ISSN 1996-3343

Asian Journal of Applied Sciences

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Asian Journal of Applied Sciences 1 (3): 259-264, 2008
ISSN 1996-3343

The Remediation Based on Old Animal Feces Effect on Radio Iodine in Uppermost Soil Layers

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Abstract: The total removed radio-iodine in surface layer of the soil demands some factors. These factors consist of detention time of animal fertilizer in the soil, amount of animal fertilizer given to soil and depth of used animal fertilizer in the soil in which were unknown and so they were determined. In the lab an experimental system reproducing the environmental conditions of the contaminated areas were designed in pots and Radio polluted sub samples soil was treated with old feces of sheep and cow which are normally used as common fertilizers. Seven days from the contamination of sub soil samples treated with animal feces as fertilizers present at the time of inoculation, a drop up to 53%, with 1/4 volume of animal fertilizer: 3/4 volume of sub sample soil in radioactivity rate was indicated, after a series of washes which simulate one year's rainfall. Radio iodine movement towards deeper soil layers, following old animal fertilizer treatments and their subsequent stabilization reduces bioavailability in the uppermost soil horizon.

Key words: Animal feces, fertilizer, radioisotopes, soil, remediation

INTRODUCTION

Some research carried on by some experts in the field results in the following: release of radio iodine in the environment is a major risk to both human and animal health as a source of irradiation and toxicity exerted at cellular level by some factors (Moysich *et al*., 2002). Radio iodine being present in the food chain (de Ruig and vander Struijs, 1992). The distribution of radio- nuclides in the soil lessens as we move down, the amount of rainfall and the passage of time are two factors in increasing radio-nuclides distribution (Carbol *et al*., 2003). Radio iodine's low mobility (Velasco *et al*., 1997). Techniques for remediation based on biological methods (Zygmunt *et al*., 1998) have been developed for soil decontamination. Potential bioremediation agents include wild plants (Sokolik *et al*., 2001), genetically engineered plants (Salt *et al*., 1995), fungi (Gray, 1998) and natural micro-organisms (Watanabe, 2001). The disposal of contaminated biomasses represents, however, a trouble and is a big limit to the methodology application. These systems are not able to reduce contamination to an acceptable level of reduction.

Contamination commonly found in polluted areas is due to Cesium 137 (^{137}Cs) and Strontium 90 (^{90}Sr) which are the only two factors polluting the soil. In the early stages of both the Chernobyl accident (which is cited extensively) and Wind Ascalea Fire (to give but two examples), radio iodine was significant. In other cases, Pu has been a factor of concern. In this experiment, radio isotope I-125 with short half-life is used as a contamination source due to (a) working with this radioisotope is both

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much more easier and feasible (b) the isotope I-125 like long half-life radioisotopes can assimilate in grass and (c) Finally, the positive effect of old animal feces as fertilizer on the reduction of radio iodine in uppermost soil layers. Based on the previous, it can be hypothesized that radio iodine may be displaced from adsorption sites, mobilized by water and then immobilized in the underlying layers of the soil. The bioavailability of radio iodine would be decreased by their transfer from the zone of leaching to the zone of accumulation. The removal of radio iodine from the most superficial soil layers renders them from herbaceous plants and grasses, allowing the production of safe hay. The major concern of this research is to assess in laboratory assays the possibility of removing the radio iodine in the superficial soil layers and fixing them into lower levels.

MATERIALS AND METHODS

The soil was collected from region around Zahedan City-Sistan and Blouchestan-Iran in April 2006. Soil sample used in this investigation were podzolic. Soil sample were collected from the O horizon containing 6% organic content. It was coarse textured containing more than 70% sand. Sample soil was collected from 40 cm side square surface and a depth of 20 cm. The soil sample was cleaned, removing grass, roots and pebbles. It was then dried at room temperature. The dried sample was kept at room temperature. Before each experiment sub-samples were re-hydrated at 10% w/w.

Three experiments were carried on in pots to assess the removing effects of animal fertilizer treatments in artificial conditions similar to natural ones and to calculate soil strength in keeping radio iodine at the more deeper layers.

