.21	0.95	0.0260
1981	1.32	1.04	1.40	1.53	2.34	1.52	0.0254
Mean	1.11	1.68	1.67	2.03	2.03	1.70	
\mathbf{P}^2	0.0116	0.0274	0.0000	0.0004	0.0000		

TABLE 10 (Continued): EFFECT OF NU EARTH APPLICATION ON NI CONCENTRATION (mg/kg) IN EDIBLE TISSUE OF VEGETABLES

¹Values for 1980 and 1981 represent data from plots receiving Nu Earth from 1977 to 1981. ²P = Significance probability of Nu Earth rate or Year.

FIGURE 5: EFFECT OF CUMULATIVE Ni LOADING ON CONCENTRATION OF Ni IN EDIBLE TISSUE OF VEGETABLES



mean Ni concentrations in the vegetables during the period were lower than maximum concentrations observed in the District's National Survey of Trace Metals in Vegetables (<u>Table 7</u>).

Lead. In most years there was no effect of Nu Earth rate on concentrations of Pb in vegetables (<u>Table 11</u>). The effect of cumulative Pb loading showed no specific trend and appeared to be due mostly to year-to-year variability in tissue Pb concentrations.

Generally, mean tissue Pb concentrations during the period annual applications were in the order: Swiss chard = spinach > beet = carrot = green beans = tomato. Except at the 100 Mg/ha Nu Earth rate for spinach, tissue Pb concentrations were lower than maximum concentrations observed for carrot and spinach in the District's National Survey of Trace Metals in Vegetables (<u>Table 7</u>). No results for the other vegetables are available in the District's National Survey (Feeney et al., 1984).

Zinc. In all years, concentrations of Zn in vegetable tissue increased with Nu Earth rate, except in spinach (1981), green beans (1977 and 1978), and tomato (1977) (<u>Table 12</u>). In all vegetables, tissue Zn concentrations changed significantly (P<0.05) with cumulative Zn loading in all treatments. The effect of cumulative Zn loading on tissue Zn concentrations tended to be linear for beet (up to approximately, 1,200 kg/ha), and carrot. There was no specific trend for the other vegetables (<u>Figure 6</u>).

Mean Zn concentrations among vegetables during the period 1977 to 1981 were generally in the order spinach > Swiss chard >> Beet > green beans = tomato > carrot. Compared to maximum concentrations found in the District's National Survey of Trace Metals in Vegetables (<u>Table 7</u>), mean Zn concentrations during the 5-year annual application period were higher for beet (20 to 100 Mg/ha Nu Earth rate), Swiss chard (20 to 100 Mg/ha Nu Earth rate), spinach (0 to 100 Mg/ha Nu Earth rate), and tomato (40 to 100 Mg/ha Nu Earth rate) from the Nu Earth garden.

Residual Effect of Nu Earth on Soil Organic Carbon and Extractable Trace Metals Concentrations in Soil and Vegetables

Generally, the trends in extractable metals in soil and metal concentrations in edible tissues of vegetables after termination of Nu Earth application were similar for the subplots receiving Nu Earth for three years and subplots receiving Nu Earth for five years, but concentrations in the latter were higher. Therefore only data for the five subplots receiving Nu Earth for five years are discussed. The cumulative Nu Earth loading rates in these subplots were 0, 100, 200, 400, and 500 Mg/ha. For evaluating the residual effect of Nu Earth on extractable metals in soil and metal concentrations in edible tissue of vegetables, the mean of concentrations for the period 1980 to 1981 were used as concentrations at the time of termination of Nu Earth application. Data after that period were analyzed as means for each of four periods, 1982 to 1985, 1986 to 1989, 1990 to 1993, and 1994 to 1998. This approach was used to minimize bias due to year-toyear variability in concentrations observed for some treatments. This year-to-year variability was also observed during the period of Nu Earth application <u>Table 6</u> and <u>Tables 8</u> through <u>12</u>.

		Annual N	Vu Earth Rat	te (Mg/ha)			
Year	0	20	40	80	100	Mean	\mathbf{P}^2
				Beet -			
1977	0.17	0.20	0.19	0.16	0.55	0.25	0.2196
1978	0.04	0.06	0.04	0.05	0.06	0.05	0.5809
1979	0.16	0.17	0.34	0.18	0.26	0.22	0.1053
1980	0.45	0.53	0.44	0.47	0.67	0.51	0.1339
1981	0.05	< 0.04	0.15	0.35	0.07	0.12	0.0031
Mean	0.17	0.19	0.23	0.24	0.32	0.23	
\mathbf{P}^2	0.0001	0.0002	0.0005	0.0108	0.0006		
1977	0.60	0.15	0.34	0.31	0.45	0.37	0.0006
1978	0.06	0.07	< 0.01	0.01	0.02	0.03	0.2778
1979	0.03	0.35	0.08	0.29	0.20	0.19	0.0204
1980	0.30	0.45	0.64	0.36		0.41	0.6870
1981	0.08	< 0.08	< 0.08	0.08	0.08	0.08	0.4747
Mean	0.20	0.20	0.21	0.20	0.21	0.20	
\mathbf{P}^2	0.0009	0.0007	0.0001	0.0011	0.0015		
				Swiss Ch	ard		
1977	ND	ND	ND	ND	ND	ND	
1978	3.07	3.60	3.37	3.16	3.55	3.35	0.5783
1979	1.49	1.43	1.12	1.49	0.95	1.29	0.0040
1980	2.41	1.90	2.24	2.43	3.04	2.40	0.1627
1981	1.63	2.38	1.45	1.64	1.64	1.75	0.1010
Mean	2.15	2.33	2.04	2.18	2.29	2.20	
\mathbf{P}^2	0.0018	0.0002	0.0001	0.0005	0.0003		
				Spinacl	า		
1977		4.49			3.84		0.0092
1978	1.90	3.02	2.49			2.42	0.002
1979	1.35	1.52	1.58	1.77		1.57	
1980	2.59	1.57	2.19			2.44	
1981	3.05		1.59			1.79	
Mean	2.34	2.32	2.22	2.40	2.51	2.36	
\mathbf{P}^2	0.0002	0.0003	0.0001		0.0002		

TABLE 11: EFFECT OF NU EARTH APPLICATIONON Pb CONCENTRATION (mg/kg) IN EDIBLE TISSUE OF VEGETABLES

		Annual N	Ju Earth Ra	te (Mg/ha)			
Year	0	20	40	80	100	Mean	\mathbf{P}^2
				Green B	eans		
1977	0.17	0.13	0.16	0.11	0.37	0.19	0.0573
1978	0.25	0.29	0.35	0.26	0.26	0.28	0.2645
1979	0.19	0.40	0.18	0.41	0.17	0.27	0.2145
1980	< 0.8	< 0.8	< 0.8	< 0.8	< 0.8	< 0.8	NA
1981	0.40	0.20	0.27	0.13	0.20	0.24	0.8002
Mean	0.20	0.20	0.19	0.18	0.20	0.20	
\mathbf{P}^2	0.0403	0.0067	0.0054	0.0013	0.0278		
				Tomat	0		
1977	ND	ND	ND	ND	ND	ND	
1978	0.14	0.27	0.21	0.23	0.15	0.20	0.6978
1979	0.16	0.49	0.63	0.36	0.50	0.43	0.2189
1980	0.18	0.15	0.02	0.13	0.36	0.17	0.0982
1981	< 0.01	0.08	< 0.01	< 0.01	0.01	0.02	1.0000
Mean	0.12	0.25	0.21	0.18	0.26	0.20	
\mathbf{P}^2	0.0253	0.0727	0.0111	0.0020	0.0372		

TABLE 11 (Continued): EFFECT OF NU EARTH APPLICATION ON Pb CONCENTRATION (mg/kg) IN EDIBLE TISSUE OF VEGETABLES

¹Values for 1980 and 1981 represent data from plots receiving Nu Earth from 1977 to 1981. ²P = Significance probability of Nu Earth rate or Year.

NA = Not applicable.

			lu Earth Ra	ζŲ,			2
Year	0	20	40	80	100	Mean	\mathbf{P}^2
				Beet	t		
1977	28.9	36.9	43.6	51.1	60.2	44.13	0.0000
1978	25.0	35.8	43.6	58.7	62.5	45.10	0.0006
1979	34.0	45.0	61.7	93.0	89.7	64.67	0.0000
1980	32.2	56.8		108	161	84.99	0.0000
1981	36.3	46.3	59.0	71.3	77.9	58.16	0.0000
Mean	31.3	44.2	54.9	76.5	90.2	59.41	
\mathbf{P}^2	0.0027	0.0000	0.0013	0.0002	0.0001		
)t		
1977	26.1	20.8	27.4	28.8	30.0	26.6	0.0010
1978	21.2	20.8	24.8	26.7	26.1	23.9	0.0146
1979	20.1	22.3	24.7	26.8	31.6	25.1	0.0055
1980	23.4	38.5	35.4	30.1	40.3	33.5	0.0040
1981	23.1	26.8	31.5	36.7	41.4	31.9	0.0000
Mean	22.8	25.8	28.8	29.8	33.9	28.2	
\mathbf{P}^2	0.0051	0.0044	0.0218	0.0000	0.0011		
				Swiss C	hard		
1977	77.7	148	234	141	225	165	0.0000
1978	52.7	159	224	289	374	220	0.0001
1979	42.9	80.7	110	148	189	114	0.0001
1980	57.5	86.8	178	264	374	192	0.0001
1981	46.5	90.6	132	227	235	146	0.0000
Mean	55.4	113	175	214	279	167	
\mathbf{P}^2	0.0004	0.0001	0.0035	0.0001	0.0005		
				Spinac	ch		
1977	167		523		368		0.0002
1978	167	243	229	233	251	225	0.0012
1979	92.2	162	166	193	194	162	0.0065
1980	125	121	142	138	152	136	0.0242
1981	263	305	313	362	317	312	0.3240
Mean	162	208	212	232	228	208	
\mathbf{P}^2	0.0003	0.0000	0.0005	0.0000	0.0052		

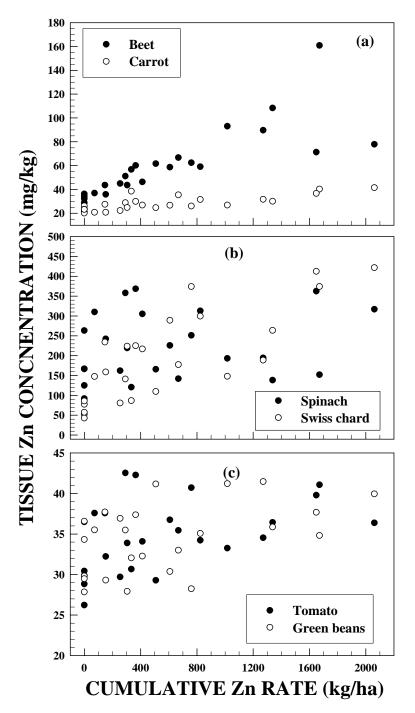
TABLE 12: EFFECT OF NU EARTH APPLICATIONON Zn CONCENTRATION (mg/kg) IN EDIBLE TISSUE OF VEGETABLES

		Annual N	u Earth Rat	e (Mg/ha)			
Year	0	20	40	80	100	Mean	\mathbf{P}^2
				Green Be	ans		
1977	34.3	35.5	37.7	35.5	37.4	36.1	0.3514
1978	27.9	29.3	27.9	30.4	28.2	28.7	0.1429
1979	36.6	36.9	41.2	41.2	41.5	39.5	0.0140
1980	29.8	32.0	33.0	35.8	34.8	33.1	0.0039
1981	29.5	32.3	35.1	37.7	40.0	34.9	0.0000
Mean	31.6	33.2	35.0	36.1	36.4	34.5	
\mathbf{P}^2	0.0000	0.0001	0.0000	0.0012	0.0001		
				Tomato			
1977	36.5	37.6	37.6	42.5	42.3	39.3	0.3240
1978	29.9	32.2	33.9	36.8	40.7	34.7	0.0000
1979	26.2	29.7	29.3	33.3	34.5	30.6	0.0085
1980	28.8	30.7	35.5	36.4	41.1	34.5	0.0016
1981	28.1	34.1	34.2	39.8	36.4	34.5	0.0312
Mean	29.9	32.8	34.1	37.8	39.0	34.7	
\mathbf{P}^2	0.0000	0.3310	0.0356	0.0072	0.0003		

TABLE 12 (Continued): EFFECT OF NU EARTH APPLICATION ON Zn CONCENTRATION (mg/kg) IN EDIBLE TISSUE OF VEGETABLES

¹Values for 1980 and 1981 represent data from plots receiving Nu Earth from 1977 to 1981. ²P = Significance probability of Nu Earth rate or Year.

FIGURE 6: EFFECT OF CUMULATIVE Zn LOADING ON CONCENTRATION OF Zn IN EDIBLE TISSUE OF VEGETABLES



Analysis of variance was conducted to test the hypothesis that the mean concentrations in periods following termination of Nu Earth application were different from the mean concentrations for the period 1980 to 1981.

Soil Organic Carbon Content. The effect of soil OC on concentrations of trace metals in edible tissue of vegetables was evaluated because soil OC is thought to be a major factor that controls solubility and bioavailability of metals in soils. After termination of biosolids application, the OC added to the soil will degrade and organically bound metals will be released into soluble forms or be bound on inorganic soil components. Soil samples collected before 1981 were not analyzed for OC, so OC content in 1981 is used to represent the level of OC at termination of Nu Earth application.

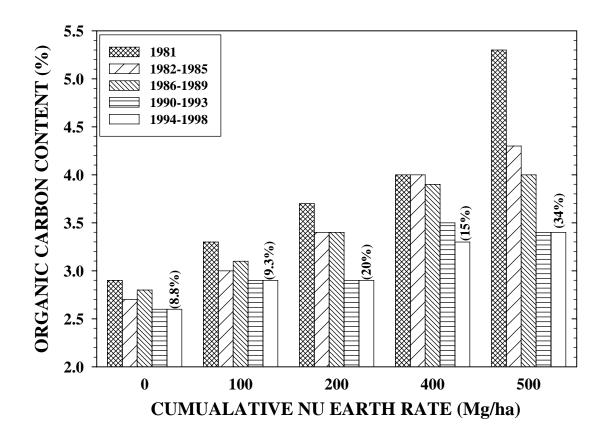
Soil OC content occurring after termination of Nu Earth application is summarized as mean concentrations over the five time periods discussed above and is presented in Figure 7. At all Nu Earth rates, soil OC content decreased with time after termination of Nu Earth application. Organic matter in the Control also tended to decrease with time. At the end of the study, soil OC in the Nu Earth treatments decreased by 9.3 to 34 percent of 1981 levels but was always higher than OC in the Control. According to the proposed "Time Bomb Hypothesis" (McBride, 1995), this significant loss of Nu Earth OC should result in the release of heavy metals associated with the OC in Nu Earth, which should increase bioavailability and uptake of these metals in plants. If this hypothesis is correct, it would lead to increased exposure to trace metals in vegetables consumed from home gardens fertilized with Nu Earth in the Chicagoland area. We therefore utilized data from this study to evaluate this hypothesis.

Extractable Trace Metal Concentrations in Soil. Because the plots used in this study were relatively small, cross-contamination between the plots during preparation of the plots for planting vegetables were likely. In 1990, the three replications of the control plots and the 500 Mg/ha cumulative Nu Earth rate were sampled at 15 locations within the plots and analyzed for 0.1 M HCl extractable metals. A summary of the variability in extractable metal concentrations is presented in <u>Tables 13</u> and <u>14</u>.

The extractable Cd and Zn concentrations in soil can be used as a marker to evaluate the possibility of cross contamination between the plots because the concentrations of these metals in Nu Earth are relatively high compared to the extractable concentrations in unamended soil. Within each replication, the ranges of Cd and Zn concentrations were relatively large and the highest coefficients of variation observed were 44 and 20 percent for Cd and Zn, respectively. The relationship between spatial variability in extractable metal concentrations within the plots (data not shown) and Nu Earth rate in proximal plots indicate that the effect of cross contamination on metal concentrations in the plots was very small. The data for the plots receiving the 500 Mg/ha Nu Earth rate also indicate that dilution of extractable metal concentrations by cross contamination from proximal plots receiving lower Nu Earth rates was minimal (Table 14).

Mean concentrations of 0.1 M HCl-extractable metals in soil over five averaging periods for the subplots that received five annual Nu Earth applications are presented in <u>Table 15</u>.

FIGURE 7: RESIDUAL EFFECT OF NU EARTH ON SOIL ORGANIC CARBON CONTENT FOLLOWING ANNUAL NU EARTH TREATMENTS APPLIED ANNUALLY DURING 1977 THROUGH 1981 (VALUES IN PARENTHESES REPRESENT REDUCTION IN 1994–1998 COMPARED TO 1981)



	Cd	Cr	Cu	Ni	Pb	Zn
			R	.ep 1		
Mean	4.5	44	42	30	48	127
Minimum	3.2	35	34	25	36	97
Maximum	5.6	55	57	39	78	172
CV^1	14	9	13	9	20	16
			R	.ep 2		
Mean	3.0	45	37	26	31	111
Minimum	1.1	31	29	18	24	88
Maximum	5.6	66	47	31	42	144
CV^1	40	21	14	15	14	14
			R	.ep 3		
Mean	4.3	49	42	25	44	124
Minimum	1.3	35	32	19	33	95
Maximum	8.1	80	57	30	64	187
CV^1	44	25	16	10	17	18

TABLE 13: VARIABILITY IN CONCENTRATION OF 0.1 M HC1-EXTRACTABLEMETALS (mg/kg) IN SOIL AT FIFTEEN LOCATIONS OF EACH REPLICATE OF
CONTROL PLOTS IN 1990

 $^{1}CV = Coefficient of variation.$

	Cd	Cr	Cu	Ni	Pb	Zn
			R	ep 1		
Mean	22.7	252	160	60	155	456
Minimum	13.0	131	106	39	112	311
Maximum	31.0	359	215	81	381	600
CV^1	21	24	17	15	30	15
			R	ep 2		
Mean	20.1	268	154	55	130	454
Minimum	12.0	183	121	43	94	346
Maximum	30.0	399	215	69	178	642
CV^1	25	23	19	11	20	20
			R	ep 3		
Mean	22.3	272	156	56	136	480
Minimum	10.4	145	98	38	79	306
Maximum	32.9	400	209	76	197	635
CV^1	31	26	21	19	25	20

TABLE 14: VARIABILITY IN CONCENTRATION OF 0.1 M HC1-EXTRACTABLEMETALS (mg/kg) IN SOIL AT FIFTEEN LOCATIONS OF EACH REPLICATE OF THE 500Mg/ha CUMULATIVE NU EARTH RATE IN 1990

 $^{1}CV = Coefficient of variation.$

Years	Cumulative Nu Earth Rate (Mg/ha)								
Averaged ¹	0	100	200	400	500				
			Cd						
1980-1981	3.0	12.0	19.9	38.1	39.8				
1982-1985	3.4 ns^3	10.9 ns	18.9 ns	33.9 ns	36.7 ns				
1986-1989	5.9 **	11.5 ns	15.8 **	30.1 *	32.6 **				
1990-1993	5.7 ns	8.9 **	12.9 **	22.6 **	22.7 **				
1994-1998	4.0 ** (-33) ⁴	9.4 ns (22)	13.4 ** (33)	22.2 ** (42)	23.9 ** (40)				
			Cr -						
1980-1981	4.2	17.4	34.7	61.9	81.9				
1982-1985	4.0 ns	15.5 ns	30.5 ns	58.0 ns	66.0 ns				
1986-1989	9.1 **	19.1 ns	28.0 *	58.4 ns	67.2 *				
1990-1993	4.4 ns	13.5 ns	19.0 **	37.6 **	38.7 **				
1994-1998	4.5 ns	12.5 ns	17.7 **	33.1 **	35.2 **				
	(-8)	(28)	(49)	(47)	(57)				
			Cu -						
1980-1981	13.9	49.9	104.4	154.4	171.2				
1982-1985	19.0 ns	56.9 ns	95.0 ns	180.2 ns	190.6 ns				
1986-1989	28.8 **	63.7 ns	87.2 **	174.3 ns	185.7 ns				
1990-1993	20.9 ns	48.6 ns	66.7 **	122.8 ns	121.2 ns				

TABLE 15: RESIDUAL EFFECT OF NU EARTH APPLICATION ON 0.1 M HCI-EXTRACTABLEMETALS (mg/kg) IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

Years		C	Cumulative Nu Earth H	Rate (Mg/ha)	
Averaged ¹	0	100	200	400	500
1994-1998	22.8 **	50.1 ns	67.9 **	114.3 ns	119.0 ns
	(-63)	(0)	(35)	(26)	(30)
1980-1981	9.0	17.4	31.0	42.9	56.9
1982-1985	8.6 ns	15.0 ns	26.1 *	44.8 ns	47.9 ns
1986-1989	11.3 ns	20.8 ns	35.8 **	42.6 ns	48.9 *
1990-1993	7.5 ns	14.0 ns	18.6 **	30.7 *	30.6 **
1994-1998	6.7 ns	12.1 *	15.5 **	23.2 **	25.1 **
	(25)	(31)	(50)	(46)	(56)
			Pb		
1980-1981	26.3	53.5	97.4	150.2	186.0
1982-1985	23.7 ns	50.3 ns	81.6 *	142.8 ns	148.0 *
1986-1989	33.2 ns	59.4 ns	75.1 **	140.6 ns	150.7 **
1990-1993	23.7 ns	46.2 ns	55.0 **	96.3 **	94.4 **
1994-1998	24.4 ns	46.4 ns	56.0 **	92.5 **	96.0 **
	(7)	(13)	(42)	(38)	(48)
			Zn		
1980-1981	49.3	161.3	356.4	541.0	689.2
1982-1985	51.8 ns	163.5 ns	278.6 ns	532.0 ns	572.8 *

TABLE 15 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON 0.1 M HCI-EXTRACTABLEMETALS (mg/kg) IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

TABLE 15 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON 0.1 M HCI-EXTRACTABLEMETALS (mg/kg) IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

Years	Cumulative Nu Earth Rate (Mg/ha)								
Averaged ¹	0	100	200	400	500				
1986-1989	84.3 **	188.0 ns	262.2 ns	467.1 ns	488.2 **				
1990-1993	67.1 **	149.4 ns	204.7 *	365.9 **	364.8 **				
1994-1998	66.5 **	135.9 ns	183.8 **	301.3 **	328.1 **				
	(-35)	(16)	(48)	(44)	(52)				

Values are average of data for years included in each averaging period. For Cd, 1980 data for the 100, 200, and 400 Mg/ha cumulative Nu Earth rates were excluded.

