Development of Composition and Technology of Additives Production for Rehabilitation of Soils Contaminated by Radionuclides and Assessment of their Application Efficiency

Project # 3189

Alexei Konoplev

Center for Environmental Chemistry

Research and Production Association "Typhoon" (Obninsk, Russia),

Leonid Moskalchuk

SSO Joint Institute for Power and Nuclear Research – "Sosny", NAS of Belarus

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The legacy of the Chernobyl accident (26 April 1986) can be used for developing environmental remediation and restoration technologies

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As a result of the Chernobyl accident, about
85×10^{15} Bq of ^{137}Cs,
54×10^{15} Bq of ^{134}Cs,
1760×10^{15} Bq of ^{131}I,
10×10^{15} Bq of ^{90}Sr and
0.07×10^{15} Bq of $^{239,240}Pu$
were released into the atmosphere and transported over long distances

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As a consequence of the accident, fallout was deposited over most of Europe; however, the largest areas of contamination were in Belarus, the Russian Federation and Ukraine

TABLE 4.1. AREAS IN EUROPE CONTAMINATED BY CHERNOBYL FALLOUT IN 1986 [4.1, 4.4]

	Area with ^{137}Cs deposition density range (km^2)			
	37–185 kBq/m^2	185–555 kBq/m^2	555–1480 kBq/m^2	>1480 kBq/m^2
Russian Federation	49 800	5 700	2100	300
Belarus	29 900	10 200	4200	2200
Ukraine	37 200	3 200	900	600
Sweden	12 000	—	—	—
Finland	11 500	—	—	—
Austria	8 600	—	—	—
Norway	5 200	—	—	—
Bulgaria	4 800	—	—	—
Switzerland	1 300	—	—	—
Greece	1 200	—	—	—
Slovenia	300	—	—	—
Italy	300	—	—	—
Republic of Moldova	60	—	—	—

CHERNOBYL EXPERIENCE

Methods of rehabilitation of radioactive contaminated soils

The most effective methods to reduce ^{137}Cs and ^{90}Sr soil-plant transfer

Application of potassium fertilizer (mainly KCl) (Belarus, Russia)
→ 2-20 times reduction of ^{137}Cs soil-plant transfer (*depending on soil type and KCl doze*)

Liming (Belarus, Russia & Ukraine)
→ Application of lime, dolomite powder, limestone, tripoli earth and other ameliorants leads to a 2-4 times reduction of ^{90}Sr and a 1,5-3 times reduction of ^{137}Cs soil-plant transfer

Application of organic fertilizer (manure, peat and others) (Belarus, Russia & Ukraine)
→ 1,5-3 times reduction of ^{137}Cs soil-plant transfer (*the highest effect on soddy-podzolic sandy soils*) and a 2-4 times of ^{90}Sr (*the highest effect under combined application of lime and organic fertilizer*)

Application of sapropels (Belarus & Ukraine)
→ Up to 6 times reduction of ^{137}Cs soil-plant transfer *(on soddy-podzolic sandy soils mainly)* and depending on soil type and the dozes of sapropel applied

Two major environmental problems to be solved

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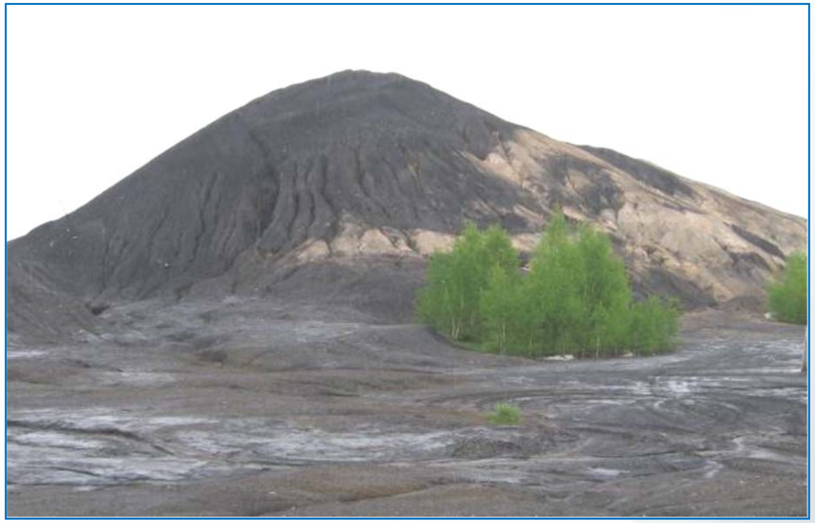
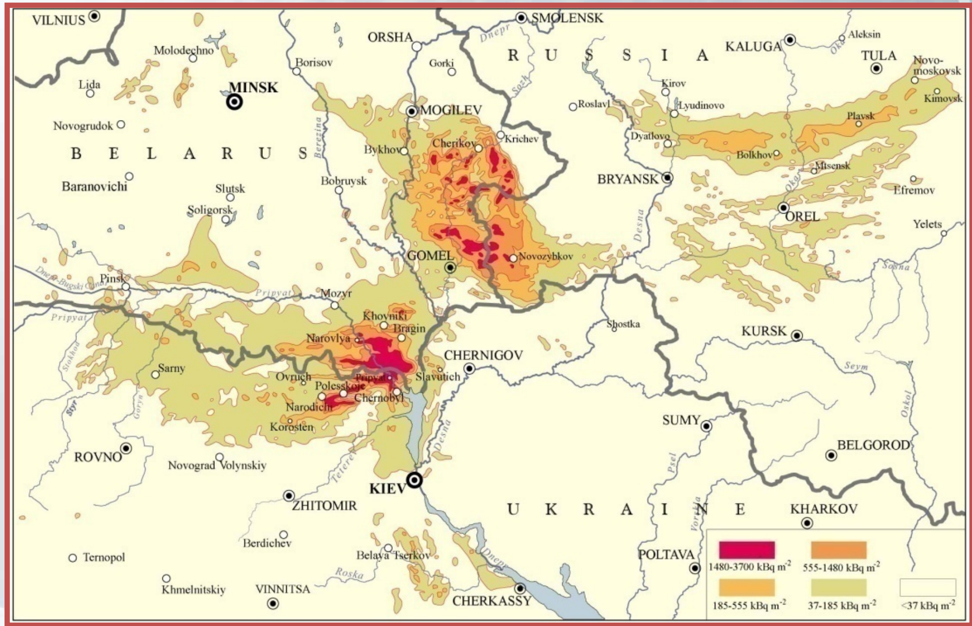
➢ Utilization of industrial wastes



