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An experimental study on mass loading of soil particles on plant surfaces

By J. G. Li, M. H. Gerzabek and K. Mück

(With 2 figures)

Summary

Radionuclide contaminated soil adhered to plant surfaces can contribute to human ingestion dose. To determine this contribution, a method of ^{46}Sc neutron activation analysis was established and tested, by which a detection limit of 0.05 mg soil per g dry plant biomass can be obtained. In the field and greenhouse experiment the mass loading of soil on ryegrass (*Lolium perenne* L.) and broad bean (*Vicia faba* L.) was investigated and the contribution from rainsplash and wind erosion were evaluated separately. Soil retained on plant surfaces in field conditions in Seibersdorf/Austria was 5.77 ± 1.44 mg soil per g dry plant for ryegrass and 9.51 ± 0.73 mg soil per g dry plant for broad bean. Estimates of contribution from rainsplash and wind erosion to soil contamination of plant during the experimental period are 68 % and 32 % for broadbean, 47 % and 53 % for ryegrass, respectively.

Mass loading results from field studies indicate that soil adhesion on plant surfaces can contribute up to 23 % of plant ^{137}Cs contamination, the transfer factors modified by mass loading decline differently, depending on ^{137}Cs concentration of the soil and the soil mass adhered to plant surfaces.

Key-words: ^{137}Cs, massloading, plant contamination, ^{46}Sc neutron activation analysis.

Ergebnisse einer experimentellen Studie über die Beladung von Pflanzenoberflächen mit Bodenpartikel

Zusammenfassung

Radioaktiv kontaminierte Bodenpartikel auf Pflanzenoberflächen können zur menschlichen Ingestionsdosis beitragen. Um diesen Beitrag quantifizieren zu können, wurde eine ^{46}Sc-Neutronenaktivierungsmethode getestet, mit der es möglich ist, die Menge von anhaftenden Bodenmengen abzuschätzen. Die mittlere Nachweisgrenze dieser Methode liegt bei 0,05 mg Boden pro g Pflanzenmasse (Trockengewicht). In einem Glashaus- und einem Freilandexperiment wurde der Beitrag luftgetragener und durch Regeneinwirkung auf die Pflanzenoberfläche gelangter Bodenpartikel an der Gesamtkontamination von Ackerbohne (*Vicia faba* L.) und Weidelgras (*Lolium perenne* L.) ermittelt. Die Bodengesamtmenge, die unter Freilandbedingungen in Seibersdorf/Österreich von dem Pflanzenbestand zurückgehalten wurde, lag bei 5,77 ± 1,44 mg Boden/g Pflanzentrockenmasse (Weidelgras) und 9,51 ± 0,73 mg (Ackerbohne). Der Anteil der windgetragenen Partikel wurde mit 32 % (Weidelgras) und 53 % (Ackerbohne) ermittelt. Der Rest ist auf die Regeneinwirkung zurückzuführen.

Messungen der Bodenkontamination an Pflanzenproben aus früheren Feldversuchen zum Boden-Pflanze-Transfer von ^{137}Cs zeigten, daß kontaminierte Bodenpartikel bis zu 23 % der gesamten ^{137}Cs-Konzentration in Pflanzen ausmachen können, wobei der Beitrag von der ^{137}Cs-Konzentration im Boden und der Menge an anhaftenden Bodenpartikeln abhängt.

Schlüsselworte: Bodenpartikel, ^{137}Cs, Pflanzenkontamination, ^{46}Sc Neutronenaktivierungsanalyse.