² Significance probability of the effect of Nu Earth rate on metal concentrations.

³ Notations following values indicate significance probability that values are different from those for the 1980-1981 period. ns = not significant. *, ** = significant at 0.05 and 0.01 probability levels, respectively.

⁴ Numbers in parentheses represent the percent decrease in average concentrations from the 1980-1981 period to the 1994-1998 period.

For Cd, the 1980 data for the 100, 200, and 400 Mg/ha cumulative Nu Earth rates were excluded from the analysis because those values were unexpectedly high (<u>Table 7</u>). Following termination of Nu Earth application, the concentration of extractable metals increased with annual Nu Earth rate (<u>Table 15</u>). Contrary to the "Time Bomb Hypothesis," extractable levels of all metals generally tended to decrease with time following termination of Nu Earth application.

The data show that the decrease in metal concentrations for the 1994 to 1998 period following termination of Nu Earth application was significant mostly at cumulative Nu Earth rates above 100 Mg/ha. The greatest relative decrease in extractable metal concentration occurred in the 500 Mg/ha cumulative Nu Earth treatment for Cr, Ni, Pb, and Zn with reductions of 57, 56, 48, and 52 percent, respectively over the 17-year period. The greatest relative decrease for Cd and Cu occurred in the 400 Mg/ha and 200 Mg/ha cumulative Nu Earth treatments, respectively, with reductions of 40 and 35 percent, respectively over the 17-year residual period.

In the control plots, metal concentrations fluctuated. In some periods concentrations in the control were significantly different from mean concentrations for 1980 and 1981. At the 100 Mg/ha cumulative Nu Earth rate, changes in levels of extractable metals following termination of Nu Earth application were insignificant compared to the 1980 to 1981 levels, except during the 1994 to 1998 period. This indicates that factors that reduce extractability of metals in soils were most likely insignificant at the metal loading rates associated with the five-year application of the 100 Mg/ha cumulative Nu Earth rate.

Concentrations of Trace Metals in Edible Tissue of Vegetables. *Cadmium*. The increase in Cd concentration in edible tissue of vegetables with Nu Earth rate observed during the period of Nu Earth application was also evident during most years following termination of Nu Earth application (P<0.05), except in green beans (<u>Table 16</u>). Except in spinach, tissue Cd generally tended to decrease with time following termination of Nu Earth application. Most instances where decreases were significant compared to the average concentrations in the 1980 to 1981 period were at the 80 and 100 Mg/ha Nu Earth rate. In spinach, Cd concentrations following termination of Nu Earth application served during the 1980 to 1981 period. The changes in concentrations followed no particular trend but fluctuated between each period.

Chromium. In all treatments, Cr concentrations in the tissue of vegetables tended to decrease steadily with time following termination of Nu Earth application (<u>Table 17</u>). The existence of this trend in the control indicates that the reduction in the biosolids amended plots can not be attributed solely to mechanisms that reduce bioavailability of biosolids applied Cr. In the 1994 to 1998 period, Cr concentrations in all Nu earth amended plots were similar to concentrations in the control plots. Similar to the observations for the period during biosolids application, there were no consistent effects of Nu earth rate on Cr concentrations in the tissue of vegetables.

Years		Cu	mulative Nu Earth	n Rate (Mg/ha)		
Averaged ¹	0	100	200	400	500	\mathbf{P}^2
				Beet		
1980-1981	1.05	2.29	3.91	5.69	6.94	0.000
1982-1985	1.13 ns^3	1.95 ns	2.74 **	3.85 ns	5.23 ns	0.000
1986-1989	0.65 **	1.41 **	2.10 **	2.87 **	4.09 **	0.000
1990-1993	0.90 ns	1.76 **	3.28 *	5.12 ns	6.98 ns	0.000
1994-1998	0.84 ns	1.84 *	3.12 *	3.88 *	6.24 ns	0.000
				Carrot		
1980-1981	1.77	3.69	5.06	4.07	4.45	0.000
1982-1985	1.03 **	2.44 *	2.99 *	3.69 ns	4.03 *	0.000
1986-1989	1.63 ns	3.34 ns	4.23 ns	5.22 ns	6.00 **	0.000
1990-1993	1.41 ns	3.13 ns	4.23 ns	5.31 ns	5.33 ns	0.000
1994-1998	1.31 ns	1.44 **	5.15 ns	4.28 ns	2.40 **	0.000
			Sv	viss Chard		
1980-1981	2.44	6.54	12.47	19.67	25.62	0.000
1982-1985	3.27 *	6.79 ns	11.35 ns	14.52 **	17.14 **	0.000
1986-1989	3.31 *	6.88 ns	13.36 ns	13.72 **	13.81 **	0.000
1990-1993	2.40 ns	5.61 ns	8.96 *	15.52 *	14.27 **	0.000
1994-1998	2.75 ns	7.81 ns	9.88 ns	13.83 **	17.71 **	0.000

TABLE 16: RESIDUAL EFFECT OF NU EARTH APPLICATION ON Cd CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

1			100		nulative Nu		· · ·			
Averaged ¹	0		100		200		400		500	\mathbf{P}^2
						S	pinach			
1980-1981	4.46		9.38		11.44		14.98		12.01	0.000
1982-1985	7.81	*	15.88	*	19.16	**	22.12	*	22.37 **	0.000
1986-1989	6.67	*	15.20	*	16.28	ns	21.81	ns	19.20 **	0.000
1990-1993	ND		ND		ND		ND		ND	
1994-1998	8.46	**	17.11	ns	15.27	ns	22.94	*	17.91 *	0.000
						Gre	en Beans -			
1980-1981	0.11		0.12		0.20		0.35		0.38	0.907
1982-1985	0.06	*	0.07	ns	0.17	ns	0.28	ns	0.34 Ns	0.596
1986-1989	0.15	ns	0.26	*	0.16	ns	0.23	*	0.31 ns	0.427
1990-1993	0.02	**	0.13	ns	0.16	ns	0.24	**	0.33 ns	0.086
1994-1998	0.02	**	0.02	**	0.04	**	0.04	**	0.06 **	0.966
						· ′	Fomato			
1980-1981	1.24		1.46		2.38		3.12		3.18	0.000
1982-1985	1.47	ns	1.95	ns	2.32	ns	2.76	ns	2.97 ns	0.000
1986-1989	1.76	*	2.18	**	2.67	ns	2.70	ns	3.29 ns	0.000
1990-1993	1.60	ns	2.12	**	2.44	ns	2.80	ns	3.48 ns	0.000
1994-1998	0.85	ns	1.40	ns	1.84	ns	1.79	**	1.97 **	0.000

TABLE 16 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON Cd CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

TABLE 16 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON Cd CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

Years		Cu	mulative Nu Earth	n Rate (Mg/ha)		
Averaged ¹	0	100	200	400	500	\mathbf{P}^2

 ¹Values are for average of data for years included in each period.
²Significance probability of the effect of Nu Earth rate on metal concentrations.
³ Notations following values indicate significance probability that values are different from those for the 1980-1981 period. ns = not significant. *, ** = significant at 0.05 and 0.01 probability levels, respectively. ND = no data.

Years						
			nulative Nu Earth	Rate (Mg/ha)		
Averaged ¹	0	100	200	400	500	\mathbf{P}^2
				Beet		
1980-1981	1.38	1.81	1.50	1.63	1.48	0.975
1982-1985	2.03 ns^3	2.36 ns	2.21 ns	2.84 ns	3.27 ns	0.081
1986-1989	0.09 **	0.26 **	0.34 ns	0.13 **	0.17 *	0.007
1990-1993	0.13 *	0.13 **	0.23 ns	0.20 *	0.42 ns	0.004
1994-1998	0.14 ns	0.23 ns	0.26 ns	0.22 ns	0.19 *	0.732
			(Carrot		
1980-1981	1.96	2.56	2.03	1.03	1.92	0.574
1982-1985	1.71 ns	1.69 ns	1.77 ns	1.57 *	2.10 ns	0.462
1986-1989	0.43 *	0.55 ns	0.33 **	0.70 ns	0.69 ns	0.003
1990-1993	0.11 **	0.14 **	1.20 ns	0.25 **	0.27 *	0.003
1994-1998	0.40 ns	0.17 *	0.43 ns	0.22 *	0.52 ns	0.380
			Sw	iss Chard		
1980-1981	1.83	2.00	1.87	1.74	2.33	0.865
1982-1985	2.00 ns	2.75 ns	2.36 ns	2.00 ns	1.98 ns	0.156
1986-1989	0.78 *	1.29 ns	1.41 ns	1.24 ns	1.05 ns	0.043
1990-1993	0.62 *	0.73 **	0.84 *	1.13 ns	0.97 *	0.475
1994-1998	0.47 **	0.63 **	0.59 *	0.70 ns	0.71 **	0.670

TABLE 17: RESIDUAL EFFECT OF NU EARTH APPLICATION ON Cr CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

Years				Cum	ulative Nu	ı Earth l	Rate (Mg/	na)		
Averaged ¹	0		100		200		400		500	\mathbf{P}^2
						Spi	nach			
1980-1981	2.82		3.12		3.47		2.72		3.36	0.904
1982-1985	3.24	ns	3.04	ns	3.19	ns	3.50	ns	3.51 ns	0.969
1986-1989	1.56	ns	2.44	ns	3.09	ns	2.15	ns	2.29 ns	0.195
1990-1993	ND		ND		ND		ND		ND	
1994-1998	0.50	*	0.75	*	0.37	**	0.64	**	0.58 **	0.758
						Gree	en Beans -			
1980-1981	0.56		0.54		0.71		0.64		0.75	0.907
1982-1985	1.24	*	1.61	**	2.01	**	1.55	**	1.85 *	0.596
1986-1989	0.10	*	0.30	ns	0.23	*	0.15	ns	0.12 *	0.427
1990-1993	0.23	ns	0.47	ns	0.24	*	0.32	ns	1.16 ns	0.086
1994-1998	0.05	*	0.07	*	0.06	**	0.05	**	0.06 *	0.966
						То	mato			
1980-1981	2.99		2.54		2.12		2.48		2.70	0.113
1982-1985	2.78	ns	3.26	ns	3.24	ns	3.18	ns	3.32 ns	0.938
1986-1989	0.06	**	0.20	**	0.08	**	0.14	**	0.11 **	0.913
1990-1993	0.06	**	< 0.01	**	0.04	**	0.04	**	0.11 **	0.171
1994-1998	0.13	**	0.07	**	0.17	**	0.10	**	0.16 **	0.482

TABLE 17 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON Cr CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

TABLE 17 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON Cr CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

Years		Cu	mulative Nu Earth	n Rate (Mg/ha)		
Averaged ¹	0	100	200	400	500	\mathbf{P}^2

 ¹ Values are for average of data for years included in each period.
² Significance probability of the effect of Nu Earth rate on metal concentrations.
³ Notations following values indicate significance probability that values are different from those for the 1980-1981 period. ns = not significant. *, ** = significant at 0.05 and 0.01 probability levels, respectively. ND = no data.

Copper. Concentrations of Cu in the tissue of vegetables tended to decrease with time following termination of Nu Earth application (<u>Table 18</u>), but did not follow a monotonic trend. After termination of Nu Earth application, the tissue of metal concentrations did not significantly decrease until after the 1982-1985 period for all of the vegetables. Some of the changes in tissue concentrations are attributed mostly to year-to-year variability, because concentrations in the control also changed significantly in some periods compared to the mean concentration for the 1980 to 1981 period.

Nickel. Concentrations of Ni in the tissue of vegetables increased during 1982 to 1985 compared to concentrations in the 1980 to 1981 period (<u>Table 19</u>), then tended to decrease. The increase was not attributed entirely to Nu Earth rate because this trend was also present in the control. Except in carrot and green beans, most of the decreases observed were significant. Similar to the effect of Nu Earth rate observed during the 5 years of Nu Earth application (<u>Table 10</u>) the concentrations of Ni in the vegetables increased (P<0.05) with Nu Earth rate in most periods after application was terminated (<u>Table 19</u>).

Lead. In some periods following termination of Nu Earth application, concentrations of Pb in the tissues of carrot, green bean, and tomato were significantly higher than mean concentrations for the 1980 to 1981 period (<u>Table 20</u>). The changes observed were most likely due to year- to-year variability because concentrations in the control also fluctuated. In most years, there was also no effect (P>0.05) of Nu Earth rate following termination of application. The Pb concentration for beet did not change significantly throughout the entire residual period, while it decreased significantly in Swiss chard and spinach in 1982-1985 and remained significantly lower than during the period 1980-1981.

Zinc. Concentrations of Zn in the tissue of vegetables tended to decrease with time following termination of Nu Earth application (Table 21). In beet and tomato, the decrease in most periods was insignificant. In spinach, concentrations in all treatments, except the control, were highest in the 1982 to 1985 period and then decreased. The high concentrations observed in this period are attributed mostly to the relatively high concentrations in 1983; up to 30 percent above concentrations in 1982 (data not shown). Over time, there were no consistent trends in concentration for carrots, green beans, and Swiss chard. Changes in Zn concentrations were not consistent from time period to time period or among Nu Earth rates. In most years, the Zn concentrations increased significantly (P<0.05) with Nu Earth rate.

Years		Cum	ulative Nu Earth	Rate (Mg/ha)			
Averaged ¹	0	100	200	400	500		\mathbf{P}^2
]	Beet			
1980-1981	9.40	10.85	10.48	13.84	14.32		0.000
1982-1985	8.65 ns^3	9.29 ns	9.23 *	10.49 **	11.83	*	0.007
1986-1989	7.64 **	7.71 **	7.81 *	8.91 **	8.61	**	0.326
1990-1993	6.67 **	5.90 **	7.59 **	8.02 **	7.87	**	0.017
1994-1998	8.98 ns	8.97 **	10.07 ns	10.42 **	9.48	**	0.070
			Ca	arrot			
1980-1981	6.84	8.61	9.11	9.01	8.40		0.034
1982-1985	8.08 ns	7.83 ns	7.71 *	7.98 ns	8.67	ns	0.422
1986-1989	6.32 ns	8.02 ns	6.64 *	9.04 ns	8.03	ns	0.109
1990-1993	5.48 *	7.29 ns	5.97 **	6.84 *	6.96	*	0.254
1994-1998	5.90 ns	5.88 *	7.25 ns	5.95 **	6.13	*	0.546
			Swis	s Chard			
1980-1981	19.73	21.42	23.85	25.84	31.15		0.000
1982-1985	19.03 ns	18.41 ns	21.01 ns	24.39 ns	27.08	ns	0.000
1986-1989	16.61 *	15.46 **	17.37 **	18.15 **	16.51	**	0.197
1990-1993	13.62 **	15.08 **	16.45 **	18.89 *	17.73	**	0.000
1994-1998	14.63 **	17.50 ns	16.74 **	21.86 *	16.11	**	0.000

TABLE 18: RESIDUAL EFFECT OF NU EARTH APPLICATION ON Cu CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

Years		Cur	nulative Nu Earth	Rate (Mg/ha)			
Averaged ¹	0	100	200	400	500		\mathbf{P}^2
			S	pinach			
1980-1981	14.18	13.82	14.48	15.68	14.67		0.633
1982-1985	12.30 ns	13.12 ns	14.17 ns	14.96 ns	14.85	ns	0.001
1986-1989	13.65 ns	14.58 ns	14.13 ns	15.49 ns	16.01	ns	0.000
1990-1993	ND	ND	ND	ND	ND		
1994-1998	10.23 ns	11.77 ns	11.10 **	12.17 *	12.77	ns	0.148
			Gre	een Beans			
1980-1981	7.66	8.95	8.41	8.73	8.46		0.030
1982-1985	8.61 **	8.38 ns	8.62 ns	8.52 ns	8.71	ns	0.900
1986-1989	7.57 ns	8.23 ns	7.58 ns	8.75 ns	8.20	ns	0.009
1990-1993	7.90 ns	8.43 ns	8.43 ns	8.59 ns	8.99	ns	0.201
1994-1998	8.39 ns	9.45 ns	9.99 ns	9.55 ns	10.08	ns	0.560
			T	'omato			
1980-1981	11.31	11.81	13.42	13.38	12.61		0.114
1982-1985	9.40 **	11.16 ns	11.84 *	10.80 **	11.26	**	0.000
1986-1989	12.30 ns	12.26 ns	11.25 **	12.35 ns	13.61	ns	0.077
1990-1993	11.89 ns	12.94 ns	12.98 ns	12.58 ns	13.45	ns	0.220
1994-1998	10.03 ns	10.46 ns	11.31 **	10.29 **	10.61	*	0.536

TABLE 18 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON Cu CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

TABLE 18 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON Cu CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

Years		Cu	mulative Nu Earth			
Averaged ¹	0	100	200	400	500	\mathbf{P}^2

 ¹ Values are for average of data for years included in each period.
² Significance probability of the effect of Nu Earth rate on metal concentrations.
³ Notations following values indicate significance probability that values are different from those for the 1980-1981 period. ns = not significant. *, ** = significant at 0.05 and 0.01 probability levels, respectively. ND = no data.

Years	Cumulative Nu Earth Rate (Mg/ha)					
Averaged ¹	0	100	200	400	500	\mathbf{P}^2
				Beet		
1980-1981	0.43	0.49	0.83	1.40	2.17	0.000
1982-1985	3.79 ** ³	4.15 **	5.16 **	5.68 **	9.98 **	0.002
1986-1989	1.01 ns	2.07 ns	0.78 *	1.01 **	1.26 **	0.000
1990-1993	0.11 **	0.21 **	0.25 **	0.40 **	0.51 **	0.000
1994-1998	1.06 ns	0.47 **	0.95 ns	0.91 *	0.90 **	0.016
			(Carrot		
1980-1981	0.53	0.70	0.94	1.59	2.96	0.000
1982-1985	2.66 **	2.04 *	2.39 **	3.28 **	4.24 ns	0.000
1986-1989	0.94 ns	1.17 ns	0.99 ns	1.70 ns	1.84 **	0.000
1990-1993	0.91 ns	0.75 ns	1.14 ns	1.49 ns	1.54 **	0.036
1994-1998	0.25 ns	0.73 ns	1.08 ns	1.60 ns	1.17 **	0.105
			Sw	iss Chard		
1980-1981	2.32	3.14	4.76	5.85	6.47	0.000
1982-1985	3.03 ns	4.34 ns	5.63 ns	4.95 ns	5.73 ns	0.015
1986-1989	3.55 ns	4.72 ns	6.14 ns	3.71 *	6.04 **	0.792
1990-1993	2.26 ns	2.71 ns	7.30 ns	3.75 ns	2.98 **	0.156
1994-1998	0.98 **	1.40 **	1.43 **	1.67 **	2.30 **	0.002

TABLE 19: RESIDUAL EFFECT OF NU EARTH APPLICATION ON NI CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

Years	Cumulative Nu Earth Rate (Mg/ha)					
Averaged ¹	0	100	200	400	500	\mathbf{P}^2
			S	pinach		
1980-1981	1.00	1.18	2.41	2.74	5.42	0.000
1982-1985	7.61 **	5.73 **	6.78 **	8.59 **	8.96 ns	0.035
1986-1989	3.42 **	4.60 ns	3.86 ns	3.45 ns	4.72 ns	0.893
1990-1993	ND	ND	ND	ND	ND	
1994-1998	0.54 *	1.33 ns	1.01 *	1.34 **	1.74 *	0.056
			G	reen Beans		
1980-1981	1.60	2.39	4.02	7.09	9.00	0.000
1982-1985	1.90 ns	2.06 ns	4.44 ns	9.05 ns	10.54 ns	0.000
1986-1989	1.51 ns	1.81 ns	2.97 ns	3.33 ns	3.92 **	0.007
1990-1993	1.57 ns	2.88 ns	2.73 *	5.30 ns	6.35 ns	0.000
1994-1998	1.62 ns	2.48 ns	2.63 ns	3.29 *	2.86 **	0.230
			T	omato		
1980-1981	1.10	1.00	1.09	1.22	1.77	0.006
1982-1985	3.53 **	3.57 **	6.37 **	3.16 **	4.29 **	0.225
1986-1989	0.09 **	0.18 **	0.31 **	0.52 **	1.25 *	0.000
1990-1993	0.35 **	0.44 **	0.58 **	0.66 *	0.83 **	0.000
1994-1998	1.22 ns	0.88 ns	1.15 ns	1.30 ns	2.81 ns	0.417

TABLE 19 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON NI CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

TABLE 19 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON NI CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

Years	Cumulative Nu Earth Rate (Mg/ha)						
Averaged ¹	0	100	200	400	500	P^2	

¹ Values are for average of data for years included in each period.

² Significance probability of the effect of Nu Earth rate on metal concentrations.

³ Notations following values indicate significance probability that values are different from those for the 1980-1981 period. ns = not significant. *, ** = significant at 0.05 and 0.01 probability levels, respectively. ND = no data.