The first experiment (experiment A), was performed to estimate the time need for initial application of animal fertilizers to the soil for the release of radio isotope from uppermost contaminated soil layers, meanwhile a second experiment, (experiment B), was carried on to represent the amount of old feces as animal fertilizer given to the sub sample soil and a third one (experiment C), was set to represent the suitable depth of giving old animal feces as fertilizer to the soil.

In experiment A, 6 conical test-tubes have been used as pots. Three pots were used as case study containing sub samples soil and fertilizer, three other pots were used as means for control study containing sub soil sample without fertilizer. Each pot was 6 cm in length and 7.5 cm in upper diameter and 4.5 cm in lower diameter. The bottom end of pot was closed by a glass stopcock, filled with cotton to prevent loss of the soil sample. Another pot was placed with the same size under the pot to collect elutes from the stopcock. Each of the six pots used in experiment A were filled with 200 g of soil sample up to 6 cm height. The soil:fertilizer mix was first treated with 5 mL of I-125 with the concentration of 2 kBq, then, 10 mL of distilled water sub was added to every pot daily. Distilled water was again added to sub-sample soil when the previous 10 mL had flown out of each pot.

The washes operation with 10 mL of distilled water was carried out in order to simulate atmospheric precipitations; the total amount of water used in this experiment corresponds approximately to 100-200 mm of rain, corresponding to the average yearly rainfall. Each addition of distilled water was delayed until every pot had completely emptied collecting. Each fraction in every pot was done as soon as the pots emptied. After reverse agitation over night and centrifugation at 2000 rpm for 20 min; 10 mL of the supernatant fraction, 10 g of sub sample soil and 10 mL eluted liquid were collected. These fractions were subjected to automatic gamma counting and the values obtained were corrected according to the initial volume of the fraction. Three times, 7, 14, 21st are the best way for the response of animal fertilizer and radio iodine in soil layer.

The three pots in experiment B were filled with sub sample soil and animal fertilizer to a height of certain centimeter. The volume ratio of sub samples soil to animal fertilizer were as 1/2 soil and 1/2 animal fertilizer, 1/4 soil and 3/4 animal fertilizer, 3/4 soil and 1/4 animal fertilizer. Then, 5 mL of I-125 was added to the surface of each sub-sample soil in control and case pots. Ten milliliters of distilled water were added to sub sample soil: animal fertilizer mix to wash every day. The radioactivity of the extracted solution and sub sample soil and elute resulting from centrifuge set which were collected and measured. The washes operation of the sub sample soil: animal fertilizer mix was followed by 10 mL of distilled water during experiment B. Like experiment A, pots in experiment B was emptied from distilled water before pouring the next 10 mL distilled water on the soil: fertilizer mixture. The resulting values were corrected. The changes of animal fertilizer and soil are considered in elute and wet soil.

The pots in experiment C were 20 cm in height and 8 cm in diameter. The old animal fertilizer with 1 cm thickness was put in every 5 cm of case pots. Control pots were the same size without fertilizer. The sub samples soil for doing experiment C were provided. Then, 5 mL of I-125 was added to the surface of each sub-sample soil in control and case pots. The washes operation of the soil: animal fertilizer mixture was followed with a 10 mL of distilled water during experiment C every day. Like experiment A, pots in experiment C were emptied of distilled water before pouring the next 10 mL distilled water on the soil: fertilizer mixtures. Sub-samples soil columns in experiment C was split into 4 segments with 5 cm in thickness. Each split was homogenized and subjected to counting. Radioactivity level of solid matrices, extracted solution on the soil and elute were determined. The resulting values were corrected. Bulk global measures on untreated sub samples soil with animal fertilizer were performed by automatic gamma counting, but the need of a very sensible and fairly rapid method, able to operate on small soil sub-samples, dictated the decision to measure radioactivity by automatic gamma counter. Moreover, as the direct counting on the sub samples soil gave heavy quenching effects that compromise the reading efficiency. The counting was carried out on liquid extracts.