1. Introduction

Radionuclides from soil particles adhered to plant surfaces can be absorbed biologically, consumed inadvertently by grazing animals, ingested and thus contribute to human dose (White et al. 1981, Green and Dodd 1988, Pinder III and McLeod 1988). Large variations in the soil-to-plant transfer factors of radionuclides (TF, as defined as the concentration of a radionuclide in plant material, divided by its concentration in the soil on which the plant was grown; Desmet and Sinnaeve (1992)) used in the dose assessment models may also result from soil contamination of plant surfaces (Gerzabek et al. 1992). Soil particles might be transferred to plant surfaces by two principle processes: rainsplash and wind erosion. Raindrops detach particles from the soil surface and splash them onto plant surfaces. Wind may either suspend soil particles into the atmosphere or cause soil particles to bounce along the soil surface and then deposite on plant surfaces (Sehmel 1980, Dreicer et al. 1984). Other factors such as soil disturbance by mechanical equipment or livestock activities may also contribute to the amount of soil transported to plants. Several studies on mass loading of soil on plant surfaces were conducted more than ten years ago (Anspaugh et al. 1975, Arthur and Alldredge 1982, Adriano et al. 1982). However, few data are available on mass loading for various types of vegetation (Pinder III and McLeod 1989). Most of the mass loading studies were conducted in arid or semi-arid regions. Humidity is expected to reduce soil resuspension (Cohen 1977). A summary of soil concentrations on plants obtained by former studies is listed in table 1. Investigated areas were mainly located in the southeastern part of the United States. The values of soil mass loading ranged from 1.1 (cabbage) to 260 (lettuce) mg soil per g dry plant biomass. For simple steady state models for the transfer of radionuclides in the environment, only general values of soil-to-plant transfer factors are needed. In this case all processes leading to plant contamination can be included in one parameter which describes the maximum transfer from soil (Köhler and Frissel 1990). For models intended to assess dose or concentration of radionuclides in the environment, more accurate soil-to-plant transfer factors will have to be used. Thus, the main processes involved should be made clear. Attempts must be made, therefore, to quantify the contribution of radioactivity from soil contamination on plant surfaces.

Table 1

A summary of soil concentrations on plant surfaces from literature

Plant	Concentration (mg soil/g dry plant)	N.	Reference
Grass	18 ± 48	26	Arthur and Alldredge (1982)
Tomatoes	17		Dreicer et al. (1984)
Lettuce	260 ± 100	4	McLeod et al. (1984 a)
Brocooli	10 ± 8.1	4	McLeod et al. (1984 a)
Turnips	32 ± 11	4	McLeod et al. (1984 a)
Cabbage	1.1 ± 1.1	4	McLeod et al. (1984 a)
Tobacco	2.1 ± 0.6	12	McLeod et al. (1984 b)
Sunflower	2.6 ± 0.9	10	Pinder and McLeod (1988)
Soybean	2.1	10	Pinder and McLeod (1989)
Wheat	4.8	10	Pinder and McLeod (1989)
Corn	1.4	10	Pinder and McLeod (1989)

Mass loading is an estimate to quantify directly the mass of soil actually deposited on plant surfaces (Hinton 1990). There are different methods to estimate the mass of soil present on plant surfaces. Arthur and Alldredge (1982) used ultrasonic washing to treat vegetation. After filtering the water through a 1.2 µm filter, the filters were ashed and weighed. Soil attached to plant surfaces was estimated by plutonium content. Green and Dodd (1988) used the titanium method which was established by Kiriyama and Kuroda (1982) to estimate the soil adhering to herbage. The procedure removes iron from the solubilised sample by ion-exchange. Subsequently there has been no interference from iron in the samples analysed. The titanium content was determined by spectrometric analysis. A detection limit of about $2.0 \cdot 10^{-3}$ mg soil/g dry plant can be obtained. Pinder III and McLeod (1989) used ^{238}Pu as a monitor of soil transport to plants and obtained a detection limit of approximately 0.5 mg/g dry plant biomass. The ^{238}Pu content of the vegetation due to soil transport was converted to grams of soil by dividing by the ^{238}Pu concentration (Bq/g) in the particles on the soil surface that could be suspended by a 6 m.s^{-1} wind velocity. Although ^{238}Pu may be measured at very low detection limits ($\sim$1 mBq per sample by alpha-spectroscopy), it was discarded as useful method in this experiment for two reasons. Firstly, the ^{238}Pu content of Austrian soils is only about 0.2 mBq/g which yields insufficiently low detection limits for mass loading of soil on plants. Secondly, the determination of ^{238}Pu by alpha-spectroscopy involves a chemical separation of the Pu which may not be 100 % with the minute amounts of soil on the plant. The chemical efficiency is not known and not correctly determinable by spiking.