57

Years		Cum	ulative Nu Earth H	Rate (Mg/ha)		
Averaged ¹	0	100	200	400	500	\mathbf{P}^2
			E	Seet		
1980-1981	0.25	0.18	0.29	0.40	0.30	0.700
1982-1985	0.24 ns^3	0.30 ns	0.26 ns	0.19 ns	0.15 ns	0.186
1986-1989	0.21 ns	0.19 ns	0.14 **	0.13 ns	0.14 ns	0.147
1990-1993	0.10 ns	0.08 ns	0.13 ns	0.09 *	0.09 ns	0.768
1994-1998	0.25 ns	0.14 ns	0.18 ns	0.10 *	0.25 ns	0.439
			······ (Carrot		
1980-1981	0.19	0.22	0.26	0.12	0.14	0.866
1982-1985	0.29 ns	0.32 ns	0.60 ns	0.53 *	0.71 **	0.066
1986-1989	0.46 ns	0.58 *	0.52 *	0.64 **	0.69 **	0.637
1990-1993	0.10 ns	0.25 ns	0.26 ns	0.48 *	0.44 **	0.000
1994-1998	0.58 ns	0.42 ns	0.51 ns	0.38 *	2.68 *	0.165
			Swis	s Chard		
1980-1981	1.90	1.72	1.89	2.23	2.42	0.200
1982-1985	0.80 **	0.87 **	0.68 **	0.66 **	0.66 **	0.791
1986-1989	1.58 ns	1.39 ns	1.13 **	1.05 **	0.99 **	0.927
1990-1993	1.25 ns	1.89 ns	0.90 **	1.27 **	1.52 ns	0.491
1994-1998	0.66 **	1.00 **	0.56 **	0.67 **	0.68 **	0.016

TABLE 20: RESIDUAL EFFECT OF NU EARTH APPLICATION ON Pb CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

Years		Cun	nulative Nu Earth	Rate (Mg/ha)		
Averaged ¹	0	100	200	400	500	\mathbf{P}^2
			Sp	inach		
1980-1981	1.90	1.16	1.74	2.08	2.26	0.050
1982-1985	0.62 **	0.77 ns	1.14 **	1.01 **	1.02 **	0.001
1986-1989	0.88 *	1.22 ns	0.91 *	1.42 *	0.91 **	0.908
1990-1993	ND	ND	ND	ND	ND	
1994-1998	0.34 *	0.70 ns	0.31 **	0.41 **	0.43 **	0.190
			Gre	en Beans		
1980-1981	0.20	0.10	0.13	0.07	0.10	0.751
1982-1985	0.05 ns	<0.01 ns	<0.01 *	0.11 ns	0.06 ns	0.898
1986-1989	0.63 ns	0.43 *	0.44 ns	0.17 ns	0.65 ns	0.528
1990-1993	0.18 *	0.19 *	0.24 ns	0.18 **	0.25 *	0.608
1994-1998	2.19 *	0.29 **	0.66 ns	0.48 **	0.50 *	0.809
			To	omato		
1980-1981	< 0.01	0.09	< 0.01	< 0.01	0.19	0.170
1982-1985	<0.01 ns	<0.01 ns	<0.01 ns	<0.01 ns	<0.01 **	0.597
1986-1989	<0.01 ns	0.23 ns	<0.01 ns	<0.01 ns	<0.01 ns	0.010
1990-1993	0.14 **	0.15 ns	0.13 **	0.14 **	0.22 ns	0.345
1994-1998	0.56 *	0.20 ns	0.25 ns	0.06 ns	0.26 ns	0.777

TABLE 20 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON Pb CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

TABLE 20 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON Pb CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

Years		Cu	mulative Nu Eartl	h Rate (Mg/ha)		
Averaged ¹	0	100	200	400	500	P^2

 ¹ Values are for average of data for years included in each period.
² Significance probability of the effect of Nu Earth rate on metal concentrations.
³ Notations following values indicate significance probability that values are different from those for the 1980-1981 period. ns = not significant. *, ** = significant at 0.05 and 0.01 probability levels, respectively. ND = no data.

Years		Cun	nulative Nu Earth	Rate (Mg/ha)		
Averaged ¹	0	100	200	400	500	\mathbf{P}^2
]	Beet		
1980-1981	34.3	51.6	62.9	89.8	119.4	0.000
1982-1985	30.5 * ³	42.2 *	48.8 *	64.7 *	76.7 **	0.000
1986-1989	21.6 **	27.1 **	28.1 **	38.6 **	48.7 **	0.000
1990-1993	28.7 *	36.2 **	44.5 **	54.8 **	66.0 **	0.000
1994-1998	45.3 ns	44.9 ns	50.8 *	57.8 **	68.4 **	0.001
			C	arrot		
1980-1981	23.3	32.6	33.0	35.1	41.0	0.000
1982-1985	25.8 ns	28.6 ns	28.8 ns	29.9 ns	34.0 *	0.051
1986-1989	26.0 *	27.8 ns	27.6 ns	31.5 ns	31.4 *	0.226
1990-1993	24.7 ns	31.9 ns	27.8 ns	33.4 ns	32.0 *	0.023
1994-1998	23.5 ns	26.2 ns	34.5 ns	31.3 ns	24.5 **	0.117
			Swis	s Chard		
1980-1981	71.7	151.8	238.5	338.0	397.7	0.000
1982-1985	61.3 ns	123.4 ns	175.8 ns	278.5 ns	347.3 ns	0.000
1986-1989	74.1 ns	126.1 ns	142.2 **	161.7 **	151.5 **	0.000
1990-1993	51.4 **	86.7 **	110.6 **	178.3 **	168.1 **	0.000
1994-1998	58.6 ns	123.9 ns	139.1 **	213.0 *	201.1 **	0.000

TABLE 21: RESIDUAL EFFECT OF NU EARTH APPLICATION ON Zn CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

Years			nulative Nu Earth			
Averaged ¹	0	100	200	400	500	\mathbf{P}^2
			St	binach		
1980-1981	207.8	258.9	244.5	272.7	250.8	0.654
1982-1985	179.7 ns	315.2 ns	305.8 ns	299.6 ns	314.0 ns	0.000
1986-1989	150.5 ns	244.0 ns	239.3 ns	261.8 ns	263.5 ns	0.000
1990-1993	ND	ND	ND	ND	ND	
1994-1998	176.2 ns	241.0 ns	196.1 ns	273.1 ns	214.2 ns	0.012
			Gree	n Beans		
1980-1981	29.6	32.2	34.0	36.8	37.4	0.000
1982-1985	28.8 ns	24.2 *	29.4 ns	28.1 ns	31.2 ns	0.683
1986-1989	24.2 **	25.0 **	24.2 **	28.5 **	27.5 **	0.000
1990-1993	32.6 ns	36.8 ns	41.4 **	39.6 ns	38.6 ns	0.003
1994-1998	37.1 *	39.4 **	20.0 *	41.7 *	43.8 *	0.179
			To	omato		
1980-1981	29.5	32.4	34.8	38.1	38.7	0.000
1982-1985	26.9 ns	29.7 ns	30.6 *	32.2 **	32.7 **	0.000
1986-1989	26.2 *	26.5 **	25.6 **	28.2 **	31.4 **	0.008
1990-1993	32.5 ns	33.2 ns	34.7 ns	35.3 ns	34.4 *	0.825
1994-1998	23.7 **	27.6 *	28.7 *	27.0 **	29.0 **	0.071

TABLE 21 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON Zn CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

TABLE 21 (Continued): RESIDUAL EFFECT OF NU EARTH APPLICATION ON Zn CONCENTRATIONS (mg/kg) IN EDIBLE TISSUE OF VEGETABLES GROWN IN SOIL RECEIVING NU EARTH ANNUALLY DURING 1977 TO 1981

Years		Cu	mulative Nu Earth	n Rate (Mg/ha)		
Averaged ¹	0	100	200	400	500	\mathbf{P}^2

¹ Values are for average of data for years included in each period.
² Significance probability of the effect of Nu Earth rate on metal concentrations.

³ Notations following values indicate significance probability that values are different from those for the 1980-1981 period. ns = not significant. *, ** = significant at 0.05 and 0.01 probability levels, respectively. ND = no data.

Trace Metal Concentrations in Vegetables Compared to USEPA Part 503 Risk Assessment Model Predictions and Cumulative Loading Limits

The USEPA Part 503 Rule on land application of biosolids is based on a risk assessment model used to predict potential human and animal exposure to metals applied in biosolids. The risk assessment included 14 exposure pathways including one protecting the home gardeners who apply biosolids to their vegetable gardens. The home gardener pathway (Pathway 2) model predicts increases in metal concentrations in edible tissue of crops that are due to cumulative metal loading from biosolids. Uptake coefficients, [UC = (mg metal/kg tissue)/(kg metal/ha)], are used to predict transfer of metals from biosolids amended soils to plant tissue. Uptake coefficients were established for Cd, Ni, and Zn in many groups of vegetables including root, leafy, and fruit. Uptake coefficients were not determined for Cr, Cu, and Pb because initial screening of pollutants indicated that Cr and Cu do not pose a hazard under the worse-case scenarios evaluated for Pathway 2 (USEPA, 1985) and no oral reference dose for Pb were available for input into the risk assessment model (USEPA, 1992).

Comparison with Risk Assessment Model Predictions. For making the comparison between increases in tissue concentrations predicted by the Part 503 model and concentrations measured in our study, predicted increases in tissue concentrations of Cd, Ni, and Zn due to biosolids were calculated according to the approach used in the Part 503 model (USEPA, 1992) using the equation:

Predicted increase in tissue concentration (mg/kg) = Cumulative metal loading (kg/ha) x UC [(mg/kg)/(kg/ha)] Eq. 1

Measured increases in tissue metal concentrations that were due to Nu Earth application were computed as tissue concentrations in Nu Earth-amended treatments minus tissue concentrations in the control (unamended soil).

The relationship between predicted and measured increases in tissue Cd concentration due to Nu Earth in the tissues of groups of vegetables, which were evaluated only for the 40 and 100 Mg/ha annual Nu Earth rates, are presented in <u>Figures 8</u> through <u>10</u>. The predicted increases in tissue Cd concentrations increased linearly from 1977 to 1981 and remained constant thereafter. This trend is the direct result of annual Nu Earth additions, which ceased after 1981.

Predicted Cd uptake due to Nu Earth application increased proportionally to metal loading during the period of Nu Earth application, then decreased thereafter (Figure 8). During the period of Nu Earth application, predicted Cd uptake in the root crops was well related to measured uptake. Thereafter, annual Nu Earth measured uptake fluctuated at levels below and above predictions. At the 100 Mg/ha biosolids rate (Figure 8b), measured Cd uptake by carrot decreased consistently after 1990 to levels well below predicted uptake.

During the period of Nu Earth application, measured increases in Cd concentrations in the tissue of the leafy vegetables at the 40 Mg/ha annual Nu Earth rate tended to be only slightly

FIGURE 8: MEASURED AND PREDICTED (PART 503 RISK ASSESSMENT) INCREASES IN Cd CONCENTRATIONS IN EDIBLE TISSUE OF ROOT VEGETABLES GROWN IN SOIL RECEIVING 40 Mg/ha (a) AND 100 Mg/ha (b) NU EARTH ANNUALLY FROM 1977 TO 1981

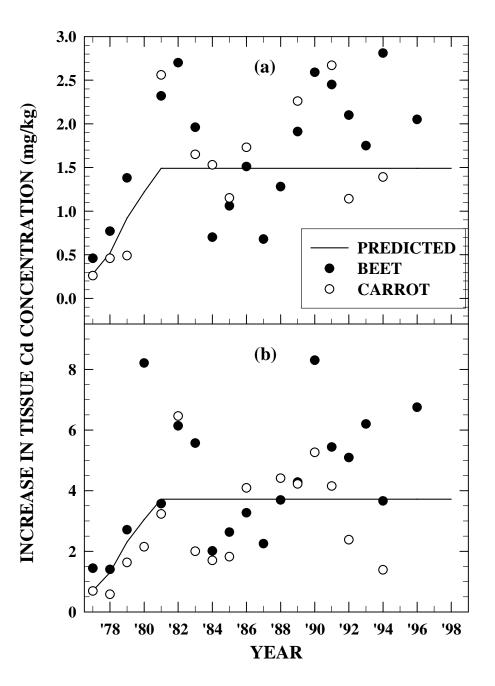


FIGURE 9: MEASURED AND PREDICTED (PART 503 RISK ASSESSMENT) INCREASES IN Cd CONCENTRATIONS IN EDIBLE LEAF VEGETABLES GROWN IN SOIL RECEIV-ING 40 Mg/ha (a) AND 100 Mg/ha (b) NU EARTH ANNUALLY FROM 1977 TO 1981

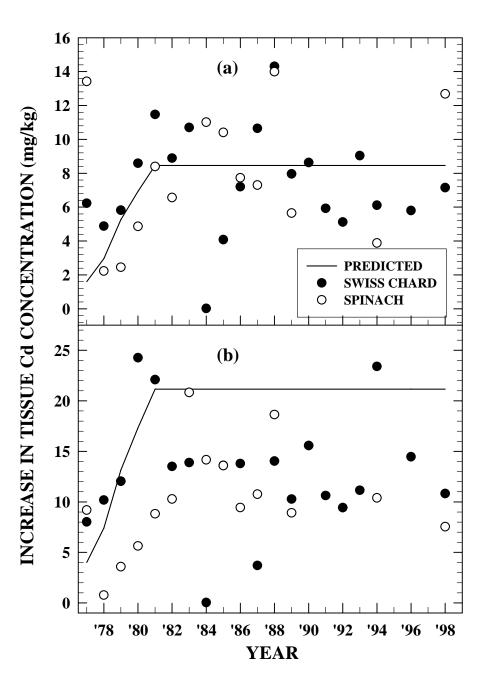
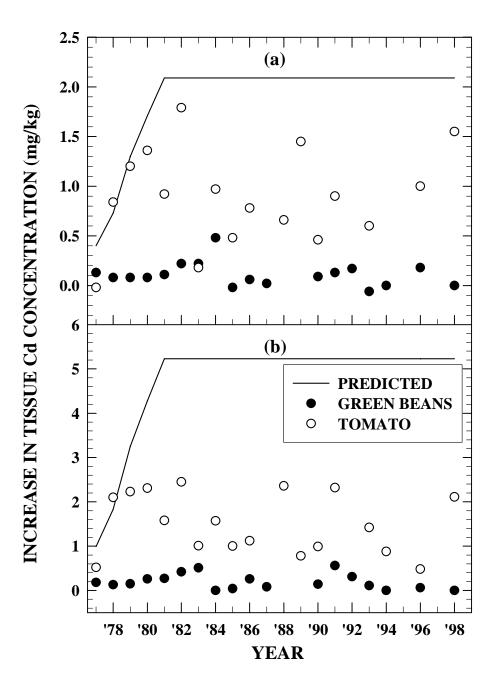


FIGURE 10: MEASURED AND PREDICTED (PART 503 RISK ASSESSMENT) INCREASES IN Cd CONCENTRATIONS IN EDIBLE FRUIT VEGETABLES GROWN IN SOIL RECEIVING 40 Mg/ha (a) AND 100 Mg/ha (b) NU EARTH ANNUALLY FROM 1977 TO 1981



higher than predicted concentrations for Swiss chard and slightly lower than predicted concentrations for spinach (Figure 9a). At the 100 Mg/ha annual Nu Earth rate, increases in Cd concentrations were mostly lower than predicted concentrations (Figure 9b). This difference in the relationship between measured and predicted increases in tissue Cd concentrations observed for the 40 and 100 Mg/ha annual Nu Earth rates indicates that increases in Cd concentrations in the leafy vegetables depend not only on cumulative loading rates but also on annual loading rates. The Part 503 risk assessment model may tend to over-estimate increases in Cd concentrations in some vegetables because the model does not account for the potential decreased bioavailability of applied metals over time.

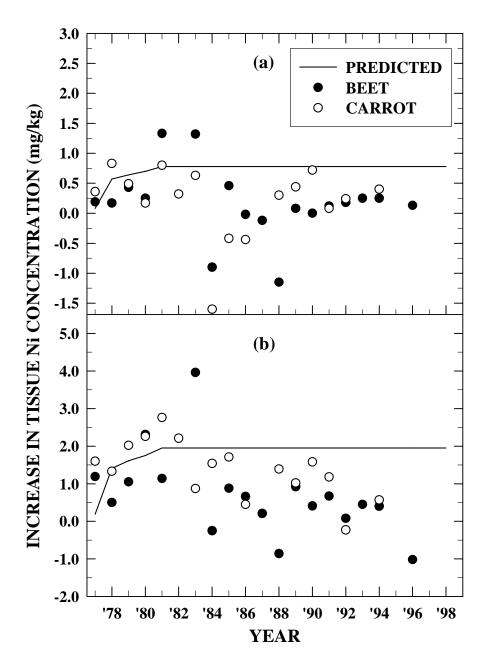
In the fruit vegetables, there was no significant effect of cumulative Cd loading on tissue Cd concentrations. The measured increases in tissue Cd concentrations were much lower than the Part 503 risk assessment predictions, except in tomato during the first three years of annual biosolids application (Figure 10).

The relationship between predicted and measured increases in tissue Ni concentrations due to Nu Earth in groups of vegetables at the 40 and 100 Mg/ha annual Nu Earth rates are presented in <u>Figures 11</u> through <u>13</u>. During the period of Nu Earth application, the measured increases in tissue Ni concentrations in the root vegetables fluctuated above and below the predicted levels (<u>Figure 14</u>). After cessation of biosolids application, the Ni concentrations tended to decline below the predicted levels. Throughout the study, measured increases in tissue Ni concentrations were mostly lower than predicted in the leaf vegetables (<u>Figure 12</u>) and in the fruit vegetables it fluctuated above and below the predicted levels (Figure 13).

The relationship between predicted and measured increases in tissue Zn concentrations due to Nu Earth in groups of vegetables at the 40 and 100 Mg/ha Nu Earth rates are presented in Figures 14 through 16. The measured increases in carrot Zn concentrations were always lower than the risk assessment model predictions. In beet, the measured increases in tissue Zn concentrations were higher than predicted only during the period of Nu Earth application and for the first two years following termination of application (Figure 14). Thereafter, the increases in beet Zn concentrations were lower than predicted by the model (Figure 14). In the leafy vegetables, measured increases in tissue Zn were lower than predicted, especially in the period following termination of Nu Earth application (Figure 15b). In most years, increases in tissue Zn concentrations in the fruit vegetables were much lower than predicted (Figure 16). The magnitude by which measured concentrations were lower than predicted was usually greater at the 100 Mg/ha rate than at the 40 Mg/ha rate.

The frequencies at which the measured increases in Cd, Ni, and Zn concentrations at the 40 and 100 Mg/ha Nu Earth rates were lower than the increases predicted by the Part 503 risk assessment model for the periods during Nu Earth application and termination of applications are presented in <u>Tables 22</u> and <u>23</u>, respectively. During the period on Nu Earth application, all measured increases in concentrations of Cd in green beans and Zn in carrot and tomato were lower than predicted (<u>Table 22</u>). Most (>50 percent) of the measured increases in concentrations of Cd in carrot, spinach, and tomato, Ni in Swiss chad and spinach, and Zn in green beans were lower than predicted.

FIGURE 11: MEASURED AND PREDICTED (PART 503 RISK ASSESSMENT) INCREASES IN NI CONCENTRATIONS IN EDIBLE TISSUE OF ROOT VEGETABLES GROWN IN SOIL RECEIVING (a) 40 Mg/ha AND (b) 100 Mg/ha NU EARTH ANNUALLY FROM 1977 TO 1981



69

FIGURE 12: MEASURED AND PREDICTED (PART 503 RISK ASSESSMENT) INCREASES IN NI CONCENTRATIONS IN EDIBLE LEAF VEGETABLES GROWN IN SOIL RECEIVING (a) 40 Mg/ha AND (b) 100 Mg/ha NU EARTH ANNUALLY FROM 1977 TO 1981

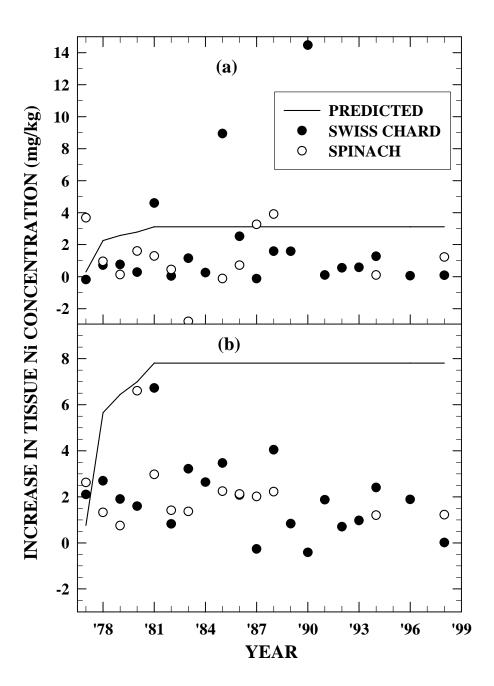


FIGURE 13: MEASURED AND PREDICTED (PART 503 RISK ASSESSMENT) INCREASES IN NI CONCENTRATIONS IN EDIBLE TISSUE OF FRUIT VEGETABLES GROWN IN SOIL RECEIVING (a) 40 Mg/ha AND (b) 100 Mg/ha NU EARTH ANNUALLY FROM 1977 TO 1981

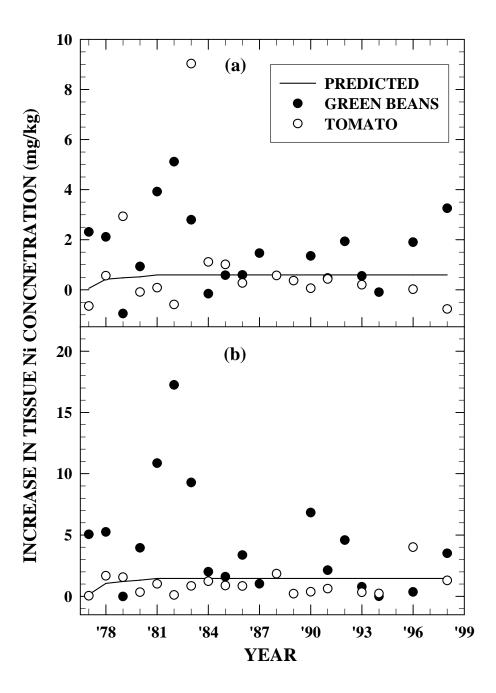


FIGURE 14: MEASURED AND PREDICTED (PART 503 RISK ASSESSMENT) INCREASES IN Zn CONCENTRATIONS IN EDIBLE TISSUE OF ROOT VEGETABLES GROWN IN SOIL RECEIVING 40 Mg/ha (a) AND 100 Mg/ha (b) NU EARTH ANNUALLY FROM 1977 TO 1981

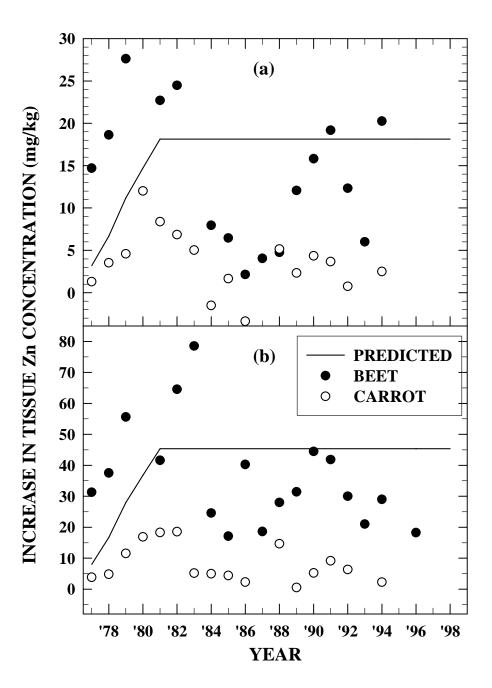


FIGURE 15: MEASURED AND PREDICTED (PART 503 RISK ASSESSMENT) INCREASES IN Zn CONCENTRATIONS IN EDIBLE LEAF VEGETABLES GROWN IN SOIL RECEIVING 40 Mg/ha (a) AND 100 Mg/ha (b) NU EARTH ANNUALLY FROM 1977 TO 1981