All solid sub sample soil matrices were subjected to extraction in order to assess the initial and the final radioactivity levels, at the end. The entire volume of each extract was submitted to counting 4 times. The average value of all results was calculated. It was also considered the Standard Radioactivity Level (SRL) for every experiment. The counts were carried out for 60 min for each sample, considering a reading window comprising all the energies (0-2000 keV counting channels). This counting method was chosen considering that it was not so important to identify the radio nuclides involved into the reactions, since the objective was to shift all the isotopes producing significant doses of radiation. The total radioactivity of each experiment with animal fertilizer was calculated as the sum of the count values of elutes belonging to each experiment series, solution extraction and soil. The changes of radio iodine in CPM of each section show the role of animal fertilizer in soil section.

RESULTS AND DISCUSSION

The A- experimental tests were carried out at room temperature, on the 7th, 14th and 21st animal fertilizer was given to the soil. The results of radioactivity were obtained using animal fertilizer for the days mentioned. The total count values of the different days carried out on case sub samples soil used in experiment A were as follows: 1310, 1580 and 1965 CPM, for the untreated contaminated sub samples soil as control sub samples soil were 600, 1455 and 1955 CPM for different days. The difference between case and control sub samples soil for the 7th, 14th and 21st are as follows respectively: 710, 105 and 10 CPM (Table 1). The oscillation in the Table 1 reports the radioactivity values following treatment of animal fertilizer and represents 53 and 31% of the initial radioactivity. The animal fertilizer removing activities of radio iodine in experiment A shows a homogeneous trend, with a decreasing tendency line reported in Table 1. The results being obtained on the 7th, 14th and 21st for experiment A had been washed continuously over this period and decay results were corrected.

Table 2 shows the amount of animal fertilizer in radio iodine absorption of sub samples soil. Radioactivity of first sub-sample was 1668 CPM in which 50% belong to elute, 27% in the mixture

Table 1: Radio iodine in surface layer of soil (CPM)

Sampling time, case and control soil (day)	Difference between case and control soil	Case of soil sub sample (CPM)	Control of soil sub sample (CPM)
7th	710±15	1310±25	600±10
14th	105±50	1560±15	1455±20
21st	10±60	1965±80	1955±20

Table 2: Radio iodine in soil sample and eluted

Sample	No. of animal fertilizer+soil	½ animal fertilizer+1/2 soil (CPM)	3/4 soil+ l/4 animal fertilizer (CPM)	3/4 animal fertilizer 1+1/4 soil (CPM)
Wet soil	200±19	400±2	800±1	300±5
Eluted	16±40	57±2	45±3	58±1

Table 3: Radio iodine in each section of soil

Depth of animal fertilizer (cm)	Case of soil sub sample (CPM)	Control of soil sub sample (CPM)
5	605±10	460±35
10	476±30	415±25
15	420±80	385±60
20	345±20	335±50
Elute	309±40	290±30

of soil and 23% solution extraction in the animal fertilizer. In the second sample, total radioactivity was 2005 CPM in which 21% in the mixture of soil and solution extraction, 56% in elute and 20% in the animal fertilizer. The total radioactivity in the third sample was 1715 CPM. This radioactivity was in mixture of soil and solution extraction as 41%, in elute as 45% and in animal fertilizer as 14%. The pot volume 1/4 animal fertilizer, 3/4 soil of experiment B carried out with different volume of animal fertilizer in different distance, demonstrates greater capacity to remove radionuclide.

The contaminated soil elute without animal fertilizer showed a radioactivity level of 225 CPM. This value, considered as background and used as the Standard Radioactivity Level (SRL), was subtracted from all the gamma counts of liquid matrices.

The results showed that radioactivity of first 5 cm in the case sub samples soil was 605 CPM, while it was 460 CPM in the control sub samples soil (Table 3). The radioactivity of depth 10 cm in the case sub samples was 475 CPM and in control sub samples as 415 CPM. It was 420 CPM and 385 CPM in case and control sub samples for 15 cm depth. Radioactivity in 20 cm depth was 345 CPM in case samples and 335 CPM in the control sub samples soil.

