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THESIS

POLONIUM-210 IN SOILS AND PLANTS

Submitted by

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In partial fulfillment of the requirements

for the Degree of Doctor of Philosophy

Colorado State University

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ABSTRACT OF THESIS

POLONIUM-210 IN SOILS AND PLANTS

Polonium-210 used as a radionuclide fuel for SNAP electrical generators may present a long term hazard if accidental release of the isotope were to contaminate agricultural soils. Absorption of $^{210}PoO_2$ in soils studied by bulk determination of distribution coefficients (K_d) indicated retention in the top soil samples was correlated with silt content. An empirical equation describing agricultural top soil absorption was derived as $\ln K_d = 3.2 + 0.046$ (percent silt). The distribution coefficients of the combined data for the A, B and C horizons combined were correlated with soil pH and silt content and could be described by the derived empirical equation $\ln K_d = -1.3 + 0.034$ (percent silt) + 0.88 (pH).

Polonium oxide was eluted through soil columns and successfully tested these distribution coefficients. Greater than 98 percent of the $^{210}PoO_2$ was retained by the first 2 cm of soil with K_d's greater than 100. Top soil samples with K_d's less than 100 indicated $^{210}PoO_2$ would be retained by the lower soil horizons. Hazards to water supplies by movement of ^{210}Po through soil would be minimal.

Chemical species of ^{210}Po in soil solution examined by electrophoresis and dialysis experiments indicated both positive and negative species existed. Dialysis experiments indicated approximately 34 percent of the ^{210}Po in soil solution passed through a semi-permeable membrane.

Plant uptake of $^{210}PoO_2$ added to three different soils was measured and expressed as a transfer coefficient, U, where

$$U = \frac{^{210}Po/g \text{ dry plant tissue}}{^{210}Po/g \text{ dry soil}}$$

Transfer coefficients of 11 different fruit, grain, leafy and root crops ranged from 10^{-2} to 10^{-6}. The transfer coefficients remained constant for the majority of plant tissues examined, although the distribution coefficients differed for the three soils used. Calculations using the transfer coefficients for estimating dietary intake of ^{210}Po from vegetables indicate a potential long-term ingestion hazard to the population.

Protein hydrolyzates of wheat and spinach were separated by paper chromatography to investigate possible organic forms of polonium in plants. One possible organic form of polonium was observed.

Measurements of naturally occurring ^{210}Po in the soil samples indicated a range of 0.22 to 3.47 pCi/g in top soils. Polonium-210 measured in the vegetation used for controls in the greenhouse experiments indicated the primary source of the isotope was the fallout of airborne ^{210}Po and its radioactive precursors.

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To my wife, Nancy Jane, must go the most sincere thanks and appreciation. Her patience, understanding and encouragement were instrumental in completion of this paper.

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CHAPTER I

INTRODUCTION AND STATEMENT OF THE PROBLEM

The element polonium was discovered by Marie and Pierre Curie in 1898 while studying the radioactivity of uranium and thorium ores. Study of the element was hampered by its low concentration in nature. Figgins (1961) estimated the concentration to be approximately 0.1 mg per ton of uranium ore. All known isotopes of polonium are radioactive; the longest lived naturally occurring isotope is ^{210}Po (RaF) with a 138.4 day half-life. Polonium is formed from the radioactive decay of uranium and it is the last radioactive element in the decay chain. Polonium-210 decays to lead-206 99.9988 percent of the time via a 5.30 MeV alpha particle. The remaining 1.2×10^{-3} percent of the decay is via a 4.50 MeV alpha particle and a 0.803 MeV gamma ray.

Kilocurie quantities of ^{210}Po have been proposed as a fuel for radioisotope electrical generators. Large quantities of ^{210}Po are prepared by neutron irradiation of pure ^{209}Bi in nuclear reactors. The ^{210}Bi which is formed has a half-life of five days and decays to ^{210}Po. The polonium-210 is encapsulated to maximize heat production from the alpha decay energy and the heat is converted to electricity through semiconductor thermoelectric elements. Radionuclide electrical generators have been named Systems for Nuclear Auxilary

Power or SNAP with major application of the units as satellite power supplies.

Use of ^{210}Po as a radioisotope fuel for electrical generators requires an evaluation of the hazards associated with accidental release of large quantities of the isotope to the biosphere. Before an evaluation of the hazards can be made, the behavior of the radioisotope in biological systems must be examined. Routes of ingested forms of ^{210}Po must be investigated for possible short-term and long-term radiation exposures of a population.

Accidental release of a large quantity of ^{210}Po as an oxide over agricultural soils may introduce a long-term hazard by contaminating soil and vegetation. Polonium oxide deposited on soil may be retained in the plant root zones or move through the soil to a water supply. Polonium-210 in the plant root zone may be available for plant uptake and entry into the food chain.

The chemical species of polonium in soil solution may control the soil retention and plant uptake. The physio-chemical properties of the soil interacting with the polonium oxide may determine the chemical species. Upon entry into the plant the chemical form may influence the availability of the isotope if ingested by animals.

Because the long-term radiological health hazards of ^{210}Po released over agricultural soils are associated with soil retention and plant uptake, this research was conducted to develop data required

for hazards analysis. Studies of ^{210}Po movement in soils had not been made previously, and investigations of ^{210}Po in plants were not conducted for evaluation of the hazards of ^{210}Po in the diet. Thus, the purpose of this study was to develop the data necessary for estimation of the hazards from deposition of ^{210}Po on agricultural soils. The objectives of the research were:

1. To develop predictive equations of soil retention of ^{210}Po in terms of a distribution coefficient (K_d).

2. To characterize the chemical species of polonium in soil solution.

3. To measure the plant uptake of ^{210}Po by the root system and express the uptake in terms which would allow estimation of ingested quantities.

4. To investigate the chemical form of the ^{210}Po in the plant tissues.

CHAPTER II

LITERATURE REVIEW

Polonium-210 in Soils and Plants

The source of ^{210}Po in soils is the radioactive decay of ^{238}U.

The decay chain leads to the formation of the rare gas ^{222}Rn. Radon-

222 has a half-life of 3.82 days which allows time for diffusion of the

gas into soil capillaries and into the air. Vohra, Subbaramu and Rao

(1964) estimated that the radon content of soils may reach 500,000

pCi/m^3. The fraction of ^{222}Rn escaping the soil into the air varied

from 18 - 87 percent for the different soil types included in their

study. The factors that influenced day to day release of radon were

temperature, air pressure, wind velocity and rainfall.

Osburn (1965) discussed the factors influencing radon generation

and migration in soil and rock structures. The highest radon genera-

tion in soils was associated with the clay fractions. In dry soil the

movement of radon gas was diffusion controlled but in water saturated

soils displacement of the gases moved radon out into the air. Hansen

et al. (1960) observed that freezing prevented radon movement in soils

during the winter months.

Radon-222 decays through several short lived daughter products

to ^{210}Pb which has a 22 year half-life. Lead-210 decays to ^{210}Bi

which decay with a five day half-life to ^{210}Po. Radon-222 decays to

its daughter products either in air or soil leads to the formation of

^{210}Pb and ^{210}Po in air and soil. The daughter products attached to airborne particles may result in fallout because of particle size or may be washed from the air by precipitation. Fallout ^{210}Po can be intercepted by plants or deposited on the top soil.

Soils and plant materials were examined for alpha activity by Mayneord, Turner and Radley (1960). Soils and plants growing on the soils were ashed to drive off ^{210}Po and the ingrowth of ^{210}Po from decay of ^{210}Pb was observed with time. The soil and plant ^{210}Po contents were not correlated. Hill (1960) examined the ^{210}Pb and ^{210}Po content of grass, soil and rain. He suggested that two routes of entry into grass were available via root uptake of ^{210}Pb and ^{210}Po from the soil or direct deposition on the grass by rain. Measurements of the grass, soil and rain indicated the levels in grass were supported by fallout in rain.

An interest in the source of ^{210}Po in human tissues and the implication that ^{210}Po may be one of the carcinogenic agents in cigarette smoke led to a more intensive study of polonium in plants and soils. Tso, Hallden and Alexander (1964) determined the ^{210}Pb and ^{210}Po content of important tobacco growing soils and found concentrations ranging from 0.26 to 1.01 pCi/g. They suggested the uptake of ^{210}Pb and ^{210}Bi from the soil could account for the ^{210}Po content of tobacco.

Berger, Erhardt and Francis (1965) suggested the source of polonium in tobacco and vegetables was from air deposition. They measured

the polonium content of vegetables, fresh tobacco, cured tobacco and the associated soils. They detected 1 to 5 pCi of ^{210}Po per gram of soil, but did not detect ^{210}Po in vegetables or fresh tobacco. Cured tobacco contained 0.96 pCi/g of polonium and cabbages left in the field over winter contained up to 6.6 pCi/g in the outer leaves. The center of the head did not contain detectable ^{210}Po activity. Tso, Harley and Alexander (1966) exposed tobacco plants to a 500-fold increase of ^{222}Rn in air in a greenhouse. Only a two-fold increase in ^{210}Pb and ^{210}Po content of the tobacco was observed after six weeks of exposure. Tso et al. concluded from the experiment that the major route of entry into the tobacco plant was root uptake of ^{210}Pb and ^{210}Po. Francis and Chesters (1967) suggested the source of ^{210}Po in tobacco could be the decay of ^{210}Pb. Their experiment followed the change of ^{210}Po activity in tobacco samples and the ingrowth of polonium was traced to ^{210}Pb through analysis of the half-lives involved. In a more detailed study Francis, Chesters and Erhardt (1968) experimented with vegetables and tobacco under greenhouse and field conditions. Plants grown under field conditions contained up to 10 times more ^{210}Po than those grown in a greenhouse. The ^{210}Po content in field plants was directly related to the rain content of ^{210}Pb and ^{210}Po at the different field stations.

Examinations of direct plant uptake of ^{210}Po from soil have been limited to movement from nutrient solution or ^{210}Po $(NO_3)_4$ mixed

into soil. Menzel (1953) reported unsupported ^{210}Po uptake by red clover, sudan grass and <u>Astragulus</u> grown in nutrient solutions. Unsupported ^{210}Po is polonium present without its radioactive precursors ^{210}Pb and ^{210}Bi. The percent of the total present in Menzel's solution detected in the plants was 0.006 percent in red clover, 0.019 percent in sudan grass and 0.055 percent in <u>Astragalus.</u> Tso and Fisenne (1968) examined tobacco plant uptake of ^{210}Po $(NO_3)_4$ mixed into soil. The plants were harvested after two weeks of growth in contaminated soil and the percent uptake of the total polonium available was measured. Upper leaves contained 0.5 percent of the activity. The lower leaves and stem each contained 0.1 percent of the activity.

Root uptake of an element from soil is governed by many factors. Several authors have reviewed the factors influencing the rate and the amount of ion entry into roots. A review of root entry of radioisotopes by Hope (1960) summarizes the internal and external factors to consider in experiments with ion uptake.

Internal plant factors that control ion entry were listed as follows:

1. Nature of the root surface area.

2. The free space available to the ion in the root.

3. The number and concentration of ion exchange centers on the root.

4. The plant transpiration rate.

5.   The age and rate of growth of the plant.

6.   The rate of plant respiration.

7.   The internal ion status of the plant.

External factors that affect root uptake of ions were generalized in the following list:

1.   Availability of the ion.

2.   The chemical nature and concentration of the ion in the soil.

3.   The temperature of the soil.

4.   The oxygen content of the soil.

5.   The water status of the soil.

6.   The presence of other ions.

7.   The soil pH (a special case of 6.).

The root uptake of polonium-210 may be governed by the availability of the chemical species in soil which is controlled by the soil retentive properties.   Rhodes (1957 a) expressed the soil retention of radioisotopes in terms of the equilibrium distribution coefficient.   The distribution coefficient (K_d) is the ratio of the amount of radioisotope per gram of adsorption media to the amount of radioisotope per ml of solution at equilibrium.   The K_d is calculated by use of the following equation discussed by Mayer and Tompkins (1947) in their investigations of bulk ion exchange measurements.

$$K_d = \frac{M_s}{M_l} \times \frac{v}{m}$$

where

M_s = the fraction of the radioisotope on the solid phase after equilibrium. $M_s = (M_{total} - M_1)/M_{total}$

M_1 = the fraction of the radioisotope in the liquid phase.

v = volume of solution.

m = soil weight.

Rhodes' investigations (1957 b) of soil factors influencing the distribution coefficient indicate that the physio-chemical properties of the soil and the radioisotope solution determine the strength of interaction. The soil cation exchange capacity, organic matter, hydrogen ion activity and the particle size distribution were correlated with changes in the K_d for plutonium and strontium.

Polonium in Relation to Man

Polonium-210 in soils and plants was implicated as one of the major contributors to mans exposure from natural radiation in early studies of radioisotopes in the diet. Mayneord and Hill (1959) identified ^{210}Po as the major source of alpha activity in grass and sheep kidneys. Hill (1962) reported that polonium accounted for the major part of the alpha activity of botanical samples and soft tissue samples of man. Polonium was present in soft tissue without its precursors, ^{210}Bi and ^{210}Pb. Studies of ^{210}Pb and ^{210}Po in human bone by Holtzman (1963) and in human tissues by Osborne (1963) suggested the

source of the isotopes was from food, water and air. Hill (1963)
suggested ^{210}Pb may contribute to the amount of ^{210}Po detected in soft
tissues, but he stated that food was probably the major source of the
isotope. Holtzman (1964) suggested food does not contain sufficient
^{210}Po to account for the amount in soft tissues. He suggested ^{210}Pb
bound in the bone tissues decays to ^{210}Po and is redistributed in the
body. Blanchard (1967) suggested either that redistribution takes place
or that errors exist in dietary approximations and transfer coefficients
from food to tissues.

Interest in the role of polonium-210 in human radiation exposure
was increased by Radford and Hunt (1964) who suggested that ^{210}Po
may be one of the carcinogenic agents of tobacco smoke. Holtzman
and Ilcewicz (1966) reported human tissues of cigarette smokers con-
tained more polonium than non-smokers.

Reports of high levels of ^{210}Pb and ^{210}Po in environmental sam-
ples in Finland and Alaska suggested the metabolism of the two
isotopes could be followed in the lichen to caribou or reindeer to
human food chain. Beasley and Palmer (1966) reported more ^{210}Po
than ^{210}Pb in caribou soft tissues. Kauranen and Miettinen (1967)
reported the ratio of ^{210}Po to ^{210}Pb ranged from 20 - 40 in elk and
reindeer meat. Other soft tissues such as kidney and liver had ratios
greater than one. Hill (1967) suggested that differences in the chemi-
cal form of ingested polonium may significantly change its

metabolic behavior. He postulated that polonium detected in meats of Arctic animals was organically bound ^{210}Po and more available to humans. Kauranen and Miettinen (1969) added weight to this suggestion with the results of their study of the Lapp food chain. They measured the ^{210}Po/^{210}Pb ratio and content of lichen, reindeer and Lapp reindeer breeders. The ratio was one for lichens which constitute the primary food of the reindeer. The ratio was 25 to 42 for reindeer meat. The amount of ^{210}Po in Lapps was much greater than could be explained by the present transfer coefficients for movement of polonium from the gastrointestinal tract to the blood to the organ of interest. The data suggest that the ^{210}Po ingested in reindeer meat was five times more available than would be predicted from the values reported by the International Committee on Radiation Protection (1959).

Although the levels of polonium-210 reported in Lapps and Eskimos are not considered dangerous, the more metabolically available forms of ^{210}Po in reindeer and caribou meat suggest ^{210}Po also may be present in plants as a more available form when ingested. Polonium oxide deposited on the soil may be absorbed by plants and incorporated into plant tissues in the organic form.

Polonium-210 as a Radionuclide Fuel

Polonium-210 is one of several radioisotopes proposed as a fuel for radionuclide electrical generators. Davis (1963) in a review of

radioisotopes for use in the space program listed the properties of ^{210}Po for use in SNAP generators. The high specific activity and the alpha decay with only 1.2×10^{-5} gamma rays per disintegration made ^{210}Po an attractive isotope for short lived space missions. Table 1 summarizes the physical properties of ^{210}Po as a radioisotope fuel The isotope has been proposed as an energy source for land based operations as well. Rodden (1967) suggested that ^{210}Po be considered as a fuel for an energy source for small manned Antarctic Experiment Stations. The system would be used for short term missions at small outposts.

Polonium-210 was first used as a fuel for radioisotope electrical generation in SNAP III. Barmat, Anderson and Bollmeir (1959) reported that SNAP III fueled with 3000 curies of ^{210}Po produced 5.3 watts of electricity. Semiconductor thermoelectric elements were used to convert the heat produced by alpha decay in the source to electricity. Polonium also was used for the construction of a radioisotope thruster-electrical power unit for space application by Jones and Austin (1966) and Leventhal (1966).

The proposed use of polonium-210 as a fuel for radioisotope generators in space requires an evaluation of the hazards that would be present if an accidental release of the fuel occurred. Lieberman and Connor (1966) stated the general safety objectives for design and use of nuclear devices in space are the use with very low probability of

Table 1. The physical properties of $^{210}_{84}$Po as a SNAP fuel.[*]

Nuclear Decay Mode	Energy	Fraction of Disintegrations Accompanied by Decay Mode
Alpha	5.298 $\pm$ 0.002 MeV	0.999988
Alpha	4.5 MeV	0.000012
Gamma	0.804 MeV	0.000012

Fuel Forms

	Energy Available (watts/gram)	Specific Activity (curies/gram)
Po metal	144	4500
Alloy GdPo	82	2573
Ta matrix with alloy		
Ta:GdPo		
10 : 1	7.5	234
50 : 1	1.6	50.4

[*] From "Pu-238 and Po-210 Data Sheets", (1967).

introducing radioisotopes into the biosphere and minimum population

radiation exposure. Harvey, Dick and Fink (1963) discussed the types

of accidents that could cause an accidental release of the radioisotope

from the fuel capsule. If a fire were to occur on a launch pad, the

fuel capsule would be exposed to shock, high temperatures and the

corrosive action of the rocket fuels. Destruction of the rocket during

an early ascent abort of the rocket could rupture the SNAP generator

by impact with land or water. Burial in soil could result in heat build-

up and rupture of the fuel capsules. Burial in sea water could result

in corrosion and fuel release. Near orbit injection rocket failure

could result in re-entry with total or partial burn-up upon impact.

Re-entry from orbit after mission completion could result in total or

partial burn-up. In each case the consequences of the release of the

fuel to the biosphere must be considered. The specific problems of the

environment surrounding the radionuclide generator in the types of

accidents described by Harvey et al. are summarized in the March,

1966 report of the Ad Hoc Committee on the Nuclear Space Program

of the Atomic Industrial Forum.

Two accidents, which are examples of possible routes for ^{210}Po

release from SNAP generators, occurred in 1964 and 1968. In

April, 1964 a navigational satellite employing a SNAP-9A generator

did not attain orbital velocity. Krey (1967) reported total burn-up

of the generator and release of fuel in the stratosphere. The ^{238}Pu

fuel was retained in the stratosphere and deposited slowly as world wide fallout. In May, 1968 intentional destruction of the rocket during launch immersed two SNAP 19 generators in the Pacific Ocean off the coast of California. Luikens (1969) reported intact recovery of the fuel capsules from the ocean floor. Corrosion removed the cooling fins and thermoelectric semiconductor elements to leave only the primary containment. Tests of the fuel capsules for leakage indicated no loss of ^{238}Pu.

Evaluation of the consequences of the release of polonium-210 fuel requires a knowledge of the probability of such an accident occurring and the probability of injury of a population. Coleman and Perez (1967) divided injury to a population into two categories. First injury from the radioisotope can occur through inhalation. Secondly injury can result from ingestion of the radionuclide through the food chain. Willis, Bramlitt, Paulson and Rooney (1967) suggested a long-term hazard may exist from the ^{210}PoO$_2$ deposited on the soil. However, at the time of their hazards evaluations no soil retention data were available. Coleman and Perez evaluated the ingestion hazard from ^{210}PoO$_2$ deposited on the plant directly. However, evaluations of the long-term foliar absorption, root absorption or movement in the soil were not included in the hazards analysis.

The Chemistry of Polonium

The chemical nature of polonium will govern the long-term soil retention and plant uptake by roots or foliar absorption. However, the chemistry of polonium is not well known because the element has no stable isotope and the chemical studies of polonium must be conducted with highly radioactive samples. Figgins pointed out that polonium-210 is the isotope most commonly used, but the high specific activity of milligram quantities leads to radiation damage of crystal structures or solutions.

Polonium is the heaviest element of Group VIA of the Periodic Table. Sulfur, selenium and tellurium are the other elements of the group. Figgins reports polonium compounds of +2 and +4 oxidation states with a preferred oxidation state of +4. Bagnall (1958, 1966) reported the formation of polonium monoxide (PoO) which rapidly oxidizes in air to polonium dioxide (PoO_2) and polonium (IV) nitrates which decomposed to the dioxide form at $130^{\circ}C$. Polonium (IV) hydroxide was prepared by Bagnall as a pale yellow precipitate formed by the addition of aqueous NH_4 OH or alkali hydroxides to acid solutions of $PoCl_4$. The composition of the hydroxide was reported as PoO $(OH)_2$ by Bagnall but Ziv and Efros (1959) suggested a formula of Po $(OH)_4$ and calculated the solubility constant as 10^{-37}.

References to the solution chemistry of polonium are few and usually the experiments have been carried out in solutions of one molar

ionic strength. $^{210}PoO_2$ as a fallout product in rain, surface of soils, or plants is likely to encounter much lower ionic strengths. Weigel (1959) studied polonium in solution between pH 1 and 14 and concluded that the polonium existed mainly as dissolved forms at pH 1-5, 7-8 and 12-14 while between pH 6-7 and 10-11 most of the polonium existed as a colloid. Haissinsky (1961) named colloidal solutions containing radioactive products in unweighable and invisible quantities radio-colloids. He reported that radiocolloidal solutions of polonium are sensitive to impurities and that polonium is readily adsorbed on powders or precipitates. Thomas and Stannard (1964) examined neutralized $^{210}PoCl_4$ solutions. Centrifuge experiments with aged solutions indicated that polonium was present as particles and application of Stoke's Law to the observed sedimentation rates indicated 95 percent of the particles in solution were greater than 0.42 microns in diameter. They suggested small amounts of impurities in solution acted as nuclei for particle formation.

The chemical form of natural polonium in soils and plants has not been explored because of the low levels detected. However, polonium may exhibit chemical properties similar to selenium which has been examined in plants and soils. Selenium in plants has been identified in protein fractions of toxic wheats and weeds. Franke and Painter (1936) linked selenium with the protein fraction of toxic wheats and Jones, Horn and Gersdorff (1936) correlated the cystine and selenium content

of toxic wheats. Beath and Eppson (1947) reported that selenium is present as both organically bound and inorganic selenite in some genera of Astragalus and Stanleya. Smith (1949) using paper chromatography identified the protein-bound selenium as seleno-cystine and seleno-methionine. Peterson and Butler (1962) also identified the above compounds in addition to 13 other compounds using paper chromatography and ^{75}Se. Rosenfeld and Beath (1964) reviewed selenium in soils, plants and animals. The selenium in soils is present as selenate, ferric selenite, and organic compounds. The methods used for experimental determination of trace quantities of selenium in plants suggests that a similar method may be used to identify polonium in plant extracts.

CHAPTER III

MATERIALS AND METHODS

Soil Sample Collection

Soil samples used in this study were obtained through the U. S.
Department of Agriculture, Soil Conservation Service; Colorado State
University, Department of Agronomy; and the U. S. Public Health
Service, Southeastern Radiological Health Laboratory. Profile sam-
ples of two Colorado soils were collected near Harmony, Colorado and
the Fraser Experimental Forest, Colorado. These soils were the Nunn
silty clay loam representing the Brown soil group and the Darling grav-
elly sandy loam, representing a Podzol. Additional profile samples of
two Gray-Brown Podzols and two Podzols were collected for this project
during soil survey activities of the Madison, Wisconsin Soil Conserva-
tion Service. The soils were Fayette silt loam and Lapeer loam from
the Gray-Brown Podzol Soil Group and Gogebic sandy loam and Onaway
fine sandy loam from the Podzol Soil Group. Profile samples of a
Brunizem were obtained from the Soil Conservation Service of Des
Moines, Iowa. Two prairie soils, Dinsdale silty clay loam and Musca-
tine silty clay loam, were collected during October, 1968 and shipped
to this laboratory. Top soil samples of five Ground-Water Podzols
and four Red-Yellow Podzols were furnished by the Southeastern
Radiological Health Laboratory of the U. S. Public Health Service.

Profile samples of the Ground-Water Podzol, Leon sandy loam, were collected by the Florida Soil Conservation Service.

Two large bulk samples of Fayette silt loam and Leon sandy loam were obtained for plant uptake experiments. The Fayette silt loam was collected at the University of Wisconsin Experiment Station at Lancaster. Bulk samples of the A_1 and A_2 horizons of the Leon sandy loam were collected by the Florida Soil Conservation Service and shipped to this laboratory.

Soil samples were collected in polyethylene bags which were opened in the laboratory and the soil allowed to air dry prior to weighing for natural polonium-210 determination or other laboratory experiments. Five hundred gram samples of each profile sample were sent to the Colorado State Soil Testing Laboratory for chemical and mechanical analyses. The mechanical analyses and profile descriptions of the soils are reported in Table A-1 located in the Appendix.

Preparation of Polonium Solutions and Suspensions

Polonium-210 tetrachloride and polonium dioxide were purchased from Monsanto Research Corporation, Nuclear Sources Division, Dayton, Ohio in quantities of 1 to 5 mCi. The chloride form was shipped as a dry deposit in a 10 ml volumetric flask. The deposit was dissolved at room temperature in 10 ml of 1.0 N HCl to form a primary solution of polonium which would not adsorb to the walls of the container. Aliquots were withdrawn from the solution as needed

for addition to soils and for distribution coefficient measurements.
Monsanto Laboratories prepared the dioxide form by thermal decomposition of a polonium nitrate deposit in a 10 ml volumetric flask. The polonium dioxide was suspended in 10 ml of filtered deionized water. The filtered water was prepared by passing deionized water through a pleated membrane cartridge having an average flow pore size of 0.45 microns. The primary $^{210}PoO_2$ suspensions had concentrations of 0.5 mCi/ml and were handled within an enclosed radiochemistry glove box.

Polonium dioxide suspensions were prepared for distribution coefficient determinations by transfering an aliquot of the primary suspension to polyethylene bottles of deionized water. Neutral polonium chloride solutions were prepared by titrating with 0.1 N NaOH in the presence of an indicator.

Equilibrium Distribution Coefficient (K_d) Determinations

The equilibrium distribution coefficient for polonium dioxide suspensions with soil was determined on air-dried samples. The soil samples were used as obtained from the field with the exception that small rocks were removed. Large clumps of soil were broken with an agate mortar and pestle and the soil sample shaken through a 2 mm screen. Two grams of soil were weighed into each of eight polypropylene centrifuge tubes. Five ml of neutral polonium chloride solution or polonium oxide suspension were pipetted into each tube.

The solutions and suspensions were measured by drying one ml aliquots
in planchets and counting them in a gas flow proportional counter. The
concentration of activity ranged from 8000 to 15,000 dpm/ml. The
tubes were sealed with polypropylene friction fit closures and placed
on a wheel which rotated about a horizontal axis at 12 rpm to agitate
them. After 20 hours of rotation, the tubes were removed and placed
in a Sorvall Model RC2-B centrifuge with a Model SS-34 rotor which
was spun at 15,000 rpm for 20 minutes. A centrifugal force of 29,000
times gravity was developed which settled particles greater than 0.02
microns as calculated from equations by Jackson (1956). The samples
were removed and 1.0 ml of the supernatant liquid from each sample
was delivered to one inch stainless steel planchets with an Eppendorf
automatic pipet. The samples were dried under a heat lamp and
counted with a Radiation Counter Laboratories or Nuclear Associates
preflushed gas flow proportional counter connected to an amplifier
and scaler.

The distribution coefficient of Nunn silty clay loam was examined
for change with different concentrations of polonium oxide suspensions
and agitation time. Distribution coefficients were determined for
suspensions of 1000 dpm/ml, 2000 dpm/ml, 6000 dpm/ml, 12,500
dpm/ml and 24,000 dpm/ml. The distribution coefficients for one
hour, two hours, four hours, eight hours, sixteen hours, twenty hours,
twenty-four hours and forty-eight hours of mixing were determined.

Distribution coefficients as a function of soil particle size were determined for the C_{Ca} horizon of the Nunn silty clay loam. Particle sizes were separated by sieving and settling techniques according to the methods of Jackson (1956). Soil particles were divided into ranges of greater than 50 microns, 50 to 20 microns, 20 to 5 microns, 5 to 2 microns and less than 2 microns. The distribution coefficients for particle size ranges were determined with polonium dioxide suspensions.

Soil Column Elution of ^{210}Po

Columns of soil having the same length as the depth of the natural horizon were prepared in 0.9 cm ID polyethylene tubing. The soil was added to the column in the dry state because slurries fractionated into particle sizes with a preponderance of clay at the top. Soil samples were ground in an agate mortar until a particle size was obtained which would allow a flow of one to two ml of water per hour through a column. One hundred ml aliquots of $^{210}PoO_2$ suspension containing 0.1 to 0.2 μCi were placed at the top of the dry soil columns. Each column was connected to a fraction collector and distilled water was supplied to the column through a siphon tube. Elution with distilled water was performed until the equivalent of one mean annual rainfall for the soil series had passed through the column. Water from the fraction collector drained into polypropylene test tubes and the eluate was

transferred to stainless steel planchets, dried, and counted for ^{210}Po

activity. Columns were cut into 1.0 cm sections and analyzed for

^{210}Po as described in the following section.

Determination of the ^{210}Po Content of Soils

Two gram soil samples were weighed into eight 12 ml polypropy-

lene centrifuge tubes and digested with 10 ml of 12 N HCl at 80°C for

40 minutes. The samples were centrifuged for 20 minutes at 15,500

rpm (RCF, 29,000 G) in a Sorvall high speed centrifuge. The super-

natant liquid was decanted into a 500 ml polyethylene bottle. The

samples were digested two more times following the same procedure.

Each sample was diluted with 30 ml of distilled water, 250 mg of

ascorbic acid were added and the solution was titrated to neutrality

with 15 N NaOH using three drops of a methyl red-bromothymol blue

indicator. The indicator solution was prepared by adding 50 mg of

methyl red and 125 mg of bromothymol blue to 85 ml of ethanol and

15 ml of water as described by Fink (1950). The indicator was orange

at pH 4, yellow at pH 5, green at pH 7 and blue at pH 8. Four ml of

12 N HCl were added to the neutral solution and the solution was

transferred with three rinses to plating cells. The cells were made

from nursing bottles as described by Baxter and Wood (1953). Dis-

tilled water was added to make up a volume of 100 ml of a 0.5 N HCl

solution. The plating cells were placed in a water filled shaker

bath at 75°C for five hours. The polonium deposited spontaneously upon nickel discs.

The polonium content of the Nunn silty clay loam and Darling gravelly sandy loam was determined using the Sorvall high speed centrifuge. The ^{210}Po content of the remaining soil samples was determined using a clinical centrifuge. Samples were spun in the clinical centrifuge for 20 minutes at 320 rpm (RCF 1610 G). The chemical yield for the extraction of ^{210}Po added to the soil was 69 ± 2 percent for the procedure using the clinical centrifuge compared to 72 ± 2 percent using the high speed centrifuge.

Addition of Polonium-210 to Soil for Greenhouse Experiments

Fort Collins silty clay loam, Fayette silt loam and Leon sandy loam were transferred to the Department of Radiology and Radiation Biology radioisotope handling facility for addition of ^{210}Po. The soil was measured with the planter pots to be used in the greenhouse into a 1.5 cubic foot cement mixer. Water was added to the soil in the mixer until a thick slurry formed. An aliquot of the polonium chloride or polonium oxide primary solution was transferred to 200 ml of distilled water and the solution added slowly with mixing to the soil slurry. Sand and peat moss were added to the Fort Collins silty clay loam and Fayette silt loam to produce mixtures which were 58 percent soil, 40 percent added sand, and 2 percent peat moss by weight. The Leon sandy loam was mixed by adding one-half of the soil to the

mixer, adding polonium oxide suspension and adding the remainder of soil. After mixing, each soil was transferred to five gallon plastic lined pails. Five random samples of approximately 20 grams were collected for polonium assay during the transfer.

The soils were moved to the greenhouse and steam sterilized by raising the temperature to $80^{o}C$ for 30 minutes. The sterilization killed weed seeds and bacteria which might flourish under greenhouse conditions. After the soil cooled it was transferred to plastic planter pots each of which contained 2500 grams dry weight of soil. Soil mixtures for control plantings were prepared in separate apparatus to prevent cross contamination.

Planting, Plant Harvest and Plant Analyses

Plants were started from seed in the control soils and polonium contaminated soils. Alfalfa, barley, celery, corn, onions, peas, potatoes, radishes, spinach, tomatoes and wheat were planted consistent with the depths and spacings between seeds listed by Knott (1955). One-half inch of vermiculite on top of the soil protected the growing plants from cross contamination of their surfaces. Plantings were started in polonium chloride contaminated Fort Collins silty clay loam in December, 1967. Plantings in polonium oxide contaminated soils were started in Fort Collins silty clay loam in May, 1968 in Fayette silt loam in March, 1969 and in Leon sandy loam in February, 1970. Table 2 contains the polonium assay data for the soils.

Table 2. The soil concentrations of ^{210}Po for each planting.

Soil Series	Chemical Form of ^{210}Po	Number of Mixes	Range of Activity/gm
Ft. Collins silty clay loam	Chloride	12	0.6 - 1 nCi
Ft. Collins silty clay loam	Oxide	10	2.2-5.2 nCi
Ft. Collins silty clay loam	(RaDEF) Nitrate	1	390 nCi
Fayette silt loam	Oxide	8	10 nCi
Leon sandy loam	Oxide	6	15-30 nCi

Figure 1 is a picture of the greenhouse in March, 1969 two weeks after planting in the Fayette silt loam. The planter pots were placed on racks in plastic lined trays containing one-inch of vermiculite to absorb any runoff during watering. The trays were relined after each crop was removed and the polonium contaminated soil was placed in the radioactive waste.

The plants were irrigated with water containing a fertilizer mixture injected into the greenhouse watering system. Table 3 lists the nutrients added to the water.

In addition to the plantings in polonium chloride and oxide contaminated Fort Collins silty clay loam, barley, radishes and peas were planted in RaDEF (^{210}Pb, ^{210}Bi, and ^{210}Po) contaminated Fort Collins soil for comparison with the data for unsupported ^{210}Po. The RaDEF added to the soil was in nitrate form.

At maturity the control plants were harvested on different days than the plants grown in contaminated soil to reduce the chance of cross contamination. The plants were transported to the laboratory in plastic bags.

Root crops were washed with a vegetable brush to remove all visible dirt. Foliage was washed with cold water to remove any soil or dust which may have accumulated. Samples were weighed to obtain wet weights and dried at 100°C for 48 hours. Checks of the dry weight with time demonstrated constant weight after 48 hours

Figure 1.   Arrangement of plantings in Fayette silt loam.
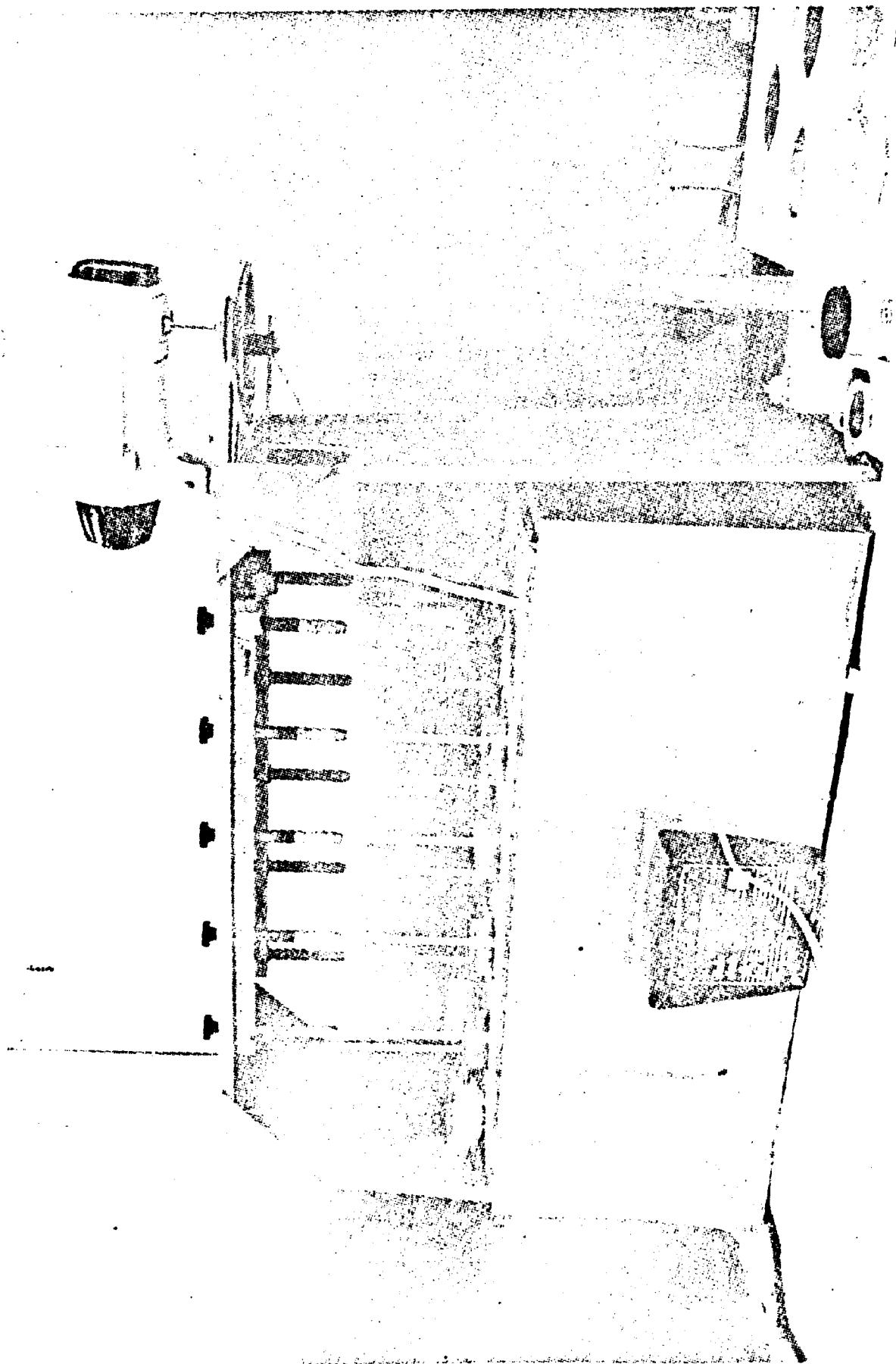
Table 3. Concentration and chemical form of plant nutrients added to greenhouse irrigation water.