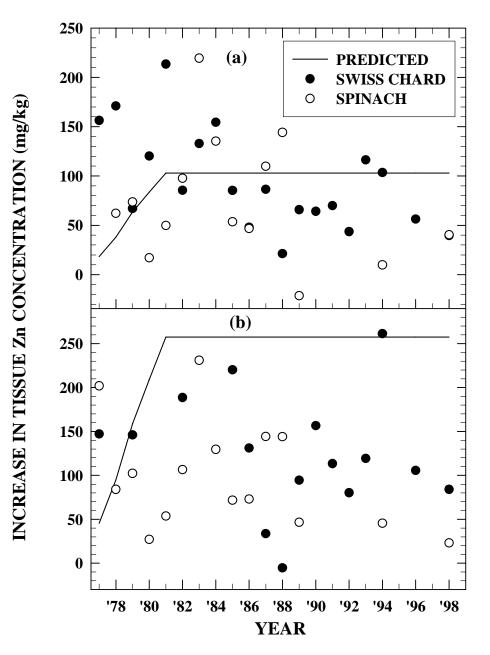


FIGURE 16: MEASURED AND PREDICTED (PART 503 RISK ASSESSMENT) INCREASES IN Zn CONCENTRATIONS IN EDIBLE TISSUE OF FRUIT VEGETABLES GROWN IN SOIL RECEIVING 40 Mg/ha (a) AND 100 Mg/ha (b) NU EARTH ANNUALLY FROM 1977 TO 1981

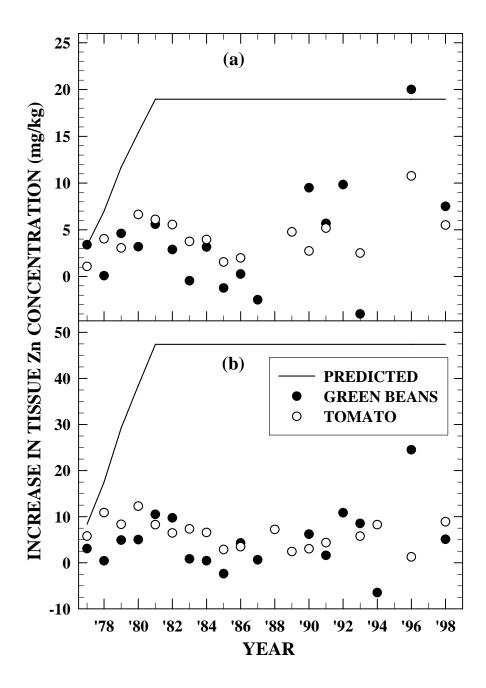


TABLE 22: FREQUENCY OF MEASURED INCREASES IN VEGETABLE TISSUE METAL CONCENTRATIONS DUE TO NU EARTH THAT WERE LOWER THAN INCREASES PREDICTED BY THE PART 503 RISK ASSESSMENT MODEL DURING THE PERIOD OF NU EARTH APPLICATION, 1977 TO 1981

Metal	Beet	Carrot	Swiss Chard	Spinach	Tomato	Green Beans
			Frequer	$(\%)^{1}$		
Cd	15	60	25	65	55	100
Ni	35	35	70	63	40	30
Zn	15	100	10	50	100	95

¹ Frequency = (count of measured tissue metal concentration increases that were lower than predicted \div total count of measured tissue metal concentration increases) x 100.

TABLE 23: FREQUENCY OF MEASURED INCREASES IN VEGETABLE TISSUE METAL CONCENTRATIONS DUE TO NU EARTH THAT WERE LOWER THAN INCREASES PREDICTED BY THE PART 503 RISK ASSESSMENT MODEL FOR THE PERIOD FOLLOWING TERMINATION OF NU EARTH APPLICATION, 1982 TO 1998

Metal	Beet	Carrot	Swiss Chard	Spinach	Tomato	Green Beans
			Freque	$ency (\%)^{1}$		
Cd	38	35	67	67	95	100
Ni	68	44	72	80	64	68
Zn	71	98	68	73	100	97

Frequency = (count of measured tissue metal concentration increases that were lower than predicted \div total count of measured tissue metal concentration increases) x 100. After termination of Nu Earth application, there was an increase in the frequencies at which the observed increases in tissue concentrations of Cd, Ni, and Zn at the 40 and 100 Mg/ha Nu Earth rates were lower than concentrations predicted by the Part 503 risk assessment model (<u>Table 23</u>). These results show that transfer of metal to tissues of plants grown in soils receiving repeated annual applications of biosolids tends to decrease with time after applications are terminated. This tendency is not considered in the Part 503 Risk Assessment model, making it conservative.

Concentrations in Vegetables Compared to Concentrations Computed Using Part 503 Loading Limits. The only metals from this study that the USEPA found to pose potentially significant risk to home gardeners were Cd, Ni, and Zn. To compare the concentrations of these metals in the edible tissues of vegetables to the concentrations computed to be safe for a 70-year consumption period based on the Part 503 risk assessment model and the cumulative loading limits (computed safety level for lifetime consumption), the following equation was used:

Tissue concentration $(mg/kg) = Cumulative metal loading (kg/ha) \times UC$ [(mg/kg)/(kg/ha)] + Control concentration Eq. 2

In this equation, it is assumed that all trace metals in the edible tissues are due to biosolids application. The metal loading limits, UC values, and the computed concentration limits in the vegetable tissues are presented in <u>Table 24</u>. The occurrences of measured tissue metal concentrations were compared to the computed safe lifetime consumption concentrations (SLCC) using the Student's *t* test.

The frequency at which metal concentrations in the vegetable tissues during the residual period were equal to or higher than the computed SLCC are presented in <u>Table 25</u>. These data show that during the residual period, except for Cd in green beans, there were several instances of the tissue concentrations of Cd and Ni that were equal to or higher than the computed SLCC. There were very few instances of higher Zn concentrations.

The frequency at which metal concentrations in the vegetable tissues were equal to or higher than the computed SLCC during the residual period were in the order Cd>Ni>Zn (<u>Table 25</u>). This trend occurred mostly because Cd concentrations in the Nu Earth and cumulative loading rates (<u>Table 3</u>) were much higher than Ni and Zn relative to the Part 503 limits. In the Control, where no biosolids were applied there were many occurrences of metal concentrations that were higher than the computed SLCC. For Cd and Ni occurrences in the control, with the exception of in Cd in green beans, incidents were in all vegetables. For Zn there was a 7 percent occurrence in the beet only.

TABLE 24: SAFE LIFETIME CONSUMPTION CONCENTRATIONS OF Cd, Ni, AND Zn IN ROOT, LEAF, AND FRUIT VEGETABLE TISSUES CALCULATED BASED ON THE PART 503 BIOSOLIDS CUMULATIVE METAL LOADING LIMITS AND UPTAKE COEFFICIENTS USED IN THE RISK ASSESSMENT MODEL

		Uptal	ke Coeffic	ient		Safe Lifetime Consumption ²						
Metal	Loading Limit ¹	Root	Leaf	Fruit	Beet	Carrot	Swiss Chard	Spinach	Green Beans	Tomato		
	kg/ha	(mg/kg)/(kg/ha)				mg/kg						
Cd	39	0.182	0.032	0.045	1.89	2.29	9.10	10.90	1.96	2.96		
Ni	420	0.016	0.004	0.003	2.09	2.22	9.84	8.72	3.32	2.37		
Zn	2800	0.125	0.022	0.023	92.9	84.4	405.4	512.0	96.0	94.3		

¹ Part 503 cumulative metal loading limit. ² Safe Lifetime Consumption = metal loading limit (kg/ha) x uptake coefficient [(mg/kg)/(kg/ha)] + control concentration.

	0.0				
		Annua	l Nu Earth Rate ((Mg/ha)	
Vegetable	0	50	40	80	100
			Cd		
Beet	29	71	100	100	100
Carrot	18	83	100	100	100
Swiss Chard	7	57	100	100	100
Spinach	30	100	100	100	100
Green Beans	0	0	9	9	0
Tomato	8	23	31	43	69
			Ni		
Beet	55	50	46	46	50
Carrot	40	33	36	58	58
Swiss Chard	13	13	29	7	13
Spinach	22	40	33	40	50
Green Beans	50	31	54	100	85
Tomato	39	39	39	39	62
			Zn		
Beet	7	0	0	8	29
Carrot	0	0	0	0	0
Swiss Chard	0	0	0	7	13
Spinach	0	0	0	9	0
Green Beans	0	0	0	0	0
Tomato	0	0	0	0	0

TABLE 25: FREQUENCY¹ OF OCCURRENCE DURING THE RESIDUAL PERIOD WHERE METAL CONCENTRATIONS IN EDIBLE TISSUE OF VEGETABLES WERE EQUAL TO OR GREATER THAN THE COMPUTED PART 503 SAFE LIFETIME CONSUMPTION CONCENTRATIONS

¹ Frequency = number of occurrences \div total count of measured tissue concentrations x 100. NA = Not applicable.

IMPLICATIONS FOR HOME GARDENS AMENDED WITH NU EARTH

The Nu Earth "Give Away" program in the Chicago area continued during the Nu Earth production era until 1979. The results of this study can be used to provide guidance in evaluating the risk of consuming vegetables produced in home gardens amended with Nu Earth. We computed Safe Lifetime Consumption (SLCC) concentrations for the vegetables grown in the study for Cd, Ni and Zn from the Part 503 risk assessment model pathway for home gardeners. These concentrations are thought to be safe for consumption by home gardeners who raise as much as 59 percent of the vegetables they consume over a 70-year period on Nu Earth amended soil. Based on the comparison between trace metal concentrations in the edible tissue of the vegetables and the computed SLCC, Cd uptake in edible tissues probably posed the highest risk for consumption of vegetables grown in Nu Earth amended soil. At the loading rate of 40 Mg/ha that was recommended for use of Nu Earth as a soil builder in the Chicago area, the data showed that there were no instances of elevated levels of Zn in any of the vegetables and there was only a 9 percent frequency of occurrence of Cd in green beans. Therefore, among the vegetables evaluated, the risk of evaluated trace metal concentrations in edible tissues is lowest for green beans. Assuming that home gardens in the Chicago area were not amended for more than five annual applications of the recommended Nu Earth loading rate, the risk associated with consumption of these vegetables is very low, because the trace metal loading limits derived from the risk assessment model were based on conservative assumptions to protect human health and the environment. One of the conservative assumptions that is useful to evaluating the risk associated with consumption of vegetables grown in Chicago area home gardens amended with Nu Earth is that 59 percent of the vegetables consumed by home gardeners during a 70-year lifespan are grown in the biosolids amended soil. In the Chicago area, such level of exposure to home gardeners is very unlikely because home gardens are very small and the growing season for vegetables is too short to allow such high consumption of homegrown vegetables.

The major risk associated with soil Pb contamination in urban areas is soil ingestion by pica children. At the recommended Nu Earth loading rate of 40 Mg/ha, the cumulative Pb loading resulting from five annual Nu Earth applications is approximately 150 kg/ha. Based on the assumption that a 15 cm depth of soil weighs 2×10^6 kg/ha, this represents a soil concentration of 75 mg/kg, which is at only 25 percent of the safe level of 300 mg/kg stipulated in the Part 503 rule to protect pica childhood behavior. Therefore, the risk to home gardeners consuming vegetables from their gardens during the era of Nu Earth use and in the long term thereafter is very low. Furthermore, the data in this study showed that plant uptake of the trace metals decrease with time following termination of Nu Earth application. Home gardeners who are still concerned about potential risks, but would like to use Nu Earth amended gardens to grow vegetables, can minimize the risk even further by maintaining soil above pH 6.5 to reduce metal bioavailability and uptake in edible tissues of vegetables.

LITERATURE CITED

- Baker, D.E. and Amacher, M. C., "Nickel, Copper, and Cadmium." *Methods of Soil Analysis*, *Part 2. Chemical and Microbiological Properties*. Agronomy Monograph no. 9 (2nd Edition). American Society of Agronomy. Madison WI. pp. 323-336, 1982.
- Basta, N. T. and Sloan, J. J., "Bioavailability of Heavy Metals in Strongly Acidic Soils Treated with Exceptional Quality Biosolids," *Journal of Environmental Quality*, 28:633-638, 1999.
- Brown, S. L., Chaney, R. L., Angle, J. S., and Ryan, J. A. Relative Uptake of Cadmium by Garden Vegetables and Fruits Grown on Long-term Sewage Sludge Amended Soils. *Environmental Science Technology* 30:3508-3511, 1996.
- Bell, P. F., James, B. R., and Chaney, R. F. L., "Heavy Metal Extractability in Long-Term Sewage Sludge and Metal Salt-Amended Soil," *Journal of Environmental Quality*, 20:481-486, 1991.
- Chaney, R. L Bruins, R. J., Baker, D. E., Korcak, R. F., Smith, J. E., and Cole, D., "Transfer of Sludge-applied Metals to the Food Chain," pp. 67-99, In *Land Application of Sewage Sludge*, Lewis Publishers, Chelsea, Michigan, 1987.
- Corey, R. B. L. D. King, Lue-Hing, C., Street, J. J., and Walker, J. M., "Effects of Sludge Properties on Accumulation of Trace Elements by Crops," pp. 25-51, In *Land Application of Sewage Sludge*, Lewis Publishers, Chelsea, Michigan, 1987.
- Feeney, S., Peterson, J. R., Zenz, D. R., and Lue-Hing, C, "National Survey of the Metals Content of Seven Vegetable Species." Metropolitan Water Reclamation District of Greater Chicago, Research and Development Department. Report No. 84-4. 1984
- Granato, T. C., Richardson, G. R., and Gschwind, J., "A Preliminary Report on the Status of the Nu Earth Garden Experiment. Results Through 1989." Metropolitan Water Reclamation District of Greater Chicago, Research and Development Department. 1990
- Jackson, A. P. and Alloway, B. J. "The Bioavailability of Cadmium to Lettuce and Cabbage in Soils Previously Treated with Sewage Sludge," *Plant and Soil*, 132: 179-186, 1991.
- Logan, T. J., Lindsay, B. J., Goins, L. E., and Ryan, J. A., "Field Assessment of Metal Bioavailability to Crops: Sludge Rate Response," *Journal of Environmental Quality*, 26:534-550, 1997.
- Logan, T. J., Goins, L. E., and Lindsay, B. J., "Field Assessment of Trace Element Uptake by Six Vegetables from N-Viro Soil," *Water Environment Research*, 69:28-33, 1997.
- McBride, M. B., "Toxic Metal Accumulation From Agricultural Use of Sludge: Are USEPA Regulations Protective?" *Journal of Environmental Quality*, 24:5-18, 1995.

- McGrath, S. P., Zhao, F. J., Dunham, S. J., Crosland, Coleman, A. R., K., "Long-Term Changes in the Extractability and Bioavailability of Zinc and Cadmium After Sludge Application," *Journal of Environmental Quality*, 29:875-883, 2000.
- Page, A. L., T. G. Logan, and Ryan, J. A., *Land Application of Sewage Sludge*, Lewis Publishers, Chelsea, Michigan, 1987.
- SAS/STAT[®] User's Guide, Volume 2, SAS[®] Institute, Inc. 5th Printing, June 1995.
- SAS[®] Procedures Guide, SAS[®] Institute, Inc. 4th Printing, June 1994.
- Sommers, L., Van Wolk, V., Giordano, P. M., Sooper, W. and Bastain, R. "Effects of Soil Properties on Accumulation of Trace Metals by Crops," pp. 5-24, In *Land Application of Sewage Sludge*, Lewis Publishers, Chelsea, Michigan, 1987.
- Sukkariyah, B. F., Evanylo, G., Zelazny, L., and Chaney, R. L. "Recovery and Distribution of Biosolids-Derived Trace Metals in a Clay Loam Soil," *Journal of Environmental Quality*, 34:1843-1850, 2005a.
- Sukkariyah, B. F., Evanylo, G., Zelazny, L., and Chaney, R. L. "Cadmium, Copper, Nickel, and Zinc Availability in a Biosolids-Amended Piedmont Soil Years after Application," *Jour*nal of Environmental Quality, 34:2255-2262, 2005b.
- United States Environmental Protection Agency, "Standards for the Use or Disposal of Sewage Sludge," *Federal Register*, 58:9387-9415, 1993.
- United States Environmental Protection Agency, "Standards for the Use or Disposal of Sewage Sludge," *Federal Register*, 60:54764-54770, 1995.
- United States Environmental Protection Agency, Technical Support Document for Land Application of Sewage Sludge, Volume 2, USEPA, Washington, DC, 1992.
- Yingming, L. and Corey, R. B., "Redistribution of Sludge-borne Cadmium, Copper, and Zinc in a Cultivated Plot," *Journal of Environmental Quality*, 22:1-8, 1993.

		ual Nu E 3-Years	Treated)	Annual Nu Earth Rate (Mg/ha) 5-Years Treated Plots				
Year	0	20	40	80	100	0	20	40	80	100
					mg/k	g				
1977	0.5	4.8	5.4	11.7	17.3					
1978	3.6	11.3	12.6	23.1	19.2					
1979	2.7	7.4	13.0	17.2	25.8					
1980	2.6	13.5	22.4	43.6	35.4	2.7	44.8	96.7	122.6	37.4
1981	2.6	7.5	16.8	20.8	31.0	3.2	12.0	19.9	38.1	42.3
1982	2.3	7.0	15.4	17.5	25.3	2.2	10.4	19.8	29.9	37.3
1983	2.5	7.2	13.6	18.9	27.8	2.8	10.4	18.9	35.3	36.5
1984	4.6	7.7	15.0	19.4	22.0	4.4	9.8	16.6	26.7	32.6
1985	4.5	10.3	17.7	27.7	31.2	4.1	13.0	20.2	43.5	40.5
986	3.4	8.2	13.3	19.3	25.0	3.4	10.7	16.1	29.3	35.8
1987	4.2	8.5	14.4	19.4	25.6	6.1	10.9	16.3	32.6	32.2
1988	4.4	7.5	12.0	23.7	27.6	8.6	9.7	14.9	30.0	31.2
1989	5.6	8.1	16.1	15.6	24.3	5.3	14.9	15.8	28.6	31.2
1990	3.7	7.7	15.0	18.0	25.7	12.0	10.2	14.8	27.3	28.7
1991	3.8	7.1	12.5	14.6	19.2	3.3	8.4	11.6	22.7	16.8
1992	4.1	7.1	11.7	17.8	18.0	4.2	7.6	12.5	14.0	16.7
1993	2.8	8.2	13.8	16.2	17.8	3.3	9.3	12.8	26.4	28.5
1994	4.3	8.4	13.8	17.8	23.8	4.4	8.9	13.7	25.6	29.4
1996	3.6	6.8	9.3	13.8	16.4	3.3	8.1	12.4	23.3	19.1
1998	5.4	12.4	13.9	16.3	21.2	4.6	12.9	15.3	21.5	25.9

TABLE AI-I

MEAN CONCENTRATION OF 0.1*M* HCI-EXTRACTABLE Cd IN SOIL

TABLE AI-II

	Ann		Earth Rat	e (Mg/ha Plots	a)			Earth Rat	ν U	a)
Year	0	20	40	80	100	0	20	40	80	100
-					mg/k	g				
1977	11.7	28.8	29.4	51.4	83.7					
1978	16.1	41.0	65.3	128.5	111.7					
1979	16.4	32.8	61.3	84.3	123.8					
1980	19.0	66.7	109.1	208.3	199.3	19.8	45.2	92.4	146.2	215.8
1981	7.1	31.1	95.4	92.3	81.8	8.1	54.7	116.3	162.7	126.5
1982	16.5	42.2	77.6	101.4	135.8	16.0	59.4	105.9	173.7	202.2
1983	18.8	42.8	75.2	107.4	154.7	20.2	61.5	106.3	205.0	206.3
1984	15.1	26.2	54.4	74.4	88.4	14.6	35.4	62.2	108.8	130.8
1985	29.1	55.9	89.0	147.0	168.5	25.1	71.3	105.8	233.3	223.0
1986	18.3	40.4	67.5	102.1	131.8	18.1	54.3	84.2	160.0	193.0
1987	21.3	46.2	76.0	110.2	147.0	22.0	59.7	91.4	196.7	190.2
1988	26.1	44.8	66.5	134.9	153.0	47.5	59.8	82.8	169.5	174.2
1989	28.9	44.4	90.2	92.2	142.3	27.5	80.8	90.3	170.8	185.3
1990	25.0	50.5	85.8	106.0	153.7	22.2	64.0	88.5	163.5	171.8
1991	27.0	43.7	72.1	88.2	112.5	24.5	52.6	68.2	138.5	101.3
1992	14.3	24.4	37.6	64.5	66.4	14.7	25.8	41.2	50.3	59.9
1993	19.3	45.6	72.2	89.8	92.8	22.0	52.1	68.9	139.0	151.7
1994	27.2	49.0	76.7	98.8	134.7	27.1	54.2	74.9	145.4	132.6
1996	21.7	39.3	52.0	77.6	90.1	21.4	47.0	68.8	121.3	106.9
1998	34.1	63.9	78.4	87.8	111.9	28.7	71.4	85.9	115.6	144.2