Scandium is geologically ubiquitous in soils but poorly absorbed by plant roots and not biologically mobile in vegetation (O'Toole et al. 1981). Therefore scandium is a perfect element for mass loading studies.

2. Materials and Methods

2.1 The method for determining mass loading

Both Ti and Sc were tested for their suitability to determine low mass loading on plants. To determine these elements, the neutron activation analysis (NAA) was chosen since this method involves no dissolving of the plant or chemical separation. Thus no losses due to imperfect chemical separation are feasible.

With Ti only the isotope ^{51}Ti with a half-life of 5.79 minutes can be used. To obtain an optimum detection limit, an irradiation of 2 seconds at a flux of $6 \cdot 10^{13}$

neutrons s^{-1} cm^{-2} was employed, followed by very short period of one minute required for the transport from the irradiation facility to the monitor and a measurement time of five minutes. Despite this optimization of discriminating against other radionuclides, due to a significant Compton background from other activated nuclides in the plant sample (^{24}Na, ^{38}Cl, ^{56}Mn, etc), the detection limit is seriously impared. Even under optimum irradiation and measurement conditions a D. L. of only 30 mg soil/g plant was obtained, not sensitive enough for the required task.

With Sc, the isotope ^{46}Sc with a half life of 83.85 days is deployed. The long half life allows a rather lengthy decay permitting a decay of the large majority of radionuclides (^{24}Na, ^{28}Al, ^{38}Cl, ^{56}Mn, etc.), which results in a low Compton background. This and the high activation cross section of ^{45}Sc results in a detection limit of 0.05 mg soil on plant if a decay time of longer than 10 days between irradiation and measurement is adopted.

The plant samples were dried at 100 °C for 48 hours, and ground by a laboratory mill. An equivalent of approximately 1.5 g of the plant was weighed into polyethylene capsules (5 ml volume) for neutron activation. Soil samples were air-dried for seven days, sieved through a 2 mm steel screen, an equivalent of 0.1 g was weighed and put into the capsules for neutron activation. The plant and corresponding soil samples were irradiated at close distances for six minutes ensuring an equal neutron flux of $8.0 \cdot 10^{13}$ neutrons cm^{-2} s^{-1}. After a storage time of two weeks, to allow the decay of the short lived activation products, samples were measured by gamma spectrometry with Ge(Li)-detector of 20 % relative efficiency. A long measurement time of 50,000 seconds for the plant and comparatively short time of 2,000 seconds for soil samples was applied resulting in the detection limit of 0.05 mg soil per g plant biomass.

2.2 Mass loading experiment

The soil used in the experiment was a Calcic Chernozem, the characteristics of which are listed in table 2. The climatic parameters of the Austrian Research Centre Seibersdorf are shown in table 3.

Table 2

The characteristics of the soil used in the mass loading experiment in Seibersdorf/Austria

% sand	% silt	% clay	pH (CaCl$_2$)	pH (H$_2$O)	% humus	% CaCO$_3$	mg K$_2$O/ 100 g
60.9	24.4	14.7	7.5	8.2	1.9	28.0	31.2

Table 3

Climatic parameters for the field experiment at the test site Seibersdorf/Austria

parameter	average over period 19 May – 1 July 1992	1993	annual average 1992	maximum value*
precipitation (mm)	80.3**	62.6**	412.8**	25.8 (20. VI.)
windspeed (m.s^{-1})	2.4	2.4	3.3	5.9 (10. VI.)
humidity (%)	63	62	66	87 (20. VI.)
temperature (°C)	17.6	19.1	11.2	33.6 (11. VI.)

* in experimental period (May 19th to July 1st 1993)
** accumulative precipitation

Two plants, ryegrass (*Lolium perenne* L.) and broadbean (*Vicia faba* L.) were grown under three conditions:

i) BG, RG: Plants were grown in pots (5 kg of soil) and in a closed cabinet located in the greenhouse. The above ground parts of the plants were carefully protected from the soil surface by polyethylene films and the cabinet was thoroughly cleaned then closed with transparent films. The cabinet was opened only for irrigation. Irrigation was applied along a glass tube (20 cm of length) deeply inserted into the pot to prevent waterdrop splash;

ii) BP, RP: Plants were grown in the field on small plots (2 × 8 m) with polyethylene film protection of the soil surface. A tube irrigation system was established under the film to avoid waterdrop splash from irrigation. The above-ground parts of the plant thus were protected from direct rain splash of the surface soil and direct wind induced soil contamination, but might receive possible indirect soil contamination from the vicinities;

iii) BF, RF: Plants were grown on small plots (2 × 8 m) without any protection, with a tube irrigation system. The performence of the experiment is shown in table 4.