Chemical Form	Concentration
KNO_3	6 meq/liter
$Ca(NO_3)_2$	3 meq/liter
$MgSO_4$	2 meq/liter
$NH_4 NO_3$	1.5 meq/liter
$Na NO_3$	0.5 meq/liter
$H_3 PO_4$	1.0 meq/liter
Borax	140 grams/5000 gallons

of drying. The samples which weighed from one to five grams dry

weight were transferred to 500 ml Erlenmeyer flasks. The tissues were

digested using the methods for polonium analysis described by

Blanchard (1966). Twenty ml of 70 percent perchloric acid and 10 ml

of concentrated nitric acid were added and samples digested at $250^{\circ}C$.

For potatoes and barley stems a mixture of 20 ml of 70 percent per-

chloric and 20 ml of concentrated HNO_3 was more satisfactory. After

the samples were dissolved and oxides of nitrogen were no longer

evolved the temperature was raised to $400^{\circ}C$. The samples were

maintained at this temperature until the solutions were clear and dense

white fumes of $HClO_4$ were present. The samples were neutralized

with 15 N NaOH using a methyl red-bromothymol blue indicator.

Eight ml of 12 N HCl were added immediately after the neutralization.

The samples were transferred with three water rinses to nursing

bottle plating cells. Distilled water was added to make up a 200 ml

0.5 N HCl solution. The plating cells were placed in a stirring appara-

tus - water bath for three hours at $85^{\circ}C$. Figure 2 illustrates the

water bath stirrer.

Using control plants or vegetable samples purchased at local gro-

cery stores the chemical yield of the polonium assay method was

determined for each type of plant sample. The chemical yield was

determined by adding known amounts of $^{210}PoO_2$ to the dry vegetation.

Digestion and plating procedures remained as described above.

Figure 2. Water bath and stirring apparatus used for spontaneous deposition of ^{210}Po.

The quantity of ^{210}Po spontaneously deposited on the nickel discs was counted on one of three preflushed gas flow proportional counters. One counter was reserved exclusively for counting control plant samples. The average background of the counter was 0.030 ± 0.004 counts per minute. The error expressed is one standard deviation of the count rate. Low counting samples were counted to a minimum preset count level to obtain a total standard deviation of the sample count rate less background not to exceed $\pm$ 10 percent.

Electrophoresis and Dialysis of Soil Solutions

Soil solutions for electrophoresis and dialysis were prepared by adding one kilogram of air dried soil to two liters of distilled water. The mixture was stirred for 48 hours and allowed to stand an additional 24 hours to permit settling. The solution was decanted and vacuum filtered through a Whatman No. 41 filter paper. The solutions were placed in refrigerated storage until used. Aliquots of solution used for wetting electrophoresis paper or for dialysis were centrifuged for 20 minutes at 15,500 rpm (29,000 G) to remove any particles that may have remained in solution. Soil solutions containing ^{210}Po were prepared by stirring 40 g of soil in 80 ml of ^{210}PoO$_2$ suspension for 48 hours. The soil solution was decanted into centrifuge tubes. After centrifugation, the soil solutions were placed in refrigerated storage or used for electrophoresis or dialysis experiments.

The electrophoresis apparatus was a Warner Chilcott Model E-800-2. The migration chamber has a water cooled migration table and can be used for paper, gel, or thin layer electrophoresis. Experiments using the above apparatus were limited to paper electrophoresis.

Whatman No. 3M paper was cut into 1" x 12" strips for use in the electrophoresis experiments. The paper was marked for center and marked every 0.5 cm from center with light pencil marks along one edge. The strips were wet with centrifuged soil solution and the electrode buffer tanks filled with the same solution. After one hour, 0.025 ml of the polonium contaminated soil solution was applied to the center of the paper strip. The electrodes were connected and the safety cover placed on the apparatus. After applying a potential of 500 volts for one hour, the paper strips were removed and air dried. The paper was cut into 0.5 cm x 2.5 cm strips and placed in 0.5 N HCl in a plating cell containing a nickel disc. The plating cell was placed in a water bath shaker for five hours at $75^{\circ}C$. The nickel discs were counted in a gas flow proportional counter. Seventy-nine percent of the $^{210}PoO_2$ or ^{210}Po in soil solution dried on paper strips was recovered in the above method.

Dialysis experiments were conducted with seamless regenerated cellulose dialysis tubing. The tubing was cut to fit between the two halves of a Chemical Rubber Company 25 ml dialysis cell. The cell was constructed with two 25 ml volumes which were separated by the

cellulose membrane. Solution was added to each chamber by means of a hypodermic syringe. The sample side of the cell was sealed with a nylon screw plug. The access to the receiving solution on the other side of the cell was through Luer-Lok Connectors. The membrane was clamped into the cell and soil solution added to presoak the system for 24 hours before use. The chambers were emptied and filled with polonium contaminated soil solution in the sample chamber and soil solution in the receiving chamber. The solution in the receiving chamber was changed every four hours during a twelve hour period. The solution was changed until less than 0.5 percent of the total activity in the system was detected in the receiving solution. The amount of time required to reduce the amount of polonium passing through the membrane into the receiving solution to 0.5 percent was three to four weeks. Each solution removed from the receiving side of the cell was stored in a polyethylene bottle with 4 ml of 12 N HCl added. The samples were plated onto nickel discs as previously described. At the completion of an experiment the solution from the sample side of the cell was removed and plated. Each chamber of the cell was rinsed three times with 20 ml of 1 N HNO_3. The acid washes and the membrane were digested in 70 percent perchloric acid and 5 ml of concentrated nitric acid. The samples were neutralized and plated onto nickel discs in 0.5 N HCl. The discs were counted for ^{210}Po on a gas flow proportional counter.

Extraction of Plant Polonium and Separation

by Paper Chromatography

Wheat and spinach were grown in sand cultures contaminated with $^{210}PoO_2$. At maturity the plants were removed and extractions of proteins or liquids were made immediately or the excess plant tissue was frozen. The protein was extracted from the spinach or wheat stems and leaves as described by Peterson and Butler (1962) and Smith (1966). The plant tissue was boiled in 100 ml of 95 percent ethanol and 5 percent 6 N HCl for 15 minutes. The ethanol was evaporated by boiling on a steam bath. The dry deposit was removed and 50 mg transferred to 10 ml of 6 N HCl. Acid hydrolysis of the extract was carried out in a boiling water bath for 40 hours. $^{210}PoO_2$ suspension was added to 10 ml of 6 N HCl and digested in a boiling water bath for 40 hours and used as a control sample. The solutions were used with paper chromatography to separate the amino-acids and peptides resulting from hydrolysis of the proteins.

Ascending paper chromatography was used with an n-butanol, acetic acid, water solution (120 ml n-butanol, 30 ml acetic acid and 50 ml water). Chromatograms were made on 30 cm x 25 cm sheets. The sheets were lightly pencil marked one inch in from a 30 cm border to mark the origin. Spots of 0.01 ml of digested plant extract and digested $^{210}PoO_2$ were dried one inch apart on the pencil line and the sheet was stapled together to form a cylinder 25 cm high.

A chromatography chamber was made from a 4-liter beaker. The walls of the beaker were lined with filter paper saturated with the liquid phase and 50 ml of solution were added to the beaker. The cylinder of paper was set in the solution with the origin at the bottom and a parafilm cover sealed the beaker and prevented evaporation. Ascent of the liquid phase required four hours to move 15 cm, then the chromatograph was removed and dried in an oven at $100^{\circ}C$. A 0.1 percent ninhydrin-acetone solution was sprayed on the chromatograph after drying. Amino acids formed violet or pink spots on the chromatograph after 24 hours. The dye spots were measured to determine the distance the amino acids moved from the origin. The paper was cut into one cm strips measured from the origin and each paper strip placed in a plating cell containing 100 ml of 0.5 N HCl and a nickel disc. The disc was counted in a gas flow proportional counter.

Errors

Experiments using radioactive materials in biological systems are subject to systematic errors in laboratory measurements. Biological variation between individuals and the randomness of radioactive decay are errors which are inherent in the systems. Statistical methods were used to estimate the variation in average values introduced by these errors.

Systematic errors may enter into each sample measurement and transcription of the data. An instrument may be read incorrectly, or

used incorrectly and part of the sample may be lost by transfer operations and chemical operations. Errors in laboratory measurements in this study were minimized by use of calibrated balances for weights and calibrated glassware for reagent volumes. Each chemical determination was calibrated for chemical yield. Data were checked for mathematical and transcription errors, but it is possible some were included in the observations used. Mathematical errors were minimized by use of computer programs for calculations of individual and averaged sample data. The programs written for the data processing are in the Appendix.

Biological variations are due to the differences between individuals and the differences that may exist in their growth environments. Biological variation between individual plants were minimized by use of the same seed supply for most species throughout the study. Exceptions were the use of Kennebec potatoes in the Fayette planting and the use of Scout, a winter wheat variety, in the $^{210}PoCl_4$ planting. Greenhouse conditions stablized the temperature and humidity environments of the plants. However, small variations of the physical environment may have existed over the length of the greenhouse bench. The biological variation introduced a source of accidental or indeterminate errors.

The use of nuclear counters to determine the concentration of ^{210}Po in each sample introduced a source of errors because of the random disintegration of radionuclides. Overman and Clark (1960) discussed

the application of the Normal distribution law and the associated statistics to radioactive decay. Their methods were used to determine the number of counts that were reasonable to accumulate for minimum error. Samples in the experiments were counted for a maximum error of 5 percent standard deviation of the net count rate in the case of plants grown in soils with added ^{210}Po. Control plant samples were counted until a 10 percent standard deviation of the net count rate was attained. The total uncertainty added to the uncertainty of the transfer coefficients because of nuclear counting was a maximum of 5 percent. The contribution of errors in the control plant ^{210}Po content were small because the activities of the plant materials grown in soils containing polonium were 10 to 1000 times greater than the control plant tissues.

Each sample measurement contains the systematic and accidental errors discussed above. Because the indeterminate sources of error are random, an estimate of the error in an average value is the Standard Error (S. E.). The errors expressed for the average distribution coefficients, the average transfer coefficients and the average ^{210}Po contents of plants and soils are Standard Errors. Texts by Snedecor and Cochran (1967) and Draper and Smith (1966) were consulted for statistical methods.

CHAPTER IV

RESULTS AND DISCUSSION

Equilibrium Distribution Coefficients (K_d)

The equilibrium distribution coefficients for two different soils

were determined with $PoCl_4$ solutions and PoO_2 suspensions. Polo-

nium oxide would be the chemical form expected from burn-up of a

SNAP radioisotope generator but polonium chloride, the more common

laboratory form, can be stored and handled in solution with more con-

venience. Table 4 lists the K_d's which were determined using both

chemical forms with Nunn silty clay loam and Darling gravelly sandy

loam. The distribution coefficients determined for the Nunn series

with polonium oxide suspension did not differ greatly from K_d's deter-

mined with polonium chloride solutions. However, for the Darling

series soil, K_d for the top soil layer changed from 104 for polonium

chloride to 24 for polonium oxide suspension. Therefore, other re-

ported distribution coefficients were determined using polonium oxide

suspensions. Table 5 lists the distribution coefficients for soils from

the Brown, Brunizem, Gray-Brown Podzol, Ground-Water Podzol,

Lateritic, Podzol and Red-Yellow Podzol Soil Groups.

The dependence of K_d on the number of hours of contact between

the soil and polonium oxide suspension is plotted in Figure 3 for Nunn

silty clay loam. A stage of equilibrium was reached after 16 hours

Table 4. Equilibrium distribution coefficients (K_d) for $^{210}PoCl_4$ and $^{210}PoO_2$

Soil Group and Location	Series and Type	Horizon	$PoCl_4$ $K_d \pm$ S.E.	PoO_2 $K_d \pm$ S.E.
Brown, Colorado	Nunn silty clay loam	A_{11}	131 ± 3	120 ± 5
		A_{12}	137 ± 6	203 ± 22
		B_1	1134 ± 56	310 ± 41
		B_{2t}	913 ± 25	766 ± 148
		B_{3Ca}	1372 ± 88	1213 ± 186
		C_{Ca}	1174 ± 65	643 ± 85
Podzol, Colorado	Darling gravelly sandy loam	A_2	104 ± 17	24 ± 1
		B_2	75 ± 23	13 ± 2
		B_{2ir}	37 ± 14	30 ± 7
		C_1	91 ± 30	66 ± 9
		C_2	$25 - 300$	75 ± 8
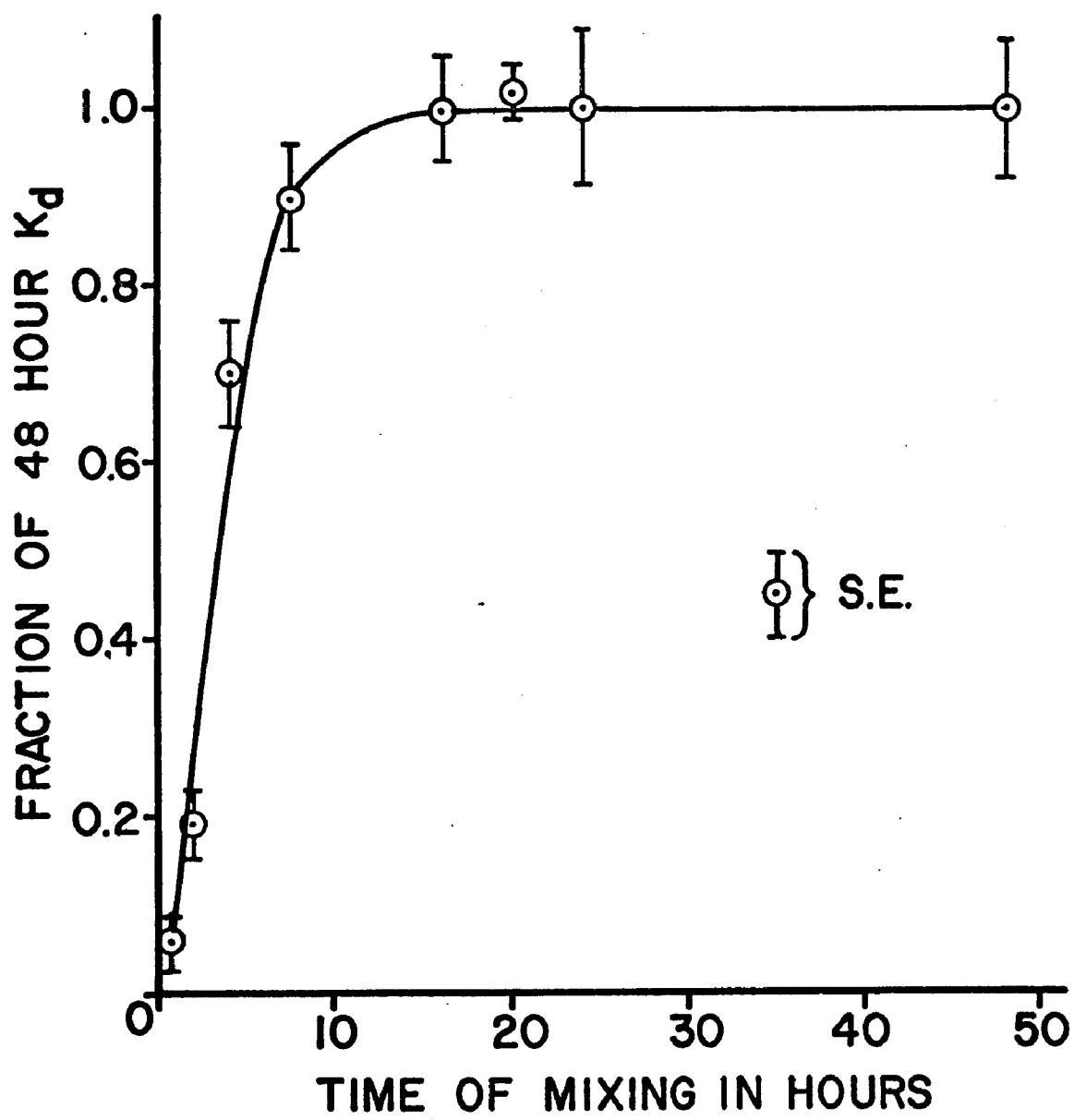
Table 5. Equilibrium distribution coefficients (K_d) for $^{210}PoO_2$.

Soil Group and Location	Series and Type	Horizon	$K_d \pm$ S. E.
Brown, Colorado	Fort Collins silty clay loam	Greenhouse mixture	230 ± 30
Brunizem, Iowa	Dinsdale silty clay loam	A	1030 ± 49
		B	976 ± 127
		C	723 ± 83
	Muscatine silty clay loam	A	1136 ± 118
		B	968 ± 32
		C	1830 ± 210
Gray-Brown Podzol, Wisconsin	Fayette silt loam	A_p	970 ± 160
		B_1	122 ± 3
		B_{21}	92 ± 3
		B_{22}	597 ± 55
		B_{23}	80 ± 2
		C_1	772 ± 29
	Lapeer loam	A_p	192 ± 26
		B_{21}	206 ± 11
		B_{22}	508 ± 34
		B_3	814 ± 42
		C_1	275 ± 9
Ground Water Podzol, Florida	Adamsville	A_1	26 ± 2
	Blanton	A_1	35 ± 3

Table 5., concluded

Soil Group and Location	Series and Type	Horizon	$K_d \pm$ S. E.
	Lakeland	A_1	25 ± 2
	Leon	A_1	17 ± 1
		A_2	15 ± 0.6
		B_n	55 ± 17
		C	77 ± 29
	Ruskin	A_1	17 ± 1
Podzol, Wisconsin	Gogebic sandy loam	A_2	254 ± 22
		Bir	371 ± 36
		Birh	137 ± 5
		B3	242 ± 25
	Onaway fine sandy loam	A_2	227 ± 20
		Birh	412 ± 150
		C_1	2248 ± 1200
		C_2	7020 ± 3600
Red-Yellow Podzol, Alabama	Amite	A_1	76 ± 11
	Congaree	A_1	405 ± 28
	Independence	A_1	188 ± 15
	Wickham	A_1	49 ± 3
Lateritic, Puerto Rico			137 ± 12

Figure 3. The change of distribution coefficient with mixing time for Nunn silty clay loam, A_{11} horizon.
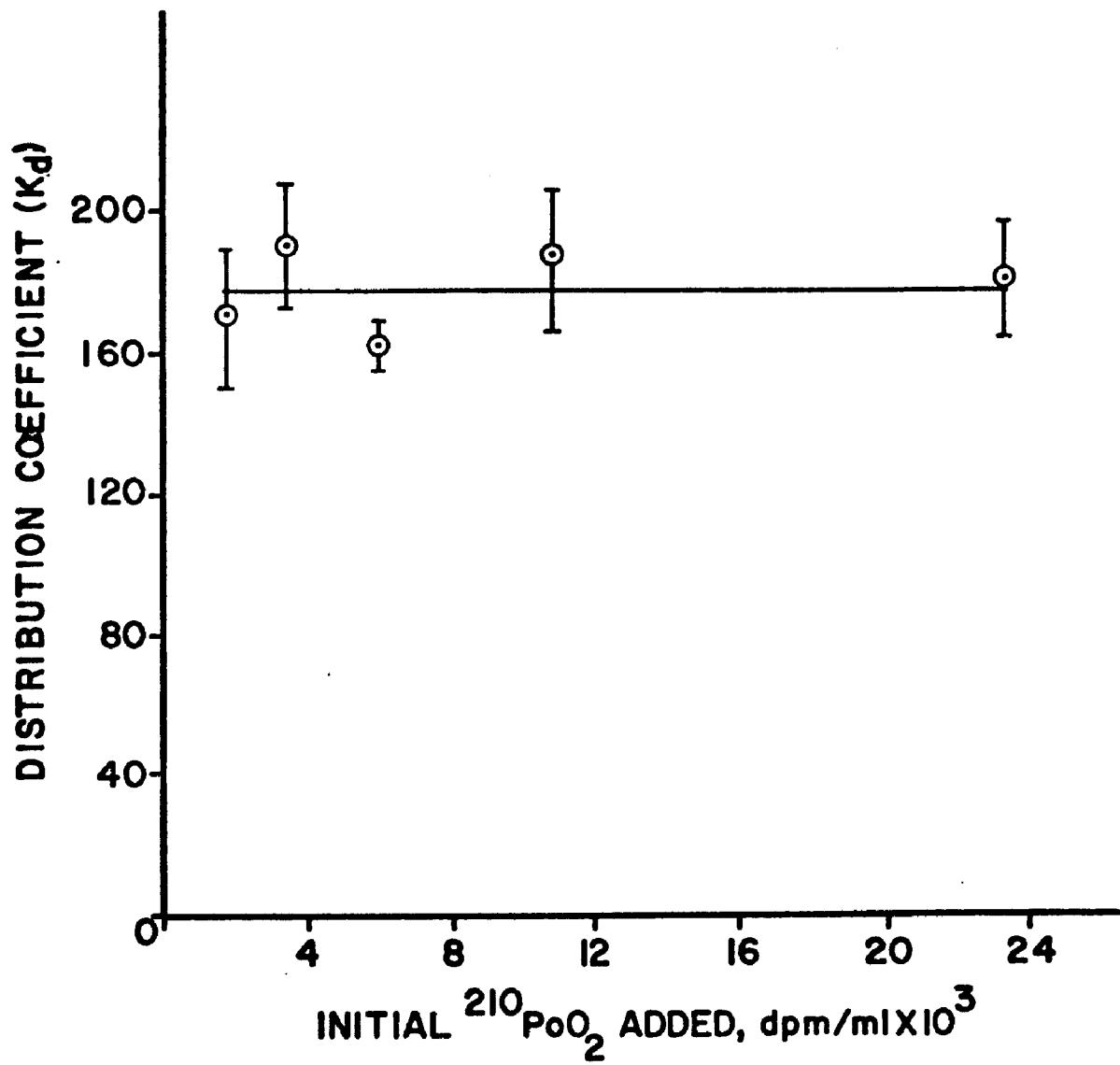
of mixing. Twenty hours of mixing were used for the distribution

coefficients listed in Table 5.

Figure 4 is a plot of the distribution coefficient versus the initial

concentration of polonium in suspension added to the soil sample. A

least squares fit indicates that the slope of the line is statistically

zero with 95 percent confidence, i. e. the distribution coefficient is

constant if determined with polonium oxide suspensions containing

between 900 to 23,500 dpm/ml. The distribution coefficients for the

soil samples in the study were determined with suspensions concentra-

tions between 10,000 and 20,000 dpm/ml.

Distribution coefficients of the soil mixtures used for the $^{210}PoO_2$

uptake study in the greenhouse were 230 $\pm$ 30 for the Fort Collins silty

clay loam and 303 $\pm$ 21 for the Fayette silt loam. The distribution

coefficients were determined using the soil, sand and peat moss mix-

tures. The 95 percent confidence intervals of the mixtures were 160 -

301 for the Fort Collins silty clay loam and 253 - 353 for the Fayette

silt loam. The confidence intervals overlap; therefore, the distribution

coefficients of the two soil mixtures were statistically equal. The dis-

tribution coefficient of the Leon sandy loam used for greenhouse experi-

ments was 17 $\pm$ 1.

Predictive equations for the distribution coefficient were developed

using the laboratory soil data of cation exchange capacity, percent

organic matter, pH, and mechanical analyses. The data were entered

Figure 4.   The distribution coefficients of Nunn silty clay loam, A_{11} horizon determined with increasing concentrations of $^{210}PoO_2$ in suspension.

into a Stepwise Multiple Regression Computer Program with the distribution coefficient considered the dependent variable. Dixon (1965) published a set of computer programs for regression analyses which includes the Stepwise Multiple Regression. The soil parameters of pH, cation exchange capacity, percent organic matter, percent silt and percent clay were considered independent variables. The first model used the data for the A horizons from 18 agricultural soil series. The statistical analysis indicated that an equation in terms of percent silt would predict the natural log of K_d. The empirical equation was

$$\ln K_d = 3.2 + (0.046 \pm 0.007) \text{ (Percent Silt)} \tag{1}$$

with a correlation coefficient, r, of 0.84. The data and equation are plotted in Figure 5.
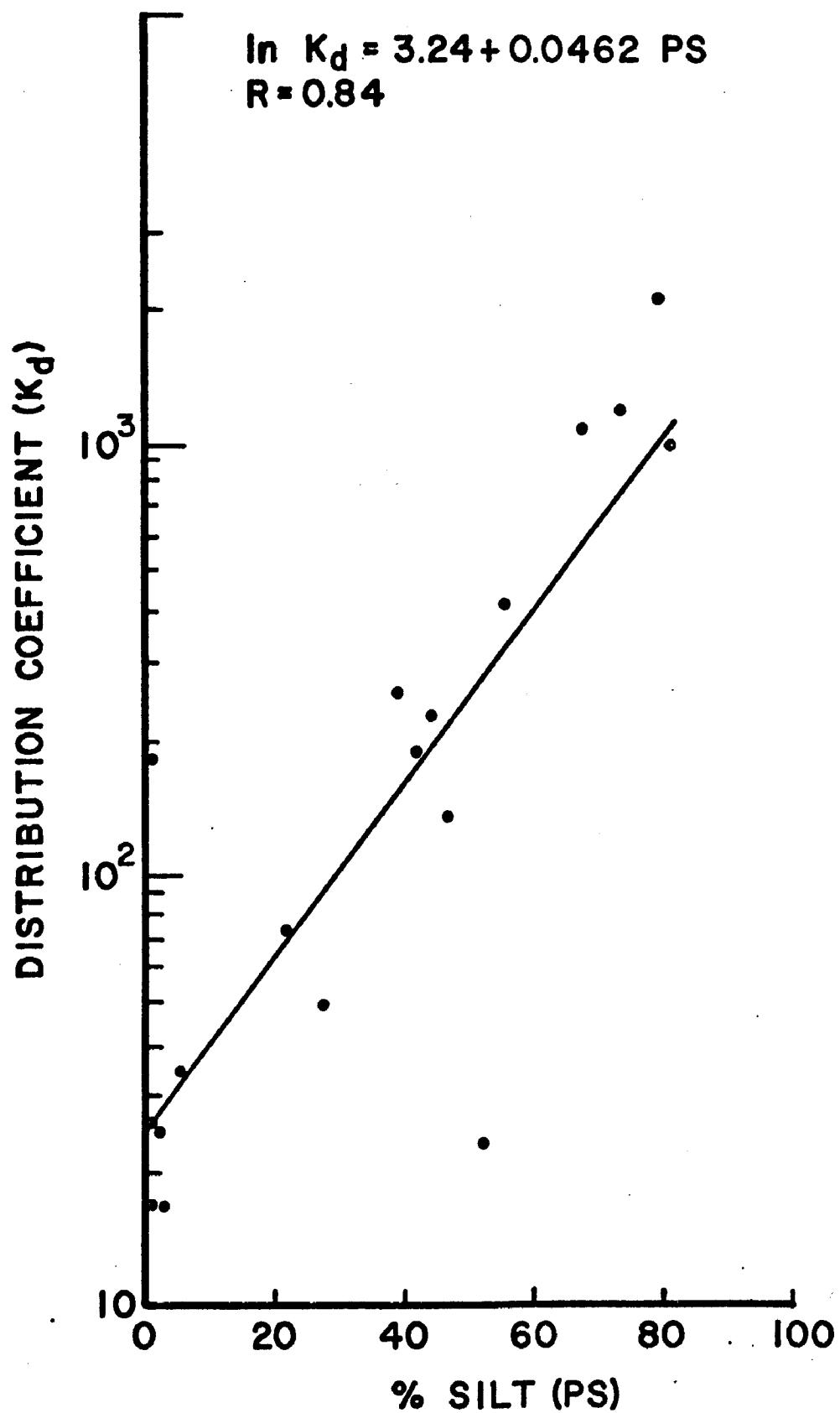
The second model which included the data for the B and C horizons indicated a $\ln K_d$ which was dependent upon the pH and silt content of the soil. The empirical equation was

$$\ln K_d = -1.3 + (0.034 \pm 0.007)\text{(Percent Silt)} + (0.88 \pm 0.20) \text{ pH} \tag{2}$$

with r = 0.74.

In most soils the ion exchange properties are usually associated with the clay fraction. Excluding the percent silt data from the input data for the Stepwise Multiple Regression Program computed an equation in terms of pH and percent clay for the A, B and C soil horizons.

Figure 5. The distribution coefficients of 18 top soils versus the silt content of the soil. The line is the least squares fit of the data.

$\ln K_d = 3.24 + 0.0462\ PS$

$R = 0.84$

The equation was

$$\ln K_d = -1.05 + (0.063 \pm 0.021)(\text{Percent Clay}) + (0.87 \pm 23) \, \text{pH} \quad (3)$$

with r = 0.65 which is lower than the multiple correlation coefficient of the model in terms of silt and pH. In the top soils the correlation between $\ln K_d$ and clay content was not statistically significant.

The lack of a correlation between the cation exchange capacity of the soil and the distribution coefficient suggests the polonium oxide is physically adsorbed. The correlation of the silt content of the soil with the adsorption of polonium may be explained by the behavior of polonium in water. Bagnall has reported the formation of an insoluble polonium hydroxide in neutral water solutions. The formula has been reported as $Po(OH)_4$ or $PoO(OH)_2$. Ziv and Efros have calculated the solubility product of $Po(OH)_4$ as 10^{-37}. Haissinsky observed polonium as a radiocolloid in solutions with pH's above 3; these solutions were very sensitive to impurities. The polonium was adsorbed on the impurities as a function of pH. Further, Thomas and Stannard demonstrated that 95 percent of the particles in a neutralized aged $PoCl_4$ solution were greater than 0.42 microns. The low solubility of the hydroxide, the adsorption of radiocolloids on solid materials as a function of pH and the particle nature of polonium suspensions would suggest the adsorption of polonium on silt or clay may be controlled by soil pH and the size of the radiocolloidal particles.

Table 6 lists the change in $^{210}PoO_2$ distribution coefficient with change in soil particle size for a sample of the C_{Ca} horizon of the Nunn silty clay loam. The K_d increased with decreasing particle size for particles to 2 microns indicating increased adsorption with greater surface area per gram. However, the K_d of particles less than 2 microns (clay) was the same as the 5 - 2 micron range of particle sizes. The change of K_d with particle size ranged from 457 to 950 for the silt and clay fractions indicating strong adsorption of $^{210}PoO_2$ in both fractions. The equations derived from the K_d data for the A, B and C soil horizons suggest either the silt or clay contents, in addition to soil pH, control the distribution coefficient of the C_{Ca} horizon. Agreement between the two methods suggests the empirical equations may be used for predicting distribution coefficients in other soils.

Equation (1) will broadly predict the soil retention of $^{210}PoO_2$ in agricultural top soils. Equation (2) based on data from all soils and soil horizons would be useful for predicting adsorption of polonium in widely differing soils. Soil groups which were not included in the study were Chestnut, Chernzems, Desert soils and Lithosols and the Intrazonal and Azonal soils. Goldberg, Fosberg, Sachet and Reimer (1965) report that 23.9 percent of the land mass of the earth is exposed rock and Lithosol type soils. The equation developed from the distribution coefficients for the combined data of the A, B and C soil horizons

Table 6. ^{210}Po oxide distribution coefficient as a function of particle size (Nunn silty clay loam Horizon C_{Ca}).

Particle Size Range (microns)	K_d
> 50	$(2.00 \pm 0.59) \times 10^2$
50 - 20	$(4.57 \pm 0.93) \times 10^2$
20 - 5	$(6.98 \pm 2.62) \times 10^2$
5 - 2	$(9.50 \pm 0.45) \times 10^2$
< 2	$(9.00 \pm 1.90) \times 10^2$

should predict the K_d's of soils that fall within the soil parameters defining equation (2). The pH range of the soil samples included in equation (2) was from 4.6 to 8.4 and the silt contents varied from less than 1 percent to 80 percent.

The distribution coefficient of a soil may not be predicted by the equations if chelates are included in fertilizers applied to agricultural soils. The behavior of polonium in soils containing chelates was not studied. Lindsay and Norvell (1959) demonstrated that the metal chelate complex is governed by soil pH and other ions in the soil solution competing for the ligand. Experiments to determine the change, if any, of the distribution coefficients for ^{210}Po in the presence of chelates are needed.

Elution of Polonium from Soil Columns

Movement of polonium dioxide through soil columns by water elution was studied in the Nunn silty clay loam, Darling gravelly sandy loam and Leon sandy loam soil samples. The column length for each sample was equal to the depth of the horizon of the soil where it was sampled. The top three horizons of the Nunn series each retained more than 98 percent of the added activity in the first two cm when eluted with one mean annual rainfall (13.5 inches of water). Distribution coefficients of the three soil horizons predict very strong retention by each horizon. The K_d of the A_{11} horizon was 120, the A_{12} horizon

203, the B_1 horizon 310. The column data for the Nunn soil is in

Table 7. Only 0.6 percent of the total activity placed on top of the A_{11}

soil column was detected in the eluate. The A_{12} and B_1 horizons had

strong retention indicating that the polonium oxide deposited on this

soil type would be retained in the first 30 cm of soil with greater than

98 percent retained within the first 2.5 cm of soil.

Table 8 contains the column data for the Darling gravelly sandy

loam which has distribution coefficients under 100 indicating the polo-

nium oxide is not strongly adsorbed. The soil is covered by a litter

layer consisting of roots, pine needles and mosses. Any polonium

deposited on the watershed (Fraser Alpine Area, Colorado) would have

to percolate through this organic material. An 8 cm column of the

litter layer eluted with one mean annual rainfall of 18.6 inches retained

only 20 percent of the polonium oxide with 80 percent of the activity

collected in the eluate. Elution of polonium oxide through the A_2 hori-

zon, the first soil layer, carried 5 percent of the activity through a

7 cm column. Analyses of the column sections indicated a large frac-

tion of the polonium had moved through the soil. Although 43 percent

of the polonium remained in the top cm of the column, the third cm of

the column contained a chromatographic peak representing 32 percent

of the total activity applied to the top of the soil. Additional elution

with water would carry the ^{210}Po in the chromatographic peak deeper

into the soil column.

Table 7.   Elution of polonium oxide through soil columns[*]

Soil Group - Brown
Series - Nunn Silty Clay Loam

Horizon	K_d	Depth (cm)	Percent of total ^{210}Po
A_{11}	170	1	97.3
		2	1.2
		3	0.6
		4-8	0.3
		Eluate	0.6
A_{12}	260	1	98.8
		2	0.8
		3	0.2
		4-12	0.2
		Eluate	<0.1
B_1	341	1	99.7
		2	0.2
		3-10	<0.1
		Eluate	Background

[*] Eluted with one mean annual rainfall at the rate of 1-2 ml/hr.

Table 8. Elution of polonium oxide through soil columns[*]

Soil Group - Podzol
Series - Darling Gravelly Sandy Loam

Horizon	K_d	Depth (cm)	Percent of total ^{210}Po
Litter layer		1	14.0
		2	1.8
		3	0.5
		4	1.0
		5	1.0
		6	0.8
		7	0.8
		8	0.6
		Eluate	80.0
A_2	24	1	42.8
		2	12.0
		3	32.4
		4	3.6
		5-8	4.2
		Eluate	5.0
B_2	13	1	95.6
		2	1.4
		3	0.8

Table 8., concluded.

Horizon	K_d	Depth (cm)	Percent of total ^{210}Po
		4	0.4
		5-12	1.3
		Eluate	0.5
B_{2ir}	29	1	95.1
		2	1.1
		3	0.6
		4	0.2
		5-10	1.0
		Eluate	2.1

* Eluted with one mean annual rainfall at the rate of 1-2 ml/hr.