MEAN CONCENTRATION OF 0.1*M* HCI-EXTRACTABLE Cu IN SOIL

	Ann		earth Rate Treated	e (Mg/ha Plots	.)	Ann		arth Rate Treated	e (Mg/ha Plots)
Year	0	20	40	80	100	0	20	40	80	100
					mg/kg	g				
1977	5.9	6.0	6.7	17.2	17.3					
1978	2.6	8.7	15.9	35.7	33.8					
1979	5.0	10.1	20.4	27.5	42.5					
1980	4.0	18.4	36.1	62.7	63.9	3.7	11.8	29.0	42.4	72.3
1981	3.7	12.7	32.1	38.6	62.7	4.7	22.9	40.4	81.3	91.6
1982	2.8	12.1	27.8	35.4	51.9	3.1	18.0	40.3	65.4	77.3
1983	3.4	11.6	24.6	35.1	52.8	4.1	18.0	37.4	70.8	71.7
1984	2.3	5.1	11.3	14.1	18.2	2.3	6.7	13.0	21.3	29.4
1985	8.4	16.9	26.9	46.0	60.7	6.4	19.4	31.5	74.6	85.6
1986	4.1	10.5	22.0	32.0	41.8	5.3	16.1	29.4	54.5	64.6
1987	6.8	15.8	29.1	39.9	56.2	6.9	20.4	34.3	73.7	73.7
1988	7.2	11.4	20.4	35.3	63.1	16.6	15.0	20.6	50.6	71.7
1989	9.0	12.6	28.1	27.0	43.8	7.8	25.0	27.9	54.8	58.9
1990	6.7	15.5	29.5	36.2	55.3	5.7	20.5	29.2	52.8	64.0
1991	7.5	13.7	23.4	29.7	41.2	6.4	17.2	22.4	50.9	38.2
1992	1.7	3.4	5.5	10.1	10.9	1.7	3.5	6.9	7.6	9.3
1993	3.0	10.8	18.2	23.6	25.6	3.9	12.7	17.5	39.0	43.5
1994	6.3	13.4	22.0	29.8	42.6	6.8	14.5	21.7	46.6	51.3
1996	5.0	10.5	13.8	23.3	27.6	4.9	13.2	20.0	38.1	32.5
1998	6.4	17.2	17.9	22.8	30.3	5.4	18.7	22.1	34.2	39.3

TABLE AI-III

MEAN CONCENTRATION OF 0.1*M* HCI-EXTRACTABLE Cr IN SOIL

	Ann	ual Nu E 3-Years			a)	An		Earth Ra s Treatec	te (Mg/ha l Plots	a)
Year	0	20	40	80	100	0	20	40	80	100
-					mg/k	g				
1977	17.7	30.9	33.4	53.9	72.9					
1978	19.5	42.9	57.6	106.5	91.9					
1979	24.5	37.9	58.0	79.4	105.2					
1980	27.6	54.3	92.7	171.2	167.5	29.7	37.6	88.8	120.4	181.8
1981	23.3	46.5	82.7	107.1	149.7	22.9	69.4	106.0	180.0	190.2
1982	22.2	45.2	72.5	95.1	122.3	22.6	59.2	95.3	149.7	172.2
1983	23.7	43.5	66.0	94.8	135.5	24.8	57.4	90.4	166.2	163.7
1984	22.3	29.7	45.8	61.2	65.2	21.2	34.4	50.0	79.2	90.3
1985	28.4	53.1	82.9	121.7	134.0	26.1	ND	90.7	176.2	166.0
1986	23.6	40.9	60.3	93.9	106.5	22.1	51.0	72.1	128.3	147.7
1987	33.7	51.8	72.6	102.6	129.3	33.2	60.5	82.3	158.0	156.7
1988	31.7	47.6	63.1	116.6	137.7	49.2	54.0	70.9	143.8	156.8
1989	28.6	44.3	76.1	76.6	117.3	28.5	72.0	75.2	132.3	141.7
1990	31.7	53.3	77.5	96.3	130.8	28.5	64.3	77.0	129.8	141.3
1991	30.7	46.0	64.9	82.9	97.2	29.8	53.2	61.4	116.3	87.2
1992	10.3	15.8	20.6	34.1	35.0	10.6	16.9	23.9	26.0	30.7
1993	23.5	43.6	62.8	78.2	76.8	25.8	50.2	57.5	113.0	118.4
1994	30.8	49.5	68.8	91.1	108.7	29.9	52.5	67.1	123.2	130.2
1996	26.1	39.8	45.6	72.8	80.5	25.4	49.8	62.8	105.9	90.7
1998	35.4	60.9	63.8	74.5	90.6	30.3	62.8	68.8	94.4	110.1

TABLE AI-IV

MEAN CONCENTRATION OF 0.1*M* HCI-EXTRACTABLE Pb IN SOIL

	AIII	ual Nu E 3-Years	Treated	. 0	.)	All		s Treated	ite (Mg/ha d Plots	ノ
Year	0	20	40	80	100	0	20	40	80	100
-					mg/k	g				
1977	6.3	10.8	12.9	21.8	28.7					
1978	6.3	11.9	18.2	29.0	39.0					
1979	7.4	10.8	16.0	20.2	28.0					
1980	9.4	18.1	31.9	47.6	50.0	10.5	15.4	31.8	32.2	53.7
1981	6.5	13.0	23.1	27.2	39.1	7.5	19.4	30.2	53.7	60.2
1982	5.8	11.3	19.0	22.4	30.2	5.5	14.7	27.6	40.1	50.2
1983	6.7	9.6	16.7	22.5	31.5	8.9	11.1	23.2	48.8	51.6
1984	9.9	11.1	17.0	21.8	26.1	9.4	13.7	20.8	30.1	39.4
1985	19.7	18.3	27.0	40.2	42.7	10.6	20.6	32.6	60.3	50.4
986	5.9	8.5	17.6	21.5	30.6	6.2	14.9	23.4	35.6	47.9
1987	8.2	15.5	24.1	29.9	37.8	9.0	18.7	73.3	51.1	49.8
1988	9.6	14.1	17.8	38.0	42.2	17.1	19.7	23.1	48.0	51.7
1989	12.3	21.4	24.1	25.4	35.2	13.0	29.9	23.4	35.5	46.2
1990	9.3	15.5	26.0	28.5	42.0	8.3	18.7	26.5	43.8	47.7
1991	9.8	13.2	21.5	23.8	30.5	9.1	16.3	20.2	38.1	29.6
1992	7.1	9.0	11.2	16.6	16.3	6.8	9.1	12.7	14.8	16.4
1993	5.7	10.8	14.8	17.3	18.1	5.9	11.7	15.0	26.0	28.7
1994	7.0	11.6	16.3	20.8	25.6	6.9	12.9	17.0	29.5	33.7
1996	7.2	9.8	11.4	16.3	17.6	6.7	11.8	15.4	24.8	21.7
1998	8.2	13.8	14.0	17.3	19.5	7.3	14.7	17.3	20.8	25.1

TABLE AI-V

MEAN CONCENTRATION OF 0.1*M* HCI-EXTRACTABLE Ni IN SOIL

	Anı	nual Nu I 3-Years	Earth Rat		Annual Nu Earth Rate (Mg/ha) 5-Years Treated Plots						
Year	0	20	40	80	100	0	20	40	80	100	
-					mg/k	g					
1977	27.4	80.7	90.8	186.7	266.8						
1978	36.2	107.9	197.2	387.3	347.0						
1979	75.5	119.2	223.3	298.0	398.1						
1980	41.8	174.8	297.8	575.5	554.0	45.2	120.6	258.2	383.7	611.7	
1981	43.8	117.1	278.2	334.2	521.7	53.4	202.0	454.7	698.3	766.7	
1982	36.5	113.7	208.5	274.2	379.2	35.7	155.3	306.5	499.0	593.(
1983	50.7	116.7	208.5	287.2	429.3	53.9	171.2	294.7	574.2	599.7	
1984	47.6	85.2	149.8	220.0	275.3	42.6	109.3	178.8	326.2	425.2	
1985	89.3	175.0	289.0	458.3	500.3	75.2	218.2	334.3	728.7	673.3	
1986	56.4	120.4	210.0	289.8	366.7	52.7	159.0	261.7	456.5	546.8	
1987	68.4	141.5	238.8	333.7	431.2	71.2	182.0	275.5	579.7	554.5	
1988	81.9	143.0	201.7	353.7	339.0	144.3	189.8	261.3	385.8	341.8	
1989	74.7	120.3	244.8	244.5	393.7	68.9	221.3	250.3	446.3	509.7	
1990	82.0	153.5	268.8	325.2	476.7	70.7	198.3	275.2	496.3	532.8	
1991	88.9	138.3	226.9	272.4	354.2	77.8	164.2	212.1	426.9	317.9	
1992	63.9	96.4	142.0	225.6	227.5	61.8	100.4	159.7	184.4	216.7	
1993	51.1	121.2	179.5	231.8	240.6	58.1	134.8	171.9	356.0	391.8	
1994	76.0	130.8	193.7	261.9	338.6	76.8	141.5	208.5	370.8	428.4	
1996	58.4	101.5	123.9	195.1	220.4	57.6	118.4	171.2	306.5	259.8	
1998	88.2	182.5	172.1	232.7	267.4	78.1	184.2	207.4	288.6	343.7	

TABLE AI-VI

MEAN CONCENTRATION OF 0.1*M* HCl-EXTRACTABLE Zn IN SOIL

TABLE AI-VII

CONCENTRATION OF 0.1*M* HCI-EXCTRACTABLE METALS IN SOIL SAMPLES COLLECTED IN 1990 AT FIFTEEN LOCATIONS WITHIN REPLICATE 1 OF THE CONTROL

Sample Location	Cd	Cr	Cu	Ni	Pb	Zn	Fe	Mn	Al
				r	ng/kg				
1	5.4	49.0	51.5	29.0	58.0	157	18778	257	15291
2	4.6	47.0	41.5	29.5	43.0	126	19744	315	17213
3	5.3	48.5	44.0	31.0	63.5	131	19322	343	17250
4	4.7	42.5	41.0	31.5	47.5	114	20438	291	18207
5	4.8	45.5	40.5	31.0	51.5	119	19407	299	16697
6	4.7	45.5	39.5	30.0	43.0	123	20410	308	16406
7	5.5	47.5	48.5	37.0	64.0	170	20325	288	16407
8	4.3	40.5	37.0	31.0	42.0	127	20953	328	21451
9	4.6	44.5	39.0	32.5	43.5	133	24638	339	16510
10	4.2	37.0	35.5	31.0	47.0	117	19201	272	19172
11	4.0	40.5	38.5	29.0	44.5	110	23306	315	25566
12	3.7	40.5	47.0	27.0	40.0	117	23935	263	19613
13	4.7	48.0	48.0	29.5	52.5	155	19219	304	19510
14	3.9	42.5	38.5	26.5	39.5	113	21675	297	21366
15	3.3	40.0	36.0	28.5	36.5	99	21516	382	19641

TABLE AI-VIII

CONCENTRATION OF 0.1*M* HCI-EXCTRACTABLE METALS IN SOIL SAMPLES COLLECTED IN 1990 AT FIFTEEN LOCATIONS WITHIN REPLICATE 2 OF THE CONTROL

Sample Location	Cd	Cr	Cu	Ni	Pb	Zn	Fe	Mn	Al
				r	ng/kg				
1	5.0	65.2	47.2	28.8	35.9	144	22760	263	18163
2	3.9	54.2	41.1	26.5	32.9	122	20529	262	16269
3	4.4	57.1	46.1	28.7	41.5	133	22466	295	18209
4	5.0	54.3	41.0	28.5	30.6	126	22675	312	17787
5	3.8	47.6	37.4	26.1	30.5	111	21447	250	17478
6	3.7	45.3	36.0	28.5	28.8	107	20913	276	16794
7	2.9	44.1	34.9	28.0	27.1	106	22123	268	17375
8	2.1	39.8	32.7	28.0	26.8	96	20050	228	16287
9	2.5	42.9	35.3	30.2	28.7	103	21326	254	17234
10	2.6	34.9	29.9	26.9	25.5	88	19188	218	14741
11	2.1	38.1	34.0	19.9	31.4	108	19441	262	18190
12	1.7	33.7	32.9	19.2	30.3	102	19029	235	17666
13	2.8	41.0	35.4	21.6	32.1	115	20060	309	18866
14	1.4	31.8	29.4	20.6	26.1	92	19347	251	18144
15	1.6	38.1	37.6	22.2	31.9	116	20238	272	18444

TABLE AI-IX

CONCENTRATION OF 0.1*M* HCI-EXCTRACTABLE METALS IN SOIL SAMPLES COLLECTED IN 1990 AT FIFTEEN LOCATIONS WITHIN REPLICATE 3 OF THE CONTROL

Sample Location	Cd	Cr	Cu	Ni	Pb	Zn	Fe	Mn	Al
				r	ng/kg				
1	5.4	52.5	35.5	19.5	34.0	109	13013	198	11215
2	6.9	66.5	46.0	25.0	47.0	143	18300	236	15143
3	7.9	77.5	57.0	29.0	63.0	187	18750	248	14955
4	5.6	59.0	45.5	27.0	41.5	128	20653	242	17037
5	6.0	61.5	49.5	26.0	44.5	135	22313	249	18593
6	5.1	52.0	47.5	26.5	44.0	129	19051	248	16324
7	3.9	43.5	42.0	24.5	38.0	108	18010	265	15349
8	4.1	47.0	43.5	26.5	40.5	117	19435	255	15846
9	5.3	51.5	49.5	25.5	52.0	141	20550	212	16990
10	3.7	38.5	40.5	24.0	36.0	103	18319	230	15705
11	2.5	35.0	33.0	24.5	36.5	98	19744	250	16483
12	2.4	36.0	36.0	22.0	42.0	111	18938	214	17533
13	1.7	37.0	33.5	22.5	39.5	105	17935	234	17309
14	2.0	38.0	35.0	23.0	45.0	115	17288	236	16596
15	2.3	41.5	41.5	24.5	51.5	127	17025	232	14880

TABLE AI-X

CONCENTRATION OF 0.1*M* HCI-EXCTRACTABLE METALS IN SOIL SAMPLES COLLECTED IN 1990 AT FIFTEEN LOCATIONS WITHIN REPLICATE 1 OF THE 100 Mg/ha NU EARTH TREATMENT

Sample Location	Cd	Cr	Cu	Ni	Pb	Zn	Fe	Mn	Al
]	mg/kg				
1	14.5	137.5	110.5	46.5	116.0	327	16918	262	12687
2	26.0	259.5	165.0	65.0	148.0	452	16169	353	12254
3	24.5	274.5	163.5	59.0	142.0	447	16313	294	12773
4	18.0	176.0	127.5	53.5	125.0	390	18707	368	14129
5	26.0	278.5	172.5	66.5	152.5	462	18159	267	14648
6	24.0	276.0	162.0	63.5	142.5	436	12736	257	11129
7	20.5	205.5	145.5	56.0	142.5	473	15938	258	12802
8	31.0	347.5	182.0	80.5	187.0	583	17380	306	14475
9	26.0	305.5	177.5	67.0	267.5	491	17178	280	14677
10	20.5	224.0	147.0	59.0	142.5	413	15765	275	13264
11	27.0	298.0	194.0	64.0	167.5	534	18505	357	13494
12	26.5	315.0	196.0	62.0	170.0	534	22025	360	17216
13	13.5	156.0	122.0	43.0	125.0	374	20178	305	16321
14	22.0	265.5	177.0	57.0	152.0	483	25486	334	20793
15	21.0	254.5	160.0	54.0	137.5	440	21996	364	18716

TABLE AI-XI

CONCENTRATION OF 0.1*M* HCI-EXCTRACTABLE METALS IN SOIL SAMPLES COLLECTED IN 1990 AT FIFTEEN LOCATIONS WITHIN REPLICATE 2 OF THE 100 Mg/ha NU EARTH TREATMENT

Sample Location	Cd	Cr	Cu	Ni	Pb	Zn	Fe	Mn	Al
-]	mg/kg				
1	20.5	216.5	123.5	47.0	109.0	375	16962	274	16471
2	18.5	210.0	122.5	46.5	96.0	368	18433	272	18895
3	18.5	232.0	129.5	51.0	105.0	379	20683	274	21577
4	20.0	296.0	164.0	59.0	134.0	479	20020	265	19241
5	22.0	281.0	156.5	54.5	129.0	457	21462	273	20337
6	18.5	290.5	141.5	54.5	108.0	413	20683	514	21490
7	23.5	322.5	185.5	62.0	163.5	558	19269	278	19788
8	28.5	388.0	210.0	66.0	168.5	631	22875	303	22529
9	27.5	348.0	191.0	60.5	152.5	560	22096	272	25010
10	26.5	354.0	198.0	60.0	175.0	587	17683	248	18837
11	18.0	244.0	149.0	57.5	133.5	436	23106	234	23568
12	13.5	184.0	124.0	55.5	100.5	347	23798	253	21058
13	20.0	251.5	159.5	58.0	145.5	487	21750	254	21289
14	13.5	206.0	126.5	52.5	110.5	367	23193	246	22731
15	12.5	202.0	129.0	46.5	114.0	372	21173	243	21808

TABLE AI-XII

CONCENTRATION OF 0.1*M* HCI-EXCTRACTABLE METALS IN SOIL SAMPLES COLLECTED IN 1990 AT FIFTEEN LOCATIONS WITHIN REPLICATE 3 OF THE 100 Mg/ha NU EARTH TREATMENT

Sample Location	Cd	Cr	Cu	Ni	Pb	Zn	Fe	Mn	Al
_]	mg/kg				
1	30.9	359.5	201.0	58.6	166.5	624	13619	287	12912
2	27.7	307.5	168.5	54.0	150.5	516	15926	266	14498
3	25.4	292.0	167.0	53.7	129.0	505	14340	261	13316
4	32.1	328.0	189.0	60.3	182.5	583	14426	273	13575
5	20.5	271.0	154.0	50.5	127.0	462	16907	270	15681
6	27.1	335.5	184.0	69.5	171.0	557	16849	270	15594
7	29.7	390.0	202.5	74.2	192.0	618	17686	324	18277
8	26.6	309.0	172.0	66.4	147.5	528	17801	266	17325
9	24.6	311.5	168.5	65.2	156.0	511	19359	254	19518
10	18.8	252.5	146.5	62.3	126.0	444	18176	237	18912
11	16.2	207.0	127.0	42.1	109.5	399	19792	255	18796
12	12.4	152.0	99.0	38.1	86.0	312	21926	268	20239
13	16.8	208.0	129.0	47.1	111.0	413	21955	257	20152
14	10.4	179.0	114.0	48.5	91.0	352	22157	261	20325
15	15.0	183.5	118.5	44.4	95.5	378	19647	238	18595

		ual Nu E 3-Years		· •	a)			arth Rate)
Year	0	20	40	80	100	0	20	40	80	100
					mg	g/kg				
1977	0.51	1.04	0.98	2.03	1.95					
1978	0.35	0.98	1.11	2.00	1.75					
1979	0.22	1.39	1.60	2.69	2.94					
1980	0.85	3.09	6.18	8.79	8.25	0.89	2.31	4.31	7.18	9.10
1981	1.10	2.13	2.22	3.42	3.63	1.20	2.27	3.52	4.21	4.78
1982	0.61	1.67	2.34	4.36	5.21	0.72	2.26	3.41	6.74	6.86
1983	0.71	1.75	1.96	1.83	4.88	0.68	2.36	2.64	3.01	6.26
1984	0.74	2.17	2.85	3.96	5.15	1.70	1.56	2.39	3.17	3.71
1985	0.67	1.55	2.01	2.31	3.59	1.45	1.63	2.50	2.38	4.08
1986	0.52	0.55	0.89	2.53	3.52	0.37	1.15	1.88	ND	3.64
1987	0.38	0.79	1.16	1.03	1.65	0.57	0.94	1.25	1.99	2.82
1988	1.06	1.47	1.80	2.54	3.30	0.93	1.94	2.21	3.18	4.62
1989	0.58	1.25	1.54	2.70	3.96	0.71	1.36	2.63	3.43	5.00
1990	1.11	3.02	3.79	6.36	6.12	1.21	1.89	3.79	6.04	9.51
1991	0.74	2.17	2.49	3.92	4.52	0.76	1.90	3.22	5.07	6.20
1992	0.67	1.51	3.25	3.43	4.80	0.73	1.53	2.84	4.24	5.82
1993	0.81	1.10	2.87	2.64	4.70	1.25	2.33	2.99	3.02	7.45
1994	0.65	1.05	1.42	1.92	3.11	0.32	1.33	3.14	3.75	3.98
1996	1.06	1.84	1.95	4.56	6.16	1.15	1.88	3.20	4.88	7.90
1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

TABLE AII-I: MEAN CONCENTRATION OF Cd IN BEET

		ual Nu E 3-Years		ν U	ia)	An		Earth Ra	ate (Mg/ha) d Plots)
Year	0	20	40	80	100	0	20	40	80	100
-					mg	/kg				
1977	1.04	1.25	1.30	1.83	1.73					
1978	0.14	0.67	0.60	0.89	0.72					
1979	0.67	1.41	1.16	1.90	2.30					
1980	1.51	3.08	3.20	3.80	3.36	1.83	4.57	6.51	2.67	3.98
1981	1.27	2.80	3.86	3.76	4.77	1.53	2.80	4.09	4.53	4.76
1982	1.71	3.80	3.85	5.42	5.78	1.34	4.02	4.86	6.68	7.80
1983	1.32	1.77	2.83	3.44	3.10	1.33	2.36	2.98	3.72	3.33
1984	0.42	1.52	2.29	1.99	3.32	0.77	1.65	2.31	2.57	2.48
1985	0.59	1.43	1.64	2.92	2.30	0.68	1.73	1.83	1.78	2.50
1986	1.09	2.71	2.83	3.85	4.48	1.30	2.74	3.03	5.33	5.39
1987	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1988	2.49	3.43	5.39	6.51	10.97	2.84	4.59	6.66	6.77	7.25
1989	ND	3.64	3.24	3.39	4.77	1.55	2.64	3.81	3.56	5.77
1990	2.68	5.59	8.60	4.70	8.65	2.69	6.51	8.53	7.85	7.95
1991	1.40	3.52	2.60	3.60	5.06	1.47	2.92	4.14	5.21	5.61
1992	1.12	1.76	1.66	2.49	3.14	1.03	2.21	2.17	2.91	3.40
1993	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1994	ND	1.48	1.72	2.07	3.55	1.31	1.50	2.70	2.73	2.71
1996	ND	1.70	2.90	6.95	5.15	ND	1.33	7.60	5.82	1.80
1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

TABLE AII-II: MEAN CONCENTRATION OF Cd IN CARROT

	Ann	ual Nu H 3-Years	Earth Ra Treated		a)	Am	nual Nu H 5-Years	Earth Rat		ι)
Year	0	20	40	80	100	0	20	40	80	100
-					m	ng/kg				
1977	5.38	11.88	18.80	16.87	14.57					
1978	2.94	4.82	5.17	7.30	3.71					
1979	3.97	7.88	6.42	9.02	7.55					
1980	2.57	4.15	7.22	6.91	7.88	2.60	3.95	7.45	8.68	8.24
1981	4.69	9.42	12.68	14.85	19.70	5.70	11.19	14.10	19.18	14.52
1982	5.75	11.25	17.17	10.56	11.79	10.92	13.90	17.48	15.98	21.20
1983	8.08	21.37	28.32	31.68	33.82	8.52	23.95	26.55	33.00	29.35
1984	3.99	12.77	12.31	15.44	30.16	5.94	11.74	16.95	16.88	20.10
1985	4.61	12.55	12.81	16.52	19.30	5.24	12.54	15.65	20.42	18.83
1986	4.55	9.20	13.15	12.95	12.67	5.07	11.05	12.80	17.97	14.52
1987	6.05	12.60	10.98	15.85	13.22	5.58	13.04	12.88	16.88	16.33
1988	7.91	22.80	25.35	27.33	28.23	9.06	20.27	23.05	29.82	27.68
1989	6.16	14.75	23.50	26.15	16.50	7.55	16.80	13.20	20.48	16.45
1990	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1991	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1992	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1993	ND	ND	ND	18.23	ND	ND	13.93	ND	25.85	ND
1994	7.45	8.70	16.94	19.24	19.35	8.60	11.04	12.48	24.88	19.00
1996	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1998	9.15	ND	19.55	16.30	ND	8.18	32.45	20.85	19.55	15.73