This study has been entirely carried out in the laboratory and is a preliminary step to field tests and only aims to investigate new approaches to the problem of probability polluted areas as removal of the radionuclide from the soil. Thus, some remediation of the polluted soils should be considered before any development for public activities. This could be achieved by mixing with animal fertilizer with soil from areas which are known to have lower radiation background (Al-Jundi and Al-Tarazi, 2008). According to experiment A, the greater the detention time, the Count Per Minute (CPM) will be increased. It means that the uptake by animal fertilizer is increased. At the end of 7th day, there is a difference between case and control soil. The difference between case and control soil are less at end of 14th day, but there is not any change between case and control soils at 21st day. It seems that CPM of case and control soils will be lessened (Table 1). This helps to know the role of animal fertilizer in absorbing the radio iodine in the soil. Although the experimental tests have demonstrated a link between old animal fertilizer and radio iodine in the soil used but other factors would affect on the results of this experiment (Bors and Martens, 1992; Shan and Javandel, 2005). The concentration of the solution extraction used here is high enough for carrying out general tests to study the elution rates. Results from the pot experiment on the contaminated soil gave the following indications. From the observation of Table 2, it may be noted how treatments of the animal fertilizer causes an immediate removal of the radio iodine, while in subsequent treatments the lower peaks of radioactivity are observed and do not correspond to the fertilizer. However, the concentration usually decreases with time as a result of different chemical and transport mechanisms.

As the Table 2, animal fertilizer should be put in soil at certain amount. Less or more animal fertilizer will be affect by the I-125 absorption. It means that if 1/2 of soil and 1/2 of animal fertilizer is used, the absorption of radio iodine is decreased. This position is also correct for 3/4 animal fertilizer and 1/4 soil or none animal fertilizer. The volume of animal fertilizer to sub sample soil used in experiment B showed a higher eluting power; the extraction ratio was in fact higher than 2:1, this ratio that was expected by the use of an amount of distilled water and highlighted a difference of action of the animal fertilizer as the used quantities vary. This rapid decrease in specific activity of elutes almost certainly results from the rapid flushing of the most soluble cationic radioisotopes. A radioactivity level ranging from 1 to 2 times higher upon the treatment of fertilizer, suggests a quadratic or cubic relationship between quantities of reactant and removed radio iodine, proposing the need for a more in-depth study of the phenomenon (Liu and Vongnten, 1988).

Evaluating the total radioactivity eluted by the animal fertilizer, the trend of the tendency line can be noted, which falls gradually as the radionuclide was removed from the soil (Table 2). The above observations lead us to believe that the slope of the tendency curve is a function of the total quantities of animal fertilizer used. The problem of avoiding radio iodine transfer into the water-bearing stratums was investigated using an uncontaminated soil by checking the mobilization of isotope following adding fertilizer to soil if the completing and buffering power of the soil is able to fix radioactive substances in the space of relatively few centimeters (Bugai *et al*., 1996). The results obtained from tests carried out on experiment C which has drawn in Table 3 showed the following indications. The depth of radio iodine absorption is 5 cm in which is better than other depth. Absorption of radio iodine in 5th layer is more than other depth. It shows that certain amount of animal fertilizer with correct demand time at good place of soil affect the uptake of radio iodine. All the elutes of washes with water, corresponding to 200 mm of rain, showed a radioactivity always similar to the value eluted from the non contaminated soil. Therefore, the entire radioactivity initially added in was held back in the mass of soil. The measured of radioactivity levels of the six fractions of soil obtained from the sectioning of the column show that the radio iodine was mobilized only several centimeters.

The acquired experimental data demonstrate that radionuclide found in contaminated soil can be removed by an animal feces commonly used in agricultural as fertilizer. The total amount of radioisotopes removed, demanding time of animal fertilizer and depth of used animal fertilizer were determined. The procedure can be further improved and optimized for large scale field use (Timms *et al*., 2004). Iodine-125 (a surrogate for (129) I) tended to accumulate at the boundary between the anoxic conditions at the base of the column and the oxic conditions above, due to its redox-dependent sorption behaviour (Ashworth and Shaw, 2006).

The reduction of radioactivity to levels matching the international recommended standards for soil and the depth to which radionuclide have to be accumulated, certainly define the cost of a field application of this methodology and have to be more investigated in further studies. This method correctly predicts the observed variation of dose-rate with elevation above the soil surface and it is shown how this method can be used to predict the reduction in surface dose-rate after remediation such as adding animal fertilizer have taken place.

ACKNOWLEDGMENT

We are grateful to Dr. Dabiri for giving permission to apply Automatic Gamma Counter and Mr. Miracki for doing analysis.

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