Elutions and analyses of columns of the next two soil horizons are also listed in Table 8. Both horizons indicate strong retention of the polonium with a small fraction escaping in the eluate. The movement of the polonium oxide through the litter and first three soil horizons would allow only approximately 10^{-6} of the total activity to reach the C horizons. However, the movement of water in a watershed soil is not usually perpendicular to the soil horizons but is a function of the slope, rainfall intensity or snow melt rate.

An experiment peripheral to the column experiments was conducted with the Darling gravelly sandy loam. The complete experimental design and the derivation of equations are in the Appendix. Polonium oxide suspension was applied to the litter layer as a 200 μCi point source one meter from water collection vessels at the base of a 30 percent simulated slope. Cores from the soil on which one mean annual rainfall (18.6 inches) of ice was melted indicated both vertical and horizontal movement of the $^{210}PoO_2$ into the soil. The run-off water collected at the base of the experimental slope indicated only 10^{-7} of the original activity moved through the soil. Data from soil cores were used to predict the movement of a uniform deposition of $^{210}PoO_2$ to a stream. The equations derived indicated that only the first 50 cm bordering a stream would contribute significant contamination to the water. The quantities deposited on surface waters would be more significant.

Elution of a 7 cm column of Leon sandy loam A_1 horizon indicated 18 percent of the added polonium oxide was moved through the column by one mean annual rainfall of 55 inches of water (Table 9). The fraction of the total activity retained by the first cm of the column was 76 percent. Comparison of the activity eluted from columns of equal lengths of Darling gravelly sandy loam (K_d = 25) and Leon sandy loam (K_d = 17) indicated 5 percent of the activity was eluted from the former and 14 percent of the total was removed from the latter by equal volumes of water.

The eluate collected from the fraction collector was examined for chromatographic peaks moving through the soil. Each column indicated a prompt pulse of activity that arrived with the water at the bottom of the soil column. The pulse contained 0.2 percent of the activity placed on the Nunn silty clay loam column, 3.3 percent of the activity placed on the Darling gravelly sandy loam and 8.3 percent of the activity placed on top of the Leon sandy loam column. The remaining eluate collected from each column contained decreasing amounts of activity with each additional aliquot and no chromatographic peak was observed passing through the soil into the eluate. However, the amount of water passed through each column (one mean annual rainfall) may not have been sufficient to elute adsorbed forms of polonium signifying the distribution coefficient in each soil was substantial. The major portion of the polonium dioxide deposited on soils with K_d's of

Table 9.   Elution of polonium oxide through soil columns[*]

Soil Group - Ground-Water Podzol
Series - Leon Sandy Loam

Horizon	K_d	Depth (cm)	Percent of total ^{210}Po
A_1	17	1	76.2
		2	1.3
		3	1.1
		4	1.1
		5	0.8
		6	0.6
		7	0.4
		8	0.2
		Eluate	18.3

[*] Eluted with one mean annual rainfall at the rate of 1-2 ml/hr.

17 or greater would be retained in the top layers of soil. Movement to a water table or aquifer would be minimal.

Natural Polonium-210 Content of Soils

The natural ^{210}Po contents of 17 top soils and 9 profiles were determined. The greatest amount of polonium detected in top soil was 3.47 $\pm$ 0.64 pCi/g and the least amount in top soil was 0.22 $\pm$ 0.02 pCi/g. Table 10 contains the data for soils examined from Alabama, Colorado, Florida, Iowa, Puerto Rico and Wisconsin. The range of ^{210}Po in top soils represents six soil groups. The average concentration in each soil group is listed in Table 11. The average concentrations indicate the distribution of ^{210}Po in soils is uniform for the soil groups tested. The range of concentrations indicate specific soil series may have characteristics that increase or decrease the ^{210}Po content.

The natural levels of polonium detected in the soil samples were in the same range of values reported by other investigators. Tso, Hallden and Alexander reported the range of ^{210}Po activity in eastern U. S. tobacco growing soils as 0.26 to 1.01 pCi/g. Berger et al. reported an average of 2.03 pCi/g of soil for mineral soils sampled in Wisconsin. Francis et al. reports a higher average value of 3.1 pCi/g for Wisconsin soils. However, Francis et al. used a H_2SO_4-$HClO_3$-HF decomposition of the soil rather than a 12 N HCl leaching process. Decomposition of the soil minerals for determination of the ^{210}Po content would release polonium trapped in crystal structures.

Table 10. Natural polonium-210 content of soils.

Soil Group and Location	Series and Type	Horizon	^{210}Po in pCi/g ± S. E.
Brown, Colorado	Fort Collins silty clay loam	A_{11}	0.48 ± 0.03
	Nunn silty clay loam	A_{11}	0.90 ± 0.04
		A_{12}	0.94 ± 0.04
		B_1	0.67 ± 0.08
		B_{2t}	0.58 ± 0.03
		B_{3Ca}	0.65 ± 0.02
		C_{Ca}	0.71 ± 0.03
Podzol, Colorado	Darling gravelly sandy loam	A_2	0.22 ± 0.02
		B_2	0.22 ± 0.04
		B_{2ir}	0.64 ± 0.06
		C_1	0.43 ± 0.05
		C_2	0.59 ± 0.05
Podzol, Wisconsin	Gogebic sandy loam	A_p	1.53 ± 0.16
		B_{ir}	0.33 ± 0.05
		B_{irh}	1.00 ± 0.10
		B_{3x}	0.23 ± 0.01
		C_1	0.17 ± 0.01
Podzol, Wisconsin	Onaway sandy loam	A_2	0.60 ± 0.02
		B	0.42 ± 0.16
		C_1	0.26 ± 0.02
		C_2	0.16 ± 0.01

Table 10, continued

Soil Group and Location	Series and Type	Horizon	^{210}Po in pCi/g $\pm$ S. E.
Gray-Brown Podzol, Wisconsin	Fayette silt loam	A_p	1.48 ± 0.06
		B_1	0.68 ± 0.12
		B_{21}	0.86 ± 0.02
		B_{22}	0.83 ± 0.05
		B_{23}	1.01 ± 0.14
		B_3	1.33 ± 0.37
		C_1	1.27 ± 0.24
Gray-Brown Podzol, Wisconsin	Lapeer loam	A_p	1.56 ± 0.09
		B_{21}	0.26 ± 0.05
		B_{22}	0.77 ± 0.17
		B_3	0.43 ± 0.08
		C_1	0.45 ± 0.05
Brunizem, Iowa	Dinsdale silty clay loam	A	1.61 ± 0.33
		B	0.78 ± 0.16
		C	0.41 ± 0.05
	Muscatine, silty clay loam	A	1.54 ± 0.30
		B	1.11 ± 0.08
		C	1.91 ± 0.74
Red-Yellow Podzol, Alabama	Amite	A	0.78 ± 0.26
	Congaree	A	3.47 ± 0.64
	Independence	A	0.63 ± 0.02

Table 10, concluded

Soil Group and Location	Series and Type	Horizon	^{210}Po in pCi/g $\pm$ S. E.
Ground-Water Podzol, Florida	Wickham	A	1.47 $\pm$ 0.31
	Adamsville	A	0.46 $\pm$ 0.07
	Blanton	A	1.35 $\pm$ 0.29
	Lakeland	A	0.89 $\pm$ 0.31
	Leon	A_1	1.47 $\pm$ 0.38
		A_2	0.29 $\pm$ 0.05
		B_n	0.33 $\pm$ 0.14
		C	0.15 $\pm$ 0.09
	Ruskin	A	1.98 $\pm$ 0.27

Table 11. Average ^{210}Po content of soils from each soil group.

Soil Group and Location	Number of Series Sampled	^{210}Po Content (pCi/g ± S. E.)	Range of ^{210}Po Content
Brown, Colorado	1	0.90 ± 0.04	
Brunizem, Iowa	2	1.57 ± 0.33	1.54 - 1.61
Gray-Brown Podzol, Wisconsin	2	1.51 ± 0.09	1.47 - 1.56
Ground-Water Podzol, Florida	5	1.23 ± 0.35	0.46 - 1.98
Podzol, Colorado, Wisconsin	3	0.78 ± 0.16	0.22 - 1.53
Red-Yellow Podzol, Alabama	4	1.59 ± 0.31	0.63 - 3.47

The source of ^{210}Po in soils is the radioactive decay of ^{238}U which decays through several radioisotopes to form ^{210}Po. Because ^{210}Po is formed from decay of ^{238}U, the polonium content of soils would be expected to correlate with the uranium content of soils. Investigations of soils for correlation between ^{238}U and ^{226}Ra, a radionuclide formed in the decay chain before ^{210}Po, indicate no correlation existed in the reports reviewed by Osburn. The lack of correlation was attributed to the chemical differences of uranium and radium. Telfair, Garrison and Smith (1960) observed that radium was not in equilibrium with its long lived parents because of soil leaching and weathering processes.

Correlations between ^{226}Ra and ^{210}Po concentrations in soil only broadly predict the ^{210}Po content. Tso et al. measured the radium and polonium activities of the same soils. Ratios of ^{226}Ra/^{210}Po calculated from the values reported by Tso et al. range from 1.1 to 3.3. They concluded ^{210}Po is not in equilibrium with ^{226}Ra because ^{222}Rn generated in the soil is partially trapped in soil or escapes to the air.

Radon-222 decaying to its daughter products either in air or soil leads to the formation of ^{210}Pb and ^{210}Po. Radon daughter products attached to airborne particles may result in fallout that is intercepted by plants or return to the top soil.

Study of the Polonium Species in Soil Solution

Investigations of soil solutions from $^{210}PoO_2$ contaminated soil

indicated a small amount of the polonium oxide remains in solution

as a soluble or suspended chemical species. Electrophoresis and

dialysis were used to investigate the movement of the species in an

electric field and through a semi-permeable membrane.

Paper electrophoresis of an aqueous suspension of polonium oxide

placed on a paper strip saturated with Nunn silty clay loam soil solu-

tion is plotted in Figure 6. Application of 500 volts across the paper

strip for one hour moved about 10 percent of the total polonium 4.5 cm

toward the positive electrode. Soil solution containing polonium in

equilibrium with the Nunn silty clay loam was applied to a paper strip

saturated with the same soil solution. Application of 500 volts for one

hour resulted in movement of polonium toward both the positive and

negative electrodes indicating the presence of negatively and positively

charged species. Figure 7 is a plot of the data indicating the fraction

of the total polonium that moved toward each electrode. In this case

there was a greater amount of polonium that moved in the electric

field. To confirm that the electric field had caused the movement, a

paper strip was prepared in the same manner, but no electrical

potential was applied. Figure 8 indicates the results of the experiment

Only 2 percent of the total activity applied moved after one hour; the

movement was to the next 0.5 cm. Figures 9, 10 and 11 contain the

Figure 6

PAPER ELECTROPHORESIS OF STOCK PoO_2 IN BROWN SOIL GROUP, NUNN SILTY CLAY LOAM, LEACHATE

SOIL SOLUTION pH 8.4
OPERATION TIME 1 HOUR AT 500 V, 4 MA

Figure 7

PAPER ELECTROPHORESIS OF PoO₂ SOIL LEACHATE

BROWN SOIL GROUP, NUNN SILTY
CLAY LOAM A₁₁ HORIZON
SOIL SOLUTION PH 7.8
OPERATION TIME I HOUR AT 500 V,
5MA
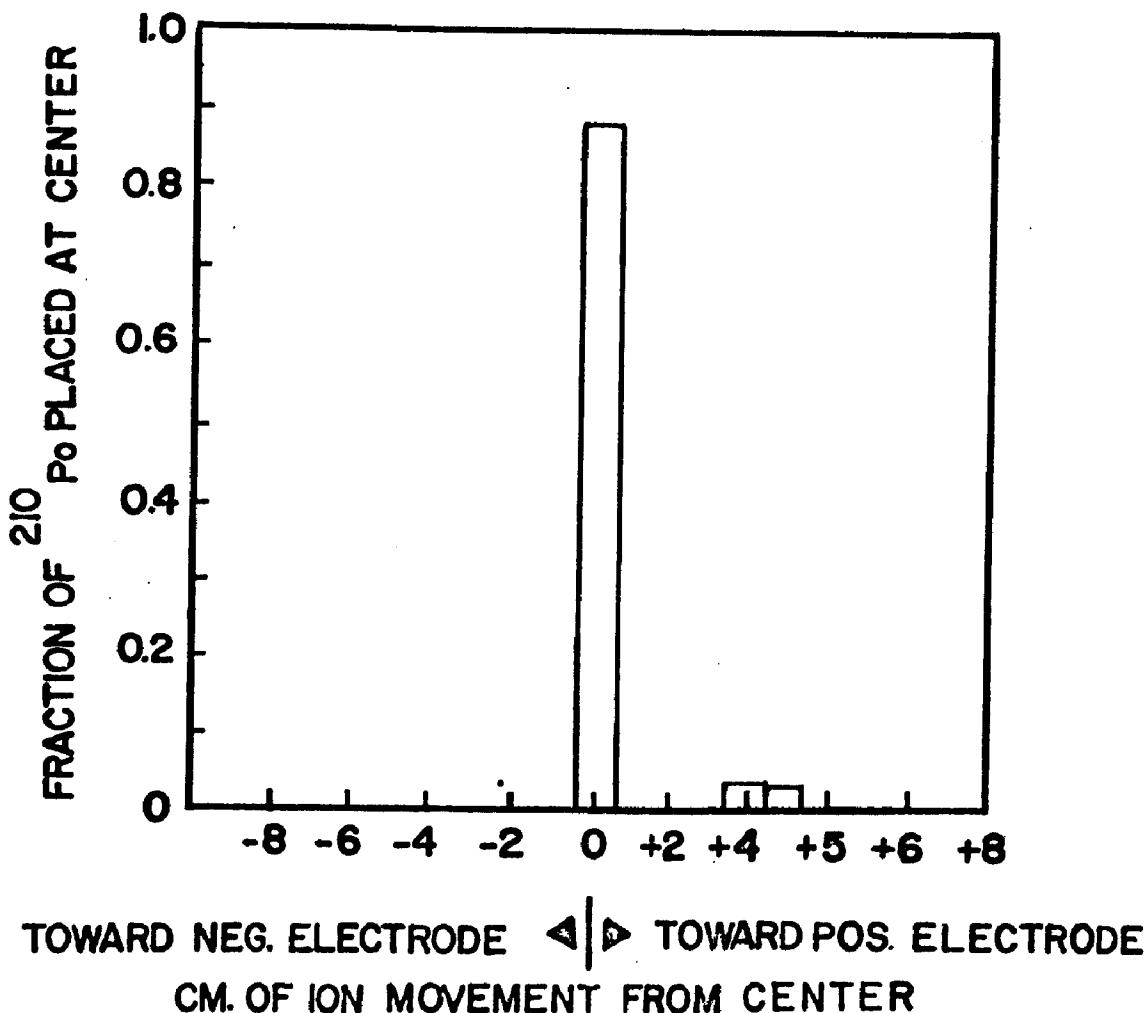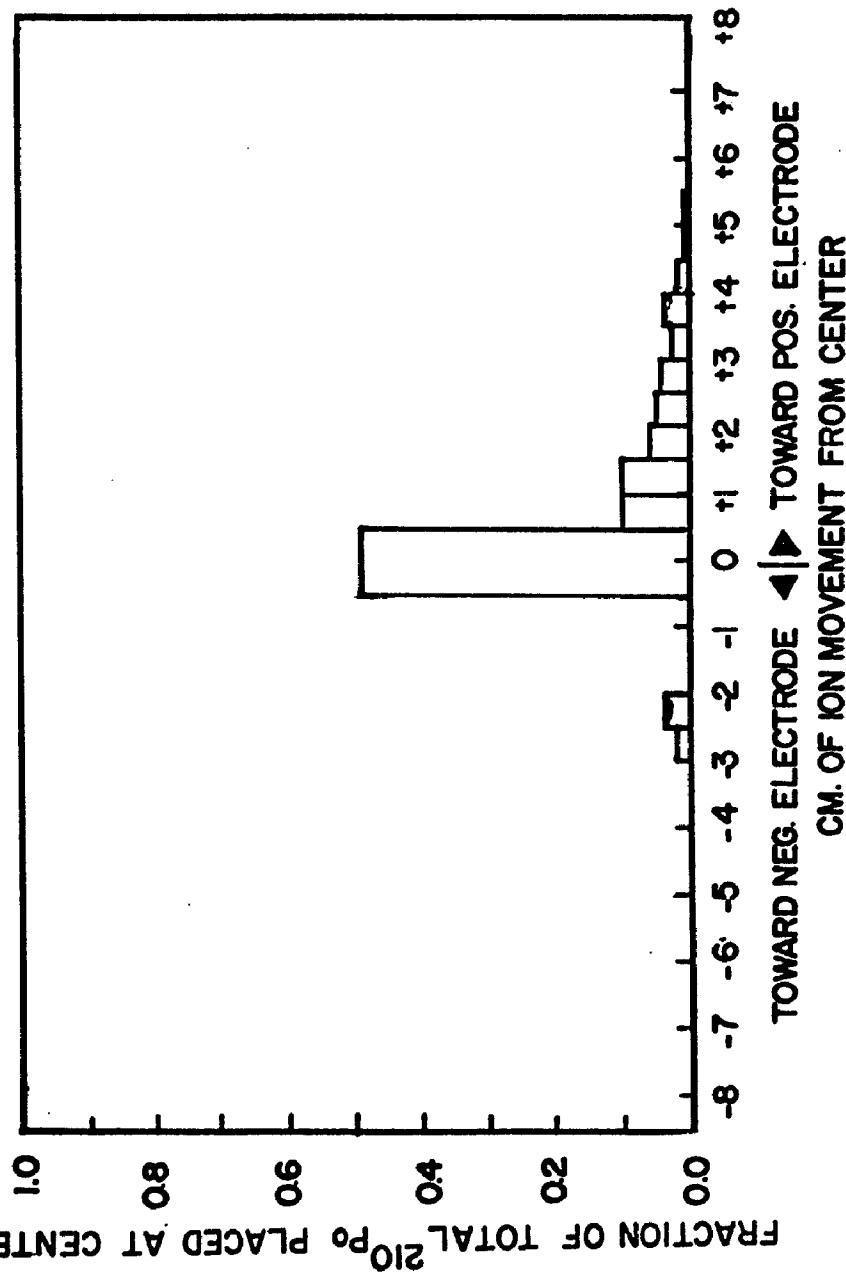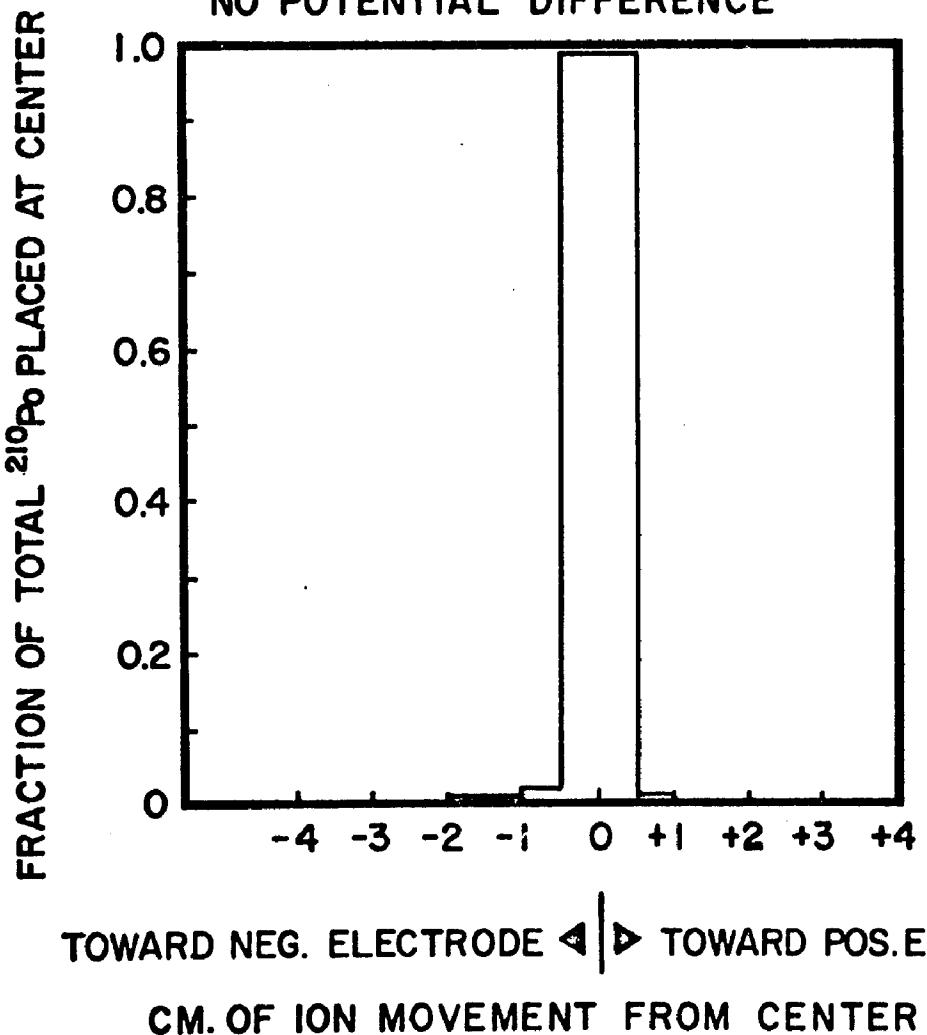
Figure 8

TEST FOR DIFFUSION OF PoO$_2$ SOLUTION

PoO$_2$ SOLUTION ON PAPER STRIP FOR I HOUR
NO POTENTIAL DIFFERENCE

Figure 9

PAPER ELECTROPHORESIS OF ^{210}PoO$_2$ SOIL LEACHATE
BROWN SOIL, FORT COLLINS SILTY CLAY LOAM
SOIL SOLUTION pH 7.6
OPERATION TIME 1hr. AT 500V 5ma
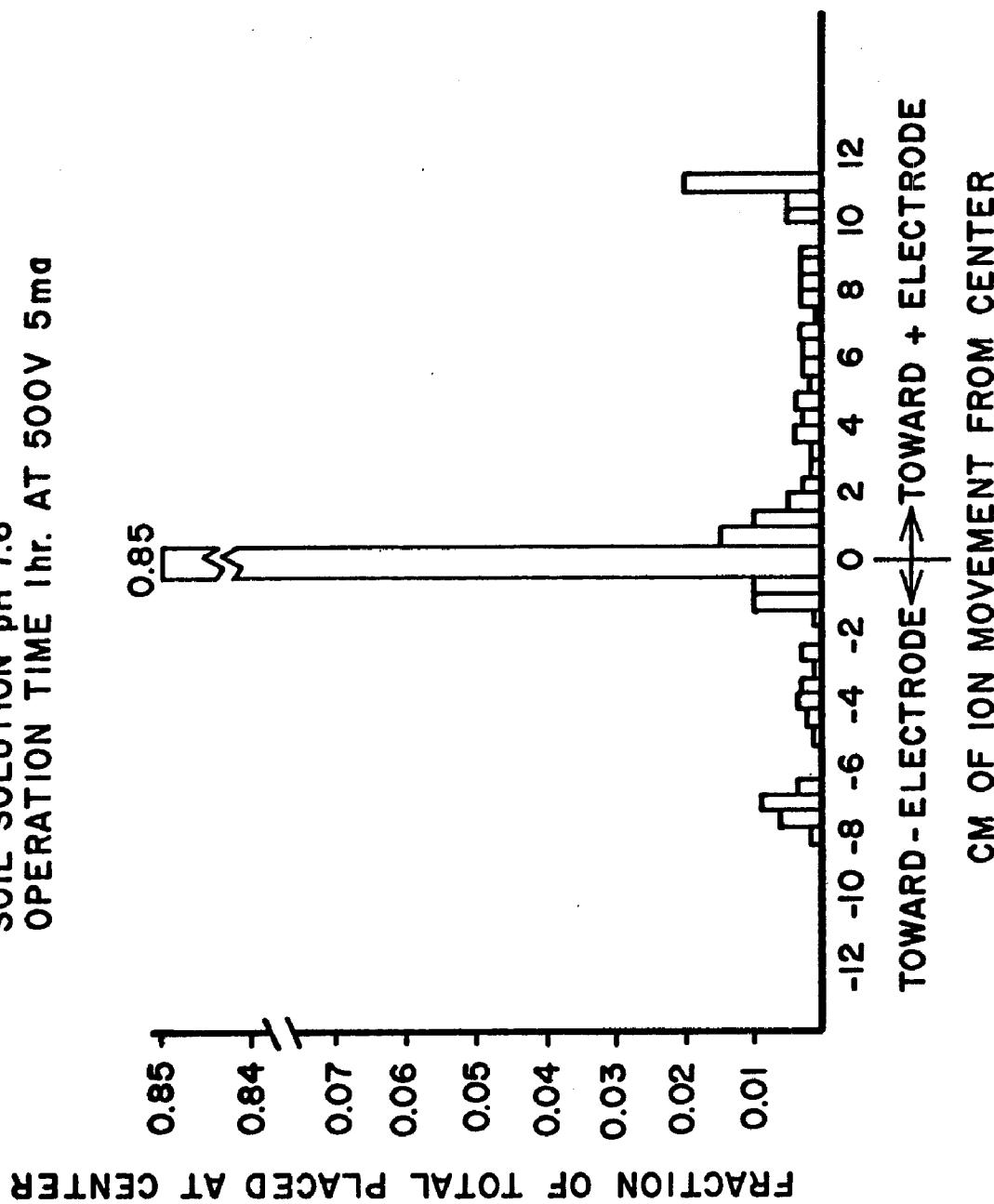
Figure 10

PAPER ELECTROPHORESIS OF $^{210}PoO_2$ SOIL LEACHATE

PODZOLIC SOIL, GOGEBIC SANDY LOAM
SOIL SOLUTION pH 6.6
OPERATING TIME 1hr AT 500V 5ma

FRACTION OF TOTAL PLACED AT CENTER

CM OF ION MOVEMENT FROM CENTER

TOWARD - ELECTRODE   TOWARD + ELECTRODE
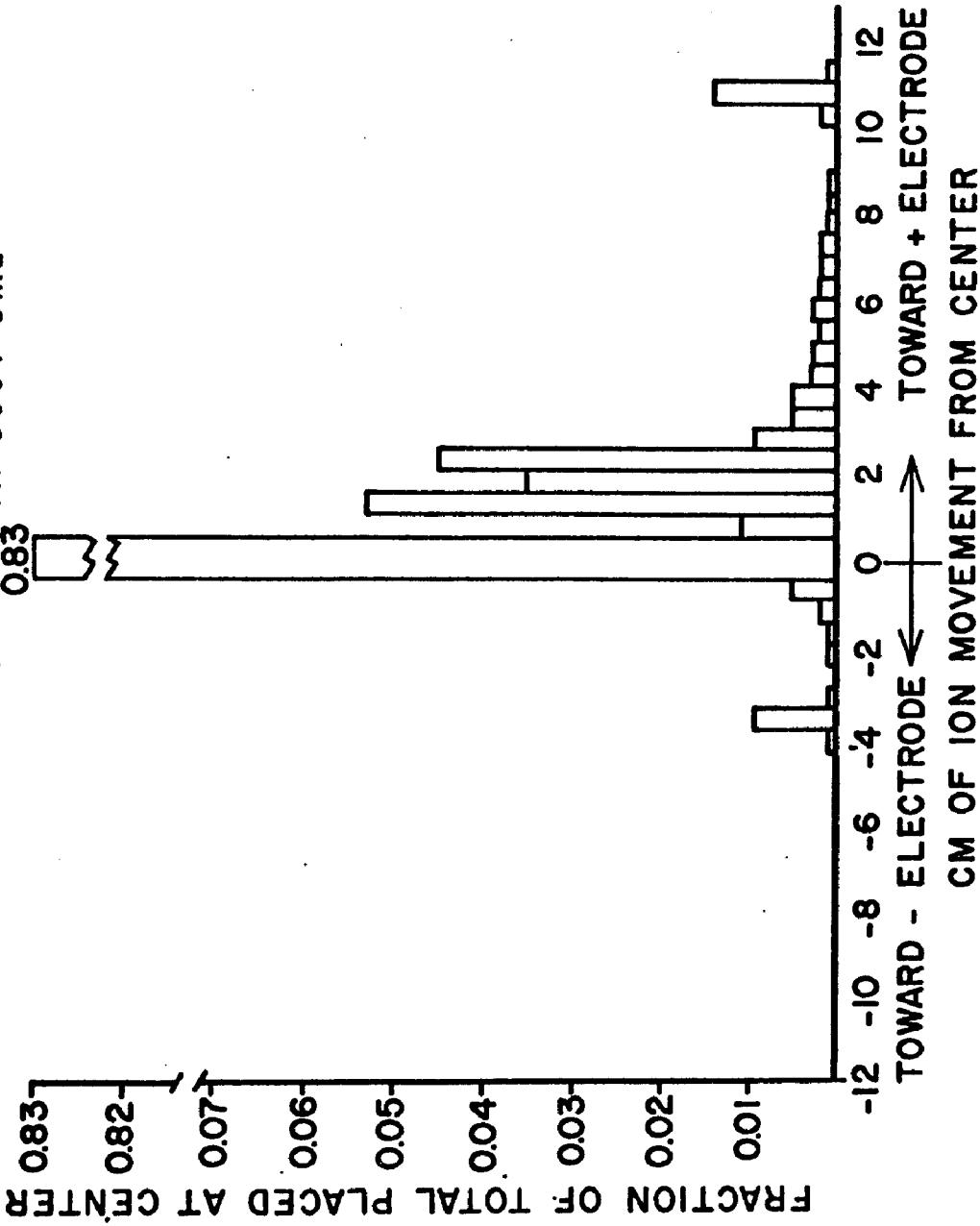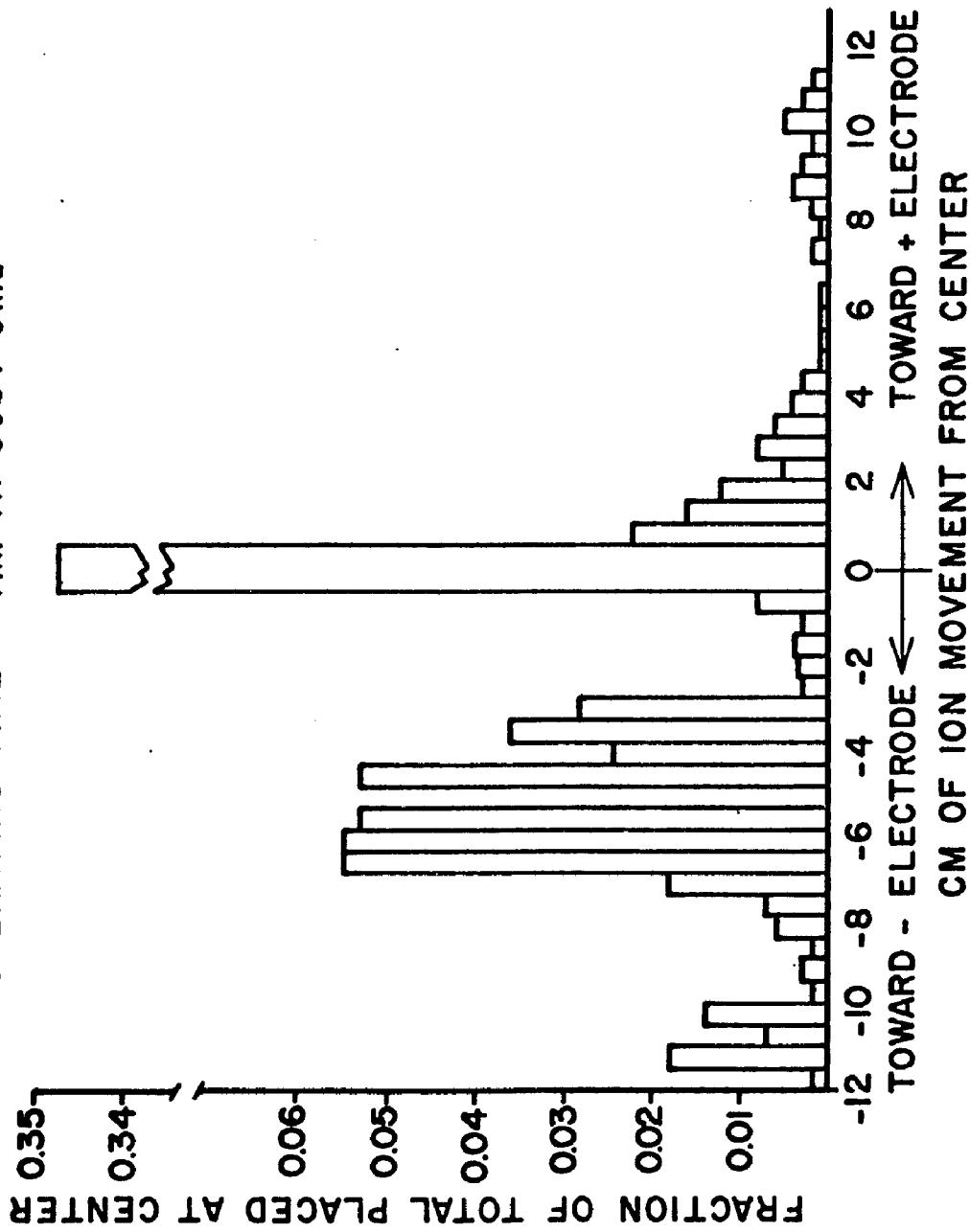
Figure 11

PAPER ELECTROPHORESIS OF $^{210}PoO_2$ SOIL LEACHATE

PODZOLIC SOIL, ONAWAY FINE SANDY LOAM
SOIL SOLUTION pH 6.8
OPERATING TIME   1hr.   AT 500V 5ma

plots of the data from $^{210}PoO_2$ contaminated Gogebic sandy loam, Fort Collins silty clay loam and Onaway fine sandy loam soil solutions.

Electrophoresis experiments indicated charged species of polonium remain in soil solution after mixing $^{210}PoO_2$ suspensions with soil samples. Positive and negative species of polonium were observed in the soil solutions tested. Polonium detected at the origin following electrophoresis may be a neutral species or ^{210}Po strongly adsorbed by the paper strips. In the Nunn, Fort Collins and Gogebic series the predominately charged species were negative. The major species in the Onaway series were positive.

The predominately positive ^{210}Po species in Onaway series soil solution may exist because of the low cation exchange capacity (C. E. C.) and low organic matter content of the soil sample. The Onaway series contained 2. 4 percent organic matter and the C. E. C. was 5. 1 meq/ 100 g. The Gogebic series sample contained 4. 0 percent organic matter and had a C. E. C. of 16. 8 meq/100 g. The pH of the Gogebic loam was 6. 6 compared to 6. 8 for the Onaway fine sandy loam. The low C. E. C. and organic matter suggest an abundant supply of cations may exist in the Onaway soil solution. Haissinsky and Weigel noted that the radiocolloidal form of polonium is most prevalent at pH 6. 9. If the radiocolloidal form were surrounded by positive ions, it may form a species able to move toward the negative electrode during electrophoresis.

Experiments using dialysis of the polonium contaminated soil solution on one side of a semi-permeable membrane with soil solution on the other side indicated ^{210}Po species were present that pass through a membrane. Dialysis data are summarized in Table 12. Soil solution from ^{210}PoO$_2$ contaminated Nunn silty clay loam used as a sample solution and uncontaminated soil solution used as the receiving solution indicated 37 percent of the polonium passed through the membrane. Using a water solution of polonium oxide dialyzing into filtered deionized water, only 6 percent of the activity was dialyzed. Soil solutions from polonium oxide contaminated Fayette, Gogebic and Onaway soils dialyzed into soil solutions from each series indicated approximately equal amounts of the soil solution polonium passed through the semi-permeable membrane. Soil solutions spiked directly with ^{210}PoO$_2$ suspension without contact with soil solid phases indicated greater amounts of the polonium passed through the membrane.

Dialysis of the soil solutions indicated approximately the same amounts of colloidal and ionic forms of polonium existed in the different soil solutions tested. However, addition of polonium oxide suspension to soil solutions without contact with the solid phases indicated an increase of ionic forms. The direct spike dialysis experiments suggest that ^{210}PoO$_2$ reacts with chemical species in soil solution. Thus, the ionic forms observed in soil solution after ^{210}PoO$_2$ suspension contact with the soil may be formed by reaction with soil solution, but

Table 12. Dialysis of $^{210}PoO_2$ in deionized water and soil solutions.

Sample	Fraction Collected in Receiving Solution
$^{210}PoO_2$ suspension	0.063
Gogebic soil solution	0.32
Fayette soil solution	0.34
Nunn soil solution	0.37
Onaway soil solution	0.35
Fayette soil solution $^{210}PoO_2$ suspension added directly	0.58
Onaway soil solution $^{210}PoO_2$ suspension added directly	0.47
Nunn soil solution $^{210}PoO_2$ suspension added directly	0.48

solid phases in the soil may preferentially adsorb specific ionic forms.

Polonium-210 Uptake by Plants

Two plantings were analyzed for polonium-210 movement from Fort Collins silty clay loam. The first crop was grown in $PoCl_4$ contaminated soil and the second planting was grown in PoO_2 contaminated soil. In addition enough soil was contaminated with RaDEF (^{210}Pb, ^{210}Bi and ^{210}Po) so that ^{210}Po uptake by barley, peas and radishes could be measured with radioactive precursors of ^{210}Po present. A third planting in the greenhouse was in a polonium oxide contaminated Fayette silt loam from the Gray-Brown Podzol Soil Group and a fourth planting was in the Leon sandy loam with added $^{210}PoO_2$.