TABLE AII-III: MEAN CONCENTRATION OF Cd IN SPINACH

			Earth Ra Treated	te (Mg/h Plots	a)	Anr		Earth Rat		ı)
Year	0	20	40	80	100	0	20	40	80	100
-					m	g/kg				
1977	2.20	4.88	8.43	7.03	10.22					
1978	1.31	5.70	6.18	11.46	11.48					
1979	1.80	5.01	7.62	10.52	13.84					
1980	1.81	9.36	12.85	21.45	23.40	2.03	4.57	10.62	17.85	26.30
1981	2.71	4.16	11.86	15.95	21.38	2.84	8.50	14.32	21.50	24.93
1982	2.14	9.91	12.40	15.81	16.83	2.65	6.22	11.53	14.90	16.15
1983	1.16	3.96	8.06	8.35	12.15	1.25	5.55	11.95	14.12	15.13
1984	0.04	0.01	0.01	0.02	0.03	0.00	0.00	0.02	0.02	0.03
1985	1.37	5.37	8.05	12.14	13.97	6.54	8.59	10.62	14.55	20.15
1986	2.81	6.13	7.76	11.19	14.20	2.76	6.99	9.96	15.02	16.55
1987	8.87	4.09	6.48	4.77	14.05	3.10	7.61	13.75	10.68	6.80
1988	3.65	8.64	12.42	15.32	14.10	3.61	8.09	17.92	15.35	17.63
1989	5.53	6.52	9.90	12.43	11.39	3.98	5.30	11.95	13.67	14.26
1990	1.77	5.80	9.77	14.53	13.55	1.78	5.65	10.41	17.30	17.35
1991	1.36	4.78	8.46	9.74	11.00	1.70	5.33	7.63	11.15	12.32
1992	4.22	3.58	8.41	10.06	15.48	3.72	5.77	8.84	18.13	13.15
1993	2.99	4.55	7.23	13.69	18.16	2.12	4.64	11.16	13.13	13.27
1994	4.13	6.74	8.39	17.42	20.85	3.28	11.01	9.39	16.74	26.67
1996	2.55	8.00	12.63	12.40	15.35	3.22	8.70	9.02	11.05	17.69
1998	2.14	7.13	5.97	8.71	9.35	2.38	6.88	9.52	15.00	13.20

TABLE AII-IV: MEAN CONCENTRATION OF Cd IN SWISS CHARD

		ial Nu Ea 3-Years '		· U)			arth Rate Treated)
Year	0	20	40	80	100	0	20	40	80	100
-					mg	/kg				
1977	1.52	1.34	1.50	1.93	2.04					
1978	0.85	1.95	1.69	2.76	2.95					
1979	1.14	1.78	2.34	2.23	3.37					
1980	1.91	1.50	3.71	4.43	3.92	1.62	1.51	2.98	3.38	3.93
1981	0.88	1.15	1.57	2.06	2.03	0.86	1.42	1.77	2.85	2.43
1982	1.38	2.56	2.92	3.46	3.44	1.35	2.54	3.14	3.83	3.80
1983	2.09	2.21	2.08	2.59	2.92	2.03	2.25	2.21	2.89	3.04
1984	1.13	1.21	1.50	2.02	2.20	0.96	1.50	1.93	2.13	2.53
1985	1.37	1.88	2.26	2.22	2.38	1.53	1.51	2.01	2.18	2.53
1986	2.02	1.95	2.73	2.40	2.50	1.80	1.92	2.58	2.77	2.91
1987	1.17	1.74	ND	1.59	ND	ND	ND	2.89	2.28	ND
1988	1.97	2.48	2.67	2.68	2.54	1.60	2.84	2.26	2.60	3.96
1989	ND	2.36	2.65	3.37	3.04	1.87	1.93	3.33	3.15	2.65
1990	1.76	2.10	2.48	3.21	2.94	2.08	1.97	2.54	3.47	3.07
1991	1.75	2.37	2.13	1.93	3.41	1.45	2.26	2.35	2.36	3.76
1992	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1993	1.44	1.40	1.96	1.62	2.44	1.44	1.20	2.04	1.68	2.86
1994	1.10	1.54	1.84	2.16	2.29	0.99	1.83	ND	1.89	1.87
1996	1.08	0.88	ND	2.25	2.29	1.02	1.21	2.03	1.96	1.50
1998	0.56	1.16	1.49	1.50	1.85	0.00	1.35	1.55	1.63	2.11

TABLE AII-V: MEAN CONCENTRATION OF Cd IN TOMATO

		ual Nu E 3-Years			a)	Ann		Earth Rate Treated I	· U /)
Year	0	20	40	80	100	0	20	40	80	100
-					mg/	/kg				
1977	0.07	0.11	0.21	0.31	0.26					
1978	0.31	0.26	0.39	0.50	0.45					
1979	0.40	0.27	0.48	0.46	0.56					
1980	0.09	0.14	0.20	0.33	0.43	0.15	0.13	0.23	0.34	0.41
1981	0.07	0.19	0.28	0.16	0.30	0.07	0.10	0.18	0.37	0.34
1982	0.12	0.23	0.34	0.37	0.39	0.13	0.15	0.34	0.47	0.54
1983	0.04	0.08	0.22	0.00	0.15	0.04	0.08	0.26	0.34	0.55
1984	0.21	0.00	0.00	0.00	0.00	0.00	0.00	0.48	0.22	0.00
1985	0.03	0.00	0.05	0.00	0.00	0.02	0.00	0.00	0.04	0.06
1986	0.29	0.33	0.27	0.23	0.26	0.17	0.38	0.23	0.32	0.43
1987	0.22	0.10	0.13	0.17	0.17	0.10	0.13	0.12	0.14	0.18
1988	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1989	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1990	0.03	0.05	0.10	0.11	0.17	0.01	0.09	0.10	0.21	0.15
1991	0.03	0.12	0.18	0.22	0.36	0.03	0.19	0.16	0.30	0.59
1992	0.03	0.09	0.20	0.18	0.27	0.02	0.09	0.19	0.19	0.33
1993	0.07	0.12	0.11	0.14	0.23	0.09	0.09	0.03	0.14	0.19
1994	0.00	0.00	0.01	0.01	0.01	0.01	0.00	0.01	0.01	0.01
1996	0.00	0.00	0.00	0.00	0.67	0.00	0.00	0.18	0.00	0.06
1998	0.00	0.00	0.50	0.00	0.29	0.00	0.00	0.00	0.00	0.00

TABLE AII-VI: MEAN CONCENTRATION OF Cd IN GREEN BEANS

		ial Nu E 3-Years		· •	a)	Anı	ual Nu E 5-Years	Earth Rate Treated I		
Year	0	20	40	80	100	0	20	40	80	100
-					r	ng/kg				
1977	0.14	0.30	0.13	0.16	0.25					
1978	0.09	0.07	0.09	0.07	0.11					
1979	0.23	0.80	0.51	0.58	0.60					
1980	2.99	3.25	2.78	3.05	2.77	2.77	3.47	2.86	3.06	2.70
1981	0.19	0.21	0.20	0.19	0.18	0.13	0.15	0.13	0.19	0.25
1982	5.21	4.91	4.24	5.67	7.98	4.62	5.48	4.41	7.34	8.88
1983	1.30	1.66	1.56	1.62	3.09	0.81	1.73	1.63	1.77	1.61
1984	0.74	1.19	1.30	1.29	1.72	0.65	0.98	0.95	1.08	1.04
1985	1.38	1.41	1.67	1.22	1.88	0.66	1.23	1.88	1.16	1.57
1986	0.05	0.04	0.09	0.12	0.14	0.10	0.06	0.20	ND	0.04
1987	0.06	0.26	0.20	0.55	0.12	0.09	0.04	1.04	0.09	0.13
1988	0.17	0.31	0.26	0.10	0.42	0.13	0.74	0.41	0.16	0.25
1989	0.04	0.09	0.10	0.25	0.20	0.14	0.08	0.07	0.16	0.22
1990	0.04	0.09	0.18	0.50	0.23	0.13	0.10	0.27	0.24	0.88
1991	0.08	0.13	0.09	0.13	0.24	0.11	0.17	0.11	0.07	0.20
1992	0.12	0.18	0.22	0.28	0.33	0.30	0.10	0.27	0.30	0.27
1993	0.15	0.20	0.15	0.57	0.20	0.15	0.25	0.10	0.10	0.30
1994	0.13	0.10	0.22	0.10	0.10	0.10	0.10	0.13	0.15	0.10
1996	0.20	0.70	0.10	0.17	0.47	0.17	0.35	0.70	0.40	0.22
1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	NE

TABLE AIII-I: MEAN CONCENTRATION OF Cr IN BEET

			arth Rate Treated	· •	a)	All		Earth Rate Treated I		
Year	0	20	40	80	100	0	20	40	80	100
-					n	ng/kg				
1977	0.14	0.14	0.07	0.12	0.14					
1978	0.02	0.03	0.01	0.02	0.01					
1979	1.74	1.77	1.81	1.49	1.90					
1980	3.88	4.14	3.19	4.01	3.95	3.07	4.78	4.30	2.43	4.20
1981	0.27	0.44	1.03	0.68	0.52	0.30	0.33	0.51	0.48	0.40
1982	1.68	1.38	1.42	1.59	1.29	1.28	1.36	1.75	1.55	2.16
1983	2.69	3.78	2.34	3.08	3.08	3.25	3.12	3.22	2.65	2.78
1984	0.60	2.19	0.52	1.18	2.66	1.13	0.88	0.83	0.64	1.59
1985	1.17	1.25	1.46	3.23	1.51	1.15	1.50	1.28	1.46	1.86
1986	1.07	0.64	0.69	0.81	0.46	0.78	0.71	0.47	0.85	0.59
1987	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1988	0.05	0.10	0.34	0.86	1.86	0.10	0.50	0.32	0.80	1.07
1989	0.29	0.16	0.25	0.32	0.25	0.18	0.15	0.42	0.44	0.52
1990	0.12	0.18	0.59	0.28	0.58	0.16	0.29	0.38	0.43	0.44
1991	0.00	0.02	5.65	0.08	2.92	0.10	0.07	2.63	0.17	0.15
1992	0.10	0.08	0.08	0.17	0.17	0.10	0.17	0.17	0.20	0.23
1993	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1994	ND	0.17	0.25	0.30	0.20	0.40	0.25	0.35	0.35	0.43
1996	ND	0.00	0.00	1.05	0.00	ND	0.00	0.50	0.10	0.70
1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

TABLE AIII-II: MEAN CONCENTRATION OF Cr IN CARROT

		ial Nu E 3-Years			a)	Anı		Earth Rate Treated I		
Year	0	20	40	80	100	0	20	40	80	100
-					m	g/kg				
1977	1.04	1.37	1.88	2.99	1.76					
1978	0.35	1.13	0.73	0.47	0.63					
1979	1.02	2.03	1.84	2.60	2.91					
1980	2.00	1.54	3.17	3.50	6.47	2.54	1.17	3.65	1.52	3.63
1981	4.86	3.81	3.17	3.20	2.85	3.10	3.77	3.35	3.52	3.18
1982	0.53	0.73	0.59	0.59	0.53	0.62	0.61	0.57	0.80	0.67
1983	8.23	6.90	8.46	7.81	8.39	7.45	6.86	7.92	8.29	7.90
1984	2.43	2.64	2.46	2.36	2.71	1.95	1.85	1.77	2.29	2.62
1985	1.79	1.99	2.19	2.45	2.58	1.91	2.44	2.51	2.92	2.84
1986	0.67	0.80	1.53	1.39	1.37	0.21	1.16	1.89	1.51	1.49
1987	0.32	0.64	0.44	0.64	0.64	0.93	2.32	2.00	0.36	1.85
1988	0.94	2.06	4.34	4.05	4.46	3.24	3.89	5.78	2.45	3.87
1989	1.18	1.67	1.65	3.32	1.76	1.75	2.15	0.84	4.44	1.25
1990	ND	ND	ND	ND	ND	ND	ND	ND	ND	NE
1991	ND	ND	ND	ND	ND	ND	ND	ND	ND	NE
1992	ND	ND	ND	ND	ND	ND	ND	ND	ND	NE
1993	ND	ND	ND	1.10	ND	ND	1.75	ND	1.45	NE
1994	0.75	0.70	0.70	0.98	0.65	0.75	0.63	0.55	0.88	0.88
1996	ND	ND	ND	ND	ND	ND	ND	ND	ND	NE
1998	0.00	ND	0.00	0.00	ND	0.00	0.00	0.00	0.00	0.00

TABLE AIII-III: MEAN CONCENTRATION OF Cr IN SPINACH

		ial Nu E 3-Years		, U	a)	Anr		Earth Rate Treated I	νų,	
Year	0	20	40	80	100	0	20	40	80	100
					m	ıg/kg				
1977	0.49	0.58	0.59	1.05	1.62					
1978	1.75	1.76	1.65	1.79	1.90					
1979	0.55	0.79	0.71	0.69	0.66					
1980	3.06	3.18	3.06	3.30	3.75	3.02	2.97	3.16	2.90	3.73
1981	0.64	0.89	0.69	0.45	0.67	0.63	0.74	0.58	0.57	0.92
1982	4.06	4.76	4.04	3.15	4.32	3.43	4.44	3.87	3.59	3.17
1983	2.78	2.80	3.92	2.45	3.36	2.26	3.29	2.74	2.27	2.07
1984	2.66	1.43	1.75	1.88	2.08	2.07	1.81	1.61	1.19	1.62
1985	0.33	1.02	1.20	1.22	1.54	0.23	1.45	1.24	0.94	1.05
1986	0.72	0.88	1.20	0.94	1.17	1.17	1.59	0.72	1.54	0.98
1987	0.72	0.39	0.80	0.85	0.85	0.35	0.92	0.93	0.92	0.90
1988	0.40	0.58	2.10	0.97	1.20	0.41	0.76	1.08	1.37	1.02
1989	1.33	1.05	1.39	0.79	1.70	0.94	1.63	2.76	1.14	1.36
1990	0.97	1.12	1.25	1.75	1.36	0.79	1.21	1.63	2.09	1.70
1991	0.46	0.57	0.46	1.03	0.47	0.47	0.34	0.36	0.80	0.69
1992	0.88	0.63	0.57	1.00	0.88	0.58	0.52	0.52	0.50	0.52
1993	0.52	1.15	0.68	0.80	1.05	0.65	0.85	1.17	0.68	1.33
1994	0.82	0.65	0.82	1.17	1.15	0.88	0.98	0.63	1.13	0.73
1996	0.73	0.32	0.70	0.98	0.55	0.38	0.70	0.55	0.65	0.80
1998	0.00	0.50	1.38	0.00	0.00	0.00	0.00	0.00	0.00	0.00

TABLE AIII-IV: MEAN CONCENTRATION OF Cr IN SWISS CHARD

		ial Nu E 3-Years			a)	Anı	ual Nu E 5-Years	Earth Rate Treated I		
Year	0	20	40	80	100	0	20	40	80	100
-					m;	g/kg				
1977	0.12	0.09	0.09	0.13	0.05					
1978	0.01	0.02	0.02	0.10	0.01					
1979	1.69	1.72	1.64	1.70	1.15					
1980	2.54	2.21	1.02	2.15	3.69	2.82	2.12	1.35	2.15	2.46
1981	2.57	2.60	2.81	2.77	2.45	3.16	2.95	2.89	2.82	2.95
1982	5.22	5.72	5.68	7.94	4.72	4.62	6.06	5.11	6.35	5.71
1983	4.40	4.68	4.55	4.54	4.05	4.50	4.51	4.32	4.44	4.27
1984	1.38	1.74	0.92	0.88	1.31	0.80	0.98	1.16	1.06	0.82
1985	1.38	1.27	2.70	1.08	1.22	1.19	1.49	2.38	0.89	1.64
1986	0.03	0.05	0.85	0.03	0.01	0.05	0.35	0.00	0.05	0.20
1987	0.04	0.02	ND	0.17	ND	ND	ND	0.10	0.33	NE
1988	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1989	ND	0.25	0.04	0.17	0.52	0.17	0.31	0.28	0.13	0.20
1990	0.05	0.04	0.09	0.10	0.18	0.25	0.08	0.55	0.09	0.73
1991	0.00	0.00	0.07	0.00	0.00	0.00	0.00	0.00	0.00	0.05
1992	ND	ND	ND	ND	ND	ND	ND	ND	ND	NE
1993	0.28	0.22	0.35	0.32	0.28	0.25	0.20	0.40	0.28	0.40
1994	0.00	0.00	0.00	0.03	0.00	0.05	0.00	0.00	0.03	0.00
1996	0.10	0.15	ND	0.05	0.10	0.22	0.08	0.25	0.10	0.3
1998	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

TABLE AIII-V: MEAN CONCENTRATION OF Cr IN TOMATO

		ıal Nu E 3-Years			a)	Anı	ual Nu E 5-Years	arth Rate	νU /	
Year	0	20	40	80	100	0	20	40	80	100
-					mg	g/kg				
1977	0.78	1.20	0.96	1.58	0.63					
1978	0.11	0.22	0.24	0.14	0.10					
1979	1.66	0.59	0.94	1.42	0.75					
1980	1.28	1.09	0.64	1.12	1.30	1.07	1.01	1.11	1.19	1.43
1981	0.17	0.12	0.25	0.07	0.05	0.05	0.07	0.31	0.09	0.08
1982	2.14	3.63	2.41	2.19	2.22	1.73	1.87	2.39	2.39	2.04
1983	1.20	2.35	2.75	0.99	2.36	0.70	1.89	2.54	0.99	2.43
1984	1.58	1.17	1.67	1.42	1.57	1.61	0.95	1.01	1.15	1.12
1985	1.64	1.40	1.28	1.58	1.57	1.43	1.32	1.44	1.53	1.33
1986	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1987	0.39	1.26	1.13	0.66	0.39	0.41	0.60	1.67	0.31	0.24
1988	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1989	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1990	0.76	1.24	0.44	0.40	0.85	0.39	0.86	0.56	0.30	1.48
1991	0.08	0.40	0.16	0.20	0.44	0.15	0.51	0.09	0.82	0.36
1992	0.18	0.35	2.87	0.20	0.08	0.15	0.18	0.17	0.00	1.37
1993	0.10	0.17	0.05	0.05	0.25	0.15	0.13	0.10	0.10	0.13
1994	0.08	0.08	0.13	0.13	0.05	0.10	0.13	0.10	0.10	0.10
1996	0.00	0.00	0.00	0.00	0.70	0.00	0.00	0.00	0.00	0.00
1998	0.00	0.00	0.00	0.00	0.26	0.00	0.00	0.00	0.00	0.00

TABLE AIII-VI: MEAN CONCENTRATION OF Cr IN GREEN BEANS

		ual Nu E 3-Years			a)	An	nual Nu l 5-Years	Earth Rate)
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	8.91	9.31	9.91	10.44	11.99					
1978	8.61	9.64	9.75	10.80	11.06					
1979	8.78	10.45	9.82	11.06	12.84					
1980	9.43	8.98	11.32	15.92	12.81	8.44	10.40	10.43	14.93	16.52
1981	9.63	11.02	10.55	12.15	11.35	10.37	11.50	10.53	12.08	12.12
1982	8.60	7.34	7.66	9.55	9.98	7.63	8.02	8.51	11.15	11.25
1983	9.83	11.92	12.25	14.20	14.13	9.66	14.13	12.82	15.07	16.90
1984	8.13	10.19	9.98	10.60	13.12	7.82	9.27	9.19	10.40	11.34
1985	10.01	8.32	7.17	6.30	6.68	9.49	5.74	6.68	5.35	7.63
1986	6.00	6.41	8.09	8.10	8.83	7.02	7.30	5.99	ND	6.78
1987	7.06	7.72	6.68	7.25	6.50	7.56	6.43	7.24	8.66	6.68
1988	8.47	10.73	9.70	10.63	10.16	9.55	11.09	10.41	10.63	12.19
1989	5.32	5.53	5.64	7.46	7.01	5.82	5.47	6.20	7.45	7.56
1990	5.58	5.52	6.07	6.77	6.43	5.91	5.44	7.13	6.99	8.35
1991	8.40	7.80	11.43	11.13	9.80	8.00	7.12	9.73	10.73	9.50
1992	5.60	5.23	6.18	6.95	5.82	6.12	5.00	5.90	6.33	6.47
1993	7.65	8.50	9.50	9.50	8.93	10.25	8.82	11.05	9.80	10.80
1994	7.55	7.45	7.57	8.57	8.38	6.45	9.65	9.15	10.68	8.20
1996	12.15	8.25	8.00	11.60	10.25	10.88	8.43	10.95	10.80	10.10
1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

TABLE AIV-I: MEAN CONCENTRATION OF Cu IN BEET

	AIIII		Earth Ra	· •	1 <i>a)</i>	All	nual Nu E 5-Years	Treated)
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	8.61	5.93	7.53	7.41	7.82					
1978	8.54	8.48	8.94	10.65	10.17					
1979	5.37	5.96	5.83	5.91	6.51					
1980	6.84	4.44	5.45	5.95	6.47	6.33	7.53	9.02	5.12	6.64
1981	8.22	8.29	8.65	7.81	8.73	7.61	9.70	9.17	10.30	9.99
1982	8.53	7.58	7.13	7.64	8.12	8.20	8.27	7.99	8.58	8.97
1983	6.71	7.78	7.54	7.75	7.65	7.87	7.88	9.03	7.16	8.09
1984	10.12	9.63	8.69	13.94	11.42	10.27	9.65	8.29	10.05	10.87
1985	5.83	5.90	4.70	6.07	7.00	5.99	5.54	5.52	6.14	6.76
1986	6.24	4.89	3.80	6.03	6.11	5.26	5.82	4.14	8.14	6.73
1987	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1988	7.14	12.20	10.23	10.26	11.85	9.37	12.55	9.36	11.72	12.30
1989	6.53	6.94	6.37	8.01	6.34	6.35	5.56	7.31	6.59	5.72
1990	7.66	11.10	11.70	8.97	9.88	8.52	10.80	9.07	8.73	8.69
1991	5.40	6.82	5.65	6.82	7.58	5.72	7.17	5.80	6.82	6.75
1992	4.33	3.33	4.75	4.57	3.83	4.30	5.27	4.60	4.97	5.43
1993	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1994	ND	5.43	7.10	6.50	5.43	5.90	6.13	6.80	6.50	6.57
1996	ND	7.00	4.50	9.50	3.15	ND	5.40	7.70	5.40	5.25
1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