Table 4

Reference data for the mass loading experiment (greenhouse and field)

Plant	Time of planting	Time of sampling	Period (days)
BG	24. 05. 1993	29. 06. 1993	36
RG	24. 05. 1993	29. 06. 1993	36
BP	19. 05. 1993	01. 07. 1993	43
RP	19. 05. 1993	01. 07. 1993	43
BF	19. 05. 1993	01. 07. 1993	43
RF	19. 05. 1993	01. 07. 1993	43

B: broadbean
R: ryegrass

G: greenhouse
P: protected field
F: field

The plant samples were taken from the above-ground parts of the plant 10 cm above the polyethylene films (or 15 cm above the soil surface). For each treatment one soil sample (collected from the top 2 cm of the plot) and four replications of plant samples were taken in a random method.

2.3 Measurements of the samples from field studies

Samples from the field studies on soil-to-plant transfer of the Austrian Research Centre Seibersdorf (HORAK et al. 1989) were investigated. Plant and soil samples were taken from different regions of Austria contaminated by the Chernobyl accident derived ^{137}Cs. The ^{137}Cs activity concentrations of the samples are given in table 5. By determining the mass loading on plant samples by ^{46}Sc neutron activation analysis (2.1) the contribution of outer contamination to the soil-to-plant transfer factors was estimated.

3. Results and Discussion

The mass loading values of soil particles on plants and their variance obtained from the experiment are shown in figure 1. Average mass loading values are given in figure 2. Vertical bars indicate 95 % confidence intervals on the mean

Table 5

^{137}Cs concentrations in plants and soils taken from Austrian areas contaminated after the Chernobyl accident (HORAK et al. 1989)

Plant No.	Bq ^{137}Cs/kg dry plant	Bq ^{137}Cs/kg dry soil
P1	0.760	23.5
P2	1.14	37.7
P3	2.55	37.7
P4	5.73	314
P5	0.88	19.7
P6	1.54	146

P1—P4: barley straw
P5—P6: wheat straw

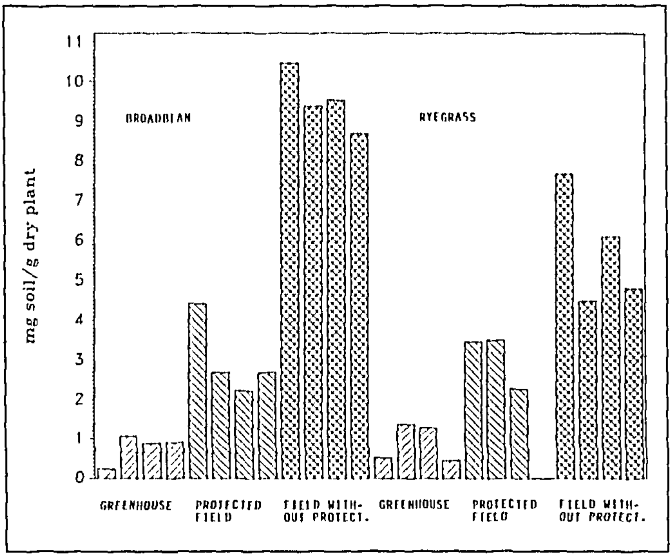

Fig. 1: Mass loading of soil on broadbean and ryegrass in three different experimental treatments