The ^{210}Po uptake by plants was calculated from the disintegration per minute of ^{210}Po per gram dry weight of plant tissue corrected for the average control plant content determined on a dry weight basis. A transfer coefficient, U, was calculated using the following relationship:

$$U = \frac{(\text{corrected dpm of } ^{210}Po \text{ per gram dry plant})}{(\text{dpm of } ^{210}Po \text{ per gram of dry soil at harvest})} \tag{4}$$

Transfer coefficients calculated for plants grown on polonium chloride contaminated soil are tabulated in Table 13. Table 14 contains the data calculated for plants grown in polonium oxide added to Fort Collins silty clay loam and Table 15 lists the data for plants grown in Fayette

Table 13. Polonium chloride transfer coefficients (U) – Fort Collins silty clay loam.

Species and Variety	Tissue	No. of Samples	Dry Wt. Transfer Coefficient ± S.E.	Wet Wt. Transfer Coefficient ± S.E.
Barley (Hordeum vulgare, Betzes)	Kernels	20	$(7.4 \pm 3.3) \times 10^{-4}$	$(6.5 \pm 2.9) \times 10^{-4}$
	Stems and Leaves	16	$(9.7 \pm 3.4) \times 10^{-3}$	$(6.6 \pm 2.2) \times 10^{-4}$
Wheat (Trictium spp., Scout)	Kernels	7	$(1.7 \pm 0.6) \times 10^{-5}$	$(9.0 \pm 4.0) \times 10^{-6}$
	Chaff	7	$(1.1 \pm 0.1) \times 10^{-3}$	$(9.4 \pm 1.5) \times 10^{-4}$
	Stems and Leaves	9	$(1.4 \pm 0.9) \times 10^{-3}$	$(7.5 \pm 0.6) \times 10^{-4}$
Onion (Allium cepa, Mountain Danver)	Bulbs	13	$(4.1 \pm 1.0) \times 10^{-4}$	$(3.7 \pm 0.9) \times 10^{-5}$
	Tops	27	$(9.0 \pm 1.4) \times 10^{-4}$	$(6.6 \pm 1.1) \times 10^{-5}$
Potato (Solanum tubersum, Norgold)	Whole	15	$(1.2 \pm 0.2) \times 10^{-3}$	$(2.5 \pm 0.4) \times 10^{-4}$
	Pulp	14	$(1.8 \pm 0.7) \times 10^{-4}$	$(6.3 \pm 2.9) \times 10^{-5}$
	Peel	8	$(8.4 \pm 0.8) \times 10^{-3}$	$(1.4 \pm 0.1) \times 10^{-3}$
	Greens	14	$(9.3 \pm 2.5) \times 10^{-3}$	$(9.2 \pm 2.5) \times 10^{-4}$
Radish (Raphanus sativus, Cavalier)	Roots	22	$(7.5 \pm 1.0) \times 10^{-3}$	$(5.7 \pm 1.1) \times 10^{-4}$
	Greens	22	$(8.9 \pm 1.9) \times 10^{-3}$	$(7.5 \pm 1.3) \times 10^{-4}$
Pea (Pisum sativum, Dwarf)	Peas	9	$(1.4 \pm 0.6) \times 10^{-4}$	$(2.9 \pm 1.4) \times 10^{-5}$
	Pod	9	$(5.2 \pm 2.1) \times 10^{-4}$	$(6.6 \pm 2.3) \times 10^{-5}$
	Vines	8	$(1.6 \pm 0.9) \times 10^{-3}$	$(3.1 \pm 1.8) \times 10^{-4}$

Table 13, concluded.

Species and Variety	Tissue	No. of Samples	Dry Wt. Transfer Coefficient ± S. E.	Wet Wt. Transfer Coefficient ± S. E.
Tomato (Lycopersion esculentum, Marglobe)	Pulp	27	$(1.5 \pm 1.4) \times 10^{-4}$	$(1.6 \pm 1.5) \times 10^{-5}$
	Juice	27	--	$(3.6 \pm 0.8) \times 10^{-5}$
	Stems	25	$(1.7 \pm 0.4) \times 10^{-3}$	$(1.8 \pm 0.4) \times 10^{-4}$
	Leaves	22	$(3.3 \pm 0.6) \times 10^{-3}$	$(4.7 \pm 0.8) \times 10^{-4}$
Spinach (Spinacia oleracea, Badger Savoy)	Stems and Leaves	15	$(2.2 \pm 0.4) \times 10^{-3}$	$(2.8 \pm 0.5) \times 10^{-4}$
Celery (Apium gravelens dulce, Golden Dwarf	Edible Stem	19	$(2.4 \pm 0.4) \times 10^{-3}$	$(2.1 \pm 0.3) \times 10^{-4}$
	Stem	18	$(2.0 \pm 0.5) \times 10^{-3}$	$(2.2 \pm 0.7) \times 10^{-4}$
	Leaves	25	$(1.0 \pm 0.1) \times 10^{-3}$	$(1.5 \pm 0.3) \times 10^{-4}$

Table 14. Polonium oxide transfer coefficient (U) - Fort Collins silty clay loam, K_d = 230.

Species and Variety	Tissue	No. of Samples	Dry Wt. Transfer Coefficient ± S.E.	Wet Wt. Transfer Coefficient ± S.E.
Barley (Hordeum vulgare, Betzes)	Kernels	24	$(2.6 \pm 0.2) \times 10^{-4}$	$(2.1 \pm 0.2) \times 10^{-4}$
	Stems and Leaves	23	$(5.1 \pm 1.7) \times 10^{-3}$	$(9.1 \pm 3.1) \times 10^{-4}$
Corn (Zea mays, Hybrid Sweet)	Kernels	10	$(1.8 \pm 0.3) \times 10^{-5}$	$(3.5 \pm 0.7) \times 10^{-6}$
	Cob	10	$(3.6 \pm 0.6) \times 10^{-5}$	$(5.0 \pm 0.8) \times 10^{-6}$
	Tassel	9	$(5.4 \pm 1.1) \times 10^{-5}$	$(1.5 \pm 0.3) \times 10^{-5}$
	Leaves (50 days)	16	$(2.1 \pm 0.4) \times 10^{-3}$	$(2.6 \pm 0.5) \times 10^{-4}$
	Leaves (130 days)	13	$(1.2 \pm 0.3) \times 10^{-3}$	$(1.4 \pm 0.4) \times 10^{-4}$
	Stems (50 days)	22	$(1.4 \pm 0.4) \times 10^{-4}$	$(1.6 \pm 0.4) \times 10^{-5}$
	Stems (130 days)	12	$(3.9 \pm 0.9) \times 10^{-4}$	$(4.8 \pm 1.1) \times 10^{-5}$
Wheat (Trictium spp., Chris)	Kernels	9	$(4.3 \pm 2.7) \times 10^{-4}$	$(3.7 \pm 2.3) \times 10^{-4}$
	Chaff	9	$(9.4 \pm 4.0) \times 10^{-3}$	$(8.7 \pm 3.7) \times 10^{-3}$
Onion (Allium cepa, Mountain Danver)	Bulb	25	$(5.6 \pm 1.2) \times 10^{-4}$	$(5.1 \pm 1.5) \times 10^{-5}$
	Tops	28	$(6.7 \pm 1.1) \times 10^{-4}$	$(4.9 \pm 0.9) \times 10^{-5}$
Peanut (Arachis hypogaea, Spanette)	Nut	12	$(1.6 \pm 0.6) \times 10^{-3}$	$(1.7 \pm 0.6) \times 10^{-4}$
	Shell	12	$(3.7 \pm 0.5) \times 10^{-2}$	$(3.6 \pm 0.5) \times 10^{-3}$
Potato (Solanum tubersum, Norgold)	Whole	14	$(4.9 \pm 1.3) \times 10^{-3}$	$(6.2 \pm 1.6) \times 10^{-4}$
	Pulp	13	$(2.7 \pm 0.9) \times 10^{-3}$	$(3.4 \pm 1.1) \times 10^{-4}$
	Peel	7	$(3.4 \pm 0.5) \times 10^{-2}$	$(5.5 \pm 0.9) \times 10^{-3}$
	Greens	16	$(4.4 \pm 0.6) \times 10^{-3}$	$(3.5 \pm 0.4) \times 10^{-4}$

Table 14, concluded.

Species and Variety	Tissue	No. of Samples	Dry Wt. Transfer Coefficient ± S. E.	Wet Wt. Transfer Coefficient ± S. E.
Radish (Raphanus sativus, Cavalier)	Roots	26	$(6.3 \pm 0.5) \times 10^{-3}$	$(5.8 \pm 0.7) \times 10^{-4}$
	Greens	23	$(5.3 \pm 1.0) \times 10^{-3}$	$(5.8 \pm 1.1) \times 10^{-4}$
Pea (Pisum sativum, Dwarf)	Peas	22	$(1.4 \pm 0.2) \times 10^{-4}$	$(3.5 \pm 0.5) \times 10^{-5}$
	Pods	27	$(3.9 \pm 0.6) \times 10^{-4}$	$(5.5 \pm 0.7) \times 10^{-5}$
	Vines	20	$(2.2 \pm 0.2) \times 10^{-3}$	$(3.3 \pm 0.3) \times 10^{-4}$
Tomato (Lycopersion esculentum, Marglobe)	Pulp	34	$(3.3 \pm 1.3) \times 10^{-5}$	$(3.7 \pm 1.5) \times 10^{-6}$
	Juice	30	--	$(3.3 \pm 0.7) \times 10^{-6}$
	Plant (30 days)	8	$(2.1 \pm 0.4) \times 10^{-3}$	$(1.7 \pm 0.3) \times 10^{-4}$
	Leaves (50 days)	26	$(1.4 \pm 0.2) \times 10^{-3}$	$(1.5 \pm 0.2) \times 10^{-4}$
	Leaves (130 days)	25	$(3.7 \pm 0.7) \times 10^{-4}$	$(5.5 \pm 1.0) \times 10^{-5}$
	Stems (50 days)	21	$(5.0 \pm 0.7) \times 10^{-4}$	$(3.0 \pm 0.4) \times 10^{-5}$
	Stems (130 days)	14	$(1.6 \pm 2.0) \times 10^{-5}$	$(1.1 \pm 2.1) \times 10^{-6}$
Alfalfa (Medicago sativa, Ranger)	Plant (1st crop)	14	$(9.2 \pm 3.3) \times 10^{-4}$	$(1.6 \pm 0.6) \times 10^{-4}$
	Plant (2nd crop)	21	$(2.9 \pm 0.9) \times 10^{-3}$	$(2.9 \pm 0.9) \times 10^{-4}$
	Plant (3rd crop)	15	$(2.9 \pm 0.6) \times 10^{-3}$	$(6.1 \pm 1.2) \times 10^{-4}$
Celery (Apium gravelens dulce, Golden Dwarf)	Edible Stem	21	$(2.7 \pm 0.3) \times 10^{-4}$	$(2.9 \pm 0.4) \times 10^{-5}$
	Stem	23	$(3.4 \pm 0.4) \times 10^{-4}$	$(4.0 \pm 0.5) \times 10^{-5}$
	Leaves	21	$(3.0 \pm 0.6) \times 10^{-4}$	$(3.2 \pm 0.6) \times 10^{-5}$
Spinach (Spinacia oleracea, Badger Savoy)	Leaves	12	$(7.3 \pm 0.5) \times 10^{-3}$	$(8.0 \pm 0.5) \times 10^{-4}$
	Stems	9	$(4.9 \pm 0.9) \times 10^{-3}$	$(5.2 \pm 1.0) \times 10^{-4}$

Table 15. Polonium oxide transfer coefficient (U) – Fayette silt loam, K_d = 303.

Species and Variety	Tissue	No. of Samples	Dry Wt. Transfer Coefficient ± S. E.	Wet Wt. Transfer Coefficient ± S. E.
Barley (Hordeum vulgare, Betzes)	Kernels	9	$(9.9 \pm 4.3) \times 10^{-4}$	$(7.5 \pm 3.3) \times 10^{-4}$
	Stems and Leaves	9	$(2.4 \pm 0.8) \times 10^{-2}$	$(1.1 \pm 0.4) \times 10^{-2}$
Corn (Zea mays, Hybrid Sweet)	Kernels	10	$(9.3 \pm 4.6) \times 10^{-5}$	$(3.6 \pm 1.8) \times 10^{-5}$
	Cob	6	$(1.7 \pm 1.2) \times 10^{-4}$	$(4.9 \pm 3.4) \times 10^{-4}$
	Leaves (50 days)	15	$(1.8 \pm 0.3) \times 10^{-3}$	$(7.0 \pm 1.0) \times 10^{-4}$
	Leaves (130 days)	11	$(1.3 \pm 0.5) \times 10^{-3}$	$(2.1 \pm 0.8) \times 10^{-4}$
	Stems (50 days)	13	$(1.0 \pm 0.2) \times 10^{-3}$	$(3.8 \pm 0.8) \times 10^{-4}$
	Stems (130 days)	11	$(2.7 \pm 0.5) \times 10^{-4}$	$(6.6 \pm 0.1) \times 10^{-5}$
Wheat (Trictium spp., Chris)	Kernels	13	$(3.7 \pm 1.2) \times 10^{-4}$	$(3.1 \pm 1.1) \times 10^{-4}$
	Chaff	13	$(2.8 \pm 1.2) \times 10^{-3}$	$(2.6 \pm 1.1) \times 10^{-3}$
Pea (Pisum sativum, Dwarf)	Peas	8	$(3.3 \pm 1.0) \times 10^{-4}$	$(7.8 \pm 2.3) \times 10^{-5}$
	Pods	8	$(2.7 \pm 1.2) \times 10^{-4}$	$(4.1 \pm 1.7) \times 10^{-5}$
	Vines	10	$(2.2 \pm 0.9) \times 10^{-3}$	$(4.9 \pm 2.0) \times 10^{-4}$
Tomato (Lycopersion esculentum, Marglobe)	Pulp	22	$(2.2 \pm 0.6) \times 10^{-4}$	$(1.8 \pm 0.5) \times 10^{-5}$
	Juice	22	--	$(1.0 \pm 0.2) \times 10^{-5}$
	Plant (7 days)	2	$(2.2 \pm 0.4) \times 10^{-3}$	$(2.5 \pm 0.4) \times 10^{-4}$
	Plant (15 days)	4	$(3.4 \pm 0.6) \times 10^{-3}$	$(3.7 \pm 0.7) \times 10^{-4}$
	Plant (30 days)	6	$(2.8 \pm 0.5) \times 10^{-3}$	$(3.0 \pm 0.6) \times 10^{-4}$
	Leaves (60 days)	14	$(1.9 \pm 0.5) \times 10^{-3}$	$(2.5 \pm 0.7) \times 10^{-4}$
	Leaves (90 days)	10	$(1.4 \pm 0.3) \times 10^{-3}$	$(1.9 \pm 0.4) \times 10^{-4}$
	Stems (60 days)	13	$(2.3 \pm 0.6) \times 10^{-3}$	$(2.5 \pm 0.7) \times 10^{-4}$
	Stems (90 days)	10	$(3.3 \pm 1.4) \times 10^{-4}$	$(3.7 \pm 1.6) \times 10^{-5}$

Table 15, concluded.

Species and Variety	Tissue	No. of Samples	Dry Wt. Transfer Coefficient ± S. E.	Wet Wt. Transfer Coefficient ± S. E.
Alfalfa (Medicago sativa, Ranger)	Plant (1st crop)	10	$(6.1 \pm 1.9) \times 10^{-2}$	$(9.7 \pm 3.0) \times 10^{-3}$
Celery (Apium gravelens dulce, Golden Dwarf)	Edible Stem	10	$(2.2 \pm 0.8) \times 10^{-3}$	$(2.7 \pm 1.0) \times 10^{-4}$
	Stem	9	$(5.7 \pm 1.3) \times 10^{-4}$	$(8.2 \pm 1.8) \times 10^{-5}$
	Leaves	12	$(8.8 \pm 1.0) \times 10^{-4}$	$(1.5 \pm 0.2) \times 10^{-5}$
Spinach (Spinacia oleracea, Badger Savoy)	Leaves	13	$(5.6 \pm 1.2) \times 10^{-3}$	$(6.8 \pm 1.4) \times 10^{-4}$
	Stems	9	$(2.1 \pm 0.3) \times 10^{-3}$	$(2.1 \pm 0.3) \times 10^{-4}$
Onion (Allium cepa, Mountain Danver)	Bulb	16	$(1.1 \pm 0.1) \times 10^{-3}$	$(9.7 \pm 1.2) \times 10^{-5}$
	Tops	15	$(5.7 \pm 1.2) \times 10^{-4}$	$(4.8 \pm 1.0) \times 10^{-5}$
Potato (Solanum tubersum, Kennibeck)	Pulp	9	$(1.1 \pm 0.6) \times 10^{-3}$	$(1.6 \pm 1.0) \times 10^{-4}$
	Peel	8	$(4.9 \pm 0.9) \times 10^{-3}$	$(5.4 \pm 1.0) \times 10^{-4}$
	Greens	9	$(1.1 \pm 0.2) \times 10^{-1}$	$(1.2 \pm 0.2) \times 10^{-2}$
Radish (Raphanus sativus, Cavalier)	Roots	21	$(7.2 \pm 1.0) \times 10^{-3}$	$(5.4 \pm 0.8) \times 10^{-4}$
	Greens	10	$(2.1 \pm 0.2) \times 10^{-2}$	$(2.1 \pm 0.2) \times 10^{-3}$

silt loam containing polonium oxide. Table 16 includes data from plants grown in Leon sandy loam contaminated with $^{210}PoO_2$. Table 17 lists the transfer coefficients for barley, peas and radishes grown in Fort Collins silty clay loam containing RaDEF.

The chemical yields for each plant tissue analysis confirm values reported by Blanchard for radishes, potatoes and leaf vegetables. Table 18 lists the chemical yields determined for each of the crops harvested from the polonium oxide contaminated soils.

Student's t test for equality of means was used to determine the differences between transfer coefficients of the chloride and oxide chemical forms for plants grown in Fort Collins silty clay loam. The following types of vegetation indicated equal uptake of the chloride and oxide forms of polonium at the 99 percent confidence level: barley, barley stems and leaves, onions, onion tops, radishes, radish greens, peas, pea pods, pea vines and leaves, and tomato pulp. Potatoes analyzed as foliage, whole, pulp or peel and spinach indicated greater transfer coefficients for the oxide form. Celery leaves, edible and non-edible celery stem, tomato juice, tomato leaves, and tomato stems had lower polonium-210 uptake from oxide contaminated soil than from soil containing the chloride form. Comparison of the transfer coefficients of barley, peas and radishes from the polonium chloride and oxide contaminated soils with the uptake of polonium-210 from soil containing RaDEF indicated the RaF (^{210}Po) did not enter the

Table 16. Polonium oxide transfer coefficient (U) - Leon sandy loam, K_d = 17.

Species and Variety	Tissue	No. of Samples	Dry Wt. Transfer Coefficient ± S. E.	Wet Wt. Transfer Coefficient ± S. E.
Radish (Raphanus sativus, Cavalier)	Roots	12	$(4.4 \pm 0.8) \times 10^{-3}$	$(3.0 \pm 0.3) \times 10^{-4}$
	Greens	11	$(1.5 \pm 0.1) \times 10^{-3}$	$(1.2 \pm 0.1) \times 10^{-4}$
	Pulp	3	$(1.2 \pm 0.2) \times 10^{-3}$	$(8.0 \pm 1.7) \times 10^{-5}$
	Peel	3	$(8.7 \pm 2.8) \times 10^{-3}$	$(3.3 \pm 1.0) \times 10^{-4}$
Spinach (Spinacia oleracea, Badger Savoy)	Leaves	3	$(2.0 \pm 0.2) \times 10^{-3}$	$(8.3 \pm 1.2) \times 10^{-5}$
	Stems	2	$(8.4 \pm 1.3) \times 10^{-4}$	$(9.5 \pm 1.3) \times 10^{-5}$

Table 17. RaDEF (^{210}Pb, ^{210}Bi and ^{210}Po chlorides) transfer coefficients – Fort Collins silty clay loam.

Species and Variety	Tissue	No. of Samples	Dry Wt. Transfer Coefficient ± S.E.	Wet Wt. Transfer Coefficient ± S.E.
Barley (Hordeum vulgare, Betzes)	Kernels	22	$(6.4 \pm 1.8) \times 10^{-5}$	$(6.0 \pm 1.4) \times 10^{-5}$
	Stems and Leaves	22	$(5.6 \pm 0.4) \times 10^{-4}$	$(2.5 \pm 0.2) \times 10^{-4}$
Pea (Pisum sativum, Dwarf)	Peas	5	$(3.4 \pm 0.1) \times 10^{-5}$	$(6.8 \pm 0.2) \times 10^{-6}$
	Pods	5	$(7.5 \pm 1.8) \times 10^{-5}$	$(5.4 \pm 1.5) \times 10^{-6}$
	Vines	5	$(8.9 \pm 2.7) \times 10^{-4}$	$(5.0 \pm 1.5) \times 10^{-5}$
Radish (Raphanus sativus, Cavalier)	Roots	10	$(5.3 \pm 0.4) \times 10^{-4}$	$(2.6 \pm 0.2) \times 10^{-5}$
	Greens	3	$(7.7 \pm 2.1) \times 10^{-4}$	$(3.8 \pm 1.0) \times 10^{-5}$

Table 18. Chemical yields of $^{210}PoO_2$ added to plant tissues.

Vegetation	Percent Added ^{210}Po Recovered $\pm$ S. E.
Alfalfa	87.9 ± 5.9
Barley	92.3 ± 0.9
Barley stems and leaves	94.2 ± 0.7
Corn	95.7 ± 1.0
Corn stems	95.0 ± 1.6
Corn leaves	94.9 ± 1.2
Celery (edible portion)	93.7 ± 1.8
Celery leaves	92.9 ± 1.8
Onion	95.8 ± 2.9
Onion tops	97.8 ± 3.1
Peas	93.0 ± 0.8
Potato (whole)	96.7 ± 1.5
Potato pulp	92.9 ± 9.7
Potato peel	98.6 ± 0.5
Radish roots	85.9 ± 1.3
Radish greens	87.1 ± 0.4
Tomato juice	100 ± 6.8
Tomato pulp	96.9 ± 2.4
Tomato leaves	93.3 ± 0.5
Tomato stems	91.3 ± 0.7

plant as readily. The difference in transfer coefficients in the latter

case may result from the presence of lead and bismuth or the nitrate

form of the RaDEF.

Comparisons between transfer coefficients of plants grown on

Fort Collins silty clay loam and Fayette silt loam indicated most

plants had equal $^{210}PoO_2$ transfer coefficients for equal soil distribu-

tion coefficients. Student's t test indicated the following transfer

coefficients differed at the 99 percent confidence level. Alfalfa,

edible celery stem, potato foliage, and radish greens grown in Fort

Collins silty clay loam had lower transfer coefficients and potato

peels from the same soil had a higher transfer coefficient than the

same plant tissues grown in Fayette silt loam.

The different transfer coefficients of the potato tissues may be a

variety difference. Kennebec potatoes were substituted for Norgold

because the former was the only available variety at the time of

planting in the Fayette silt loam. Potato pulp transfer coefficients for

both varieties were equal for both soil mixtures. The surface charac-

teristics of the potato tuber varieties may have introduced a difference

that changed the potato peel uptake. However, the potato pulp trans-

fer coefficients would be the values used for dietary estimates of

ingested polonium-210. The increased transfer coefficients for the

alfalfa, edible celery stem and radish greens grown in Fayette silt

loam may result from contamination of the vegetation by the soil

polonium, different transpiration rates between plantings or the chemical differences of the two soils. Contamination of the vegetation by soil would have increased the Standard Error of each determination but the Standard Errors were not unusually large. Increased transpiration also could have carried more polonium into the plant.

If the soil distribution coefficients and the plant transfer coefficients correlated, the transfer coefficients for plants grown in Leon soil would be expected to be greater than those of the Fort Collins and Fayette soils. Comparison of data for radish roots, radish greens, spinach leaves and spinach stems grown in Leon sandy loam with the same vegetation from the previous two plantings indicated the transfer coefficient may be independent of the soil distribution coefficient. Table 19 lists the dry weight transfer coefficients for plants grown in the three different soils. Transfer coefficients of radish roots and tops removed from Leon sandy loam (K_d = 17) were equal to the transfer coefficients for the Fort Collins silty clay loam and equal or less than the transfer coefficients of the same vegetation types from Fayette silt loam. The transfer coefficient for spinach leaves harvested from the Leon soil was less than the transfer coefficients for the two earlier plantings. The transfer coefficients for spinach stems were equal for all three soils. The comparisons suggest plant transfer coefficients are independent of soil distribution coefficients.

Plant transfer coefficients which are independent of the soil distribution coefficients suggest that factors other than soil retention control

Table 19. Dry weight transfer coefficients for vegetation grown in three soils.

Plant and Tissue	$^{210}PoO_2$ Dry Weight Transfer Coefficient		
	Fort Collins (K_d = 230)	Fayette (K_d = 303)	Leon (K_d = 17)
Radish Roots	$(6.3 \pm 0.5) \times 10^{-3}$	$(7.3 \pm 1.0) \times 10^{-3}$	$(4.4 \pm 0.8) \times 10^{-3}$
Radish Greens	$(5.3 \pm 1.0) \times 10^{-3}$	$(2.1 \pm 0.2) \times 10^{-2}$	$(1.5 \pm 0.1) \times 10^{-3}$
Spinach Leaves	$(7.3 \pm 0.5) \times 10^{-3}$	$(5.6 \pm 1.2) \times 10^{-3}$	$(8.4 \pm 1.3) \times 10^{-4}$
Spinach Stems	$(4.9 \pm 0.9) \times 10^{-3}$	$(2.1 \pm 0.3) \times 10^{-3}$	$(2.0 \pm 0.2) \times 10^{-3}$

plant uptake of $^{210}PoO_2$. Transpiration may control the amount of ^{210}Po entering the plant roots. Uptake by plant transpiration suggests a passive uptake rather than active uptake. However, the observed equal transfer coefficients for the same plant species grown in three soil series suggest soil or plant factors which change the $^{210}PoO_2$ in soil solution to a common chemical form at the root surface.

Rovira (1962) reported plant roots excrete a variety of substances which may combine with ions in the region of the plant root. Chemical changes of the polonium species in the region of the plant root may form equal amounts of a ^{210}Po form which is common to a plant species. Assuming the greenhouse conditions equalized the amount of transpiration for the plantings in the three soil series, passive uptake of the chemical form of ^{210}Po common to a plant species may be combined with factors that equalize or lower the transfer coefficients of plants grown in the Leon sandy loam.

The concentration of polonium-210 in plants depended upon the period the plant had been growing. The data for tomato plants grown in Fort Collins silty clay loam show a decrease in transfer coefficients with growth time. At 30 days of growth the plants had a transfer coefficient of 2.1×10^{-3}. At 50 days of growth the leaf transfer coefficient was 5.0×10^{-4}. After 130 days of growth the leaf transfer coefficient was 3.7×10^{-4} and the stem transfer coefficient was 1.6×10^{-5}. Examination of the data for the other plant types indicated that the

plants which matured in 30 to 60 days had transfer coefficients that exceeded 2×10^{-3} and plants maturing in 130 to 140 days had transfer coefficients that were less than 2×10^{-3} independent of the radioactive decay of the polonium-210.

The decrease in transfer coefficient with time may be related to several factors. Polonium-210 may enter the plant at the early stages of growth and be diluted by added growth. As the plants matured, the root supply of ^{210}Po may have been depleted by uptake in early stages of growth and diffusion rates of the ^{210}Po species in the soil may not have maintained the original concentrations of polonium in the region of the roots.

The transfer coefficients for a given type of vegetation showed greater concentrations of ^{210}Po in the stems and leaves than in the fruit. Concentrations in barley kernels, corn kernels, peas, tomatoes and wheat kernels were less than the corresponding concentrations in the stems and leaves of the same plants. Root crops such as potatoes and radishes indicated higher concentrations in the peels than in the pulp. The differences were an order of magnitude or smaller. The reduced transfer coefficient for fruits again suggests the major polonium uptake may be passive.

Use of the transfer coefficients from edible plants grown in Fort Collins soil containing $^{210}PoO_2$ indicated ingestion hazards may be significant to populations obtaining all of their vegetable diet from

a contaminated agricultural area. Calculations by Watters and Hansen (1970) indicated if one megacurie of ^{210}Po were distributed over 62 square miles of alkaline agricultural land, ^{210}Po could produce a one year population dose of 0.5 rem to the kidney from ingestion of vegetable products contaminated by root uptake. Transfer coefficients of plants grown in the Fayette silt loam were generally equal to those of the Fort Collins silty clay loam indicating the ingestion hazard for vegetables grown on Gray-Brown Podzols would be the same. Table 20 lists the ranges of the observed transfer coefficients for the three ^{210}PoO$_2$ plantings. The equations and assumptions used for estimation of the ingestion hazards are in the Appendix.

Control Plant Polonium-210 Content

The ^{210}Po concentrations of the control plants grown in Fort Collins silty clay loam (Table 21) are similar to those reported by Francis et al. in their study of the ^{210}Po content of plants grown under greenhouse conditions. Comparison of the data from the plantings in Fort Collins silty clay loam using Student's t test for equality of means indicated the ^{210}Po content per gram was the same at the 95 percent confidence level for all vegetation types except barley stems and leaves, potato pulp, peas, pea pods, wheat kernels, wheat chaff, and wheat stems and leaves. Use of a 99 percent confidence interval allowed acceptance of equality between the means of all vegetation types except the peas and pea pods and the wheat tissue samples.

Table 20. Range in the observed dry weight transfer coefficients for $^{210}PoO_2$.

Plant Type and Tissue	Range for Fort Collins, Fayette and Leon Soils
Radish roots	4.4×10^{-3} to 7.3×10^{-3}
Radish greens	1.5×10^{-3} to 2.1×10^{-2}
Spinach leaves	8.4×10^{-3} to 7.3×10^{-3}
Spinach stems	2.0×10^{-3} to 4.9×10^{-3}

Plant Type and Tissue	Range for Fort Collins and Fayette Soils
Alfalfa	9.2×10^{-4} to 6.1×10^{-2}
Barley kernels	2.6×10^{-4} to 9.9×10^{-4}
Barley stems and leaves	5.1×10^{-3} to 2.4×10^{-2}
Celery, edible stem	2.7×10^{-4} to 2.2×10^{-3}
Celery leaves	3.0×10^{-4} to 8.8×10^{-4}
Celery stems	3.4×10^{-4} to 5.7×10^{-4}
Corn kernels	1.8×10^{-5} to 9.3×10^{-5}
Corn cob	3.6×10^{-5} to 1.7×10^{-4}
Corn leaves	1.2×10^{-3} to 1.3×10^{-3}
Corn stems	2.7×10^{-4} to 3.9×10^{-4}
Onion bulbs	5.6×10^{-4} to 1.1×10^{-3}
Onion tops	5.7×10^{-4} to 6.7×10^{-4}
Peas	1.4×10^{-4} to 3.3×10^{-4}
Pea pods	2.7×10^{-4} to 3.9×10^{-4}
Pea vines and leaves	2.2×10^{-3}
Potato pulp	1.1×10^{-3} to 2.7×10^{-3}
Potato peel	4.9×10^{-3} to 3.4×10^{-2}
Potato greens	4.4×10^{-3} to 1.1×10^{-1}
Tomato pulp	3.3×10^{-5} to 2.2×10^{-4}
Tomato juice*	3.3×10^{-6} to 1.0×10^{-5}
Tomato stems	1.6×10^{-5} to 3.3×10^{-4}
Tomato leaves	3.7×10^{-4} to 1.4×10^{-3}
Wheat kernels	3.7×10^{-4} to 4.3×10^{-4}
Wheat chaff	2.8×10^{-3} to 9.4×10^{-4}

* Wet weight transfer coefficient.

Table 21. Control plants ^{210}Po content - Fort Collins silty clay loam.

Species and Variety	Tissue	1st Planting ^{210}Po pCi/g dry weight ± S.E.	2nd Planting ^{210}Po pCi/g dry weight ± S.E.
Barley (Hordeum vulgare, Betzes)	Kernels	0.030 ± 0.008	0.013 ± 0.004
	Stems and Leaves	0.050 ± 0.007	0.13 ± 0.03
Corn (Zea mays, Hybrid Sweet)	Kernels		Not Detected
	Cob		Not Detected
	Leaves		0.12 ± 0.02
	Stems		0.025 ± 0.004
Wheat (Trictium spp., Chris)	Kernels	0.043 ± 0.035	0.30 ± 0.06
	Chaff	0.076 ± 0.007	15 ± 1.5
	Stems and Leaves	0.040 ± 0.009	54 ± 15
Onion (Allium cepa, Mountain Danver)	Bulbs	0.13 ± 0.05	0.11 ± 0.09
	Tops	0.13 ± 0.02	0.051 ± 0.014
Potato (Solanum tubersum, Norgold)	Whole	0.027 ± 0.008	0.020 ± 0.004
	Pulp	0.16 ± 0.04	0.017 ± 0.005
	Peel	0.054 ± 0.011	0.040 ± 0.005
	Greens	0.45 ± 0.15	0.14 ± 0.02
Peanut (Arachis hypogaea, Spanette)	Nuts		0.033 ± 0.020
	Shells		0.63 ± 0.10
Radish (Raphanus sativus, Cavalier)	Roots	0.069 ± 0.011	0.097 ± 0.034
	Greens	0.11 ± 0.02	0.082 ± 0.019

Table 21, concluded.

Species and Variety	Tissue	1st Planting ^{210}Po pCi/g dry weight ± S. E.	2nd Planting ^{210}Po pCi/g dry weight ± S. E.
Alfalfa (Medicago sativa, Ranger)	Plant (1st crop) Plant (2nd crop) Plant (3rd crop)		0.17 ± 0.02 0.115 ± 0.02 0.095 ± 0.02
Celery (Apium gravelens dulce, Golden Dwarf)	Edible Stem Stem Leaves	0.055 ± 0.015 0.11 ± 0.04 0.068 ± 0.006	
Spinach (Spinacia oleracea, Badger Savoy)	Leaves Stems	0.086 ± 0.024	0.078 ± 0.020 0.099 ± 0.019
Pea (Pisum sativum, Dwarf)	Peas Pod Vines	0.57 ± 0.21 0.42 ± 0.14 0.29 ± 0.10	0.025 ± 0.015 0.025 ± 0.007 0.16 ± 0.03
Tomato (Lycopersion esculentum, Marglobe)	Pulp Juice Leaves Stems	0.089 ± 0.021 Not Detected 0.038 ± 0.005 0.078 ± 0.009	0.028 ± 0.004 Not Detected 0.16 ± 0.05 0.19 ± 0.05

Comparison of the polonium content of the plants grown in Fayette silt loam (Table 22) with the previous plantings indicated higher levels of ^{210}Po in the control plants. Wheat tissues and alfalfa contained much greater amounts of polonium than detected in the other plants. During the mid-point of the growth of the alfalfa and wheat in Fayette silt loam and the wheat in the Fort Collins silty clay loam a sprayer system was installed above a parallel bench in the greenhouse. The spray system emitted a fine mist that drifted over the plants grown for examination of polonium-210 content. Wheat stems and leaves removed from the Fort Collins silty clay loam contained 54 ± 15 pCi/g, chaff contained 15 ± 1.5 pCi/g, but the wheat kernels only contained 0.30 ± 0.06 pCi/g with the chaff removed. Wheat stems and leaves grown in Fayette silt loam contained 67 ± 27 pCi/g, wheat chaff had 22 ± 1.8 pCi/g and wheat kernels 0.66 ± 0.12 pCi/g. Alfalfa grown during the same period in Fayette silt loam contained 30 ± 15 pCi/g compared to 0.17 ± 0.02 pCi/g for plants grown in Fort Collins soil previous to the installation of the sprayer system. Hill observed that rainfall accounted for the ^{210}Po levels in grass. The sprayer systems acted as scrubbers to wash polonium from the air and deposit it on the plants. The plantings in the Leon sandy loam (Table 23) were moved to a different greenhouse.

The source of the natural levels of ^{210}Po in plants has been attributed to root uptake by Tso et al. Assuming the natural polonium in

Table 22. Control plants ^{210}Po content – Fayette silt loam.

Species and Variety	Tissue	^{210}Po pCi/g dry weight $\pm$ S. E.
Barley (Hordeum vulgare, Betzes)	Kernels	0.033 ± 0.008
	Stems and Leaves	0.48 ± 0.34
Corn (Zea mays, Hybrid Sweet)	Kernels	Not Detected
	Cob	0.21 ± 0.08
	Leaves	0.18 ± 0.05
	Stems	0.20 ± 0.04
Wheat (Trictium spp., Chris)	Kernels	0.66 ± 0.12
	Chaff	22 ± 1.8
	Stems and Leaves	67 ± 27
Onion (Allium cepa, Mountain Danver)	Bulb	0.11 ± 0.07
	Tops	0.39 ± 0.16
Potato (Solanum tubersum, Kennebec)	Pulp	0.033 ± 0.005
	Peel	0.22 ± 0.15
	Greens	0.49 ± 0.02
Radish (Raphanus sativus, Cavalier)	Roots	0.12 ± 0.01
	Greens	0.24 ± 0.08
Pea (Pisum sativum, Dwarf)	Peas	0.14 ± 0.07
	Pod	0.054 ± 0.016
	Vines	0.37 ± 0.08

Table 22, concluded.