TABLE AIV-II: MEAN CONCENTRATION OF Cu IN CARROT

	Ann	ual Nu H 3-Years			na)	An	nual Nu E 5-Years	Earth Rate Treated		
Year	0	20	40	80	100	0	20	40	80	100
						- mg/kg				
1977	21.07	28.80	35.60	26.75	32.43					
1978	18.22	19.28	19.65	22.85	24.33					
1979	14.80	13.00	15.97	19.32	18.20					
1980	16.97	18.35	19.77	25.58	27.07	17.12	17.42	19.93	20.38	29.32
1981	22.80	23.23	24.38	27.98	33.40	22.33	25.42	27.77	31.30	32.98
1982	14.82	15.12	19.47	20.85	20.97	13.43	14.08	17.10	19.55	19.32
1983	23.38	26.58	27.58	26.68	29.42	25.18	22.05	29.35	31.38	36.22
1984	0.02	0.02	0.02	0.02	0.03	0.02	0.02	0.02	0.02	0.03
1985	15.95	16.25	19.88	20.05	19.92	18.48	17.40	16.58	22.23	23.67
1986	14.95	14.50	16.03	15.87	17.23	15.43	16.00	15.93	18.83	17.28
1987	18.52	18.50	20.50	17.73	21.32	20.28	17.65	23.30	21.55	20.62
1988	9.90	11.68	11.55	10.22	12.16	12.45	13.05	12.34	12.10	11.15
1989	16.43	15.80	16.08	20.43	17.42	16.90	16.60	19.90	20.10	17.27
1990	12.48	14.95	13.22	15.45	16.57	13.25	15.58	14.97	16.70	19.58
1991	10.00	14.88	14.23	14.21	15.09	11.42	11.90	15.77	18.02	16.13
1992	15.48	16.98	18.03	17.50	18.35	16.20	16.70	18.62	21.95	17.47
1993	15.93	16.98	17.13	20.75	20.38	16.10	13.25	19.10	22.38	15.88
1994	15.75	17.00	19.05	25.98	25.00	17.20	22.58	20.23	24.42	20.50
1996	13.05	19.25	15.15	23.80	12.20	14.05	21.30	14.95	21.92	15.60
1998	10.70	13.82	11.96	10.90	11.68	11.18	12.88	11.80	15.55	12.48

TABLE AIV-III: MEAN CONCENTRATION OF Cu IN SWISS CHARD

	Ann	ual Nu I 3-Years	Earth Ra	. 0	na)	Ar	nual Nu 1 5-Years	Earth Rates Treated)
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	12.95	16.95	20.10	18.17	18.15					
1978	13.40	15.67	15.33	14.63	14.77					
1979	8.37	8.63	10.55	11.52	11.70					
1980	12.88	12.28	12.70	12.70	11.85	10.77	11.13	11.68	12.35	12.63
1981	13.75	14.73	16.52	16.17	17.03	16.45	15.17	16.35	18.15	16.03
1982	11.23	12.50	12.25	12.35	12.67	12.33	12.95	13.32	13.80	13.65
1983	12.35	14.27	15.05	16.28	17.45	12.27	14.37	15.63	17.78	17.55
1984	10.90	13.40	16.90	14.80	15.55	ND	ND	ND	ND	ND
1985	11.45	12.65	12.65	12.45	12.48	12.29	12.03	13.57	13.30	13.36
1986	14.10	13.20	16.43	17.53	15.75	13.63	14.55	13.45	15.63	15.92
1987	14.45	15.60	17.10	13.98	13.80	14.95	16.42	16.40	15.50	16.98
1988	11.85	13.92	13.97	16.10	15.20	12.00	13.90	14.07	14.60	14.78
1989	11.25	12.40	15.20	15.20	12.35	14.70	13.00	11.80	16.60	17.05
1990	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1991	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1992	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1993	ND	ND	ND	12.05	ND	ND	14.40	ND	14.40	ND
1994	11.88	9.90	9.60	12.30	10.68	9.30	10.02	10.45	10.93	11.98
1996	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1998	10.95	ND	12.05	13.10	ND	12.10	12.65	12.40	12.31	14.35

TABLE AIV-IV: MEAN CONCENTRATION OF Cu IN SPINACH

	16.84 16.47 18.73 25.60 18 9.50 11.45 10.73 11.91 12 13.13 16.52 14.40 16.13 16 10.27 11.87 14.60 13.93 1 11.14 11.63 11.98 13.08 13 9.74 11.78 11.12 10.67 16 9.83 12.42 11.93 11.02 1 8.02 9.18 11.19 10.49 16					An	nual Nu I 5-Years	Earth Rate)
Year	0	20	40	80	100	0	20	40	80	100
						- mg/kg				
1977	16.84	16.47	18.73	25.60	18.40					
1978	9.50	11.45	10.73	11.91	12.87					
1979	13.13	16.52	14.40	16.13	16.58					
1980	10.27	11.87	14.60	13.93	11.32	11.10	10.38	13.22	12.32	11.37
1981	11.14	11.63	11.98	13.08	13.07	12.35	13.23	13.63	14.43	12.88
1982	9.74	11.78	11.12	10.67	10.44	8.82	11.92	11.26	12.82	10.93
1983	9.83	12.42	11.93	11.02	11.97	10.39	10.87	11.12	11.63	11.92
1984	8.02	9.18	11.19	10.49	10.87	8.88	11.40	11.61	9.89	11.06
1985	10.15	11.27	12.20	11.11	10.93	9.49	10.45	13.40	10.53	30.27
1986	11.95	12.10	12.55	13.07	11.87	12.05	12.28	12.52	15.30	12.60
1987	6.28	8.44	ND	7.28	ND	ND	ND	7.91	8.82	ND
1988	13.22	14.73	14.47	13.65	12.45	13.48	13.45	10.40	12.74	15.72
1989	ND	12.01	10.34	11.71	10.47	11.39	11.44	12.30	12.53	11.48
1990	11.40	14.27	12.40	13.27	12.72	15.05	13.47	13.40	13.27	12.85
1991	12.07	13.07	12.45	12.55	14.27	10.83	12.42	12.55	12.12	14.05
1992	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1993	11.65	10.95	12.65	10.02	11.05	10.98	9.70	11.70	9.77	13.25
1994	8.15	9.25	9.00	10.05	8.45	8.20	9.75	ND	8.77	7.50
1996	10.77	14.10	ND	13.02	14.25	11.32	12.32	13.05	13.25	11.93
1998	9.32	9.88	10.25	9.93	10.00	8.73	10.05	10.06	9.38	11.09

TABLE AIV-V: MEAN CONCENTRATION OF Cu IN TOMATO

	Ann		Earth Ra	te (Mg/ł l Plots	na)	An		Earth Rate	e (Mg/ha) Plots)
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	9.88	10.38	9.87	9.40	9.27					
1978	8.40	10.09	8.70	8.59	7.53					
1979	9.69	10.03	10.36	9.88	9.61					
1980	8.69	8.63	8.82	9.56	8.65	7.83	9.67	9.21	9.70	8.98
1981	6.24	8.04	7.36	7.40	7.53	7.49	8.23	7.61	10.39	7.94
1982	6.86	7.16	9.56	7.14	8.63	8.66	8.16	8.18	8.43	9.80
1983	8.14	9.11	7.61	8.64	7.99	8.48	8.60	8.68	7.73	8.18
1984	8.64	10.22	9.32	9.38	8.60	9.90	8.68	9.38	8.15	8.53
1985	7.68	8.39	8.56	8.13	7.94	8.27	6.87	9.33	9.54	8.20
1986	7.54	8.15	7.49	8.35	8.26	7.23	8.37	7.58	8.86	7.62
1987	9.59	8.22	8.36	7.99	8.52	8.26	8.10	7.57	8.64	8.78
1988	ND	ND	ND	ND	ND	ND	ND	ND	ND	NE
1989	ND	ND	ND	ND	ND	ND	ND	ND	ND	NE
1990	8.47	9.48	10.21	8.82	9.07	9.61	9.31	9.58	8.61	9.19
1991	8.34	9.25	10.24	8.92	7.85	7.65	9.25	8.15	9.57	9.90
1992	7.25	7.17	8.02	7.33	9.40	7.67	7.03	7.93	7.92	8.20
1993	8.55	9.05	8.75	9.60	9.73	8.35	10.57	8.95	9.85	11.32
1994	8.63	9.55	8.80	10.60	9.35	10.80	8.95	9.10	10.57	10.40
1996	10.90	12.17	12.25	11.73	12.25	8.30	12.90	15.85	11.53	12.75
1998	7.05	5.22	6.31	7.26	7.00	7.24	7.09	6.95	6.74	7.19

TABLE AIV-VI: MEAN CONCENTRATION OF Cu IN GREEN BEANS

	Ann	ual Nu E 3-Years		. 0	a)	Anr		Earth Rate Treated	e (Mg/ha) Plots)
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	0.38	0.58	0.57	1.08	1.56					
1978	0.34	0.38	0.51	0.76	0.85					
1979	0.46	0.54	0.90	1.37	1.52					
1980	0.65	0.67	0.86	2.01	1.47	0.53	0.47	0.78	1.28	2.84
1981	0.32	0.61	0.57	0.87	0.71	0.32	0.50	1.66	1.51	1.47
1982	3.94	16.68	9.28	10.17	7.78	2.68	4.96	7.64	12.64	22.65
1983	2.49	2.64	2.83	3.25	6.58	2.05	3.64	3.38	5.13	6.01
1984	5.33	7.47	6.91	8.22	8.41	6.78	5.41	5.88	6.83	6.53
1985	7.97	2.48	4.62	2.85	3.61	3.28	2.80	3.73	3.08	4.15
1986	0.10	0.05	0.13	0.30	0.39	0.09	7.63	0.08	ND	0.75
1987	0.32	0.58	0.34	0.53	0.50	0.62	0.32	0.50	0.61	0.83
1988	2.11	8.88	5.02	1.24	1.77	2.83	1.43	1.68	1.76	1.97
1989	0.22	0.10	0.19	0.46	0.67	0.22	0.16	0.31	0.68	1.15
1990	0.06	0.06	0.06	0.14	0.06	0.06	0.42	0.06	0.07	0.47
1991	0.08	0.16	0.16	0.20	0.26	0.08	0.07	0.20	0.62	0.75
1992	0.15	0.17	0.28	0.33	0.42	0.30	0.18	0.48	0.47	0.38
1993	0.10	0.10	0.40	0.10	0.05	0.10	0.85	0.35	0.25	0.55
1994	0.20	0.25	0.35	0.35	0.55	0.22	0.32	0.47	0.73	0.63
1996	0.73	1.08	0.50	0.98	2.38	2.38	0.25	2.50	1.75	1.35
1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

TABLE AV-I: MEAN CONCENTRATION OF Ni IN BEET

		ial Nu E 3-Years			a)	Anı		Earth Rate Treated I	e (Mg/ha) Plots	
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	1.10	0.92	1.46	1.69	2.71					
1978	0.64	0.64	1.47	1.63	1.96					
1979	0.03	0.36	0.52	1.43	2.05					
1980	0.74	0.91	0.87	1.84	1.85	0.81	0.99	0.98	0.61	3.07
1981	0.56	0.73	0.83	0.70	0.88	0.12	0.41	0.92	1.92	2.88
1982	1.75	1.35	2.05	1.33	2.50	1.37	1.05	1.69	3.10	3.58
1983	0.52	0.66	0.77	1.21	1.01	0.90	1.00	1.53	1.63	1.77
1984	5.27	6.11	3.50	4.55	6.82	5.21	4.42	3.61	4.62	6.75
1985	2.92	2.68	2.69	4.81	4.05	3.14	2.52	2.72	3.77	4.86
1986	1.44	1.06	1.32	1.69	1.53	1.63	1.55	1.19	2.46	2.09
1987	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1988	1.54	1.30	1.09	1.38	3.08	0.91	1.01	1.21	1.67	2.30
1989	ND	0.37	0.53	0.93	0.67	0.26	0.39	0.70	0.97	1.28
1990	0.40	0.75	1.11	0.97	1.71	0.40	1.15	1.12	1.63	1.98
1991	0.47	0.68	0.50	0.73	1.60	0.65	1.00	0.73	1.55	1.83
1992	1.43	0.17	0.90	0.92	1.13	1.33	0.62	1.58	1.27	1.10
1993	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1994	ND	0.43	0.50	0.75	0.80	0.25	0.52	0.65	1.20	0.82
1996	ND	1.55	1.80	1.55	1.60	ND	1.15	1.50	2.00	1.85
1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

TABLE AV-II : MEAN CONCENTRATION OF Ni IN CARROT

		ial Nu E 3-Years			a)	Annual Nu Earth Rate (Mg/ha) 5-Years Treated Plots						
Year	0	20	40	80	100	0	20	40	80	100		
					m	g/kg						
1977	1.89	1.34	1.69	2.79	3.99							
1978	2.28	2.45	2.99	3.60	4.97							
1979	0.80	0.86	1.56	2.20	2.70							
1980	2.91	2.76	2.26	4.57	4.60	3.31	2.07	3.59	4.22	4.90		
1981	1.86	3.02	2.84	3.47	4.84	1.33	4.20	5.93	7.47	8.05		
1982	7.43	6.28	6.96	8.59	9.09	7.31	6.99	7.35	7.62	8.13		
1983	1.63	2.30	2.44	2.12	3.31	0.81	1.93	1.97	3.71	4.02		
1984	1.00	1.30	2.56	1.62	2.87	1.09	1.38	1.34	1.60	3.71		
1985	3.09	6.46	4.57	4.07	5.02	2.92	7.06	11.86	6.88	6.38		
1986	3.08	6.72	6.04	3.77	5.64	5.53	4.63	8.04	4.68	7.59		
1987	0.99	0.47	1.12	1.50	1.52	1.77	0.71	1.63	1.20	1.50		
1988	7.43	2.07	6.50	5.96	6.38	8.49	8.36	10.08	6.02	12.53		
1989	2.41	2.11	1.78	5.42	2.60	1.72	2.49	3.30	2.92	2.55		
1990	4.32	4.51	2.83	8.50	4.91	4.96	5.69	19.43	7.37	4.54		
1991	0.10	0.25	0.38	0.44	0.35	0.07	0.72	0.16	0.91	1.94		
1992	2.10	2.68	1.53	4.38	1.80	1.75	1.05	2.30	2.98	2.45		
1993	0.35	0.95	1.05	1.45	1.90	0.40	0.68	0.98	1.50	1.38		
1994	0.57	0.75	1.55	1.77	1.98	0.50	1.23	1.77	1.83	2.90		
1996	2.03	2.75	2.55	3.25	1.90	1.75	2.00	1.80	1.88	3.63		
1998	1.04	1.13	1.14	0.78	1.65	1.28	1.69	1.35	1.32	1.29		

TABLE AV-III: MEAN CONCENTRATION OF Ni IN SWISS CHARD

	Ann		Earth Rat	. 0	na)	An	nual Nu E 5-Years	Earth Rate Treated)
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	5.59	5.91	9.27	9.33	8.21					
1978	1.48	1.46	2.43	1.89	2.80					
1979	0.90	0.96	1.04	1.71	1.65					
1980	1.07	ND	ND	2.73	2.59	1.17	0.55	2.77	2.13	7.77
1981	1.27	1.21	1.73	1.33	2.75	0.89	1.40	2.18	3.04	3.86
1982	2.45	2.12	2.54	2.70	2.82	3.13	2.69	3.57	3.86	4.54
1983	8.11	5.77	8.80	5.96	9.95	10.64	5.58	7.84	10.05	12.00
1984	12.14	10.00	11.50	9.89	11.60	ND	ND	ND	ND	ND
1985	9.85	9.11	8.25	8.76	9.57	9.06	8.42	8.95	11.84	11.30
1986	2.13	1.62	2.35	3.41	4.47	1.44	2.14	2.15	3.69	3.56
1987	2.33	2.71	4.50	1.68	2.55	3.80	7.60	7.06	2.91	5.80
1988	1.65	1.63	3.48	6.01	5.01	3.78	5.64	7.68	1.60	6.00
1989	1.10	1.50	1.02	1.17	0.88	6.34	2.87	0.08	6.39	1.10
1990	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1991	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1992	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1993	ND	ND	ND	0.50	ND	ND	1.35	ND	1.50	ND
1994	0.80	0.63	0.65	1.05	0.98	0.43	1.13	0.52	1.08	1.63
1996	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1998	2.66	ND	1.98	1.73	ND	0.76	1.73	1.98	1.52	1.98

TABLE AV-IV: MEAN CONCENTRATION OF Ni IN SPINACH

			Earth Rat		a)	Anr		Earth Rate	e (Mg/ha) Plots	
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	1.71	1.31	1.06	2.15	1.75					
1978	0.46	0.88	1.02	1.50	2.14					
1979	1.17	4.22	4.09	4.08	2.72					
1980	0.70	1.04	0.75	1.21	1.13	0.88	0.96	0.79	0.90	1.21
1981	1.02	0.75	1.04	1.01	1.61	1.32	1.04	1.40	1.53	2.34
1982	1.15	2.80	1.54	1.74	1.85	2.29	2.20	1.70	3.37	2.39
1983	4.03	7.10	27.13	6.90	8.24	6.83	7.00	15.87	10.26	7.67
1984	3.53	3.42	4.29	3.46	4.48	3.13	2.36	4.24	3.72	4.35
1985	1.92	1.95	3.60	2.21	2.16	1.89	2.54	2.90	2.39	2.76
1986	0.47	0.33	0.43	0.37	0.51	0.14	0.33	0.41	0.62	0.97
1987	0.06	0.29	ND	0.20	ND	ND	ND	0.34	0.64	ND
1988	0.41	0.15	0.50	0.08	0.89	0.02	0.01	0.59	0.51	1.86
1989	ND	0.24	0.50	0.40	0.36	0.28	0.14	0.64	0.71	0.48
1990	0.04	0.31	0.34	0.38	1.12	0.31	0.50	0.37	0.69	0.68
1991	0.60	0.72	0.68	0.68	1.00	0.37	0.37	0.80	0.63	0.98
1992	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1993	0.35	0.30	0.45	0.77	0.73	0.38	0.32	0.57	0.63	0.70
1994	0.28	0.25	0.35	0.43	0.38	0.15	0.32	ND	0.40	0.38
1996	1.58	0.85	ND	1.27	1.15	1.52	0.73	1.55	1.98	5.53
1998	1.20	2.09	1.50	1.42	1.26	2.30	2.16	1.52	2.21	3.59

TABLE AV-V: MEAN CONCENTRATION OF Ni IN TOMATO

		ıal Nu E 3-Years		e (Mg/h Plots	a)	Annual Nu Earth Rate (Mg/ha) 5-Years Treated Plots					
Year	0	20	40	80	100	0	20	40	80	100	
						mg/kg					
1977	2.22	2.93	4.53	6.31	7.27						
1978	1.18	1.60	3.29	5.53	6.43						
1979	3.67	1.34	2.72	3.92	3.65						
1980	2.08	2.17	2.26	3.81	5.05	1.83	2.10	2.75	3.09	5.78	
1981	1.37	1.83	2.96	4.25	6.57	1.38	2.68	5.30	11.08	12.23	
1982	1.87	1.97	7.34	6.19	8.47	2.01	2.05	7.12	19.30	19.25	
1983	2.42	2.09	2.86	5.07	4.68	2.27	2.33	5.07	7.08	11.55	
1984	2.45	2.32	2.65	2.88	3.24	2.78	2.35	2.62	2.66	4.78	
1985	1.49	1.62	1.46	1.96	2.09	1.16	1.70	1.75	2.88	2.75	
1986	1.07	0.89	1.58	2.00	2.98	1.07	1.73	1.66	3.44	4.43	
1987	6.34	6.18	3.81	4.23	2.79	2.38	1.88	3.85	3.21	3.40	
1988	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	
1989	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	
1990	4.18	5.96	3.31	3.34	7.52	2.16	5.30	3.51	5.92	8.98	
1991	1.55	2.37	1.83	2.08	2.45	1.73	2.70	2.20	6.13	3.85	
1992	1.32	4.17	2.85	4.17	4.72	0.82	1.45	2.75	5.62	5.40	
1993	2.10	1.35	2.10	2.75	2.67	1.70	1.83	2.25	3.90	2.47	
1994	1.45	1.42	2.45	2.15	2.25	1.95	1.90	1.85	3.22	1.95	
1996	0.30	0.93	0.55	0.43	0.50	0.30	0.45	2.20	2.97	0.65	
1998	2.75	1.88	4.00	5.50	7.38	1.75	4.75	5.00	3.00	5.25	

TABLE AV-VI: MEAN CONCENTRATION OF Ni IN GREEN BEANS

		ual Nu E 3-Years			a)	Anr		Earth Rate Treated I	· •	
Year	0	20	40	80	100	0	20	40	80	100
]	mg/kg				
1977	0.17	0.20	0.19	0.16	0.55					
1978	0.04	0.06	0.04	0.05	0.06					
1979	0.16	0.17	0.34	0.18	0.26					
1980	0.51	0.94	0.47	0.48	0.39	0.45	0.53	0.44	0.47	0.67
1981	0.21	0.11	0.14	0.00	0.01	0.05	0.00	0.15	0.35	0.07
1982	0.36	0.34	0.29	0.30	0.24	0.46	0.52	0.27	0.36	0.27
1983	0.13	0.30	0.21	0.25	0.18	0.36	0.43	0.36	0.21	0.32
1984	0.14	0.19	0.16	0.48	0.24	0.13	0.09	0.14	0.19	0.00
1985	0.34	0.15	0.30	0.00	0.00	0.15	0.17	0.52	0.00	0.00
1986	0.08	0.16	0.10	0.17	0.16	0.13	0.16	0.10	ND	0.16
1987	0.18	0.29	0.11	0.16	0.14	0.28	0.13	0.21	0.12	0.10
1988	0.19	0.28	0.30	0.16	0.32	0.25	0.36	0.17	0.20	0.21
1989	0.07	0.10	0.10	0.10	0.16	0.14	0.07	0.09	0.08	0.10
1990	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1991	0.10	0.07	0.09	0.05	0.05	0.08	0.11	0.10	0.05	0.05
1992	0.08	0.10	0.07	0.11	0.10	0.13	0.05	0.15	0.12	0.11
1993	0.00	0.00	0.02	0.01	0.00	0.00	0.07	0.03	0.00	0.05
1994	0.40	0.23	0.47	0.40	0.32	0.34	0.34	0.35	0.31	0.32
1996	0.00	0.05	0.00	0.07	0.36	0.27	0.00	0.00	0.00	0.28
1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