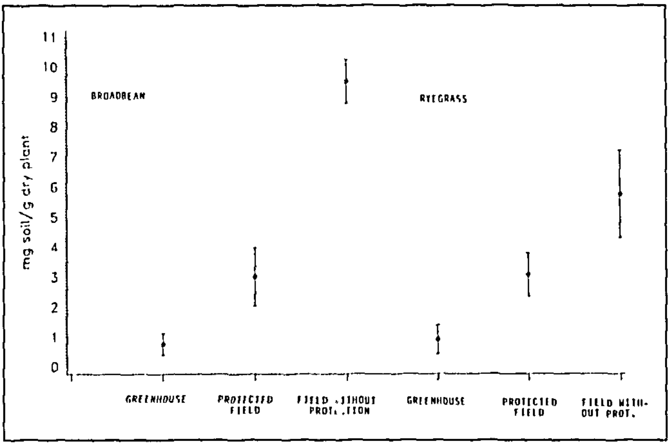

Fig. 2: Average soil mass loading values on broadbean and ryegrass in different experimental treatments with 95 % confidence intervals

value of observations (dark circles). Obviously, the concentration of soil on plants increases from protected plants in the greenhouse (BG, RG) to the plants in the field, but protected against rainsplash (BP, RP), and finally to the unprotected plants in the field (BF, RF).

An analysis of variance of the differences between mean mass loading values is given in table 6. The differences of mean mass loading values are statistically significant between treatments in both broadbean and ryegrass. The means of BF and RF are as well significantly different. This indicates that broadbean and ryegrass are distinctly different in soil adhesion and retention characteristics.

Table 6

An analysis of variance of the differences in mean concentrations of soil on plant surfaces from the mass loading experiment in Seibersdorf/Austria

Comparison	Lower Confidence Limit	Difference Between Means	Upper Confidence Limit
BF-BP	5.2148	6.5075	7.8002***
BP-BG	0.9423	2.2350	3.5277***
BF-BG	7.4498	8.7425	10.0352***
RF-RP	1.2937	2.6900	4.0863***
RP-RG	0.7737	2.1700	3.5663***
RF-RG	3.5673	4.8600	6.1527***
BF-RF	2.4523	3.7450	5.0377***
BP-RP	− 1.4688	−0.0725	1.3238 n.s.
BG-RG	− 1.4302	−0.1375	1.1552 n.s.

*** denotes $p < 0.001$
n.s. denotes not statistically significant

The soil concentrations on broadbean and ryegrass in the field without protection are both higher than those in the field with plastic film protection. The precipitation in Seibersdorf during the experimental period was 62.6 mm (see table 3). Because the plants grown in the protected field were well protected from rainsplash, rough estimates of the contribution from rainsplash and wind erosion to plant contamination can be calculated (table 7), assuming that ^{46}Sc-contents in plants from the greenhouse are due to small soil contaminations instead of root uptake of ^{46}Sc. For broadbean 68 % of soil contamination are from rainsplash, only 32 % from wind erosion. For ryegrass 47 % soil adhesion appears to be from rainsplash and 53 % from wind erosion. These results might be due to the differences in plant anatomy and leave structure.

Table 7

Contribution of pathways to soil contamination on plant surfaces obtained from the mass-loading experiment

Plant	Soil concentration on plant surfaces (mg soil/g dry plant) Wind erosion	Rainsplash	Total
Broadbean	3.01 (32 %)	6.50 (68 %)	9.51
Ryegrass	3.08 (53 %)	2.69 (47 %)	5.77

Data in brackets are the percentage of soil concentration compared to the total concentration.

The second part of the paper deals with the practical use of mass loading values in soil-to-plant transfer studies (2.3). The transfer factors do not discriminate the contribution of soil particles on plant surfaces between the radionuclide contamination of crops from that of root uptake. The transfer factors due to pure root uptake may be estimated by subtracting the mass loading contribution according to the formula:

$Ar = At - As \cdot Ms / 1{,}000$

Ar: Activity concentration of plant due to root uptake (Bq/kg);
At: Activity concentration in plant determined by transfer measurements (Bq/kg);
As: Activity in soil (Bq/kg);
Ms: Mass loading value (mg soil/g plant)

The corrected ^{137}Cs transfer factors from field experiments (Horak et al. 1989) are given in table 8 in comparison to the original transfer factors.