Species and Variety	Tissue	^{210}Po pCi/g dry weight $\pm$ S. E.
Tomato (Lycopersion esculentum, Marglobe)	Pulp	0.28 $\pm$ 0.12
	Juice	Not Detected
	Leaves	0.21 $\pm$ 0.07
	Stems	0.091 $\pm$ 0.025
Alfalfa (Medicago sativa, Ranger)	Plant (1st crop)	30 $\pm$ 15
Celery (Apium gravelens dulce, Golden Dwarf)	Edible Stem	0.16 $\pm$ 0.05
	Stems	0.65 $\pm$ 0.47
	Leaves	0.45 $\pm$ 0.17
Spinach (Spinacia oleracea, Badger Savoy)	Leaves	0.30 $\pm$ 0.14
	Stems	1.50 $\pm$ 0.51

Table 23. Control plants ^{210}Po content - Leon sandy loam.

Species and Variety	Tissue	^{210}Po pCi/g dry weight $\pm$ S. E.
Radish (Raphanus sativus, Cavalier)	Roots Greens	0.23 ± 0.04 0.65 ± 0.12
Corn (Zea mays, Hybrid Sweet)	Leaves (30 days) Stems (30 days)	0.53 ± 0.16 0.20 ± 0.01
Tomato (Lycopersion esculentum, Marglobe)	Plants	0.50 ± 0.17

soil acts similarly to PoO_2 in soil, apparent transfer coefficients were calculated from the ratio of the ^{210}Po concentration in the control plants grown in Fort Collins soil to the concentration in the soil (0.48 pCi/g). The values for the "apparent" transfer coefficients are listed in Table 24. Comparison of these coefficients with the values in Table 14 for plants grown in artificially contaminated soil indicated that the calculated apparent uptake in control plants is one to two orders of magnitude higher.

Studies of ^{210}Pb uptake by Tso et al. suggest ^{210}Pb and ^{210}Po together in soil would lead to a transfer coefficient that is higher than the transfer coefficient for unsupported ^{210}Po. However, analysis of plants grown in Fort Collins soil contaminated with an equilibrium mixture of ^{210}Pb, ^{210}Bi and ^{210}Po (RaDEF) did not indicate an increased ^{210}Po transfer coefficient. Barley, peas, and radishes grown in RaDEF soils had transfer coefficients of $(6.3 \pm 1.7) \times 10^{-5}$, $(3.4 \pm 0.1) \times 10^{-5}$ and $(5.3 \pm 0.4) \times 10^{-4}$, respectively. Comparison of the above transfer coefficients with those for the same vegetation type in Table 25 indicated a statistically equal or lower uptake in each case. The observations indicated that the ^{210}Po concentration in the control samples was not because of root uptake and that the major source of natural polonium in plants would be as suggested by Hill, Berger et al. and Francis et al. The radioactive decay of ^{222}Rn in the air leads to contamination of the plant foliage by fallout of ^{210}Pb and decay to ^{210}Po.

Table 24. Polonium-210 uptake by plants based on control plants - Fort Collins silty clay loam.

Vegetation Type	Apparent Transfer Coefficient $\pm$ S. E.
Grains	
Barley	0.047 ± 0.008
Barley Stems and Leaves	0.47 ± 0.06
Corn	0.0
Corn Cob	0.0
Corn Leaves	0.25 ± 0.04
Corn Stems	0.052 ± 0.008
Leafy Vegetables	
Spinach Leaves	0.21 ± 0.04
Spinach Stems	0.16 ± 0.04
Celery (edible stem)	0.11 ± 0.031
Celery Leaves	0.14 ± 0.01
Celery Stems (above first leaf node)	0.23 ± 0.08
Root Crops	
Onions	0.23 ± 0.18
Onion Tops	0.11 ± 0.03
Potato (whole)	0.042 ± 0.008
Potato Pulp	0.032 ± 0.010
Potato Peel	0.15 ± 0.02
Potato Greens	0.29 ± 0.04
Radish Roots	0.20 ± 0.07
Radish Greens	0.17 ± 0.04
Edible Fruit	
Peas	0.052 ± 0.031
Pea Pods	0.052 ± 0.002
Pea Vines and Leaves	0.33 ± 0.06
Tomato Pulp	0.058 ± 0.008
Tomato Juice	0.0
Tomato Leaves	0.40 ± 0.01
Tomato Stems	0.33 ± 0.01

Table 25. Dry weight transfer coefficients for plants grown in Fort Collins silty clay loam containing $^{210}PoO_2$ and RaDEF.

Plant and Tissue	^{210}Po Dry Weight Transfer Coefficients	
	$^{210}PoO_2$ Added	RaDEF Added
Barley kernels	$(2.6 \pm 0.2) \times 10^{-4}$	$(6.4 \pm 1.8) \times 10^{-5}$
Barley stems and leaves	$(5.1 \pm 1.7) \times 10^{-3}$	$(5.6 \pm 0.4) \times 10^{-4}$
Peas	$(1.4 \pm 0.2) \times 10^{-4}$	$(3.4 \pm 0.1) \times 10^{-5}$
Pea pods	$(3.9 \pm 0.6) \times 10^{-4}$	$(7.5 \pm 1.8) \times 10^{-5}$
Pea vines	$(2.2 \pm 0.2) \times 10^{-3}$	$(5.3 \pm 0.4) \times 10^{-4}$
Radish roots	$(6.3 \pm 0.5) \times 10^{-3}$	$(5.3 \pm 0.4) \times 10^{-4}$
Radish greens	$(5.3 \pm 1.0) \times 10^{-3}$	$(7.7 \pm 2.1) \times 10^{-4}$

Paper Chromatography of Plant Extracts

Paper chromatographs of separated plant protein extracts indicated an organic form of polonium-210. Figures 12 and 13 are plots of the fraction of ^{210}Po in a sample moved by the chromatography solvent versus the R_f, the ratio of the distance moved by a spot of activity to the distance moved by the solvent front. Movement of ^{210}Po in 6 N HCl protein digest extracted from wheat leaves and stems is plotted in Figure 12. Movement of polonium oxide digested in 6 N HCl is plotted as a control. Figure 13 contains the plot of the movement of ^{210}Po in 6 N HCl protein digest extracted from spinach leaves and stems. The width of a bar is the uncertainty of the R_f of the movement of ^{210}Po activity.

R_f's of maximum ^{210}Po activity in extracts of wheat leaves and stems were detected between 0.42 and 0.48, 0.54 and 0.60, 0.84 and 0.90. Extracts of spinach leaves and stems indicated maximum ^{210}Po activity between R_f's of 0.18 and 0.24, 0.36 and 0.42. Polonium oxide digested in 6 N HCl moved to R_f s of maximum activity between 0.36 and 0.42, 0.54 and 0.60. The ^{210}Po activity R_f between 0.84 and 0.90 in extract of wheat leaves and stems moved the greatest distance and no corresponding peak of ^{210}Po control activity was present at the same R_f. Locations of other peaks of ^{210}Po activity in the plant extracts were identical or close to peaks of ^{210}Po activity in the control solution. The peak of activity between 0.84 and 0.90 may be an

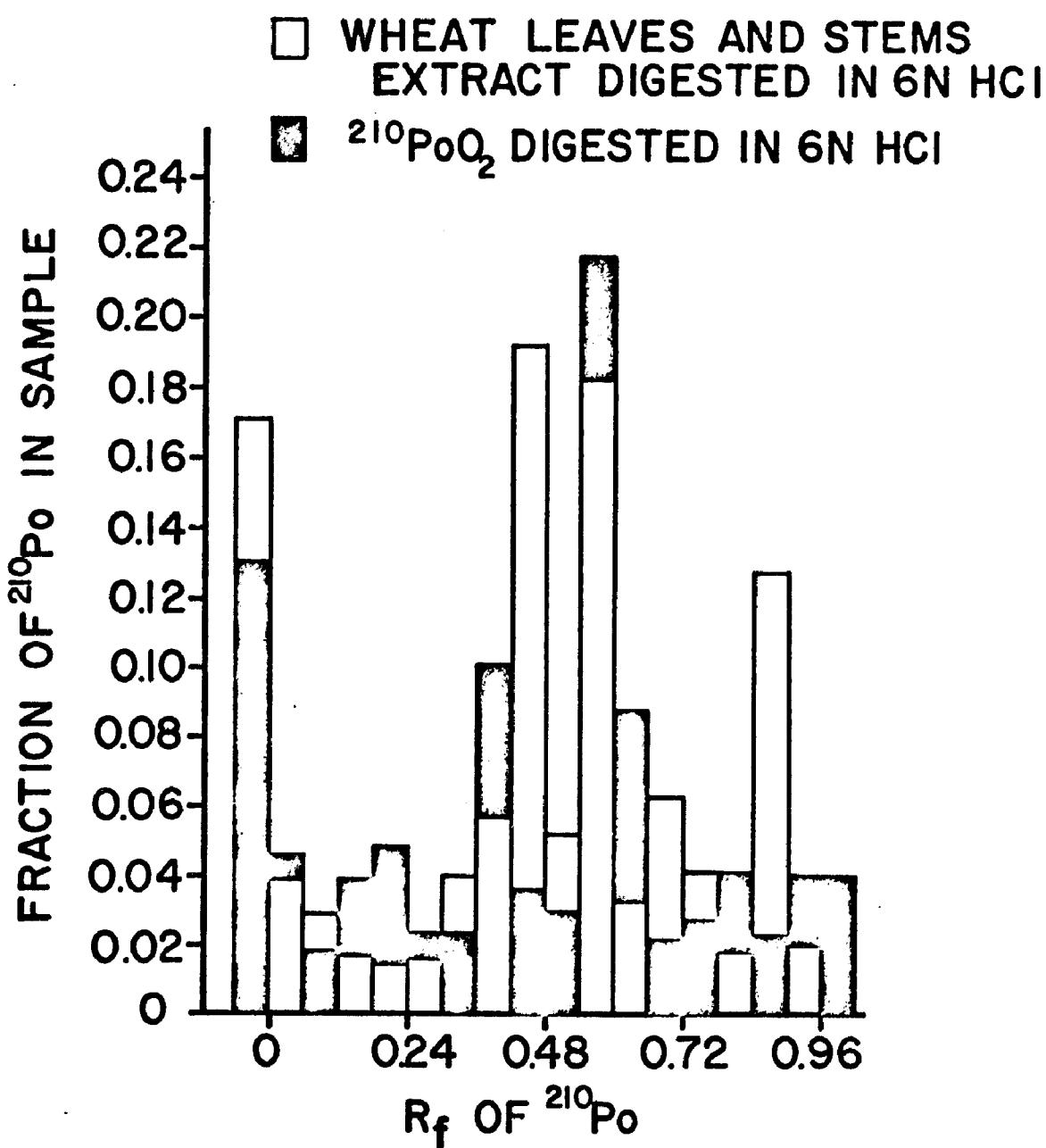Figure 12.   Paper chromatography of wheat leaves and stems protein extract and $^{210}PoO_2$ digested in 6 N HCl.
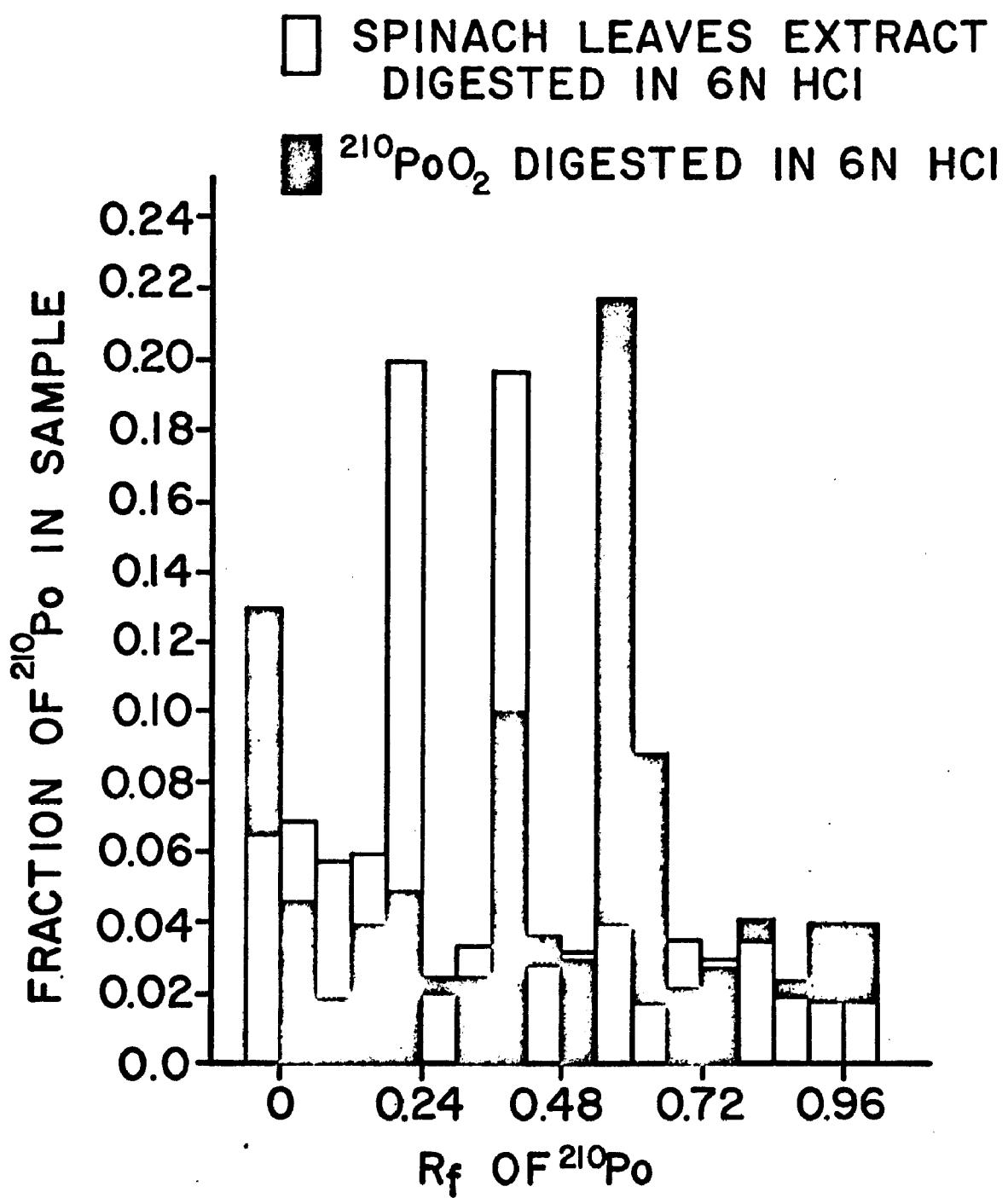
Figure 13.   Paper chromatography of spinach leaves and stems protein extract and $^{210}PoO_2$ digested in 6 N HCl.

organic form of ^{210}Po containing 12 percent of the total activity. Other organically complexed forms of ^{210}Po may exist in plant tissues, but chemical processing to obtain protein hydrolyzates could destroy organically complexed or weakly bound ^{210}Po. The organic form of ^{210}Po observed may be strongly bound or complexed to a molecule which did not undergo changes during chemical digestions and separations.

Ratios of ^{210}Po activities in plant extracts to the ^{210}Po activities in control solutions at the same R_f's indicate the plant materials may alter the relative amounts of ^{210}Po existing as different chemical species. Table 26 lists the ratios calculated from the data plotted in Figures 12 and 13. At the R_f between 0.18 and 0.24 spinach hydrolyzate contained greater amounts of ^{210}Po than the control solutions or wheat extract. Wheat extract contained greater relative amounts of ^{210}Po than spinach extract at R_f's between 0.54 and 0.60 and 0.84 and 0.90. At R_f's between 0.54 and 0.60 the form of polonium in the control solution was present in the greatest relative quantity. Differences between the ratios observed in spinach and wheat extracts indicate forms of ^{210}Po in plants may vary with plant species.

R_f's of organic ^{210}Po in plant extracts may change with soil-plant interactions. The spinach and wheat samples used for protein extractions were grown in the same soil. Electrophoresis experiments indicated the relative concentrations of charged ^{210}Po species in soil solution changed with the soil series. Plants grown in different soil

Table 26. Ratios of ^{210}Po activity in plants extract digest to the ^{210}Po activity in the control digest.

R_f	Ratio of $\dfrac{\text{Wheat }^{210}\text{Po}}{\text{Control }^{210}\text{Po}}$	Ratio of $\dfrac{\text{Spinach }^{210}\text{Po}}{\text{Control }^{210}\text{Po}}$
0.18 - 0.24	0.3	4.2
0.36 - 0.42	0.6	2.0
0.54 - 0.60	0.8	0.2
0.84 - 0.90	5.3	0.8

types may contain other organic forms of ^{210}Po or the relative amount of a given chemical form may increase or decrease.

Polonium-210 detected in plants grown in soils containing added ^{210}PoO$_2$ may enter roots as inorganic ions and undergo chemical change within the plant, or an active uptake mechanism in the root may form an organic species which is translocated to the aerial plant tissue. Transfer coefficients would indicate passive uptake of ^{210}Po is the route of entry into the plant. Rosenfeld and Beath suggested selenium, a possible chemical analog of polonium, entered plants as an inorganic ion. Selenite (SeO$_3^=$) translocated to the leaves of plants was identified as the source of selenium in plants. Figgins speculated that polonite (PoO$_3^=$) ion may exist. If polonite ion exists in soil solution it may act like selenite and enter plants through the root system undergoing chemical changes in the leaves of the plants.

An organic form of ^{210}Po in plants suggests the isotope may be more available for ingestion into soft tissues of men or other animals ingesting the plants. An organic form of ^{210}Po in plants may act similar to organic forms postulated to exist in reindeer and caribou meats. The transfer of ^{210}Po from meat through the gastrointestinal tracts of Lapps and Eskimos has been found to be five times greater than would be predicted from data based on experiments where inorganic forms of ^{210}Po were administered to animals.

Possible organic forms of ^{210}Po in plants suggest further experiments are needed to characterize the organic polonium and its behavior in a food chain. Additional chromatographic experiments with plant extracts and additional solvent separation systems may aid in the study. An experiment which feeds animals with plants containing ^{210}Po may act similarly to postulated organic forms in caribou and reindeer meats. A trophic level study would use a byproduct of the plant to animal study, such as milk, to trace the behavior of polonium in a food chain.

CHAPTER V

SUMMARY OF RESULTS AND CONCLUSIONS

Kilocurie quantities of ^{210}Po have been proposed as a radioisotope

fuel to meet projected satellite power requirements. Accidental

release of 1 MCi of ^{210}Po over an agricultural or watershed area may

introduce the isotope into food and water supplies as an oxide and result

in the exceeding of the maximum safe concentration in vegetables

grown in contaminated soils.

Adsorption of ^{210}PoO$_2$ in soils was studied by bulk determination

of distribution coefficients (K_d) of 18 different top soils and 9 soil

profiles from 6 soil groups. Distribution coefficients were correlated

with the silt content of the top soils. An empirical equation describing

agricultural top soil adsorption was derived as $\ln K_d = 3.2 + 0.046$

(Percent Silt). The distribution coefficients of the combined data for

the A, B and C horizons of all soil samples were correlated with soil

pH and silt content. The empirical equation derived was $\ln K_d =$

$- 1.3 + 0.034$ (Percent Silt) $+ 0.88$ (pH). Polonium oxide eluted

through soil columns with water indicated the K_d predicted soil reten-

tion of ^{210}PoO$_2$. The soil adsorption indicated movement of ^{210}PoO$_2$

to a water supply would be minimal.

The chemical species of ^{210}Po in soil solution were characterized

by electrophoresis and dialysis experiments. Polonium-210 in soil

solution subjected to an electric field indicated positive and negative

species in soil solution. The relative concentrations of the species varied with soil type. Dialyzed soil solutions indicated approximately 34 percent of the ^{210}Po in soil solution would pass through a semi-permeable membrane. The addition of polonium oxide directly to soil solutions indicated soil solid phases alter the dialyzed forms.

Plants grown under greenhouse conditions were examined for accumulation of natural and added ^{210}Po from three different soils. Alcoholic protein extracts of wheat and spinach were separated by paper chromatography to investigate possible organic forms of polonium.

Plant uptake of ^{210}PoO$_2$ added to soil was measured and expressed as a transfer coefficient, U, which is the ratio of the amount of ^{210}Po per gram of dry plant material to the amount of ^{210}Po per gram of dry soil at the date of harvest. Transfer coefficients of 11 different fruit, grain, leafy and root crops ranged from 10^{-2} to 10^{-6}. The transfer coefficients were independent of the soil distribution coefficients. Calculations using U to estimate the ingestion hazards indicated 1 MCi deposited over 62 Mi^2 could produce maximum permissible body burdens of ^{210}Po. Paper chromatography indicated one organically complexed form of polonium-210 which represented 12 percent of the total in the wheat protein hydrolyzate. An organic form of ^{210}Po may increase the ingestion hazard associated with plant uptake of polonium from soil.

Measurements of the naturally occurring ^{210}Po in the soil samples indicated a range of 0.22 to 3.47 pCi/g in top soils. Measurements of

the ^{210}Po in the vegetation used for controls in the greenhouse experi-
ments indicated the source of naturally occurring ^{210}Po in plants was
the fallout of the isotope and its radioactive precursors.

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APPENDIX

^{210}PoO$_2$ Movement in a Mountain Watershed Soil[*]

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Abstract

Uniform contamination of a Rocky Mountain watershed with ^{210}PoO$_2$ from a SNAP generator would find the major contamination retained by the soil. A simulated snow-melt run-off experiment was conducted with a mountain Podzol soil from the Fraser Alpine Area, Colorado. ^{210}PoO$_2$ was applied to the litter layer as a point source one meter from water collection vessels at the base of a 30 percent slope. Cores from the soil on which one mean annual rainfall (18.6 inches) of ice was melted indicated both vertical and horizontal movement of ^{210}PoO$_2$ into the soil. The run-off water collected at the base of the slope at the bottom of the litter layer and A$_2$ horizon indicated only 10^{-7} of the original activity moved through the soil. Data from soil cores were used to predict the movement of a uniform deposition of ^{210}PoO$_2$ to a stream. The equations derived indicate only the first 50 cm bordering a stream contribute significant contamination to the water.

Introduction

The proposed use of ^{210}Po as a fuel in radioisotope electrical generators requires that the hazards associated with an accidental release of the radionuclide into the biosphere must be carefully examined. Dispersal of the fuel material as an oxide into an alpine watershed area presents a potential for contaminating water supplies used for human consumption. The water collected from the large watersheds is primarily melting snow while rain produces only a small part of the annual run-off. Because the soils are porous, most of the snow melt moves into and through the soil and surface run-off occurs only at peak periods of snow melt during two or three hours on warm afternoons.[1] Therefore, the adsorption of PoO_2 on soil and litter was expected to reduce considerably the amount which would ultimately reach the streams draining a watershed.

Experimental Methods

The Darling soil series from the Fraser Alpine Area of Colorado (see Fig. 1) was selected for study of the movement of PoO_2 through the soil under simulated run-off conditions. The Darling series is a coarse textured Podzol occurring at elevations between 9500 feet and timberline.[1] The profile of the Darling gravelly sandy loam under a forest cover of spruce and fir is described in Table 1.[1] Samples of the litter layer, the surface soil and the subsoil were obtained from the watershed during the month of July after the major snow melt.

Table 1

Description of the Darling series soil horizons.

Litter layer -- A Horizon

4 inches to 0, dark-brown layer of organic material made up of needles, twigs, roots and moss.

Surface soil -- A_2 Horizon

0 to 3 inches, light brownish-gray to light gray loam or gravelly sandy loam; very strongly acid, pH of approximately 4.7.
* K_d = 24.

Subsoil -- B_2 and B_{2ir} Horizons

3 to 38 inches, brown to dark brown, gravelly, coarse sandy loam; very strongly to strongly acid; pH approximately 4.8 to 5.2. Average K_d = 21.

Substratum -- C_1 and C_2 Horizons

38 to 80 inches, light yellowish-brown to light olive-brown in deeper part, gravelly coarse sandy loam; an estimated 70 to 80 percent of layer is stone and gravel; medium acid, pH of approximately 5.5 to 5.8.

$$* \quad K_d = \frac{^{210}Po/gm \text{ soil}}{^{210}Po/ml \text{ solution}}$$

The soil sample was removed from a 30 percent slope 200 yards above a water diversion dam on East St. Lewis Creek.

At the laboratory the three layers of soil were packed into a box divided into two sections, each section 100 x 5 x 30 cm deep (see Fig. 2). The soils were packed to approximate their natural depth and density. Ten cm of the subsoil were first added to the bottom of the box to give an average bulk density of 1.3 g/cm^3.[2] This layer was covered with 10 cm of surface soil and packed to an average bulk density of 1.15 g/cm^3. A plastic sheet covered the last 20 cm of length of the subsoil to act as a water collecting surface for the soil above it. Finally a 10 cm layer of litter was placed on top of the soil. Again a plastic sheet covered the last 20 cm of length of the surface soil to act as a collecting area for the litter layer. One section of the box was used for the ^{210}Po experiment and the other section was used as a control to determine any displacement of natural ^{210}Po (RaF). The box was set on a frame at a 30 percent slope. Two ml of ^{210}PoO$_2$ suspension (100 $\pm$ 14 μCi/ml) were pipetted onto the litter 2.5 cm from the high end of the box. The aliquot was centered relative to the walls of the container and covered approximately 1 cm^2.

To simulate snow melting on the slope, crushed ice was added evenly over the surface of the soil at 0.5 kg every three hours for 15 hours. A total 18.6 inches of water representing one mean annual rainfall was melted through the entire surface over a period of 15 days.

Run-off from the three collection levels was collected on a daily basis and assayed for ^{210}Po.

After drainage had ceased, one side panel from the box was removed and 1 cm diameter, 5 cm deep (the width of the box) soil cores were removed for assay. The cores were spaced 5 cm apart in the vertical and horizontal directions for the first 50 cm from the source. Ten cm spacings were cored at distances greater than 50 cm. Although the original polonium suspension was applied on the soil as a centered spot of activity, the lateral movement of the water was expected to distribute the activity across the 5 cm width of the box, as well as along the slope. The core samples were taken across the width of the soil so that the total amount at any distance from the starting point could be determined.

Polonium was recovered from the soil by leaching with 12 N HCl as described by Berger et al.[3] The samples were assayed by spontaneous deposition onto Ni and counted in gas proportional counters. The results were expressed as fractions of the total ^{210}Po activity per 5 cm core.

Results

Since the box was limited in size, some of the results of the experiment were distorted and unusable. The water which was collected at the bottom of the subsoil appeared to be due mostly to the channeling of water down the bottom plastic surface. This led to a higher transport

of ^{210}Po in the drainage water from the subsoil than if unimpeded

vertical drainage had occurred. Therefore, the results for the subsoil

horizon and its drainage water were not considered useful.

The litter layer and surface soil layer appeared to be free of this

perturbation. The fraction of the applied ^{210}Po in the water from the

litter layer was 4.2×10^{-7} in 17 percent of the total water collected.

The fraction of radioactivity in the water collected from the A_2 soil

horizon was 5.9×10^{-7} in 19 percent of the total water collected.

These data indicate that a rather small portion of the source activity

moved parallel to the soil horizons. The water collected from the

contro section indicated only 1 percent of the total activity was from

natural ^{210}Po (RaF).

The soil analyses confirmed that only a small fraction of the ^{210}Po

moved more than a few centimeters along the slope. Figure 3 shows a

^{210}Po activity contour map for the soil after drainage ceased. A

material balance calculated from the contours accounted for the ^{210}Po

added. Major displacement of the original deposition was downward

and downslope. The 10^{-6} contour was distorted because of the imper-

vious bottom of the box. However, in no direction did more than 10^{-5}

of the deposited ^{210}Po activity appear beyond 30 cm.

The decreasing amounts of ^{210}Po activity at distances downslope

from the original deposition were described by empirical equations

for the litter layer and the A_2 surface soil. Two least squares fit

equations were computed from the data for the 5 cm and 15 cm soil depths. These equations allow prediction of the fraction of the applied polonium that would be detected at a distance from the vertical at that depth below the original deposition. They describe the distribution as if it were moving down the slope in a corridor with a cross section of 1 cm^2.

(1) Litter layer at 5 cm from surface

$$\ln F = (-6.2 \pm 1.3) - (0.22 s \pm 0.04) s \qquad r = 0.93$$

where

F = fraction of the original activity at point s

s = distance from the vertical below the surface deposition

(2) A_2 layer at 15 cm from the surface

$$\ln F = (-9.5 \pm 0.2) - (0.22 s \pm 0.01) s \qquad r = 0.99$$

The slopes of the two equations are equal at the 95 percent confidence level using Student's t test for equality. Equality of the slopes indicates that movement parallel to the surface in both soil layers was the same. This is not surprising since the equilibrium distribution of ^{210}Po between the solid phase and solution phase were similar in both layers.

The fraction of activity that moved to point s in the soil at a 5 cm depth can be calculated from the equation:

$$\ln F = -6.2 - 0.22\,s$$

$$F = e^{-6.2}\, e^{-0.22\,s}$$

$$F = 0.0020\, e^{-0.22\,s} \tag{1}$$

However, the quantity 0.0020 is the concentration directly below the point source when $s = 0$. Because the slopes of the equations are equal in both soil horizons a more general case can be stated where V_d is the fraction of the total activity moving to depth d when $s = 0$. The equation is then expressed

$$F = V_d\, e^{-0.22\,s} \tag{2}$$

The total fraction of the activity, F_T^d, at depth d can be obtained by integrating the equation between 0 and s.

$$F_T^d = V_d \int_0^s e^{-0.22\,s}\, ds$$

$$F_T^d = \frac{V_d}{0.22}\,(1 - e^{-0.22\,s}) \tag{3}$$

To obtain values of V_d at different depths, d, in the soil horizons, a least squares fit equation of the vertical movement of the ^{210}Po in the

experiment was computed. The resultant equation $\ln V_d = -4.3 - 0.29\,d$ had a correlation coefficient of 0.91. Simplifying the equation, $V_d = 0.014\,e^{-0.29\,d}$. The total fraction of the activity that moved to depth d and distance s from the point source becomes

$$F_T^d = 0.064\,e^{-0.29\,d}\,(1 - e^{-0.22\,s}).\tag{4}$$

The total activity moved throughout the soil slab to depth d and distance s then becomes

$$F_T = \sum_{1=1}^{d} 0.064\,e^{-0.29\,d}\,(1 - e^{-0.22\,s}).\tag{5}$$

This equation provides a method for estimating the amount of ^{210}Po that would enter a stream which is fed by a snow field. For a uniform distribution of polonium over the surface of a slope above a stream the contribution from each square centimeter to the stream can be calculated.

Equation (1) was evaluated for the value of s where the fraction of activity moving to that point was negligible. This value was calculated to be 3×10^{-8} of the original deposition at 50 cm from the square centimeter source. Equation (1) is based on data from the experiment between 2.5 and 66 cm from the vertical. The total polonium that has moved from the source out into the soil solution in the corridor 20 cm deep and 50 cm long can be obtained by evaluating equation (5). For a

uniform deposition of $^{210}PoO_2$ on the soil, any point greater than 50 cm distance upslope from a stream would not contribute significant contamination to the water. For the 1 cm^2 source 49 cm upslope from the stream, the water reaching the stream would contain the fraction of activity retained by the 50 cm long corridor of soil less the fraction of activity retained by the 49 cm long corridor of soil. The total eluted fraction, E_T, would be the sum of the contributions from each cm^2 between 50 cm and the stream bank.

$$E_T = \sum_{j=1}^{s} \left[F_T(\text{at 50 cm}) - \sum_{i=1}^{d} 0.064 \, e^{-0.29 \, d} \, (1 - e^{-0.22 \, s}) \right] \quad (6)$$

For an initial surface concentration of C_o the activity, A, entering the stream through a 1 cm wide corridor is $A = C_o E_T$. The assumption of a 1 cm corridor without lateral diffusion seems reasonable since diffusion from adjacent areas should compensate each other. Evaluation of the above summation for a 20 cm soil depth produces $A = 1.03 \, C_o$. For the experimental case, if 200 μCi/cm^2 were deposited on the slope and the depth of soil contributing to stream water were 20 cm an estimate of the amount reaching the stream would be 206 μCi/cm times the length of deposition along the stream. This quantity would be small compared to that deposited on surface waters.

Discussion

A uniform contamination of a Rocky Mountain watershed with $^{210}PoO_2$ would find most of the contamination held in the soil. Only that amount of activity deposited close to water diversion systems would add to that deposited on water surfaces. The latter may be more significant for most cases. Use of equation (6) for calculating an estimated contribution from $^{210}PoO_2$ deposited on soil results in a converging summation. The contribution to the total activity from points 20 cm or greater from the stream bank add only 10^{-2} to the 1.03 of the original deposition reaching a stream.

The small fraction of activity observed in the water collected from the litter layer and A_2 horizon may be due to a species of polonium that is only weakly retained by the soil. Column experiments with the A_2 horizon indicate the fraction is 0.3 percent of the total polonium placed on the column. Because of the low retention the pulse of activity moved further into the soil and was eluted in the water. This activity would be in addition to the fraction moved through the soil as predicted by equation (6). However, this small fractional contribution would not be significant in the case of a uniform distribution of $^{210}PoO_2$. Equation (6) was evaluated for contributions of activity from the stream bank to 50 cm upslope. If each square centimeter added a rapidly moving fraction of activity equal to that collected, the total would be

5×10^{-5} added to 1.03 of the original deposition. The nature of this rapidly moving fraction is under investigation.

The $^{210}PoO_2$ moving through the soil to a stream would be reduced by radioactive decay since the half-life is 138.4 days. The retention time in the soil would govern the amount of decay. The peak snow melt period in the Fraser Alpine Area occurs during the months of June and July.[1] Activity deposited between the months of September and the following May would be trapped in snow and ice until the snow melt started water movement in the soil. The movement of the contaminant to a stream on a slope of greater than 30 percent would increase the transport parallel to the horizons while on shallower slopes the movement would be less. However, the terrain near stream banks tend to be fairly gentle slopes. Slopes less than 30 percent would reduce the movement of ^{210}Po along the horizons as vertical movement of water into the soil becomes more pronounced.

Studies of the possible contamination of ground water from radioactive materials have used a distribution coefficient (K_d) for predicting the movement of isotopes in soil.[4] The distribution coefficient is the ratio of the amount of material adsorbed per gram of solid phase (soil) to the amount in solution (soil solution) at equilibrium.[5] In this study the $K_d = {}^{210}Po/g$ soil$/^{210}Po/ml$ solution. The Darling series top soil had a K_d of 24 for $^{210}PoO_2$ indicating a small fraction of the activity remained in solution. Vertical movement through level soil

can be predicted by use of the distribution coefficient and the number of theoretical ion exchange volumes developed from elution of soil columns.[5] The amount of $^{210}PoO_2$ available to one soil exchange volume in the Darling soil is 4 percent of the total activity entering the previous exchange volume. Thus, through the repeated distribution of the diminishing ^{210}Po soil solution concentration in each lower layer of soil the amount which would move to any distance away from the surface deposition is very small.

Summary

A uniform contamination of a Rocky Mountain watershed with $^{210}PoO_2$ would find the major contamination retained by the soil. Snow melt on mountain slopes would carry the major portion of the surface activity into the lower soil horizons. Water percolating from the soil horizons into streams would carry only the activity moved from soil within 50 cm of the stream.