TABLE AVI-I: MEAN CONCENTRATION OF Pb IN BEET

		ial Nu E 3-Years		e (Mg/h Plots	a)	Anr		Earth Rate Treated I		
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	0.60	0.15	0.34	0.31	0.45					
1978	0.06	0.07	0.00	0.01	0.02					
1979	0.03	0.35	0.08	0.29	0.20					
1980	0.39	0.25	0.12	0.34	0.19	0.30	0.45	0.64	0.36	0.30
1981	0.00	0.22	0.00	0.03	0.01	0.02	0.00	0.00	0.04	0.06
1982	0.31	0.52	0.73	0.61	0.64	0.39	0.52	0.68	0.48	0.56
1983	0.01	0.20	0.01	0.06	0.46	0.01	0.00	0.06	0.10	0.23
1984	0.71	0.88	1.02	1.01	1.26	0.51	0.63	1.17	1.23	1.69
1985	0.26	0.38	0.10	0.30	0.22	0.21	0.24	0.44	0.32	0.34
1986	0.30	0.51	0.65	0.96	0.35	0.56	0.58	0.58	0.89	0.82
1987	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1988	0.28	0.95	0.57	0.92	1.04	0.76	0.81	0.59	0.68	0.84
1989	ND	0.38	0.34	0.63	0.25	0.26	0.13	0.42	0.34	0.46
1990	0.10	0.27	0.32	0.29	0.49	0.09	0.37	0.31	0.30	0.35
1991	0.05	0.45	0.16	0.41	0.23	0.13	0.20	0.38	0.41	0.64
1992	0.04	0.02	0.26	0.20	0.13	0.08	0.25	0.13	0.77	0.36
1993	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1994	ND	0.36	0.44	0.74	0.57	0.58	0.47	0.60	0.43	0.55
1996	ND	0.35	0.55	0.43	0.32	ND	0.32	0.43	0.34	6.95
1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

TABLE AVI-II: MEAN CONCENTRATION OF Pb IN CARROT

		ial Nu E 3-Years		, U	a)	Annual Nu Earth Rate (Mg/ha) 5-Years Treated Plots					
Year	0	20	40	80	100	0	20	40	80	100	
						mg/kg					
1977	ND	ND	ND	ND	ND						
1978	3.07	3.60	3.37	3.16	3.55						
1979	1.49	1.43	1.12	1.49	0.95						
1980	1.74	2.12	2.09	2.25	1.83	2.41	1.90	2.24	2.43	3.04	
1981	1.39	1.38	1.58	1.72	1.53	1.42	1.73	1.54	2.03	1.79	
1982	1.75	2.15	1.78	1.34	1.25	1.63	2.38	1.45	1.64	1.64	
1983	0.75	0.85	1.38	0.95	0.55	0.69	0.88	0.75	0.67	0.74	
1984	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
1985	0.36	0.82	0.70	0.69	0.75	0.87	1.04	0.51	0.35	0.28	
1986	0.14	0.46	0.71	1.41	0.69	2.18	1.37	1.20	1.63	1.06	
1987	0.80	0.71	1.00	1.03	0.86	0.62	1.23	0.93	1.03	0.85	
1988	0.42	0.49	0.80	0.63	0.69	0.06	0.55	0.58	0.96	0.49	
1989	1.65	1.73	1.51	1.94	1.55	2.44	2.32	1.74	1.54	1.54	
1990	2.41	2.18	1.97	3.72	2.75	2.06	3.66	2.44	2.73	3.31	
1991	0.43	0.73	0.30	1.28	0.56	0.81	0.43	0.25	1.13	0.60	
1992	0.70	0.65	0.85	0.48	1.25	0.89	0.72	0.90	0.54	0.65	
1993	0.71	1.42	0.40	0.62	0.69	0.31	1.41	0.33	0.38	0.59	
1994	0.56	0.56	0.58	0.58	0.64	0.54	0.63	0.49	0.72	0.54	
1996	0.70	0.90	0.93	0.86	0.44	0.74	0.82	0.46	0.82	0.79	
1998	0.48	0.71	2.86	0.68	1.19	1.06	1.12	0.93	0.84	0.81	

TABLE AVI-III: MEAN CONCENTRATION OF Pb IN SWISS CHARD

		ıal Nu E 3-Years			a)	Anı		Earth Rate Treated I		
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	2.84	4.49	3.25	3.44	3.84					
1978	1.90	3.02	2.49	2.41	2.28					
1979	1.35	1.52	1.58	1.77	1.63					
1980	2.35	2.18	2.80	2.88	2.36	2.59	1.57	2.19	2.82	3.06
1981	0.92	1.02	1.00	1.24	1.16	3.05	1.03	1.59	1.59	1.72
1982	1.38	0.91	1.33	1.01	1.22	0.75	0.93	1.15	1.40	1.51
1983	0.74	0.59	1.98	0.77	0.49	0.49	0.71	0.99	0.50	0.56
1984	0.18	0.86	2.36	1.15	0.96	0.51	0.39	1.34	1.07	0.77
1985	0.63	0.92	1.15	1.08	1.13	0.68	0.90	1.10	1.08	1.24
1986	0.00	0.00	2.39	0.04	0.49	0.00	0.32	0.00	0.00	0.00
1987	0.82	0.91	1.38	0.84	0.80	1.45	1.91	1.22	0.92	1.34
1988	0.60	0.90	1.41	1.79	1.27	0.75	1.10	1.45	0.69	0.91
1989	1.59	2.12	3.39	2.58	2.55	2.24	2.94	1.40	5.15	2.34
1990	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1991	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1992	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1993	ND	ND	ND	0.51	ND	ND	0.96	ND	0.75	ND
1994	0.56	0.63	0.56	0.83	0.58	0.51	0.84	0.38	0.64	0.64
1996	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1998	0.00	ND	0.00	0.00	ND	0.00	0.17	0.17	0.00	0.00

TABLE AVI-IV: MEAN CONCENTRATION OF Pb IN SPINACH

		ıal Nu E 3-Years			a)	Anr		Earth Rate Treated I	, U	
Year	0	20	40	80	100	0	20	40	80	100
]	mg/kg				
1977	ND	ND	ND	ND	ND					
1978	0.14	0.27	0.21	0.23	0.15					
1979	0.16	0.49	0.63	0.36	0.50					
1980	0.05	0.10	0.13	0.23	0.04	0.18	0.15	0.02	0.13	0.36
1981	0.05	0.28	0.00	0.00	0.00	0.00	0.08	0.00	0.00	0.01
1982	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.05
1983	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1984	0.00	0.26	0.02	0.06	0.03	0.02	0.04	0.11	0.00	0.00
1985	0.00	0.00	0.00	0.00	0.00	0.07	0.00	0.00	0.00	0.00
1986	0.01	0.13	0.01	0.00	0.00	0.01	0.60	0.01	0.00	0.01
1987	0.00	0.00	ND	0.05	ND	ND	ND	0.00	0.05	ND
1988	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
1989	ND	0.06	0.00	0.00	0.00	0.05	0.09	0.09	0.00	0.00
1990	0.21	0.26	0.22	0.21	0.20	0.22	0.25	0.22	0.27	0.32
1991	0.11	0.10	0.06	0.06	0.06	0.11	0.04	0.04	0.05	0.12
1992	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1993	0.03	0.05	0.10	0.10	0.06	0.06	0.07	0.13	0.09	0.08
1994	0.12	0.13	0.12	0.26	0.08	0.29	0.15	ND	0.16	0.12
1996	1.25	0.30	ND	0.25	1.05	1.76	0.59	1.00	0.00	0.74
1998	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

TABLE AVI-V: MEAN CONCENTRATION OF Pb IN TOMATO

		ual Nu E 3-Years		· •	a)	Annual Nu Earth Rate (Mg/ha) 5-Years Treated Plots					
Year	0	20	40	80	100	0	20	40	80	100	
					m	g/kg					
1977	0.17	0.13	0.16	0.11	0.37						
1978	0.25	0.29	0.35	0.26	0.26						
1979	0.19	0.40	0.18	0.41	0.17						
1980	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
1981	0.33	0.27	0.87	0.26	0.40	0.40	0.20	0.27	0.13	0.20	
1982	0.89	0.00	0.09	0.01	0.12	0.17	0.14	0.00	0.00	0.26	
1983	0.00	0.00	0.05	0.00	0.05	0.07	0.14	0.04	0.10	0.12	
1984	0.12	0.00	0.04	0.04	0.08	0.00	0.00	0.00	0.04	0.00	
1985	0.00	0.14	0.00	0.00	0.00	0.09	0.00	0.00	0.24	0.01	
1986	1.74	0.10	0.60	0.41	0.00	0.93	0.57	0.97	0.32	1.16	
1987	0.03	0.03	0.02	0.04	0.04	0.03	0.29	0.07	0.02	0.09	
1988	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	
1989	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	
1990	0.12	0.10	0.16	0.35	0.14	0.13	0.15	0.15	0.08	0.13	
1991	0.24	0.22	0.20	0.28	0.43	0.20	0.29	0.22	0.29	0.49	
1992	0.15	0.42	0.28	0.19	0.29	0.19	0.13	0.30	0.49	0.19	
1993	0.00	0.01	0.07	0.20	0.02	0.00	0.03	0.02	0.08	0.01	
1994	0.14	0.11	0.21	0.12	0.09	0.17	0.14	0.14	0.13	0.14	
1996	0.16	0.41	0.50	0.40	0.62	0.41	0.46	0.23	0.67	0.35	
1998	0.99	1.61	0.95	1.10	1.79	4.75	0.63	2.78	1.09	1.44	

TABLE AVI-VI: MEAN CONCENTRATION OF Pb IN GREEN BEANS

		ıal Nu E 3-Years		te (Mg/ha Plots	a)	Anr		Earth Rate Treated	e (Mg/ha) Plots)
Year	0	20	40	80	100	0	20	40	80	100
					n	ng/kg				
1977	28.9	36.9	43.6	51.1	60.2					
1978	25.0	35.8	43.6	58.7	62.5					
1979	34.0	45.0	61.7	93.0	89.7					
1980	30.1	59.8	75.0	136.2	96.8	32.2	56.8	66.8	108.3	160.8
1981	32.0	45.5	42.3	57.9	58.4	36.3	46.3	59.0	71.3	77.9
1982	25.1	41.2	33.5	56.1	64.3	26.9	38.6	51.4	81.3	91.4
1983	39.2	55.1	58.5	80.2	96.9	39.2	70.1	73.7	101.1	117.8
1984	26.3	37.6	36.3	48.5	60.6	26.5	32.9	34.5	44.6	51.1
1985	22.3	24.2	30.5	35.2	40.5	29.4	27.0	35.8	32.0	46.5
1986	18.7	22.5	26.8	36.4	49.2	21.3	30.1	23.4	ND	61.5
1987	21.0	23.0	23.2	24.1	29.3	20.2	26.1	24.3	34.6	38.8
1988	22.4	29.5	26.5	35.6	38.8	25.9	32.7	30.7	39.8	53.9
1989	16.4	20.3	20.9	32.7	42.0	17.6	20.2	29.7	41.4	49.0
1990	22.5	27.2	32.7	52.5	52.8	24.6	30.5	40.5	51.0	69.1
1991	40.2	48.2	52.8	55.5	70.2	34.2	48.0	53.3	62.3	76.0
1992	26.5	29.0	41.8	45.3	47.3	27.3	28.2	39.7	51.2	57.3
1993	29.5	32.5	47.5	43.5	51.8	49.5	42.3	55.5	46.8	70.5
1994	28.5	32.5	29.0	45.5	48.0	27.3	42.0	47.5	60.8	56.3
1996	54.8	46.5	37.5	65.3	69.3	61.3	50.5	52.5	66.0	79.5
1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

TABLE AVII-I: MEAN CONCENTRATION OF Zn IN BEET

		ıal Nu E 3-Years			a)	Anr		Earth Rate Treated I		
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	26.1	20.8	27.4	28.8	30.0					
1978	21.2	20.8	24.8	26.7	26.1					
1979	20.1	22.3	24.7	26.8	31.6					
1980	23.6	22.9	25.2	33.5	36.6	23.4	38.5	35.4	30.1	40.3
1981	21.9	27.5	24.4	26.8	31.4	23.1	26.8	31.5	36.7	41.4
1982	27.4	30.0	28.3	34.8	39.3	29.0	34.3	35.9	45.0	47.6
1983	27.0	33.0	30.2	31.0	33.3	30.7	33.4	35.7	30.8	35.8
1984	25.3	25.8	23.9	24.1	29.4	25.6	27.3	24.1	26.9	30.6
1985	16.1	20.4	15.6	19.2	19.6	17.8	19.4	19.4	17.1	22.1
1986	21.8	21.0	19.4	28.1	23.4	23.5	22.3	20.1	29.5	25.8
1987	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1988	24.0	46.8	38.3	32.1	53.4	31.8	40.1	37.0	40.3	46.5
1989	ND	24.2	22.4	30.8	29.5	26.5	19.9	28.9	24.6	27.0
1990	26.7	36.0	36.2	36.3	37.3	31.1	36.2	35.5	33.5	36.4
1991	23.8	30.8	34.0	31.8	36.0	28.3	35.0	32.0	39.2	37.5
1992	18.8	15.3	20.3	23.0	19.8	19.0	23.3	19.8	24.5	25.3
1993	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1994	ND	20.8	23.5	23.8	25.5	23.5	22.5	26.0	26.3	25.8
1996	ND	32.5	25.5	58.0	31.5	ND	33.5	43.0	36.3	22.0
1998	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND

TABLE AVII-II: MEAN CONCENTRATION OF Zn IN CARROT

	Ann	ual Nu I 3-Years	Earth Ra	ν U	na)	An		Earth Rates Treated	νų,)
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	77.7	147.8	234.0	141.3	224.8					
1978	52.7	159.0	223.7	289.0	374.0					
1979	42.9	80.7	109.8	148.2	188.9					
1980	51.2	154.7	165.0	315.7	382.3	57.5	86.8	177.7	263.7	374.0
1981	86.8	170.3	222.5	321.2	405.0	85.9	216.8	299.3	412.3	421.3
1982	55.5	99.5	177.3	242.2	232.3	46.5	90.6	132.0	226.8	235.0
1983	56.0	91.1	120.2	128.7	251.0	62.0	101.3	194.8	235.8	392.5
1984	70.1	169.0	177.2	272.8	424.5	79.8	168.0	234.2	297.2	484.5
1985	60.3	99.2	87.3	190.3	202.8	57.0	133.6	142.3	354.0	277.2
1986	47.4	66.8	72.1	106.1	145.8	43.3	71.3	91.2	157.2	174.3
1987	124.6	79.3	95.2	65.0	131.7	70.0	90.1	156.5	116.5	103.6
1988	104.3	206.9	136.7	124.7	199.6	157.5	212.0	178.7	174.2	152.2
1989	101.3	96.8	113.0	189.5	175.0	81.2	107.0	147.0	198.8	175.7
1990	44.6	93.9	103.9	165.7	176.2	46.0	98.1	110.1	189.8	202.5
1991	40.3	76.5	107.3	123.0	141.8	51.7	77.8	121.5	133.8	165.0
1992	75.5	79.7	118.8	123.7	171.2	56.7	81.3	100.2	211.2	136.8
1993	70.0	81.5	113.8	210.0	203.8	59.5	87.5	175.8	252.3	178.8
1994	77.8	126.0	127.3	238.3	297.5	64.0	183.5	167.5	253.3	325.3
1996	62.8	147.3	143.5	235.3	118.8	70.8	153.0	127.0	175.5	176.3
1998	39.8	79.1	57.6	90.2	96.5	40.3	71.5	80.0	128.8	124.3

TABLE AVII-III: MEAN CONCENTRATION OF Zn IN SWISS CHARD

	Ann	ual Nu I 3-Years	Earth Ra	. 0	na)	An	nual Nu l 5-Years	Earth Rate)
Year	0	20	40	80	100	0	20	40	80	100
						- mg/kg				
1977	166.5	310.0	523.0	357.8	368.3					
1978	167.0	242.5	229.2	232.8	251.0					
1979	92.2	162.2	165.8	193.3	194.3					
1980	118.5	122.7	132.3	127.8	136.5	124.9	120.5	142.0	138.3	152.0
1981	171.8	283.5	294.3	298.7	341.0	263.0	305.0	312.8	362.3	316.7
1982	168.5	276.0	315.3	226.5	258.3	200.5	312.0	298.2	267.5	307.0
1983	201.8	433.5	403.7	450.8	472.0	208.5	401.0	427.8	430.3	439.5
1984	146.3	286.0	298.4	292.2	370.0	175.0	320.3	310.3	317.5	304.5
1985	109.2	209.0	169.2	178.8	211.2	133.3	179.8	186.8	183.0	205.0
1986	153.2	198.3	218.0	237.8	209.3	156.8	213.2	203.5	249.7	229.8
1987	130.3	262.8	227.5	253.0	225.0	121.7	231.8	231.5	251.3	265.8
1988	140.3	300.7	319.2	310.8	309.0	155.3	284.7	299.5	299.0	299.3
1989	166.0	256.5	268.0	336.5	281.0	203.5	239.0	182.0	234.5	250.0
1990	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1991	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1992	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1993	ND	ND	ND	184.5	ND	ND	216.0	ND	253.5	ND
1994	152.0	195.3	236.8	313.8	302.0	197.8	233.0	207.5	335.5	243.3
1996	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1998	154.0	ND	236.0	203.5	ND	133.0	282.0	173.4	220.6	156.0

TABLE AVII-IV: MEAN CONCENTRATION OF Zn IN SPINACH

		ual Nu E 3-Years			a)	Anr		Earth Rate Treated I	, U	
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	36.5	37.6	37.6	42.5	42.3					
1978	29.9	32.2	33.9	36.8	40.7					
1979	26.2	29.7	29.3	33.3	34.5					
1980	29.0	30.5	37.1	40.2	33.4	28.8	30.7	35.5	36.4	41.1
1981	25.9	28.5	29.1	32.5	36.2	28.1	34.1	34.2	39.8	36.4
1982	23.0	29.3	27.9	28.1	26.2	23.6	27.9	29.2	32.0	30.0
1983	24.3	32.5	30.7	29.4	32.0	26.5	30.2	30.2	31.8	33.8
1984	26.3	29.5	30.9	30.6	32.0	26.7	28.8	30.6	30.5	33.2
1985	28.5	30.8	31.8	34.2	34.2	30.9	31.8	32.5	34.3	33.8
1986	24.9	23.5	24.9	26.4	25.5	25.0	23.0	27.0	28.8	28.5
1987	18.5	21.0	ND	25.2	ND	ND	ND	17.9	23.4	ND
1988	27.3	30.9	32.6	36.4	27.7	29.8	46.3	24.7	30.2	37.0
1989	ND	29.4	16.7	27.7	24.6	23.7	25.5	28.5	30.4	26.1
1990	26.5	28.7	30.3	26.7	27.3	27.2	29.9	29.9	28.5	30.2
1991	38.0	36.2	39.8	37.8	45.2	34.3	36.5	39.5	39.8	38.7
1992	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1993	28.3	25.5	33.0	28.5	30.3	29.3	27.8	31.8	29.8	35.0
1994	22.5	30.0	32.0	32.0	35.0	23.5	32.8	ND	27.0	31.8
1996	24.0	30.5	ND	31.8	32.3	25.8	28.0	36.5	31.3	27.0
1998	18.4	20.5	19.8	23.4	19.4	16.3	21.9	21.8	20.0	25.1

TABLE AVII-V: MEAN CONCENTRATION OF Zn IN TOMATO

	Annual Nu Earth Rate (Mg/ha) 3-Years Treated Plots					Annual Nu Earth Rate (Mg/ha) 5-Years Treated Plots				
Year	0	20	40	80	100	0	20	40	80	100
						mg/kg				
1977	34.3	35.5	37.7	35.5	37.4					
1978	27.9	29.3	27.9	30.4	28.2					
1979	36.6	36.9	41.2	41.2	41.5					
1980	30.0	31.6	30.6	34.7	32.7	29.8	32.0	33.0	35.8	34.8
1981	28.5	32.0	32.0	31.6	34.1	29.5	32.3	35.1	37.7	40.0
1982	31.9	28.1	38.4	32.2	37.5	32.5	31.8	35.4	38.5	42.3
1983	35.8	35.0	33.1	34.7	34.8	37.6	35.2	37.1	35.8	38.4
1984	27.7	29.3	34.3	30.4	32.8	33.1	27.1	36.3	27.0	33.5
1985	10.5	12.9	19.0	8.7	9.5	14.7	9.8	13.5	10.3	12.3
1986	22.8	23.5	24.3	22.3	27.9	22.5	23.5	22.8	27.8	26.7
1987	33.3	25.7	26.5	24.7	28.1	27.6	26.6	25.1	29.3	28.3
1988	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1989	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
1990	27.3	30.8	35.3	32.2	32.7	27.2	30.6	36.7	34.7	33.4
1991	40.3	37.2	40.8	35.5	38.3	38.7	43.0	44.3	40.3	40.3
1992	34.3	37.2	42.2	38.3	42.3	31.8	34.7	41.7	41.0	42.7
1993	38.0	39.8	29.5	33.0	44.5	36.5	43.3	32.5	39.5	45.0
1994	38.0	39.0	36.3	40.3	37.0	48.5	35.0	37.5	46.3	42.0
1996	34.0	40.7	42.5	41.3	40.5	32.0	43.5	52.0	44.7	56.5
1998	32.6	27.8	35.2	39.2	37.0	33.0	37.8	40.5	37.1	38.1

TABLE AVII-VI: MEAN CONCENTRATION OF Zn IN GREEN BEANS