Table 8

Soil adhesion on plant shoots from field studies after the Chernobyl fallout in Austria obtained by ^{46}Sc neutron activation method and its impact on soil-to-plant transfer factors

Sample *	Concentration mg soil/g dry plant	D. L.$^{a)}$	n	Bq ^{137}Cs$^{b)}$/kg dry plant (% of total)	TF1$^{c)}$	TF2$^{d)}$
P1	6.01	0.050	6	0.141 (18.34)	0.029	0.023
P2	1.33	0.045	3	0.063 (5.53)	0.027	0.025
P3	2.31	0.048	2	0.087 (3.41)	0.059	0.057
P4	4.23	0.076	1	1.328 (23.18)	0.016	0.012
P5	1.16	0.028	1	0.023 (2.61)	0.039	0.038
P6	1.53	0.029	3	0.223 (14.48)	0.009	0.008

* See note of table 5
a) Detection limit (mg soil/g dry plant);
b) Radioactivity derived from soil adhesion;
c) Transfer factor (Bq/kg fresh plant) / (Bq/kg dry soil) for ^{137}Cs;
d) Transfer factor modified by the contribution of soil adhesion.

Table 8 also shows the relative importance of soil adhesion to the plant surface ^{137}Cs contamination. Based on the soil concentrations of the plants from this experiment, the soil fraction on plant surfaces (without considering the distribution differences of ^{137}Cs in soil particles, Gerzabek et al. 1992) would contribute up to 23.2 % to ^{137}Cs contamination in plant samples. For sample P4 (barley straw), the transfer factor (TF) with soil massloading correction is 0.012, thus decreasing by 23 %, compared to the original TF (0.016). This change is due to the high ^{137}Cs concentration of the soil (314 Bq/kg soil) and the high soil mass loading value (4.23 mg soil/g dry plant). For sample P1, despite the fact that the soil mass loading was highest among the measurements (6.01 mg soil/g dry plant), as a result of the low ^{137}Cs soil concentration, the transfer value with soil correction decreased only by a rate of 18 %.

Because of the fact that radionuclide concentration in resuspendable soil particles (< 125 µm in diameter, Dreicer et al. 1984) is higher than that of the entire soil (Livens and Baxter 1988, Pinder III and McLeod 1989), it is possible that the radionuclide contamination of plants from soil adhesion may be underestimated in table 8. For example, the ^{137}Cs concentrations in soil particles smaller than 2 µm can be 2.7 times higher than that in the entire soil (Gerzabek et al. 1992).

The importance of soil in contributing ^{137}Cs contamination (or other pollutants) to plant surfaces depend upon the type of plants, the concentration of ^{137}Cs in the soil, and the resuspension and retention characteristics of the particles. Other factors such as weather conditions (wind speed, precipitation, humidity), agricultural activities (plowing, irrigation, mechanical harvest) etc., may be as well important to the soil adhesion (Arthur and Alldredge 1982, Dreicer et al. 1984, Nicholson 1988, Pinder III and McLeod 1988).

A limitation of the proposed method arises from the fact that it is not possible to obtain information on radionuclide uptake from contaminated particles on the plant surface into plant tissue and the subsequent translocation. That means there may be a further contribution of soil adhesion to plant contamination by radionuclide plant uptake into shoots during the vegetation period through contaminated particles, which may not be present at harvest due to weather effects.

4. Conclusions

Mass loading of soil on plant surfaces as an approach to estimate the effect of soil contamination is advisable, because it integrates the effects of all soil transport mechanisms and it is the one that requires the least additional data to obtain an estimate of contamination onto plant surfaces (Pinder III and McLeod 1989, Hinton 1990). From the present experiment it seems that rainsplash and wind erosion contribute to soil mass loading of plants in the same order of magnitude.

^{46}Sc neutron activation analysis is an effective and practical method for soil mass loading research. It is also an undestructive analysis and the quantity of samples needed keep to the minimum.

Mass loading research should be intensified and directed to various types of vegetation, climates and soils in order to meet necessities of radioecological models. Further research should include bioavailability of radionuclides in soil particles adhered to plant surfaces, the weather effects (short-term, intermediate-term, long-term), seasonal variations, and the role of different soil particle sizes retained on plant surfaces.

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