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Figure A-1.   Location of the Fraser Alpine Area, Colorado.
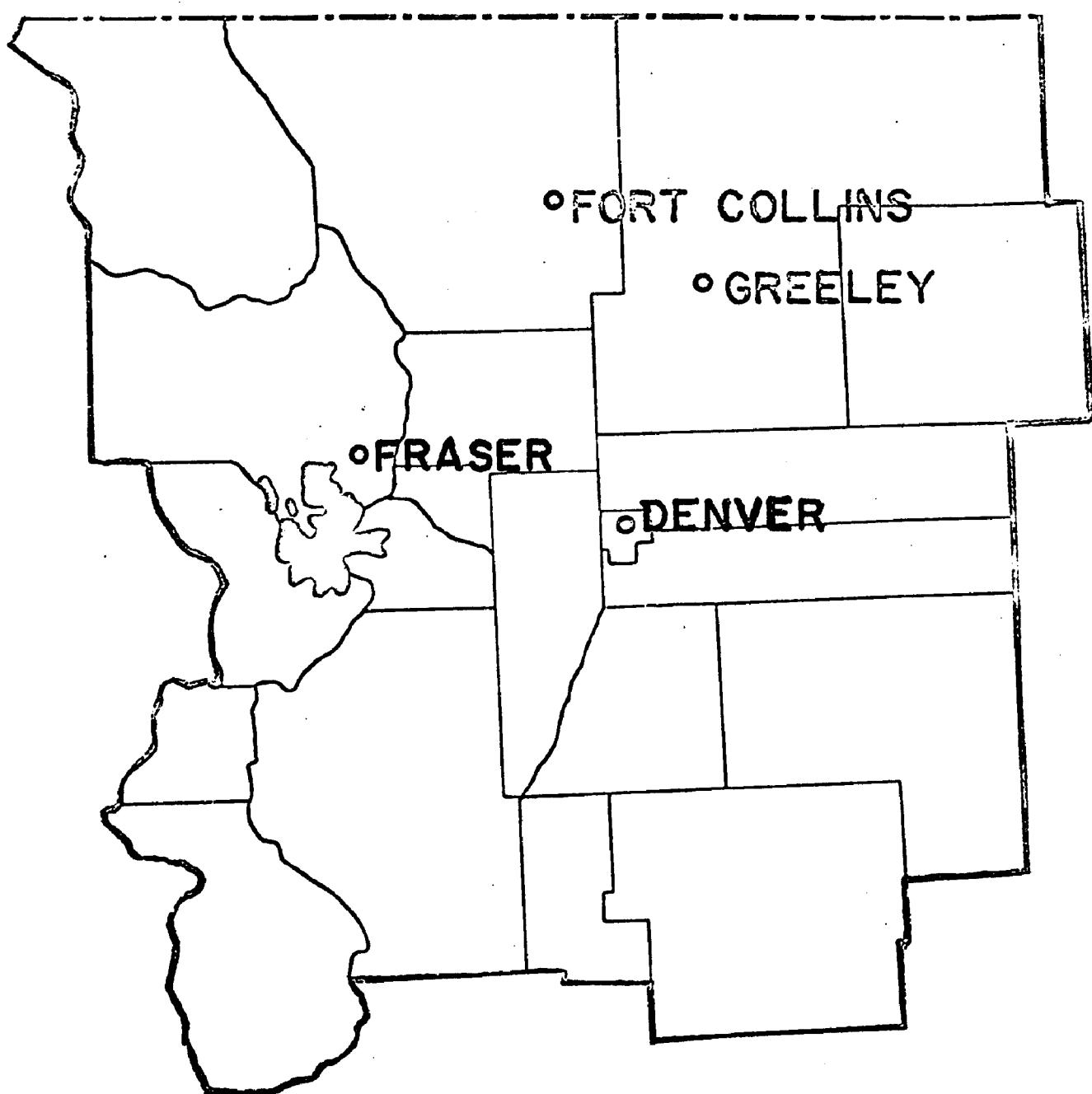
Figure A-2.   Soil in Box, positioned for water collection.
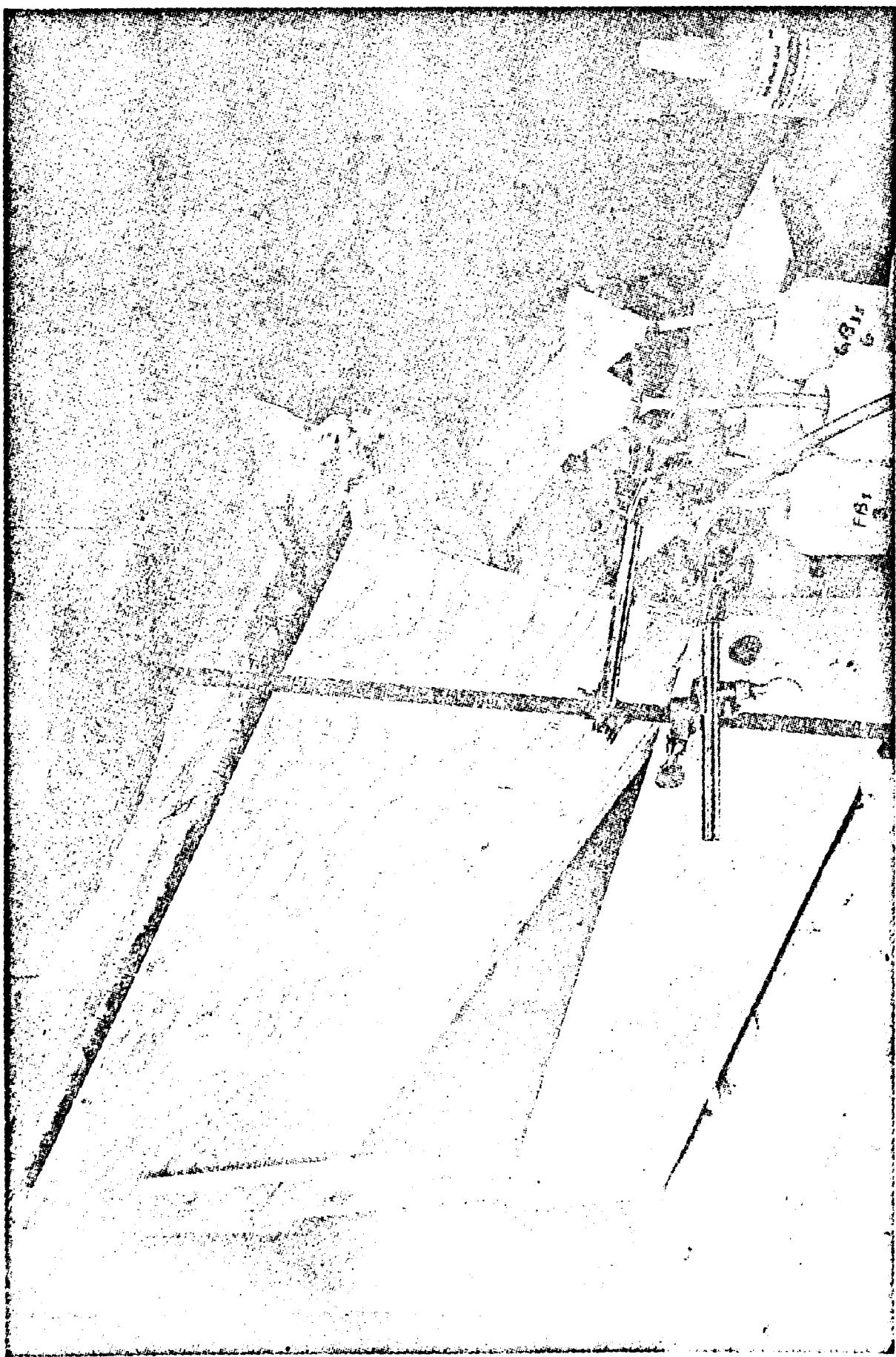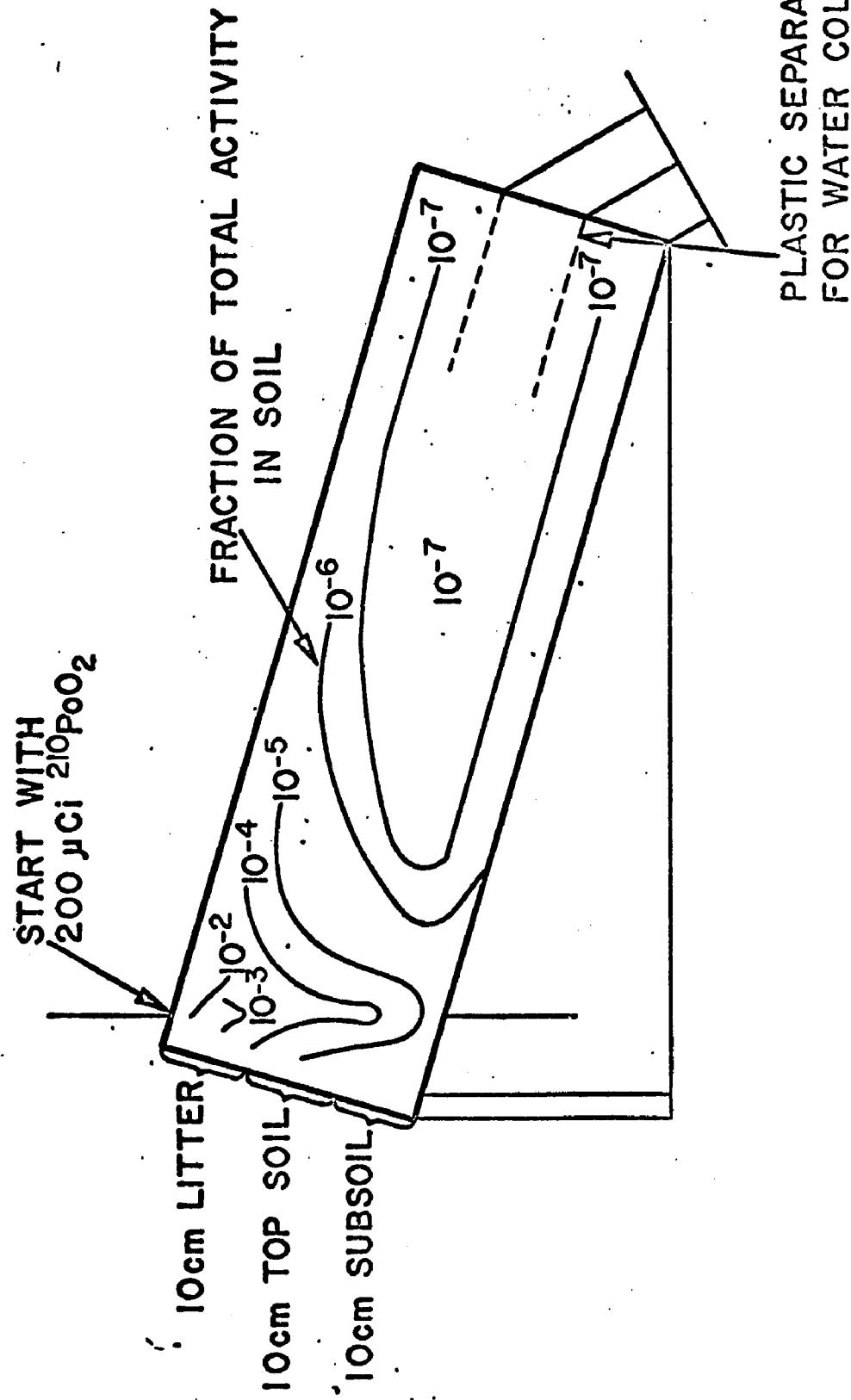
Figure A-3.   Contour map of $^{210}PoO_2$ distribution after one mean annual rainfall as melting ice.

START WITH 200 μCi $^{210}PoO_2$

FRACTION OF TOTAL ACTIVITY IN SOIL

10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-7}
10^{-7}
10^{-7}

10cm LITTER

10cm TOP SOIL

10cm SUBSOIL

PLASTIC SEPARATORS FOR WATER COLLECTION

THE HAZARDS IMPLICATION OF THE TRANSFER

OF UNSUPPORTED ^{210}Po FROM ALKALINE SOIL TO PLANTS*

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Abstract--A study of plants grown in ^{210}PoO$_2$ contaminated alkaline

Brown soil indicated a small but definite uptake through the root

system. The concentration in various tissues varied with species

and growing time. A hazards analysis revealed that a deposition of

^{210}PoO$_2$ in megacurie quantities onto such an agricultural soil could

produce a radiological health problem for more than one year because

of root uptake in edible vegetables.

INTRODUCTION

Polonium-210 has been suggested as a radionuclide fuel for large SNAP generators.[1] Kilocurie to megacurie quantities of the radioisotope would be required for projected satellite power requirements. An accidental release of the fuel from the radioisotope generator could occur if encapsulation of the isotope failed during a launch pad abort, failure to attain orbit, or re-entry from orbit. Although such a failure has a low probability, the hazards that would be presented must be evaluated.

The immediate hazards from the rupture of radioisotope generator fuel capsules with total or partial burnup have been evaluated by several studies.[2] The models used for evaluation of the population exposure include inhalation dose and ingestion of the fallout particles on foodstuffs and ingestion through milk products.[3] The Department of Radiology and Radiation Biology at Colorado State University has been interested in the longer term effects of ^{210}Po contamination on large agricultural areas. Data for transfer of the isotope from soil to plant are needed for this type of hazards analysis.

METHODS

The Nuclear Sources Department, Monsanto Research Corporation, Dayton, Ohio prepared the polonium oxide used in this study by

thermal decomposition of polonium nitrate. The dry oxide was dissolved in water which was previously distilled, deionized and filtered with a 0.45 micron membrane filter. The solution thus formed is referred to as a polonium oxide solution; although it probably represents complex ionic and colloidal forms. This solution was the contaminating agent for the potting soil.

The potting soil consisted of a mixture of Colorado alkaline (pH 7.6) Brown soil (58% by wt.), sand (40% by wt.), and sphagnum peat moss (2% by wt.). This was prepared and contaminated by placing seven kilograms of soil in a small batch cement mixer and adding water to produce a slurry. As the mixer agitated the wet soil, a solution of polonium oxide was added slowly and allowed to mix thoroughly. Sand was added to the soil while mixing continued and, finally, the peat moss was added and thoroughly mixed. Five soil samples for ^{210}Po assay were taken from each batch of soil as it was transferred to its storage container. This potting soil sat for at least two weeks so that distribution of the ^{210}Po between the soil and soil solution would reach equilibrium. Then the soil was steam sterilized to kill any weed seeds and harmful bacteria. The control potting soil had the same composition without the ^{210}Po and underwent the same treatment.

The soil was transferred to seven inch diameter, plastic, azalea pots, which were placed on racks in containment trays (Figure 1). The trays contained polyethylene liners upon which lay one cm of vermiculite to absorb any leached ^{210}Po. The racks produced a separation between pot and tray which prevented root movement to the vermiculite.

A variety of vegetable species were grown from seeds in this soil, with the exception of the wheat, which was grown in polonium chloride contaminated soil (Table 1). The soil surface in the pot was covered with one cm of vermiculite after planting to reduce the possibility of soil contamination on the vegetation surfaces. The temperature of the greenhouse ranged from 20°C to 29°C during the growth period and the plants were simultaneously irrigated and fertilized with a nutrient solution[*].

Plant harvest occurred at maturity except for some time dependency studies. The plants, with the exception of root vegetables, were harvested by cutting them at least two cm above the vermiculite surface. The harvest of the control plants occurred on days other than the harvest of the plants grown in the ^{210}Po soil to reduce the possibility of cross contamination.

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[*] The nutrient solution contained 6 meg/l KNO_3, 3 meg/l $Ca(NO_3)_2$, 2 meg/l $MgSO_4$, 1.5 meg/l NH_4NO_3, 0.5 meg/l H_3PO_4 and 140 g Borax/5000 gal.

All of the harvested plants grown in contaminated soil were washed with detergent and thoroughly rinsed in distilled water to remove any possible soil particles. The plants then were sectioned into leaf, stem, seed and/or fruit samples. They were digested in concentrated nitric acid and 70% perchloric acid until all organic matter was oxidized. The solution was evaporated to fumes of perchloric acid made 0.5 N in hydrochloric acid and analyzed by spontaneous deposition onto nickel by the method of Blanchard.[5] The control samples were analyzed by the same method.

The contaminated soil samples and control soil samples were leached three times for one hour with 12 N HCl at 95°C. The leach liquor was adjusted to 0.5 N HCl and analyzed by spontaneous deposition.

All samples were counted on gas flow proportional counters. Low counting samples were counted to a minimum preset count level to obtain a total standard deviation of the sample count less background not to exceed $\pm$ 10%. The minimum detectable activity at the 95% confidence level for the counting systems was 0.008 pCi of ^{210}Po.

RESULTS

The amount of ^{210}Po transferred from soil to plant tissue at time of harvest was expressed as a transfer coefficient, U:

$$U = \frac{\mu Ci \ ^{210}Po/g \ vegetation}{\mu Ci \ ^{210}Po/g \ dry \ soil \ at \ time \ of \ harvest} \quad (1)$$

This coefficient provides a ratio which may be used to calculate the concentration of ^{210}Po which would exist in plants grown in similar soil at various ^{210}Po contamination levels. This study has shown a constancy in transfer coefficients over a soil concentration range from 0.5 nCi to 5 nCi/g.

The transfer coefficients for a variety of plant tissues are shown in Table 2. In general, the leaves and stems have higher transfer coefficients than do the fruit and seeds for individual plant types. For the root vegetables, the stem and leaf concentrations are comparable to the edible root concentrations. An interesting point to note is that plants which mature in thirty to forty days tend to have higher transfer coefficients for ^{210}Po than plants which mature in 120 to 140 days. For example, the transfer coefficient for the onion, a slow maturing plant, is an order of magnitude lower than the potato or radish and celery is an order of magnitude lower than spinach.

A temporal sampling of tomato and corn plants throughout their growing season showed that the transfer coefficients became smaller with increasing growth time. This evidence lends support to the observation that the longer growing species tend to have lower transfer coefficients at harvest than do more rapidly growing plants. The reason for this tendency has not been established at this time.

DISCUSSION

The transfer coefficients for ^{210}Po in edible vegetation are quite small with the exception of potato peels, which are a minor item in the diet; yet, even with the relatively small uptake which occurs, ingestion hazards from ^{210}Po may be significant to populations obtaining all of their vegetable diet from a highly contaminated agricultural area.

To estimate the ^{210}Po concentration level in soil at time of harvest which could lead to a radiological hazard, these assumptions were used:

1. A population obtained all of its garden vegetable products from the contaminated area.

2. Grain products, except corn, would not be grown in the same geographic area as the truck garden products.

3. The U. S. teenager diet [4] was typical of the population consumption (Table 3).

4. The experimental vegetables were typical of the diet categories.

5. The diet was equally weighted where applicable for short and long maturity vegetables.

6. No other source of intake was present.

7. The critical organ was the kidney[6] and the suggested allowable organ radiation dose was one-thirtieth of the occupational maximum permissible dose or 0.5 rem/year.[7]

The number of microcurie days, Q, which would produce 0.5 rem dose to the kidney is given by[6]:

$$Q = \frac{Dm}{(1.6 \times 10^{-6} \text{ ergs/MeV})(\frac{\text{g-rad}}{100 \text{ ergs}})(3.2 \times 10^9 \text{ dis/day})(\sum EF(RBE)n)}$$

$$= 5.3 \times 10^{-2} \ \mu\text{Ci-days} \tag{2}$$

D = dose to organ in rem = 0.5 rem

m = mass of organ in g = 300 g

$\sum EF(RBE)n$ = effective energy deposited in organ in MeV

= 55 MeV/dis

but

$$Q = \int_0^t q_c (t) \ dt \tag{3}$$

where q_c, the critical organ burden at time t, is given by

$$q_c = \frac{I_i}{\lambda_c - \lambda_r} (e^{-\lambda_r t} - e^{-\lambda_c t}) \tag{4}$$

I_i = intake in μCi on the first day to critical organ

$$\lambda_r = \frac{0.693}{T_{phys}} = \frac{0.693}{138 \text{ days}} = 5.03 \times 10^{-3} \text{ days}^{-1}$$

$$\lambda_c = \frac{0.693}{T_{eff}} = \frac{0.693}{40 \text{ days}} = 1.73 \times 10^{-2} \text{ days}^{-1}$$

and

$$Q = \int_{t=0}^{t=365} \frac{I_i}{0.0123} (e^{-0.00503t} - e^{-0.0173t}) \, dt$$

$$= 8.9 \times 10^3 \ I_i \ \text{days}^2$$

then

$$I_i = \frac{Q}{8.9 \times 10^3 \text{ days}^2} = \frac{5.3 \times 10^{-2} \text{ μCi days}}{8.9 \times 10^3 \text{ days}^2}$$

$$= 6.0 \times 10^{-6} \text{ μCi/day} \tag{5}$$

Now, with the simplifying assumption that all harvests occur at the same time and consumption begins directly after harvest, the intake term is:

$$I_i = \sum_{j=1}^{j=n} f_w \ R_j \ C_j \tag{6}$$

where

f_w = fraction which reaches the kidney from the intestine

= $0.006.^{(6, 7)}$

R_j = the rate that the jth food type is consumed (g/day).

C_j = radioactive concentration in j^{th} food type ($\mu Ci/g$) = ZU_j

Z = soil concentration ($\mu Ci/g$ of dry soil).

U_j = transfer coefficient for j^{th} food type.

Substitution and rearrangement yields the equation:

$$I_i = 0.006 \, Z \sum_{j=1}^{j=n} R_j \, U_j \tag{7}$$

Table 3 shows that the summation $\sum R_j \, U_j = 0.082$.

Solving for Z, the soil concentration, we obtain

$$Z = \frac{I_i}{(0.006) \sum R_j \, U_j} = \frac{6.0 \times 10^{-6} \; \mu Ci/day}{(0.006)\,(0.082 \; g \; soil/day)}$$

$$= 1.2 \times 10^{-2} \; \mu Ci/g$$

The surface concentration to produce the average soil concentration can be estimated with the assumptions of 8 inch (20 cm) cultivation depth and a soil bulk density of 1.3 g/cm^3.[8]

Surface concentration = (20 cm) (1.3 g/cm^3) (1.2 x 10^{-2} μCi/g)

$$= 0.31 \ \mu Ci/cm^2 = 3.1 \times 10^{-3} \ Ci/m^2$$

If one megacurie of ^{210}Po is assumed to deposit with the average surface concentration and if harvest occurs one half life after deposition then the area affected would be

$$Area = \frac{(1.0 \times 10^6 \ Ci) \ (0.5)}{3.1 \times 10^{-3} \ Ci/m^2 \ (2.59 \times 10^6 \ m^2/mi^2)} = 62 \ mi^2$$

Agricultural soil contamination by ^{210}Po could lead to a radiological health problem for more than one year. Studies of the movement of ^{210}Po through alkaline soil columns indicate that it is retained strongly by the soil.[9] The passage of the equivalent of 14 inches of water through these soils has moved the bulk of the polonium no more than one centimeter; therefore, radioactive decay would be the principal natural means of contamination removal. This would reduce the ^{210}Po by a factor of 6 each year.

The transfer coefficients for cereals, i.e. wheat and corn, and the daily average intake of grain products indicates a fractional intake rate which is a factor of 60 less than from the truck garden vegetables.

With the rather low yield of grain products per acre, the contamination of wheat and corn areas probably does not represent a radiological hazard from root uptake.

In summary, a study was made of the transfer coefficients for ^{210}Po from an alkaline soil to edible vegetables representative of the U. S. teenager diet. The results indicated that if one megacurie of ^{210}Po were distributed over 62 sq miles of an alkaline agricultural land, it could produce a one year population dose of 0.5 rem to the kidney from ingestion of vegetable products contaminated by root uptake. Polonium-210 does not migrate from the soil and could represent a hazard for more than one planting season.

This evaluation is for vegetation grown on contaminated soil and does not consider the ingestion of foliar contaminated vegetation nor does it consider the ingestion from dairy products produced in the contaminated area. Either of the above may produce a greater exposure hazard.[3]

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Table 1. List of edible vegetables grown in ^{210}Po contaminated soils.

Vegetable	Diet Type*	Maturity Time (days)
Celery (Golden Dwarf)	Leafy	130
Corn (Northrup King Hybrid)	Smooth Grain	130
Onions (Mountain)	Root	120
Peas (Dwarf)	Smooth	60-90
Potato (Norgold)	Potato	40
Radish (Cavalier)	Root	25-30
Spinach (Badger Savoy)	Leafy	30-40
Tomato (Marglobe)	Fruit	110-140
Wheat[+] (Scout)	Grain	210

* According to teen-ager diet categories in Radiological Health Data (4).

[+] Grown in polonium chloride contaminated soil.

Table 2. Transfer coefficient for edible portions of plant grown in ^{210}Po contaminated soil.

Vegetable	Transfer Coefficient $\dfrac{\mu Ci \ ^{210}Po/g \ veg.}{\mu Ci \ ^{210}Po/g \ dry \ soil}$
Celery	2.9×10^{-5}
Corn	3.5×10^{-6}
Onions	5.1×10^{-5}
Peas	3.5×10^{-5}
Potatoes	6.2×10^{-4}
Radish	5.8×10^{-4}
Spinach	8.0×10^{-4}
Tomatoes	1.2×10^{-6}
Wheat*	9.0×10^{-6}

* Data obtained from $PoCl_4$ contaminated soil.

Table 3. Estimated teen-ager daily vegetable diet[*] and fractional intake rate for ^{210}Po.

Vegetable Type	Consumption Rate (g/day)	Representative Experimental Vegetable	Weighted Transfer Coefficient	Fractional Intake Rate g of Soil Equivalent/ day
Leafy Vegetable	38	Spinach Celery	4.1×10^{-4}	1.6×10^{-2}
Smooth Vegetable including beans	71	Corn Peas	1.9×10^{-5}	1.3×10^{-3}
Root Vegetable	23	Onions Radishes	3.2×10^{-4}	7.4×10^{-3}
Potato	92	Potatoes	6.2×10^{-4}	5.7×10^{-2}
Fruit	166	Tomato	1.2×10^{-6}	2.0×10^{-4}

$$\sum_{j=1}^{j=n} R_j U_j$$

8.2×10^{-2}

[*] According to teen-ager diet categories in Radiological Health Data (4).

Figure A-1.2. A view of the planting pots in the greenhouse
environment.
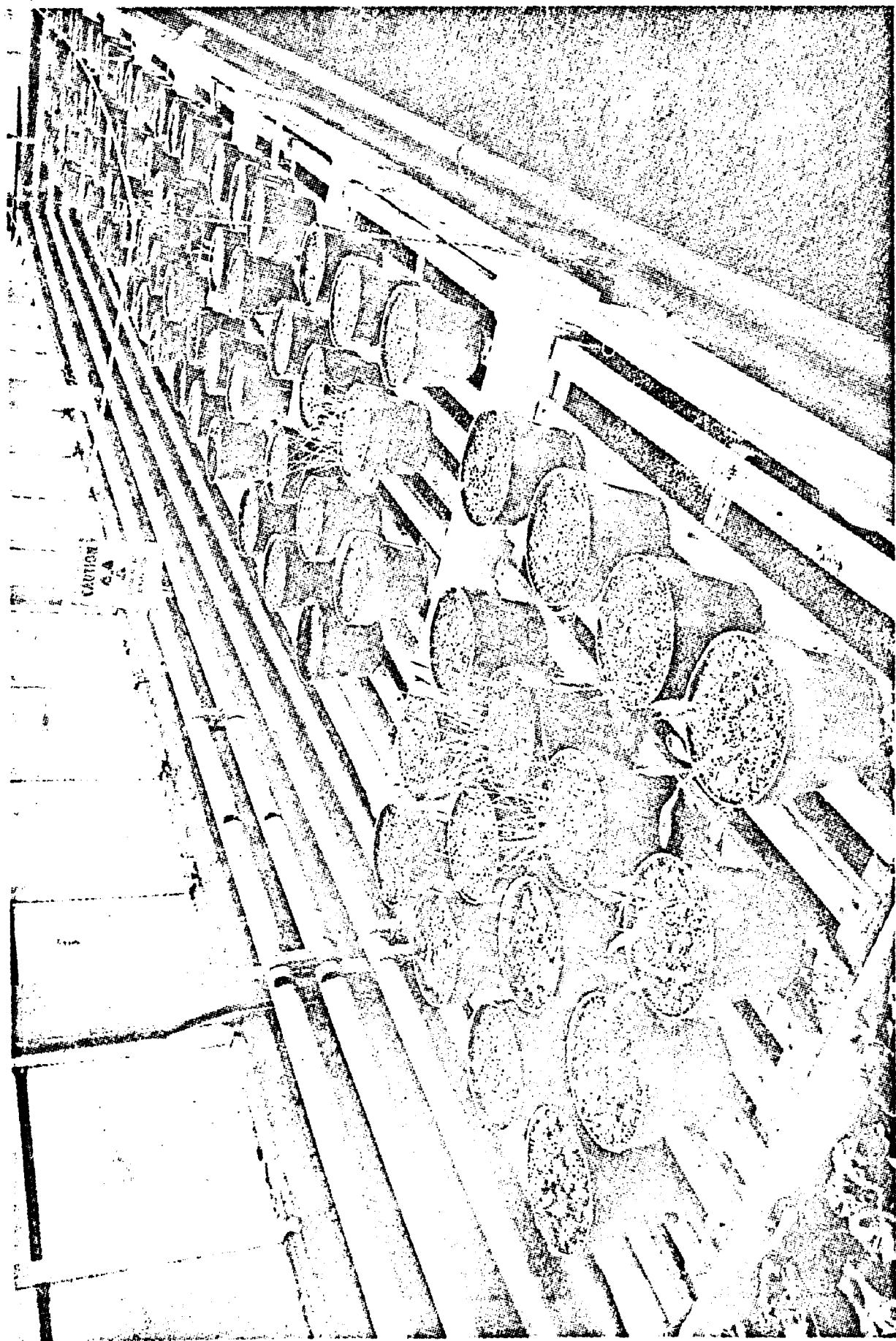
Table A-1. Mechanical analyses pH of soils used for distribution coefficient determinations.

Soil Group and Location	Series and Type	Horizon	pH	Percent Sand	Percent Silt	Percent Clay
Brown, Colorado	Nunn silty clay loam	A_{11}	7.6	42	32	26
Brunizem, Iowa	Dinsdale silty clay loam	A	5.8	11	68	21
		B	5.6	17	55	28
		C	7.9	40	31	39
	Muscatine silty clay loam	A	5.5	3	73	24
		B	5.9	10	61	29
		C	7.8	11	65	24
Gray-Brown Podzol, Wisconsin	Fayette silt loam	A_p	5.9	10	80	10
		B_1	6.2	8	71	21
		B_{21}	6.1	5	66	29
		B_{22}	5.6	2	66	32
		B_{23}	5.3	5	65	30
		B_3	5.3	6	66	28
		C_1	5.5	5	66	29
	Lapeer loam	A_p	6.5	45	42	13
		B_{21}	6.7	54	22	24
		B_{22}	5.5	59	20	21
		B_3	5.7	62	15	23
		C_1	7.8	72	18	10
Ground Water Podzol	Adamsville	A_1	5.9	95	0	5
	Blanton	A_1	5.4	84	6	10

Table A-1., continued

Soil Group and Location	Series and Type	Horizon	pH	Percent Sand	Percent Silt	Percent Clay
	Lakeland	A_1	5.5	95	2	3
	Leon	A_1	5.5	97	1	2
	Ruskin	A_1	5.6	96	2	2
Podzol, Colorado	Darling gravelly sandy loam	A_2	5.1	33	52	15
		B_2	5.5	57	30	13
		B_{2ir}	5.7	74	17	8
		C_1	6.0	80	16	4
		C_2	6.0	74	22	4
Podzol, Wisconsin	Gogebic sandy loam	A_2	6.6	49	39	12
		B_{ir}	5.5	57	30	13
		B_{irh}	6.8	47	29	24
		B_3	5.9	68	27	5
	Onaway fine sandy loam	A_p	6.8	46	44	10
		B_{irh}	6.9	46	43	11
		C_1	8.2	67	20	13
		C_2	8.4	67	20	13
Red-Yellow Podzol, Alabama	Amite	A_1	6.3	82	5	13
	Congaree	A_1	5.5	27	55	18
	Independence	A_1	5.0	91	0	9

Table A-1., concluded

Soil Group and Location	Series and Type	Horizon	pH	Percent Sand	Percent Silt	Percent Clay
	Wickham	A_1	5.6	63	27	10
Lateritic, Puerto Rico		A	4.6	13	47	40

DESCRIPTION OF SOILS

Darling Gravelly Sandy Loam

Location: NW 1/4 of Sec. 8, T2S, R76W; Grand County, Colorado

West St. Louis Creek.

Physiographic Position: Upland.

Topography: Steeply sloping mountain side facing north.

Drainage: Well drained.

Vegetation: Spruce - fir.

Use: Presently in National Forest Service lands.

Collected and Described by: John L. Retzer, September 5, 1952.

Profile Description

A_o-A_{oo} 4 to 0 inches. Grayish brown (10YR 5/2 dry) to very dark

gray (10YR 3/1 moist); a horizon of undecomposed and par-

tially decayed forest litter made up of vaccinium roots,

needles, twigs, and a few scattered leafy plants. The upper

1 inch is predominantly the undecomposed forest litter.

A_2 0 to 4 inches. Light gray (10YR 7/2 dry) to grayish brown or

light brownish gray (10YR 5.5/2 moist) gravelly loam; soft

when dry, very friable when moist; weak to moderate fine

platy structure breaking to moderate fine granules; extremely

acid, approximate pH 4.2; lower boundary clear and smooth.

B_2 4 to 14 inches. Brown (10YR 5/3 dry) to brown to dark

brown (10YR 4/3 moist) gravelly sandy loam; slightly hard

when dry, very friable when moist; weak medium subangular blocky structure breaking to moderate very fine subangular blocks; extremely acid, approximate pH 4.2; lower boundary gradual and smooth.

B_3 14 to 30 inches. Yellowish brown (10YR 5/4 dry) to dark yellowish brown (10YR 4/4 moist) gravelly sandy loam; slightly hard when dry, very friable when moist; massive or weak medium subangular blocky structure; very strongly acid, approximate pH 4.8; lower boundary gradual and smooth.

C_1 30 to 52 inches. Light brownish gray (2.5Y 6/2 dry) to grayish brown (2.5Y 5/2 moist) gravelly sandy loam; hard when dry, friable when moist; massive; strongly acid, approximate pH 5.3; lower boundary gradual and smooth.

C_2 52 to 74 inches. Light yellowish brown (2.5Y 6/4 dry) to light olive brown (2.5Y 5/3 moist) very gravelly sandy loam; loose when dry or moist; single grain; very strongly or strongly acid, approximate pH 5.0; this horizon is partially weathered bedrock.

D_r 74 to 94 inches. Fractured gneiss and schist only slightly altered by weathering.

Nunn Silty Clay Loam

Subgroup: Typic Argiustolls.

Family: Fine, montmorillonitic, mesic.

Diagnostic Horizonation: Mollic epipedon, argillic horizon.

Mean Annual Soil Temperature: 49^OF.

Mean Summer Soil Temperature: 69^OF.

Mean Annual Precipitation: 14 inches, approximately 9 inches falling

during the months of April through August.

Parent Material: Aeolian sediments water-modified in places.

1938 Yearbook Classification: Brown soil.

Profile Description

A_{11} 0 to 3 inches. Grayish-brown (10YR 5/2 dry) silty clay loam,

very dark grayish-brown (10YR 3/2 moist); moderate to strong

medium and coarse granular structure; slightly hard dry, very

friable moist; many grass roots; noncalcareous; pH 7.6; clear

smooth boundary.

A_{12} 3 to 8 inches. Grayish-brown (10YR 5/2 dry) silty clay loam,

very dark grayish-brown (10YR 3/2 moist); very weak medium

prismatic structure breaking to moderate to strong fine sub-

angular blocks; the soil mass is slightly hard dry, very

friable moist, but the individual aggregates are hard dry; non-

calcareous, pH 7.6; clear smooth boundary.

B₁   8 to 13 inches. Brown (10YR 5/3 dry) heavy silty clay loam, dark brown (10YR 3/3 moist); weak medium prismatic structure breaking to moderate to strong fine subangular blocks; the soil mass is slightly hard dry, very friable moist, but the individual aggregates are hard dry; moderate numbers of thin patchy clay films on both horizontal and vertical faces of the soil aggregates; strong worm activity in this horizon, noncalcareous, pH 7.8; clear smooth boundary.

B₂t   13 to 23 inches. Brown (10YR 5/3 dry) silty clay, brown or dark brown (10YR 4/3 moist); moderate to strong angular and subangular blocks; horizon is hard dry, very friable moist, but individual aggregates are very hard to extremely hard dry; thin continuous clay films on the surfaces of the soil aggregates; noncalcareous, pH 7.8; clear wavy boundary.

B₃Ca   23 to 33 inches. Pale brown (10YR 6/3 dry) heavy silty clay loam, brown (10YR 5/3 moist); weak medium prismatic structure breaking to weak to moderate medium subangular blocks; the soil mass is hard dry, very friable moist and individual aggregates are very hard dry; a few thin patchy clay films on the faces of the soil aggregates; this is a weak Ca horizon with some visible secondary calcium carbonate occurring as small concretions and in thin seams and streaks; calcareous, pH 8.0; gradual wavy boundary.

C_{Ca} 33 to 50 inches. Pale brown (10YR 6/3 dry) light silty clay loam, brown (10YR 5/3 moist); massive or very weak coarse subangular blocky structure; hard dry, very friable moist; a weak to moderate Ca horizon with some visible secondary calcium carbonate occurring as concretions and in thin seams and streaks; calcareous, pH 8.0.

Fayette Silt Loam

Classification: Fine silty, mixed, mesic family of Typic Hapludalfs.

Location: NW 1/4 of NW 1/4, Sec. 12, T4N, R4W; Grant County, Wisconsin.

Position and Relief: Sloping upland ridges, bedrock controlled. Four percent convex slope with southwest aspect.

Drainage and Permeability: Well drained; moderately permeable.

Parent Material: Thick loess from Mississippi and Wisconsin River terraces and bottoms.

Vegetation: Old alfalfa

Described by: Carl Glocker

Sampled by: J. Kubota and C. Glocker, September 30, 1968.

Profile Description

A_p 0 to 6 inches. Dark grayish-brown (10YR 4/2) silt loam; moderate fine granular structure; friable; roots plentiful; abrupt smooth boundary.

A$_2$ 6 to 8 inches. Brown (10YR 5/3) silt loam; moderate fine platy structure, friable, roots plentiful; abrupt smooth boundary.

B$_1$ 8 to 13 inches. Dark-brown (10YR 4/3) heavy silt loam; moderate fine subangular blocky structure; friable; roots plentiful; clear wavy boundary.

B$_{21t}$ 13 to 20 inches. Dark-brown (10YR 4/3) silty clay loam; moderate fine subangular blocky structure; firm; roots plentiful; few thin patchy clay films; clear wavy boundary.

B$_{22t}$ 20 to 29 inches. Dark-brown (10YR 4/3) silty clay loam; moderate fine subangular blocky structure; firm; thick silt coats of grayish-brown (10YR 5/2) and thin patchy dark-brown (10YR 3/3) clay films on ped faces; roots plentiful; clear wavy boundary.

B$_{23t}$ 29 to 36 inches. Yellowish-brown (10YR 5/4) silty clay loam; moderate medium subangular blocky structure; firm; thin patchy clay films; few fine distinct mottles of dark brown (7.5 YR 4/4); roots plentiful; clear wavy boundary.

B$_3$ 36 to 46 inches. Yellowish-brown (10YR 5/4) light silty clay loam; weak coarse subangular blocky structure; firm; few roots; few clay films on vertical ped faces; few medium distinct grayish-brown (10YR 5/2) and dark-brown (7.5YR 4/4 mottles; clear wavy boundary.

C_1 46 to 60 inches. Yellowish-brown (10YR 5/4) silt loam; massive; friable.

Remarks: Mottling in lower part of sola is not uncommon in this series, especially where impervious material occurs below the profile.

Gogebic Loam

Classification: Coarse-loamy, mixed, frigid family of Typic Fragiorthods.

Location: NE 1/4 of NE 1/4, Sec. 19, T45N, R3E; 200 feet west of Highway 51 and 500 feet south of farmstead; Iron County, Wisconsin.

Position and Relief: Sloping glacial ground moraine. Four percent convex slope with east aspect.

Drainage and Permeability: Moderate to well drained. Moderately permeable.

Parent Material: Sandy loam acid glacial till.

Vegetation: Two-year red clover stand.

Described by: A. Klingelhoets

Sampled by: J. Kubota and A. Klingelhoets, September 25, 1968.

Profile Description

A_p 0 to 8 inches. Dark-brown (7.5YR 3/2) loam; weak to moderate fine subangular blocky structure; friable; roots plentiful; abrupt smooth boundary.

B_{irh} 8 to 13 inches. Dark reddish-brown (5YR 3/2) light loam; weak medium subangular blocky structure; friable; roots plentiful; clear irregular boundary.

B_{ir} 13 to 23 inches. Dark reddish-brown (5YR 3/3) sandy loam; weak fine subangular blocky structure, friable; roots plentiful; gradual wavy boundary.

B_{irx} 23 to 28 inches. Reddish-brown (5YR 4/3) sandy loam; coarse platy breaking into weak medium fragmental blocky structure; slightly firm and brittle when dry; friable when moist; few plant roots; clear wavy boundary.

B_{3x} 28 to 44 inches. Reddish-brown (5YR 5/4) sandy loam; moderate coarse plates breaking into moderate medium fragmental blocks; firm and brittle when dry, slightly firm when moist; vesicular; few roots; clear wavy boundary.

C_1 44 to 60 inches. Reddish-brown (2.5YR 4/4) sandy loam or fine sandy loam; massive; friable; many feet thick.

Remarks: This soil is quite stony with a high proportion of dark minerals.

Lapeer Loam

Classification: Fine loamy, mixed, mesic family of Typic Hapludalfs.

Location: SW 1/4 of SE 1/4, Sec. 16, T5N, R10E, cut on south side of road; Dane County, Wisconsin.

Position and Relief: Rolling ground moraine. Three percent convex

slope with northwest aspect.

Drainage and Permeability: Well drained; moderately permeable.

Parent Material: Sandy loam, calcareous glacial till of Cary Age.

Vegetation: Oak-Hickory woodlot.

Described by: Carl Glocker, SCS

Sampled by: Carl Glocker, SCS, October 1, 1968.

Profile Description

A_1 0 to 3 inches. Dark-gray (10YR 4/1) loam; moderate fine

granular structure; friable; roots plentiful; medium acid;

abrupt smooth boundary.

A_2 3 to 9 inches. Brown (10YR 5/3) loam; weak medium platy

structure; friable; roots plentiful; medium acid; abrupt smooth

boundary.

B_{21t} 9 to 14 inches. Dark-brown (7.5YR 4/4) sandy clay loam;

weak medium subangular blocky structure; firm; few thin patchy

clay films; roots plentiful; medium acid; clear wavy boundary.

B_{22t} 14 to 20 inches. Dark-brown (7.5YR 4/4) sandy clay loam;

moderate medium subangular blocky structure; firm; thick

patchy clay films; roots plentiful; clear wavy boundary.

B_{23t} 20 to 30 inches. Dark-brown (7.5YR 4/4) sandy clay loam;

moderate medium subangular blocky structure; firm; thin

patchy clay films; strongly acid; roots plentiful; clear wavy boundary.

B$_3$ 30 to 38 inches. Strong-brown (7.5YR 5/6) sandy clay loam; weak medium subangular blocky structure; firm; few roots; medium acid; gradual wavy boundary.

C$_1$ 38 to 60 inches. Yellowish-brown (10YR 5/4) sandy loam; weak medium fragmental blocks; friable; moderately calcareous.

Remarks: Typical profile of Hapludalf soils developed in the calcareous drift of southern Wisconsin.

Onaway Fine Sandy Loam

Location: SE of SW, Sec. 20, T27N, R20E, 120 feet north of road and 12 feet west of line fence.

Vegetation and Use: Mixed hardwood and conifer woodlot.

Slope and Land Form: Undulating relief (3 percent convex). West aspect, undulating glacial ground moraine.

Drainage and Permeability: Well drained with medium to rapid runoff and moderate internal drainage. Moderate permeability.

Parent Material: Highly calcareous gravelly loam to sandy loam glacial till.

Collected by: R. B. Grossman, Leo Juve, George Kwiroga, A. Klingelhoets, R. Fox, E. Link, August 22, 1968.

Described by: A. Klingelhoets, R. Fox, E. Link, August 22, 1968.

Colors are for moist conditions unless otherwise stated.

Profile Description

O   1/2 to 0 inches. Mat of partially decomposed hardwood leaves, twigs, stems with some grasses.

A_1 0 to 2 inches. Very dark gray (10YR 3/1) fine sandy loam; moderate fine crumb structure; friable; many of the sand grains have been stripped of their color coatings; roots abundant; neutral in reaction; abrupt wavy boundary.

A_2 2 to 5 inches. Grayish-brown (10YR 5/2) fine sandy loam; weak medium platy structure; friable; roots plentiful; slightly acid in reaction; clear wavy boundary.

B_{irh} 5 to 12 inches. Dark-brown (7.5YR 4/4) fine sandy loam; weak medium subangular blocky structure; friable; 5 percent of volume made up of stones 3/4 to 3 inches in diameter and 3 percent of larger stones; roots plentiful; slightly acid; gradual wavy boundary.

A'_2 12 to 16 inches. Brown (7.5YR 5/2) sandy loam; moderate coarse platy structure; slightly hard and brittle when dry (weak fragipan); friable when moist; vesicular; 5 percent of volume comprised of stones 3/4 to 3 inches in diameter and 3 percent of larger stones; roots plentiful to few; slightly acid; abrupt wavy boundary.

B_{2t} 16 to 23 inches. Reddish-brown (5YR 4/4) light clay loam; moderate medium subangular blocky structure; firm when moist; thin patchy clay films with dark reddish-brown (5YR 3/3) colors; 5 percent of volume made up of stones 3/4 to 3 inches in diameter and 3 percent of larger stones; roots plentiful; slightly acid; clear wavy boundary.

B_3 23 to 32 inches. Reddish-brown (5YR 4/4) grading to (5YR 5/4) in lower part, loam; weak to moderate medium subangular blocky structure; friable; 5 percent of volume is composed of rocks 3/4 to 3 inches in diameter and 3 percent of larger stones; roots plentiful, mildly alkaline; clear irregular boundary.

C_1 32 to 39 inches. Reddish-brown (5YR 5/4) sandy loam; weak medium fragmental blocky structure (basal till or weak fragipan ?); friable when moist; 5 percent of volume made up of stones 3/4 to 3 inches in diameter and 3 percent of larger stones; few roots; strongly calcareous; abrupt wavy boundary.

IIC_2 39 to 51 inches. Light brown and strong brown (7.5YR 6/4 and 5/6) sand; single grained; loose; weakly stratified; moderately calcareous; abrupt wavy boundary.

$IIIC_3$ 51 to 61 inches. Brown (7.5YR 5/4) light loam; moderate medium fragmental blocky structure; slightly firm and brittle when dry; 5 percent of volume comprised of stones 3/4 to

3 inches in diameter and 3 percent of larger stones; highly calcareous.

Remarks: Soil was dry in the solum and moist in the substratum when sampled. The glacial till has a high proportion of dolomite rocks and rock flows. The $IIIC_3$ horizon becomes very compact and has some of the characteristics associated with basal till or a fragipan. The IIC_2 horizon is an inclusion in this profile. The original vegetation is identical to the present vegetative cover on this pedon. Classification is fine-loamy (coarse loamy?), mixed, frigid family of Alfic Haplorthods.

A sample of the plow layer from a field 240 feet south of the road had the following characteristics.

A_p 0 to 7 inches. Very dark grayish-brown (10YR 3/2, moist) or light brownish-gray (10YR 6/2, dry) fine sandy loam; weak medium very fine subangular blocky structure; friable; roots plentiful; neutral in reaction; abrupt smooth boundary.

Remarks: The face of this pit at which this profile was described and sampled also had some evidence of solifluction. Tongues of A'_2 and B_{2t} occurred in the 2 to 3 foot depth and in some cases were truncated at the upper part.

Dinsdale Silty Clay Loam

Location: NE 1/4, Sec. 20, T88N, R15W, 437 feet east and 639 feet south of NW corner; Grundy County, Iowa.

Vegetation: Alfalfa hay field.

Parent Material: Wisconsin loess over Iowan till.

Physiographic Position: Upland till plain on a convex south-facing

ridge with 3 to 4 percent side slopes.

Slope: 2 percent

Drainage: Well drained.

Permeability: Moderate in loess; moderately slow in till.

Ground Water: None within 70 inches.

Moisture: Slightly moist.

Described by: R. I. Turner, October 14, 1960.

Profile Description

A_{1p} 0 to 6 inches. Black (10YR 2/1)[*] light silty clay loam; cloddy

breaking to weak fine subangular blocky structure and then to

weak fine granules; slightly firm; common fine tubular pores;

gradual boundary.

A_3 6 to 12 inches. Very dark brown (10YR 2/2) light silty clay

loam; moderate very fine subangular blocky and moderate

fine granular structure; friable; a great deal of black (10YR

2/1) mixing as ped coats and worm casts; kneaded color very

dark brown (10YR 2/2); abundant fine tubular inped pores;

gradual boundary.

B_1 12 to 16 inches. Dark brown (10YR 3/3) light silty clay loam;

moderate fine and very fine subangular blocky structure;

friable; few fine black (10YR 2/1) stains on peds; abundant fine tubular inped pores; gradual boundary.

B_{21}' 16 to 21 inches. Dark brown (10YR 4/3) medium silty clay loam; moderate fine subangular blocky structure; friable; kneaded color dark brown (10YR 4/3); abundant fine tubular inped pores; a few faint discontinuous clay films on ped faces, a very few fine soft dark oxide concretions; gradual boundary.

B_{22} 21 to 26 inches. Dark brown (10YR 4/3) medium silty clay loam; weak medium prismatic structure breaking to medium fine subangular blocks; friable; ped interiors are dark brown (10YR 4/3) and ped surfaces 1/4 to 1/2 unit lower in chroma; kneaded color dark brown (10YR 4/3); abundant fine tubular inped pores; a few faint discontinuous clay films on ped faces; very few fine soft dark oxide concretions; gradual boundary.

B_{23} 26 to 30 inches. Dark brown (10YR 4/3) light silty clay loam; weak medium prismatic structure breaking to weak to moderate fine subangular blocks; friable; kneaded color dark brown (10YR 4/3); abundant fine tubular inped pores; a very few very faint clay flows in some root channels; a very few soft dark oxide concretions; fine sand content increases with depth; clear boundary.

IIB_{31} 30 to 37 inches. Brown (10YR 5/3) and yellowish brown (10YR 5/4) heavy loam; weak medium prismatic structure breaking

to weak medium and coarse subangular blocks; slightly firm; brown (10YR 5/3) on ped exteriors and yellowish brown (10YR 5/4) interiors with a few fine faint yellowish brown (10YR 5/6) and a few fine distinct strong brown (7.5YR 5/8) mottles; common fine tubular inped pores; a slight concentration of stones 1 to 3 inches in diameter in the upper 3 inches of this horizon; gradual boundary.

IIB_{32} 37 to 44 inches. Dark brown (10YR 4/3) and yellowish brown (10YR 5/4) heavy loam; weak medium to coarse prismatic structure breaking to very weak coarse subangular blocks; slightly firm; dark brown (10YR 4/3) on exteriors of peds and yellowish brown (10YR 5/4) ped interiors with a few fine distinct strong brown (7.5YR 5/8) mottles; common fine tubular inped pores; gradual boundary.

IIB_{33} 44 to 48 inches. Same as IIB_{32}.

IIC_1 48 to 58 inches. Yellowish brown (10YR 5/6) medium loam; massive with discontinuous vertical cleavage; slightly firm; few fine distinct grayish brown (2.5Y 5/2) and strong brown (7.5YR 5/6) mottles; common medium dark reddish brown (5YR 3/2) on stains on cleavage faces, common fine tubular inped pores; calcareous; common white lime filaments; gradual boundary.

IIC$_2$ 58 to 66 inches. Yellowish brown (10YR 5/6) loam; massive with faint discontinuous vertical cleavage; friable; common fine distinct grayish brown (2.5YR 5/2) and a few fine distinct strong brown (7.5YR 5/8) mottles; common medium distinct very dark gray (10YR 3/1) stains on cleavage faces; gradual boundary.

IIC$_3$ 66 to 70 inches. Same as above.

Notes: A few clear uncoated quartz grains are on ped faces from 0 to 30 inches. Roots plentiful 0 to 12 inches, common from 12 to 21, few from 21 to 48, and practically absent below 48 inches. Horizons A$_1$, B$_{21}$-B$_{22}$, IIB$_{32}$, and IIC$_1$ were sampled for the Bureau of Public Roads.

Muscatine Silty Clay Loam

Location: 130 feet south and 126 feet west of NE corner of NW 1/4 SW 1/4 NE 1/4 Sec. 20, T87N, R17W, Grundy Co., Iowa.

Vegetation: Meadow

Parent Material: Wisconsin loess

Physiographic Position: Nearly level upland on a very slightly convex east-facing slope.

Slope: About 1 percent.

Drainage: Imperfectly drained.

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* Munsell colors for moist soil.

Permeability: Moderate.

Ground Water: None within 70 inches.

Moisture: Moist.

Described by: D. F. Slusher and R. L. Buckner, October 13, 1960.

Profile Description

A_{1p} 0 to 7 inches. Black (10YR 2/1)[*] light silty clay loam; cloddy, breaking to weak fine subangular blocky structure and then to weak fine granules; slightly firm; gradual boundary.

A_{12} 7 to 14 inches. Black (10YR 2/1) light silty clay loam; moderate fine granular structure; friable; gradual boundary.

A_3 14 to 18 inches. Black (10YR 2/1) light silty clay loam; moderate very fine and fine subangular blocky structure; friable; few fine distinct very dark grayish brown (10YR 3/2) and dark grayish brown (10YR 4/2 to 2.5Y 4/2) worm casts and mixings; kneaded color very dark gray (10YR 3/1); gradual boundary.

B_1 18 to 23 inches. Mixed very dark gray (10YR 3/1) and dark grayish brown (10YR 4/2 to 2.5Y 4/2) medium silty clay loam; moderate fine and very fine subangular blocky structure; friable; kneaded color very dark grayish brown (10YR 3/2); common fine tubular pores in peds; gradual boundary.

B_{21} 23 to 30 inches. Dark grayish brown (10YR 4/2 to 2.5Y 4/2) and olive brown (2.5Y 4/4) medium silty clay loam; moderate fine and very fine subangular blocky structure; friable; dark

grayish brown (10YR 4/2 to 2.5Y 4/2) on ped surfaces, olive brown (2.5Y 4/4) inside peds; a few very dark gray (10YR 3/1) worm casts and mixings from rodents; kneaded color olive brown (2.5Y 4/4); common fine tubular pores in peds; a few fine iron-manganese concretions, a few distinct patchy clay films on vertical ped surfaces, gradual boundary.

B_{22} 30 to 35 inches. Dark grayish brown (10YR 4/2 to 2.5Y 4/2) and grayish brown (2.5Y 5/2) to light olive brown (2.5Y 5/4) light to medium silty clay loam; weak to moderate fine sub-angular blocky structure; friable; dark grayish brown (10Y 4/2 to 2.5Y 4/2) on ped surfaces; grayish brown (2.5Y 5/2) to light olive brown (2.5Y 5/4) with a very few fine faint yellow-ish brown (10YR 5/6) mottles inside peds; common fine tubular pores in peds; a few faint patchy clay films; gradual boundary.

B_{31} 35 to 41 inches. Grayish brown (2.5Y 5/2) and dark grayish brown (10YR 4/2) to grayish brown (10YR 5/2) weak medium subangular blocky structure; friable; dark grayish brown (10YR 4/2) to grayish brown (10YR 5/2) with common fine faint dark brown (10YR 4/3) and a few fine faint yellowish brown (10YR 5/6) mottles on ped faces; grayish brown (2.5Y 5/2) with common fine distinct yellowish brown (10YR 5/6) and a few fine distinct strong brown (7.5Y 5/6) mottles inside peds; common fine tubular pores in peds; a very few faint dark gray

(10YR 4/1) clay accumulations as streaks on ped faces; common fine soft iron-manganese concretions; gradual boundary.

B_{32} 41 to 47 inches. Grayish brown (2.5Y 5/2) light silty clay loam; weak medium prismatic structure breaking to weak medium subangular blocks; friable; grayish brown (2.5Y 5/2) with common fine faint brown (10YR 5/3) mottles on ped surfaces; grayish brown (2.5Y 5/2) with common fine distinct yellowish brown (10YR 5/6) and a few fine distinct strong brown (7.5YR 5/6) mottles inside peds; common fine tubular pores in peds; a few fine soft iron-manganese concretions; gradual boundary.

B_{3C1} 47 to 53 inches. Grayish brown (2.5Y 5/2) silt loam; weak coarse prismatic structure; friable; grayish brown (2.5Y 5/2) with common fine distinct yellowish brown (10YR 5/6) and a very few fine and medium distinct strong brown (7.5YR 5/8) mottles inside prisms; grayish brown (2.5Y 5/2) on prisms surfaces; common fine tubular pores in peds; a few fine distinct iron-manganese concretions; weakly calcareous; gradual boundary.

C_2 53 to 60 inches. Light brownish gray (2.5Y 6/2) silt loam; massive; friable; many fine to medium prominent yellowish brown (10YR 5/6) and a few fine distinct strong brown (7.5YR 5/8) mottles; common very fine pores; a few soft

iron-manganese concretions; a few soft very pale brown (10YR 8/3) lime concretions; calcareous; gradual boundary.

C_3 60 to 70 inches. Light brownish gray (2.5Y 6/2) silt loam; massive; friable; many fine to medium prominent yellowish brown (10YR 5/6) and a few medium distinct yellowish red (5YR 4/8) mottles; common very fine tubular pores; a few fine soft iron-manganese concretions; a very few soft very pale brown (10YR 8/3) lime concretions in upper part; calcareous.

Notes: Roots are plentiful from 0 to 14 inches, common from 14 to 23, few from 23 to 41, and very few from 41 to 70 inches. A few clear uncoated quartz grains are on ped surfaces from 0 to 23 inches. The loess thickness is greater than 120 inches and depth to carbonates is 80 inches at a point about 100 feet southwest of sample site. Loess thickness at sample site is considered to be about the same. Horizons A_{1p}, B_{21}, and C_2 were sampled for the Bureau of Public Roads.

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*   Munsell colors for moist soil.

The computer programs listed on the following pages were written for calculation of the natural polonium-210 levels in soils and plants, calculation of a transfer coefficient for plants grown in polonium-210 contaminated soils and calculation of a distribution coefficient for polonium-210 solutions added to soil. The programs are adaptable to use for other radioisotopes by changing the decay constant in the source deck and by changing the subroutine for calculations of elapsed time.

Program SOILBK calculates the natural polonium-210 levels in soil.

Program CONTRO calculates the polonium-210 content of plants on the date of removal from the soil.

Program SPPL calculates the uptake of polonium-210 from soils in dpm/g and a transfer coefficient for wet and dry weights of vegetation.

Program SOILKD calculates the distribution coefficient for soils equilibrated with radioisotopes. The program is not specific for polonium-210.

```
      PROGRAM SOILBK    FORTRAN EXTENDED VERSION 2.0        05/01/70

          PROGRAM SOILBK (INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)
C         PROGRAM SOILBK CALCULATES THE INDIVIDUAL AND AVERAGE SOIL CONTENT
C         OF POLONIUM 210
          DIMENSION SOPO(60)
          WRITE(6,3)
        3 FORMAT(1H1,20X,23HNATURAL PO 210 IN SOILS)
C         READ THE NUMBER OF DIFFERENT SOILS
          READ(5,1)N
        1 FORMAT(I2)
          DO 30 I=1,N
          WRITE(6,2)
        2 FORMAT(1H ,6HSAMPLE,14H PCI/G OF SOIL)
C         READ THE NUMBER OF SAMPLES FOR THE PARTICULAR SOIL HORIZON
          READ(5,1)M
          DO 20 J=1,M
C         READ THE INDIVIDUAL SAMPLE DATA
          READ(5,4)SAM,PM,PD,PY,CM,CD,CY,YMN,COUNT,WT,BKG
        4 FORMAT(A6,6F3.0,2F5.0,2F5.3)
          CALL DAY(PM,PD,PY,TP)
          CALL DAY(CM,CD,CY,TC)
          TDC=TC-TP
          X=-0.00502*TDC
          DEC=EXP(X)
          SMPC=(COUNT/YMN)-BKG
          IF(SMPC.GT.0.01)GO TO 41
          DPMS=0.001
          GO TO 42
       41 DPMS=SMPC/(DEC*0.69*0.51)
       42 DPMGS=DPMS/WT
          SOPO(J)=DPMGS/2.22
C         WRITE THE INDIVIDUAL SAMPLE POLONIUM 210 CONTENT
          WRITE(6,5)SAM,SOPO(J)
```

```
    5 FORMAT(1H ,A6,E11.4)
   20 CONTINUE
C     CALL STAT(M,SOPO,PCIG,SDPC,SEPC)
      WRITE THE AVERAGE SOIL POLONIUM 210 CONTENT IN PCI/G AND S.E.
      WRITE(6,6)SAM,PCIG,SEPC
    6 FORMAT(1H ,/A6,13H SOIL HORIZON,E11.4,14H AVERAGE PCI/G,E11.4,16H
     1S.E. OF AVERAGE/)
   30 CONTINUE
      END



SUBROUTINE STAT      FORTRAN EXTENDED VERSION 2.0      05/01/70

      SUBROUTINE STAT (MX,VAL,AV,SD,SE)
C     SUBROUTINE STAT CALCULATES THE AVERAGE, STANDARD DEVIATION, AND
C     THE STANDARD ERROR OF DATA SUPPLIED FROM THE MAIN PROGRAM IN ARRAY
      DIMENSION VAL(60)
      SUM=0.0
      DO 99 I=1,MX
   99 SUM=SUM+VAL(I)
      ZNUM=MX
      AV=SUM/ZNUM
      YY=0.0
      DO 98 I=1,MX
   98 YY=YY+VAL(I)**2
      SSDEV=YY-(SUM**2)/ZNUM
      VAR=SSDEV/(ZNUM-1.0)
      SD=SQRT(VAR)
      SE=SD/SQRT(ZNUM)
      RETURN
      END
```

SUBROUTINE DAY   FORTRAN EXTENDED VERSION 2.0   05/01/70

```
        SUBROUTINE DAY (XM,DA,YR,TD)
C       VERSION 1 OF SUBROUTINE DAY. CALCULATES THE NUMBER OF DAYS IN
C       THE DATE SENT TO IT FROM THE MAIN PROGRAM
C       XM=MONTH OF YEAR, DA=DAY OF YEAR, YR=THE YEAR IN TENS AND UNITS
        TD=XM*30.+DA+YR*365.
        RETURN
        END
```

05

PROGRAM CONTRO   FORTRAN EXTENDED VERSION 2.0   05/01/70

```
            PROGRAM CONTRO (INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT,PUNCH)
      C     PROGRAM CONTRO COMPUTES THE RADIOISOTOPE CONTENT OF PLANTS BASED
      C     ON THE CONTENT AT DATE OF REMOVAL FROM THE SOIL. ISOTOPE CONTENT
      C     IS COMPUTED AS DPM AND PICOCURIES PER GRAM WET WEIGHT AND DRY
05    C     WEIGHT. PERCENT WATER IS CALCULATED WHERE  THE DATA IS AVAILABLE.
            DIMENSION FRCHOH(60),DRYDPM(60),PCIDRY(60),WETDPM(60),PCIWET(60)
            ISUNUM=0
            WRITE(6,1)
          1 FORMAT(1H1,20X,27HCONTROL PLANT 210PO CONTENT)
10    C     READ THE TOTAL NUMBER OF DIFFERENT PLANT TISSUES
            READ(5,8)N
          8 FORMAT(I2)
            DO 30 J=1,N
            WRITE(6,2)
15        2 FORMAT(1H ,14H SAMPLE NUMBER,1X,14HFRACTION WATER,1H   DPM/G DRY,1
           1H   PCI/G DRY,1H  DPM/G WET,1H  PCI/G WET)
      C     READ THE TOTAL NUMBER OF SAMPLES FOR THE TISSUE
            READ(5,7)MX
          7 FORMAT(I2)
20          DO 20 K=1,MX
            ISUNUM=ISUNUM+MX
      C     READ THE DATA FOR THE TISSUE SAMPLE . THIS IS A SINGLE SAMPLE
            READ(5,3)INUM,SAM,DHM,DHD,DHY,CM,CD,CY,CTIM,COUNT,EFF,YLD,BKG,WETW
           1T,DRYWT
25        3 FORMAT(I3,A3,6F3.0,2F5.0,3F5.3,F6.3,F8.5)
            FRCZ=0.90
      C     IF NO WET WEIGHT IS PRESENT IN THE DATA , A FRACTION WATER IS
      C     SUBSTITUTED AS 90 PERCENT WATER
            IF(WETWT.LT.0.002) GO TO 31
30          GO TO 32
         31 WETWT=DRYWT/(1.0-FRCZ)
         32 FRCHOH(K)=(WETWT-DRYWT)/WETWT
```

```
      CALL DAY(DHM,DHD,DHY,VARDA)
      CALL DAY(CM,CD,CY,CTL)
35    TOTL=CTL-VARDA
      X=-0.00502*TOTL
      DECA=EXP(X)
      SPCT=(COUNT/CTIM)-PKG
C     THIS IF STATMENT CHECKS THE SAMPLE ACTIVITY TO TEST IF IT IS
C     GREATER THAN THE BACKGROUND OF THE COUNTER
40    IF(SPCT.GT. 0.004) GO TO 41
      DPM=0.0001
      GO TO 44
41    DPM=SPCT/(EFF*YLD*DECA)
44    DRYDPM(K)=DPM/DRYWT
45    PCIDRY(K)=DRYDPM(K)/2.22
      WETDPM(K)=DPM/WETWT
      PCIWET(K)=WETDPM(K)/2.22
      WRITE(6,4)INUM,SAM,FRCHOH(K),DRYDPM(K),PCIDRY(K),WETDPM(K),PCIWET(
     1K)
50  4 FORMAT(1H ,2X,I3,A3,I3X,F5.3,4X,4E11.4)
20    CONTINUE
      CALL STAT (MX,FRCHOH,HOHAV,SDHOH,SFHOH)
C     WRITE THE AVERAGE WATER CONTENT OF THE VEGETATION TYPE
55    WRITE(6,5)HOHAV,SDHOH
    5 FORMAT(1H ,20HAVERAGE FRACTION HOH,F5.3,16HS.E.FRACTION HOH,E11.4)
      CALL STAT (MX,DRYDPM,DRDPA,SDDD,SEDD)
      CALL STAT (MX,PCIDRY,PCDA,SDPD,SEPD)
      CALL STAT (MX,WETDPM,WTDPA,SDWD,SEWD)
      CALL STAT (MX,PCIWET,PCWA,SDPW,SEPW)
60    WRITE THE AVERAGE DPM/G WET WT,AVERAGE DPM/G DRY WT, AVERAGE PCI/
C     G, WET WT, AND THE AVERAGE PCI/G DRY WT
C     WRITE(6,6)SAM,WTDPA,SDWD,SEWD,DRDPA,SDDD,SEDD,PCWA,SDPW,SEPW,
     1PCDA,SDPD,SEPD
```

```
65    6 FORMAT(1H ,A3,30H AVERAGE    S.D.       S.E.    /2X,3E11.4,10H DPM/
        1G WET/2X,3E11.4,10H DPM/G DRY/2X,3E11.4,10H PCI/G WET/2X,3E11.4,10
        1H PCI/G DRY/)
        PUNCH 51,SAM,DRDPA,SEDD,HOHAV
     51 FORMAT(A3,E11.4,E11.4,F5.3)
70   30 CONTINUE
        WRITE(6,100)ISUNUM
    100 FORMAT(13HTOTAL SAMPLES,I6)
        END


SUBROUTINE STAT    FORTRAN EXTENDED VERSION 2.0       05/01/70

        SUBROUTINE STAT (MX,VAL,AV,SD,SE)
     C  SUBROUTINE STAT CALCULATES THE AVERAGE, STANDARD DEVIATION, AND
     C  THE STANDARD ERROR OF DATA SUPPLIED FROM THE MAIN PROGRAM IN ARRAY
        DIMENSION VAL(60)
05      SUM=0.0
        DO 99 I=1,MX
     99 SUM=SUM+VAL(I)
        ZNUM=MX
        AV=SUM/ZNUM
10      YY=0.0
        DO 98 I=1,MX
     98 YY=YY+VAL(I)**2
        SSDEV=YY-(SUM**2)/ZNUM
        VAR=SSDEV/(ZNUM-1.0)
15      SD=SQRT(VAR)
        SE=SD/SQRT(ZNUM)
        RETURN
        END
```

```
SUBROUTINE  DAY          FORTRAN EXTENDED VERSION 2.0          05/01/70

         SUBROUTINE DAY (XM,DA,YR,TD)
      C     VERSION 1 OF SUBROUTINE DAY CALCULATES THE NUMBER OF DAYS IN
      C     THE DATE SENT TO IT FROM THE MAIN PROGRAM
      C     XM=MONTH OF YEAR, DA=DAY OF YEAR, YR=THE YEAR IN TENS AND UNITS
         TD=XM*30.+DA+YR*365.
  05     RETURN
         END
```

```
PROGRAM   SPPL    FORTRAN EXTENDED VERSION 2.0          05/01/70

          PROGRAM SPPL    (INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)
      C   THE PROGRAM SPPL CALCULATES THE POLONIUM CONTENT OF  PLANTS
      C   GROWN IN SOILS CONTAINING UNSUPPORTED POLONIUM. THE PERCENT UPTAKE
      C   IS CALCULATED FROM THE CONTENT OF THE SOIL AND THE PLANT CONTENT
   05 C   CORRECTED FOR THE CONTENT OF CONTROL PLANTS GROWN UNDER THE SAME
      C   CONDITIONS.
          DIMENSION FRCHOH(60),DPMUR(60),TCJDR(60),DPMWT(60),TCUWT(60)
          TSUNUM=0
      C   READ THE YEAR OF SOIL ASSAY,PLANT HARVEST AND SAMPLE COUNT
   10     READ(5,5)AY,HY,CY
        5 FORMAT(3F3.0)
      C   READ THE NUMBER OF DIFFERENT PLANT TISSUES
          READ(5,1)N
        1 FORMAT(I2)
   15     DO 20 I=1,N
          WRITE(6,4)
        4 FORMAT(1H ,6HSAMPLE,6X,13H FRACTION HOH,2X,12H DPM/G DRY  ,12HTRAN
         1 CO DRY ,12H DPM/G WET  ,12HTRAN CO WET )
      C   READ THE SAMPLE TYPE, CONTROL PO 210 CONTENT, ERROR IN CONTROL
   20 C   PO 210 CONTENT AND FRACTION OF WATER
          READ(5,8)AMS,CON,CONEP,FRCZ
        8 FORMAT(A3,2E11.4,F5.3)
      C   READ THE NUMBER OF SAMPLES OF A TISSUE TYPE
   25     READ(5,1)MX
          TSUNUM=TSJNUM+MX
          DO 20 K=1,MX
      C   READ THE INDIVIDUAL SAMPLE DATA
          READ(5,10)INUM,SAM,SOPU,AM,AD,HM,HD,CM,CD,CTIM,COUNT,EFF,YLD,BKG,D
         1RYWT
   30  10 FORMAT(I3,A3,F8.0,6F3.0,2F5.0,3F4.3,F8.5)
       31 WETWT=DRYWT/(1.0-FRCZ)
       32 FRCHOH(K)=(WETWT-DRYWT)/WETWT
          CONWT=CON*(1.0-FRCHOH(K))
```

```fortran
         CALL DAY(AM,AD,AY,AT)
         CALL DAY(HM,HD,HY,HT)
         TSD=HT-AT
         CALL DAY(CM,CD,CY,CTL)
         TOTL=CTL-HT
C        CORRECT THE SOIL FOR RADIOACTIVE DECAY TO DATE OF HARVEST
         X=-0.00502*TSD
         SODF=EXP(X)
         TSOC=SODD*SODF
C        CORRECT THE PLANT TISSUE COUNT TO THE DATE OF HARVEST
         Y=-0.00502*TOTL
         PLDF=EXP(Y)
         SMPC=(COUNT/CTIM)*HKG
         IF (SMPC.GT.0.001) GO TO 41
         DPMT=0.0001
         GO TO 44
   41    DPMT=SMPC/(PLDE*FFF*YLD)
   44    DPMDR(K)=DPMT/DRYWT
         TCUDR(K)=(DPMDR(K)-CON)/ISOC
C        THIS IF STATMENT CORRECTS FOR NEGATIVE VALUES
         IF(TCUDR(K).GT.*0.1E-08)GO TO 53
         TCUDR(K)=0.1E-08
         GO TO 53
   53    DPMWT(K)=DPMT/WETWT
         TCUWT(K)=(DPMWT(K)-CONWT)/ISOC
         IF(TCUWT(K).GT.*0.1E-09)GO TO 55
         TCUWT(K)=0.1E-09
         GO TO 55
C        WRITE THE INDIVIDUAL SAMPLE DATA
   55    WRITE(6,9)INUM,SAM,FRCHOH(K),DPMDR(K),TCUDR(K),DPMDR(K),DPMWT(K),TCUWT(K)
    9    FORMAT(1H ,2X,I3,2X,A3,F5.3,9X,4E11.4)
   20    CONTINUE
         CALL STAT (MY,FRCHOH,HOHAV,SFHOH)
C        WRITE THE AVERAGE WATER CONTENT  AS A FRACTION WATER.  THE
C        FRACTION WATER CALCULATED IS BASED ON THE CONTROL PLANT DATA
C        READ INTO THE PROGRAM AND THE DRY WEIGHT OF THE SAMPLE.
```

```
      WRITE(6,7) HOTAV,SEHOH
    7 FORMAT(1H ,20HAVERAGE FRACTION HOT,F5.3,7H   SE  ,E11.4)
      CALL STAT (MX,DPMDR,AVDR,SEDR)
      CALL STAT(MX,TCDDR,AVTD,SETD)
      CALL STAT (MX,DPMWT,AVDW,SEDW)
      CALL STAT (MX,TCHWT,AVTW,SETW)
C     WRITE THE AVERAGE DPM/G AND TRANSFER COEFFICIENTS
C     AVDR=AVERAGE DPM/G DRY WEIGHT,AVTD=AVERAGE DRY WT TRANSFER
C     COEFFICIENT,AVDW=AVERAGE WET WT DPM/G,AVTW=AVERAGE WET WEIGHT
C     TRANSFER COEFFICIENT
      WRITE(6,6)SAM,AVDR,SEDR,AVTD,SETD,AVDW,SEDW,AVTW,SETW
    6 FORMAT(1H ,A2,7HAVERAGE,3X,4HS E./2E11.4,1CH DPM/G DRY/2E11.4,27H
     1DRY WT TRANSFER COEFFICIENT/2E11.4,10H DPM/G WET/2E11.4,28H WET WT
     1TRANSFER COEFFICIENT)
   30 CONTINUE
      WRITE(6,100)TSUNIM
  100 FORMAT(13HTOTAL SAMPLES,I6)
      END


SUBROUTINE   DAY      FORTRAN EXTENDED VERSION 2.0          05/01/70

      SUBROUTINE DAY (XM,DA,YR,TD)
C     VERSION 1 OF SUBROUTINE DAY CALCULATES THE NUMBER OF DAYS IN
C     THE DATE SENT TO IT FROM THE MAIN PROGRAM
C     XM=MONTH OF YEAR, DA=DAY OF YEAR, YR=THE YEAR IN TENS AND UNITS
      TD=XM*30.+DA+YR*365.
      RETURN
      END
```

```
      SUBROUTINE STAT (MX,VAL,AV,SE)
C     SUBROUTINE STAT CALCULATES THE AVERAGE, STANDARD DEVIATION, AND
C     THE STANDARD ERROR OF DATA SUPPLIED FROM THE MAIN PROGRAM IN ARRAY
      DIMENSION VAL(60)
      SUM=0.0
      DO 99 I=1,MX
99    SUM=SUM+VAL(I)
      ZNUM=MX
      AV=SUM/ZNUM
      YY=0.0
      DO 98 I=1,MX
98    YY=YY+VAL(I)**2
      SSDEV=YY-(SUM**2)/ZNUM
      IF(SSDEV.GT.1.0E-12)GO TO 97
      SSDEV=1.0E-12
97    VAR=SSDEV/(ZNUM-1.0)
      SE=SQRT(VAR/ZNUM)
      RETURN
      END
```

```
PROGRAM    SOI-KD    FORTRAN EXTENDED VERSION 2.0        05/01/70

           PROGRAM SOILKD (INPUT,OUTPUT,TAPE5=INPUT,TAPE6=OUTPUT)
    C      PROGRAM SOILKD CALCULATES THE DISTRIBUTION COEFFICIENT FOR
    C      SOIL EQUILIBRATED WITH RADIOISOTOPES.  THE PROGRAM ASSUMES THE
    C      SAMPLES ARE COUNTED WITH THE SAME RADIATION DETECTOR WITH THE
05  C      SAME GEOMETRY FOR BOTH THE SPIKE SAMPLES AND SOIL SOLUTION SAMPLES
           DIMENSION XKD(8)
    C      THE FOLLOWING DATA STATMENT ADDS THE BACKGROUND OF THE RADIATION
    C      DETECTOR.  THE VOLUME OF SPIKE SOLUTION ASSAYED, AND THE VOLUME
    C      OF THE SOIL SOLUTION ASSAYED TO THE PROGRAM
10         DATA BKG,SVOL,VOLSP/0.05,0.97,0.97/
           WRITE(6,7)
    7      FORMAT(1H1,49X,27HPOLONIUM 210 STUDY IN SOILS)
           WRITE(6,6)
    6      FORMAT(1H ,44X,37HEQUILIBRIUM DISTRIBUTION COEFFICIENTS)
15         WRITE(6,8)
    8      FORMAT(1H ,6HSAMPLE,15X,2HKD)
    C      READ THE NUMBER OF DIFFERENT SOILS IN THE DATA DECK
           READ(5,111)IK
11)        FORMAT(I2)
20         DO 30 J=1,IK
    C      READ THE DATA FOR THE SPIKE PLATES INTO THE PROGRAM
           READ(5,1)SPI,SPK,SPE,SPP,SPS,ZN
    1      FORMAT(5(F7.1),F2.0)
           SPIKE=(SPI+SPK+SPE+SPP+SPS)/ZN
25         VAR=((SPI-SPIKE)**2+(SPK-SPIKE)**2+(SPE-SPIKE)**2+(SPP-SPIKE)**2+(
           1SPS-SPIKE)**2)/(ZN-1.0)
           SDSP=SQRT(VAR)
           SERSP=SDSP/SQRT(ZN)
    C      WRITE THE AVERAGE OF THE SPIKE PLATE ACTIVITY AND THE S.D. AND S.E.
30         WRITE(6,5)SPIKE,SDSP,SERSP
    5      FORMAT(1H ,13HSPIKE AVERAGE,F4.2,8H    S.D.,F7.2,8H    S.E. ,F8.2)
           XNUM=0.0
    C      READ THE NUMBER OF INDIVIDUAL SAMPLES FOR THE SOIL TYPE
```

```
      READ(5,9)M,XNUM
    9 FORMAT(I2,F2.0)
      DO 20 K=1,M
C     READ THE INDIVIDUAL SAMPLE DATA
      READ(5,2)SAMPL,AMASS,CTIM,COUNT
    2 FORMAT(A7,F7.5,F3.0,F5.0)
      CPMS=COUNT/CTIM
      XML=((CPMS-BKG)*SVOL)/((SPIKE-BKG)*VOLSP)
      ZMS=1.0-XML
      XKD(K)=(2.45*4.96)/(XML*AMASS)
C     WRITE THE INDIVIDUAL SAMPLE KD
      WRITE(6,3)SAMPL,XKD(K)
    3 FORMAT(1H ,A7,9X,E10.3)
   20 CONTINUE
      SUM=0.0
C     THE FOLLOWING CALCULATES THE S.D. AND S.E. AND THE AVERAGE KD
      DO 21 K=1,M
   21 SUM=SUM+XKD(K)
      AVKD=SUM/XNUM
      XSUM=0.0
      DO 31 K=1,M
      DIF=XKD(K)-AVKD
      VAL=DIF**2
   31 XSUM=XSUM+VAL
      XVAD=XSUM/(XNUM-1.0)
      SDKD=SQRT(XVAD)
      SERKD=SDKD/SQRT(XNUM)
C     WRITE THE AVERAGE KD AND THE STANDARD DEVIATION (S.D.) AND
C     STANDARD ERROR (S.E.)
      WRITE(6,4)AVKD,SDKD,SERKD
    4 FORMAT(1H ,10HAV RAGE KD,E10.3,7H  S.D. ,E10.3,7H  S.E. ,E10.3)
   30 CONTINUE
      END
```

explanation. First, it would appear to be at odds with a least one other feature of the results. The majority of wars which historians allege were waged for reasons of economic expansion during the period since 1825, as well as virtually all the colonial and imperial (extra-systemic) wars, were fought during the nineteenth century. If the economic imperialism model were the correct one, we might therefore expect that the relationship between differential industrial growth and war would be stronger in the earlier than in the later part of the temporal domain. In fact, we find precisely the reverse: for the 1825-1919 period, the strength of the relationship is <u>below</u> that for the 140-year period as a whole; for the 1850-1964 period, the observed association is <u>above</u> that for the entire period.

Second, just as in the case of the other two models, it is not at all clear whether the presence or absence of a relationship between differential industrial mobility and war constitutes sufficient evidence either to confirm or deny the hypothesis. It might be equally plausible, for example, that economic expansion would not cause much conflict when only a limited number of nations were engaged in rapid industrialization and differential growth was high, since under these conditions there would be a relatively large "pie" to divide amongst a few (Rosecrance, 1963, pp. 232-267).

If, however, many nations began to engage in rapid indus-

trialization, (and, as a consequence, industrial mobility

was comparatively low), conflict might rapidly develop as

the competition for markets, resources, and capital outlets

became more intense and the pool of available resources

scarcer. Wars of economic imperialism may therefore just

as plausibly be associated with high mean growth, even if

there are only small differences between national growth

rates.

Thus, once again it would seem that any definitive

statements regarding the applicability of the model will

require further tests, in this case those relating particular

configurations or "profiles" of industrial growth with the

amount of war begun in the system.

Summary

In short, there would seem to be a number of plausible

models which could account for the observed direct connection

between war and mobility on the industrial capability

dimension. All have some difficulties; this may indicate

that none of the explanations set forth above are adequate,

or, perhaps, that there exists more than one causal sequence

linking differential industrial mobility and war. At all

events, none can be offered as more than tentative at this

stage, requiring much additional evidence for conclusive demonstration. A common feature of this evidence seemed to be the need to move beyond simple summary indices of the distribution of capability change within the system as a whole to those which measure more complex configurations of system structure.

Having examined some possible explanations for the observed relationship between industrial mobility and war, let us now turn to examine the apparent causal connection between status inconsistency and the amount of war begun in the international system.

Status Inconsistency and War: Alternative Models

On the face of it, the task of accounting for the strong and apparently non-spurious relationship between status inconsistency and war would be much easier than explaining the rather unexpected _direct_ relationship between differential status mobility and war discussed above. The line of reasoning which has led many to suspect a connection between status inconsistency was carefully outlined in the first two chapters, and, given that the very positive findings would appear to confirm the hypothesis, one might suppose that the only remaining tasks are to draw the appropriate conclusions for future research and discuss the

implications of the findings for the conduct of policy.

Such, however, is not the case, for two reasons. First,

as noted in earlier discussion and as will be amply detailed

below, the original model is by no means the _only_ plausible

causal sequence which could account for the observed rela-

tionship between status inconsistency and war. Second, there

are, as we shall see, some features of the results which might

lead to an alternative interpretation of the findings.

What follows, then, is a re-examination of the earlier model

in light of the evidence, followed by some plausible alter-

natives suggested by the findings, and concluding with a

weighing of their relative probabilities and suggestions for

further research which might provide more definitive answers.

The Frustration-Agression Model

Let us begin the discussion with a brief recapitula-

tion of the model outlined in Chapter 2. There it was argued

that a nation (or, more accurately, national decision-makers

and their attentive publics) will adjust status expectations

to capability. If the status attributed to that nation

by the other national actors in the system is lower than its

self-perceived capability ranking, decision-makers and publics

will believe this to be unjust and inequitable. In short,

it is hypothesized that status inconsistency creates

dissatisfaction because it violates the crucial system norm that nations receive rewards in proportion to their position on the capability pecking order. The frustration thus engendered, coupled with the leverage possessed by such a nation in having relatively high capability status, create both the motive and opportunity for increasingly aggressive international behavior, resulting in war if the discrepancy remains uncorrected.

To a very large extent, this model is compatible with the observed findings. To begin with, it accounts for the strong relationship observed between status inconsistency and war when no time lag was employed. If status inconsistency did not operate directly to stimulate hostile or aggressive attitudes on the part of national elites and their internal clientele, we would not expect to observe so strong a bivariate relationship except when a long time-lag was introduced.

Second, this model would explain why the apparent effect of status inconsistency on the amount of war begun in the system was greater when we examined the later portion of the temporal domain. If status inconsistency results in a sense of frustration amongst those within a nation who perceive it, one might expect such discrepancies to exert a greater influence on national behavior during the period in which the circle of those attentive to foreign policy

matters expanded to include mass publics,and foreign policy
decision-makers were no longer able to insulate themselves
from the demands of such publics. Although there are,of
course,enormous variations from nation to nation, for most
nations in the system the attentiveness and influence of
mass publics increased sharply during the late nineteenth
and early twentieth centuries.

Finally, as noted above, this model would explain to a
large extent why the particular _indices_ of status inconsist-
ency which predict best to war vary with the time period
examined. If national elites and attentive publics were
sensitive to discrepancies between perceived national capa-
bility and attributed status, then it might be expected that
the indices of status inconsistency which predicted best to
war would be those based on capability measures most closely
corresponding to current beliefs about national power. Such
appears to have been the case; with the exception of the
iron and steel production index discussed above, we see a
gradual shift in the nature of the indices which predict
best to war, those which measure mere size (total population
and number of military personnel) gradually giving way to
those which are sensitive to a nation's sophistication and
level of development (urban population and military expendi-
tures). This shift could be interpreted as reflecting the

growing awareness (engendered in part by contacts with large, non-European societies such as Turkey and China) that capability was based on far more than sheer size.

However, if this model fits well with most of the observed results, it does not accord so well with one crucial feature of the findings. Specifically, it does not explain why an even stronger relationship between status inconsistency and war was discovered when a 10 to 15 year time lag was employed. To be sure, by itself such a long time lag would not necessarily cast doubt on the validity of the model. It could be argued that many national decision-makers, at least in the pre-World War II era, were very slow to perceive and react to status changes within the system, being as always prisoners of inherited conventional wisdom; examples might be the surprise with which Japan's victory over Russia in 1905 was greeted and the equal astonishment which accompanied the sudden fall of France in 1940. Perceptions of national status inconsistency may likewise become apparent only very gradually, accounting for the long observed lag between the existence of status inconsistency and its culmination in the onset of war.

Unfortunately, if we choose to explain the existence of a strong lag effect in this way, we immediately run into two difficulties. First, it is difficult to see how we can

have it both ways; if we assume that status perceptions change very slowly, we can no longer explain the existence of a relationship when no time lag is employed. Second, we can no longer account so well for the observed differences in the particular indices of status inconsistency which predict well to war in the different time periods; it is scarcely probable that national decision-makers will be very slow to react to important changes in the international pecking order while at the same time very quick to respond to changes in the national attributes which go to make up this rank-ordering.

On balance, then, while the "frustration-aggression" model may partly account for the findings, it does not appear to be a wholly satisfactory explanation. Let us, then, see to what extent other possible models could better account for the observed results.

The Status Confusion Model

One possible alternative line of reasoning would stress the possibilities for confusion and judgmental error which might attend status discrepancies in the international system. This argument would run as follows. A common feature of several popular models of the international system is the emphasis they place on a decision-maker's

ability to estimate accurately the capabilities and status

position of other nations vis-a-vis one's own. In the balance

of power model, such finely tuned perceptions--particularly

with regard to military capability--are vital if statesmen

are to react quickly enough to counteract potential threats to

the balance (Gulick, 1955, pp. 24-29). In the power transi-

tion model, misperceptions about capability status on the

part of the "challenger" can lead him to initiate hostilities

in the mistaken belief that he can overturn the established

order (Organski, 1968, pp. 371-373).

If such confusion and misperception about status re-

present an important source of danger to peace and stability

in the system, it is easy to see how status inconsistency may

generate such misperceptions. Even the most sagacious

statesman will be influenced to some extent in his evalua-

tion of a nation's capability by the judgments of his counter-

parts in other nations. If the collective judgment of the

comity of nations concerning the importance of a given

national actor--what we have termed the attributed status

of a nation--is seriously at variance with its capabilities,

errors in evaluating these capabilities would undoubtedly

be greater in magnitude and more frequent in occurrence.

Recent examples of such distorted evaluations due to status

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Recent examples of such distorted evaluations due to status
inconsistency might be the serious underrating of Soviet

capabilities prior to the launch of Sputnik, and the similar

misestimate of Chinese capabilities prior to the explosion

of that nation's first atomic device.

To some extent, this model fits quite well with the

observed results. First, it accounts well for the existence

of a relationship when no time lag is employed, since pre-

sumably any confusion generated by status inconsistency

will tend to dissipate with time as decision-makers receive

feed-back concerning their evaluations. Second, it accounts

even better than the previous model for the observed var-

iations in the particular indices of status inconsistency

which predict best to war in different time periods; ac-

cording to this model, what is important are the discrepan-

cies between attributed status on the one hand and real, not

perceived, capability on the other. It might therefore be

expected that the results would be quite sensitive to over-

time changes in the attributes which comprise a nation's power

base. Finally, this model is congruent with results in a

still more attractive sense: since it is compatible with

both the balance of power and power transition models which

were advanced as possible explanations of the observed <u>direct</u>

relationship between differential status mobility and war,

it leaves open the intriguing possibility that one of these

or perhaps some similar model might be capable of explaining

both of the major patterns in our findings.

There are, however, two important drawbacks to this "confusion" model linking status inconsistency and war. First, for precisely the reason it offers a good explanation for the existence of a relationship in the no-lag case, it would appear unable to account for the relationship observed with a very long lag; after 10 or 15 years, any confusion in evaluating national capability generated by status inconsistency should have lessened, not grown stronger. Second, and more important, it is basically at odds with still another finding. If status inconsistency led to conflict by creating confusion regarding national capabilities, we would expect that discrepancies between the basic dimensions of national capability themselves (demographic, military, and industrial) would prove to exercise a strong influence on war, since (even more than discrepancies between the various capability indices and attributed status), such inconsistencies would make difficult the evaluation of national capability. This, however, did not prove to be the case; in pretests run prior to undertaking this study, the relationship between these sorts of discrepancies and war was found to be virtually nil.

In short, then, the status confusion model does not seem to do any better, and in some respects fares rather

worse than the original. While later evidence may demonstrate the contrary, on the basis of the present findings it can scarcely be considered a strong alternative.

The Arms Race Model

A second alternative model may be constructed which represents only a slight modification on the original "frustration-aggression" approach, yet which, in some respects, gives a better fit. According to this line of reasoning, status inconsistency does indeed exercise a frustrating and disturbing influence on a nation, but it is unlikely that in all cases this will result immediately in overtly aggressive behavior. It is more probable that a nation will first attempt to redress the perceived injustice by putting pressure of one sort or another on the member or members of the international system held responsible for the low attributed status. Given the mores of the international system, however, such pressure will no doubt often involve at least a show of military force, and, in order to exert it, the aggrieved nation may embark upon a program of military expansion.

Moreover, it is possible that status discrepancies may induce such expansion in another way. If a nation's military capability should drop below the other dimensions in its status set, and if it has the industrial and demographic

capability to rectify the situation, the inconsistent nation may embark upon a rapid military build-up to balance its rankings. In either case, if other nations have reason to feel threatened by such an expansion, an arms race is not an improbable outcome, and, of course, the explosive properties of such self-feeding spirals of military might have been amply attested to in the history of the international system (Singer, 1958).

This model fits the findings well in certain key respects. First, it would easily account for the 10 to 15 year time-lag which must be introduced to optimize the status inconsistency-war relationship; not only is there likely to be a lag between the perception of status inconsistency and the launching of an arms-backed foreign policy drive to rectify it, but there may be a further delay before such a posture provokes an arms race, and an additional lapse of time before such an arms race becomes sufficiently serious to result in war. Second, it would appear to account well for the observed increase in the relationship over time. In the age of advanced weaponry an arms race is likely to produce a greater increase in the magnitude and severity of war than when the implements of war are relatively primitive. Third, it would account for the decrease in the optimal time-lag from the earlier to the later portion of the temporal

domain. Presumably, both the genesis and the process of an arms race are likely to proceed with greater rapidity in an era when weapons become obsolete more rapidly.

But despite this good fit, the arms race model is not without liabilities as an explanation of war. First, the indices of status inconsistency which are most closely related to war do not correspond to the predictions we would make from this model. If the arms race hypothesis were the correct one, we would expect that inconsistencies between _military_ capability and attributed status would be the best predictors. While indices of inconsistency based on military capability generally predict _well_, they are the _best_ predictors only in the 1850-1964 period using a 10-year lag, and, furthermore, they do not tend to make as significant a contribution to the total variance in the amount of war explained as do indices based on other dimensions of capability. Second, while this model explains the existence of a relationship with a long lag, it cannot account for the no-lag results, as even in the nuclear age a near instantaneous arms race is hardly likely.

Finally, if this was indeed the correct explanation for the observed connection between status inconsistency and war, one might expect the onset of war to be preceded by the rapid buildup of military activity on the part of the nations

involved. This should result in a strong _direct_ relationship between differential rate of change on military capability and war, with very little lag. As can be seen from Tables 5 and 7, however, we do not observe such a relationship. Thus while the arms race model, unlike the previous ones discussed, can adequately account for the 10 to 15 year lagged relationship, it too has its flaws, and once again cannot be considered a complete explanation for the observed findings.

The Alliance Model

Up until now, the models put forward have dwelled more or less exclusively on the _direct_ effects of status inconsistency on the behavior of nation-states. Such discrepancies, however may conceivably act to increase the magnitude and severity of war in quite a different way: they may alter the patterns of formal or informal affiliation and interaction in the international system, and these changed structural configurations may be more war-prone than those they replace. One structural configuration which may play this role is the pattern of alliances between and among the members of the system.

If we assume that status inconsistency does indeed put discomforting pressures on a nation, it will of course

seek changes in at least that aspect of the international status quo which is understood to be at the root of the discrepancy. But it is doubtful that unilateral action of an aggressive nature would always be the first method employed to achieve the necessary changes, except perhaps in the case of international "pariahs" which do not have the normal opportunities for relationships with other nations. In most cases, a better strategy will be to attempt such changes by acting in concert with other nations:

> "States (struggle) for what they regard as appropriate places in the distribution of power...(by the) "artificial" method of linking themselves to the strength of other states. Indeed, this is the only method available to the bulk of states in the actual circumstances of modern history" (Claude, 1963, p. 89).

In other words, we might suppose that a high level of status inconsistency in the international system will result in an increased level of alliance activity directed towards altering the status quo. Another line of reasoning leading us to the same hypothesis is suggested by the theory of coalition formation. Since the problem facing a status inconsistent nation may be viewed as that of obtaining certain types of outcomes which are consistent with its capabilities, one would expect national decision-makers to seek new interaction opportunities which produce such outcomes. Russett (1968) has suggested that, as a working

hypothesis, it is not unreasonable to assume that the dis-
tribution of payoffs within an alliance will be some function
of the capabilities of the partners. If so, it might be
supposed that another attraction of alliances for status
inconsistent nations would be that the alliance bond would
result in an upgrading of ascribed status within the frame-
work of the alliance, as well as assisting the nation in
achieving a higher status in the system as a whole. One
might expect further that such coalitions would stimulate
the formation of defensive pacts or agreements on the part of
those whose interests are threatened by such activity.
The combination of these two mutually-reinforcing tendencies
should result in an increase in the proportion of nations
entering into alliance bonds.

The reader has no doubt anticipated that this causal
chain may readily be extended directly to the onset of war.
International relations scholars are by no means in agreement
over whether alliance bonds are negatively associated with
war in that they reduce the temptations for potential ag-
gressors, or positively in that they polarize conflict in the
system, but the empirical evidence indicates that the over-
all relationship is positive, though weak and probably non-
additive (Singer and Small, 1968). This suggests the pos-
sibility of a two-stage causal link between status

inconsistency and war via alliance aggregation.

For several reasons the alliance model would appear
an attractive explanation of the lagged relationship be-
tween status inconsistency and war. As noted by Singer and
Small, the relationship between alliance aggregation and war
is optimized only when a three-year time lag is introduced.
Since it would no doubt be some time before status incon-
sistency resulted in the creation of formal alliances, a 10
to 15 year time lag for the two-step sequence is scarcely
surprising. However, if we compare the Singer-Small findings
with those obtained here on two other dimensions important
difficulties arise.

First, while they found a _positive_ relationship between
alliance aggregation and war in the twentieth century, Singer
and Small discovered that the relationship in the nineteenth
century is _negative_. If the primary effect of status incon-
sistency was to increase alliance aggregation, it might be
expected that the status inconsistency-war relationship for the
earlier period would be negative or vanishing. While it is
somewhat lower for the 1825-1919 period than for the temporal
domain as a whole, it is still quite strong and positive.

Second, if alliance aggregation were the sole, or even
the primary intervening link between status inconsistency
and war, we would predict that the observed magnitude of

the association between alliance aggregation and war would be greater than that between status inconsistency and war. Such, however, is not the case; even with no time lag, the observed association between status inconsistency and war is considerably the larger, and this difference is accentuated if a longer time-lag is employed. Thus, while alliance aggregation may form part of one causal sequence linking status inconsistency and war, the alliance model is evidently not a sufficient explanation for the findings.

The Intergovernmental Organization Model

Another way in which status inconsistency may act on the structure of the international system to increase the likelihood of war is by affecting the institutional structure designed to reduce the level of conflict within the system; status inconsistency may well seriously interfere with the formation and functioning of intergovernmental organizations. If the reasoning set forth thus far is correct, status inconsistency will both increase tensions and result in a greater degree of stress being placed on the competitive aspects of the relationships between nations. In such an atmosphere, major initiatives of a cooperative nature between nations will be relatively less frequent. One would expect this to be especially true of those acts of

co-operation which imply formal, long-term commitments at a time when the status quo is under attack. Indeed, a continuing theme in the history of many international organizations has been the assaults made upon them by "revisionist" nations. In other words, status inconsistency is likely to have a strong negative impact on the creation, development and functioning of international organizations, regardless of whether they are oriented towards political objectives or are designed to fulfill a purely technical role. And if, as most of us believe, the existence of such organizations does in some way help to reduce the likelihood of international war, we may hypothesize the existence of a causal link based on the tendency of status inconsistency to inhibit the growth of organizations which serve to promote co-operation and reduce tension in the international system.

Like the previous model, the intergovernmental organization hypothesis would account for the long time lag required to optimize the status inconsistency-war relationship. On the whole, however, it is even less satisfactory than the alliance model as an explanation of the results, for two reasons. First, while intergovernmental organizations have become increasingly associated with efforts to prevent war and promote international co-operation in the twentieth century, it is highly unlikely that they were either numerous

or strong enough to play such a role in the nineteenth.

As noted by Wallace and Singer (1970), it was not until

1885 that there were more than a dozen intergovernmental

organizations, and by the end of the First World War

they still numbered no more than 41. Thus, even if the inter-

governmental organization hypothesis could explain the find-

ings for the 1850-1964 period, it could scarcely account for

the existence of strong relationship between status incon-

sistency and war for the 1825-1919 period.

A second and more damaging criticism is that the

model does not square with the existing systematic evidence

about the relationship between intergovernmental organization

and war. Singer and Wallace (1970) have shown that for the

1820-1964 period as a whole there appears to be virtually

no association between the amount of interstate war begun

and either the amount of intergovernmental organization

extant in the previous five and ten years, or the amount

established during these periods of time. While of course

the amount of such organization may be only a crude indica-

tor of its impact and effectiveness, these findings would

appear to effectively rule out the intergovernmental organ-

ization model, at least for the present.

An Evaluation

Having examined in some detail a number of different alternative models which would explain the patterns and trends in the findings, it appears difficult to draw anything but the most ambivalent and tentative conclusions. On the one hand, we saw that the status inconsistency model as originally formulated was by itself inadequate to account for all the observed findings. On the other hand, the various alternative models scarcely fared any better, and in many cases appeared to be far less satisfactory than the original both in accounting for the findings of this study and in meshing them with the systematic evidence already obtained. One possible inference we may draw from this, of course, is that some entirely different explanation is needed to account for the results; given the paucity of hard evidence in this area, and the tentative and incomplete character of the study, we are in no position to rule out such an interpretation. It may well be that further research will enable the construction of a more adequate theoretical model.

An alternative conclusion is equally plausible, however. It may be that status inconsistency and war are linked by not one, but several different causal sequences. It might be, for example, that status inconsistency acts

both _directly_ to increase aggressive national behavior by stimulating national frustration, while at the same time acting to increase the probability of war _indirectly_ by stimulating arms races or by altering some aspect of the structure of the international system such as its alliance configurations. Assuming a multiplicity of causal sequences permits us to account for the existence of a strong status inconsistency-war relationship both with and without a time lag. Moreover, such an interpretation of our findings accords more nearly with our intuitive belief that the onset of war involves not a single sequence of processes, conditions, and behaviors, but the overlap and confluence of many. To speculate further on the particular combination of processes and events which may prove to be the correct intervening sequence would be foolhardy without further evidence, but it is to be hoped that the various lines of reasoning advanced here may point to the next steps for further research in this area.

Some Overall Conclusions on Interpretation

It is difficult to summarize briefly the diverse and often conflicting interpretations which may be given the findings produced by this study. Nor is it easy to come to any definitive conclusions concerning which, if any,

of the various models outlined represent the most acceptable explanations of these findings. Throughout the chapter, however, two basic themes have continually recurred, and it might be well to restate and emphasize them prior to discussing the policy implications of the findings.

First, although this study began as the test of a relatively straightforward hypothesis derived from social psychology, as the results were examined closely it became increasingly clear that the relationships discovered between and among the various indices were indicative of anything but a clear-cut set of answers to the questions posed at the outset. We saw that only by making the most farfetched assumptions or relying on very implausible inferences was it possible to subsume all the findings under a single, simple explanatory rubric. In other words, although the magnitude of the various correlation and dependence coefficients, and the explanatory power of the regression equations, all indicate that there are indeed strong relationships between war and both status inconsistency and differential status mobility, far from writing finis to the matter this merely raises new and more challenging questions about the whole conception of status in the international system and its relation to war.

A second recurrent theme has been the extent to which

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status relationships can make to that task, it is scarcely
too much to hope that some of these opportunities may be
pursued.

But if the importance and urgency of accounting for
the genesis of international war points to the need for fur-
ther research, it also demands that we make the best of what
we have. In other words, even though the results are not
yet all in, we must operate on the assumption that some
knowledge is better than none at all, and not hesitate
in extracting from the findings and their interpretation
all possible implications they may have for the search to
reduce the level of violence in the relations between and
among nations in the international system. It is to this
task that we turn in the next--and final--section.

Policy Implications

If our findings have important implications for
future studies in the field, they also appear to shed some
important light on the problem of achieving a reduction in
the level of war in the international system. At first
blush, however, the insights so generated would seem to have
succeeded less in finding a solution to the problem of war
than in illuminating with still greater clarity the intrac-
tability of the situation. The reason for this rather

pessimistic conclusion lies in the evident difficulties that stand in the way of alleviating the conditions which apparently breed war. How, precisely <u>can</u> one go about reducing differential status mobility and status inconsistency? Neither of the two most obvious answers seem either plausible or attractive.

The Manipulation of Attributed Status

The first and most obvious procedure would be to increase the level of recognition, benefits, and prestige accorded by the system to those nations whose attributed status is markedly lower than their national capability, in short, adjusting attributed status to status expectations. In this way it might be possible to halt or check any animosities that they might develop or stop any war-generating sequence of events that may have been set in motion. However, there would appear to be three important disadvantages to this strategy.

To begin with, it is most unlikely that the collectivity of nations can be persuaded to permit status adjustments to take place quite so readily. As mentioned earlier, since reputational status is almost inevitably a constant-sum attribute, there can be little doubt that attempts at "re-apportionment" will usually meet with tremendous

resistance on the part of those who would be on the losing side of the equation. Such a redistribution could still take place if the superpowers, secure in _their_ status and aware of the dangers of inconsistency, were to act so as to promote status equilibration. Unfortunately, in view of the attitudes which both have displayed towards that most prominent of inconsistent nations, China, it would seem unlikely that they can be relied upon to play this role. Of course, it might be possible to arrange some system of trade-offs or side-payments whereby the "aggrieved" nation is rewarded on some other dimension. Yet, this might backfire if the low dimension remained salient to the national actor and the "reward" had the effect of increasing national capability; the result would then be to increase status inconsistency. In short, it seems unlikely that manipulation of attributed status by itself can be expected to make much impact in reduced status inconsistency. It goes without saying, however, that any additional flexibility shown by nations in the granting of higher status to inconsistent nations would pay some modest dividends in reducing the likelihood of war.

A second possible objection to this course of action is an ethical one. By advocating the granting of attributed status (and the rewards which accompany it) on the basis

of capability, we would be overlooking the fact that there are often very pressing ethical reasons for <u>not</u> granting such status. The clearest examples are those nations which are governed by regimes which practice totally abhorrent and morally unacceptable policies within their borders, such as Nazi Germany and South Africa. In such cases, the danger of war which may result from withholding attributed status must surely be balanced against the great harm that may be caused by lending support and sanction to such regimes.

But even leaving aside such obvious outliers, the strategy of attributing status and reward solely on the basis of capability clearly contains within it a basic amoralism which is scarcely an attractive basis for an international community. National size and power have historically borne little relationship to the quality of life for the inhabitants of the nation, and to reward national decision-makers for their successful pursuit of ever-larger increments of national capability may easily have the effect of re-inforcing the all-too-prevalent tendency of national leaders to seek international reputation at the expense of their population.

A third and final difficulty lies in the fact that the overt acceptance of the principle of national inequality as a peace strategy runs squarely up against one of the most

cherished beliefs held by the leaders of nations and inter-

national organizations alike--that the sovereign equality of

nations is a good in and of itself, and constitutes the

basis for a sound international order. Traditionally, most

have believed that justice and order amongst nations have

been more or less synonymous with a basic equality between

them; although as noted at the outset the old doctrine of

sovereign equality is honored more in the breach than in the

performance, it has nevertheless been commonly assumed that

a basic equality of rights and privileges among nations is

the ideal state of affairs to which all those concerned

with international justice should aspire (Lagos, 1963, pp.

22-23). Thus, hierarchy in the international system has

been viewed in somewhat the same way as sin: While

perhaps inevitable, it is nonetheless not to be accepted

but rather struggled against.

Moreover, not only has a large measure of equality

in relations amongst nations in the international system

been regarded as a good in itself, but it has usually been

understood that such a state of affairs is more conducive

to peace than one in which a more strict hierarchy prevails

and where the rewards and honors bestowed by the system are

more closely controlled by those at the top of the hierarchy.

Peace, for some, has been almost defined as equivalent to

the rights of small nations, as is well illustrated in Wilson's Fourteen Points.[1]

This attempted juxtaposition of the equal rights of states and the achievement of peace is usually associated with the philosophy and practice of the League of Nations, but it seems clear that the same assumption undergirds much of the philosophy and operation of the United Nations system as well, even to the present day (Nicholas, 1963, pp. 35-37). Thus, the implementation of a status equilibration strategy would almost inevitably meet great resistance from the very organizational leadership most committed to a search for international peace, and, as a consequence, the prospect of initiating and implementing such a strategy through organizations representing the world community as a whole does not appear very promising.

The Manipulation of Capability Status

The second obvious strategy to deal with status inconsistency would be to reduce or eliminate the differences

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[1]In introducing these famous statements, Wilson made this explicit: "An evident principle runs through all the programme I have outlined. It is the principle of justice for all peoples and nations and their right to live on equal terms of liberty and safety with one another, whether they be strong or weak" (cited in Lagos, 1963, p. 22).

in rates of capability change. As we saw, such differential

changes were in large measure responsible for generating

status inconsistency and, in some cases, exercised a strong

independent influence on war by themselves. Thus, by

"smoothing out" the differences in national growth rates it

might be possible to lessen the risk of war. But clearly,

the disadvantages of the status equilibration strategy pale

beside the drawbacks that would be encountered in any attempt

to manipulate national capability.

First and most obviously, it is virtually impossible

to conceive how such a manipulation could, in fact, be

successfully undertaken. Some of the factors to be regulated,

such as population growth and economic expansion, have

usually proved difficult or impossible to adjust either up-

wards or downwards even when a major policy initiative is

undertaken <u>within</u> a nation. The likelihood that such a manip-

uulation can be undertaken successfully from the outside--

possibly with the active opposition of the national leader-

ship--would seem virtually nil. With regard to <u>military</u>

capability, years of fruitless disarmament talks have shown

the difficulties of manipulating that dimension of national

status.

Second, the strategy of equalizing rates of capa-

bility change has a fatal political and ethical drawback in

that it would deny relative mobility and thus, in effect, freeze the status quo amongst nations. Such a solution would, of course, be acceptable to the superpowers (and indeed, there are many who would accuse them of planning towards such an eventuality), but it is almost certain to be unacceptable to those on the bottom of the existing pecking order. Moreover, it will be least acceptable to those to whom it must needs be most quickly applied: the rapidly upwardly mobile.

Finally, even if it could be applied, such a strategy would produce grotesque and almost certainly unacceptable results within the various national societies, particularly on the economic dimension. This is an almost inevitable consequence of the vast absolute disparities in the levels of economic development between and among nations. On the one hand, for the nations at the bottom of the ladder, a relatively high rate of economic growth is a vital necessity to achieve acceptable standards of material well-being. On the other hand, the developed nations are becoming increasingly aware of the need for a lower rate of economic growth if only to protect the physical environment. Clearly, these needs would seem to rule out any uniform, stabilized global rate of growth in the near future.

Salience Manipulation-A Possible Way Out

Since the manipulation of status inconsistency and differential status mobility appears neither possible nor desireable, must we then conclude that it is impossible to deal with what may very well be one of the underlying roots of war, and content ourselves with devising strategies to cope with and control only their overt manifestations of tension, crisis, and agression? While such symptomatic treatment is clearly indispensible for the moment, it would be premature to conclude that status mobility and status inconsistency must result in the endless generation of crises and violence within the system; there may very well be one effective approach to coping with them.

As was discussed earlier, most of the plausible causal sequences linking status inconsistency and war take as their point of departure the sensitivity of national de-cision-makers, along with their domestic clientele and mass publics, to considerations of national status. If ways could be found of reducing this sensitivity and focusing the attention of both decision-makers and publics in other, safer directions, the tension-producing effects of inconsist-encies and mobilities might be considerably reduced.

That such a strategy might work at least in principle

is at least arguable from anecdotal evidence. During periods
of severe domestic disorder or stress, policy elites have
been forced by necessity towards what Wolfers has referred
to as a strategy of "self-abnegation", and it has typically
resulted in at least limited periods of peace despite high
status inconsistency and mobility. An example might be
the Soviet Union during the 1920's and '30's, forced by its
internal circumstances to react less strongly than it other-
wise might have to its very low attributed status.

Obviously, to generate such abnegatory behavior in the
absence of massive internal strife will be no easy matter.
As long as both elites and mass publics continue to iden-
tify their personal security with national power, and, as a
consequence, evaluate outcomes in the international system
almost totally in terms of the success of their particular
nation, any diffusion of attention away from national status
and towards more the productive tasks of international co-
operation and internal development would appear a very im-
probable occurrance. Yet, as the past few years have shown,
there is a growing awareness in many parts of the world--
not the least in this country--that a major shift in emphasis
along these lines is essential not only for the achievement
of peace but to free resources for many other urgent goals
as well. If the findings of this study are to be believed,

this movement is extremely encouraging; if it succeeds, it may signal the beginning of the end of the zero-sum game of international status seeking that has led to so much bloodshed.

APPENDIX

DATA SOURCES FOR THE INDICES OF THE INDEPENDENT VARIABLES

General

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