Waste generation (million tons/year):
1. Clay-salt slimes = 2.0
2. Hydrolized lignin = 0.24
3. Phosphogypsum = 0.42



➢ Rehabilitation of territories contaminated by ^{137}Cs and ^{90}Sr as a result of the Chernobyl Accident

Objectives of Project 3189

- To develop efficient and ecologically safe additives based on industrial waste and natural raw materials for remediation of soils contaminated by ^{137}Cs and ^{90}Sr;
- To develop optimum compositions of additives ;
- To develop methods and models for prediction of efficiency of such countermeasures as part of remediation.

Goals of Project 3189

- Determination of the main parameters of soils, sorbents and additives (composition of soil solution, sorption properties of the solid phase, kinetic and equilibrium parameters of radionuclides fixation);
- Development of the methods to estimate the key parameters of soil-additives mixtures using properties and ratio of the components;
- Assessing the efficiency of additives application for typical soils of contaminated areas in Belarus and Russia and developing recommendations for their use.

Goal of the research

Sapropel are fresh water lakes bottom sediments formed from fossil plant and animal bodies, mineral substances of biochemical origin and derived mineral components.

Hydrolyzed lignin is a wood processing product, containing organic matter (90-95%), and a large-capacity waste of chemical industry.

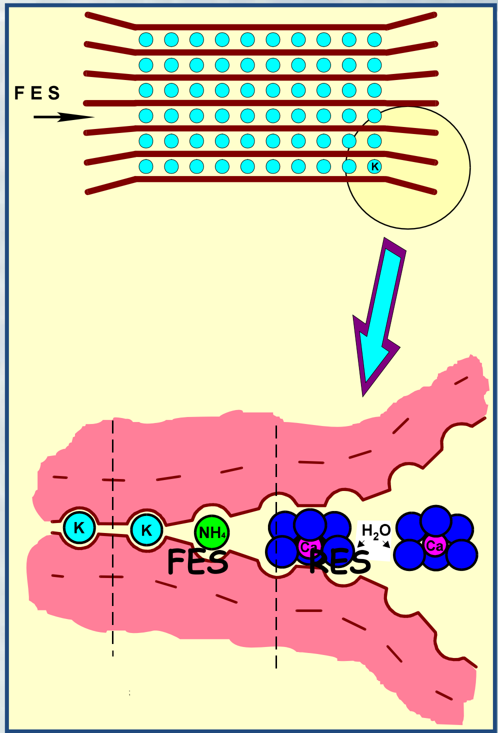
Clay-salt slimes are the waste of potassium industry produced as a result of sylvinite ore reprocessing at the factories of the “Belaruskali” Plant.

additives were prepared on their basis

Selective sorption and fixation of radiocesium

High retention of radiocesium in soils is caused by two main processes:

- selective reversible sorption on illitic clay minerals;
- fixation



$$FES-M+{}^{137}Cs^{+}\xleftrightarrow{K_c^{FES}(Cs/M)}FES-{}^{137}Cs+M^{+}$$

$$RIP^{ex}(M)=K_d^{ex}(Cs)\times m_M=K_c^{FES}(Cs/M)\times[FES]$$

Distribution coefficients K_d of radiocesium and radiostrontium in soils

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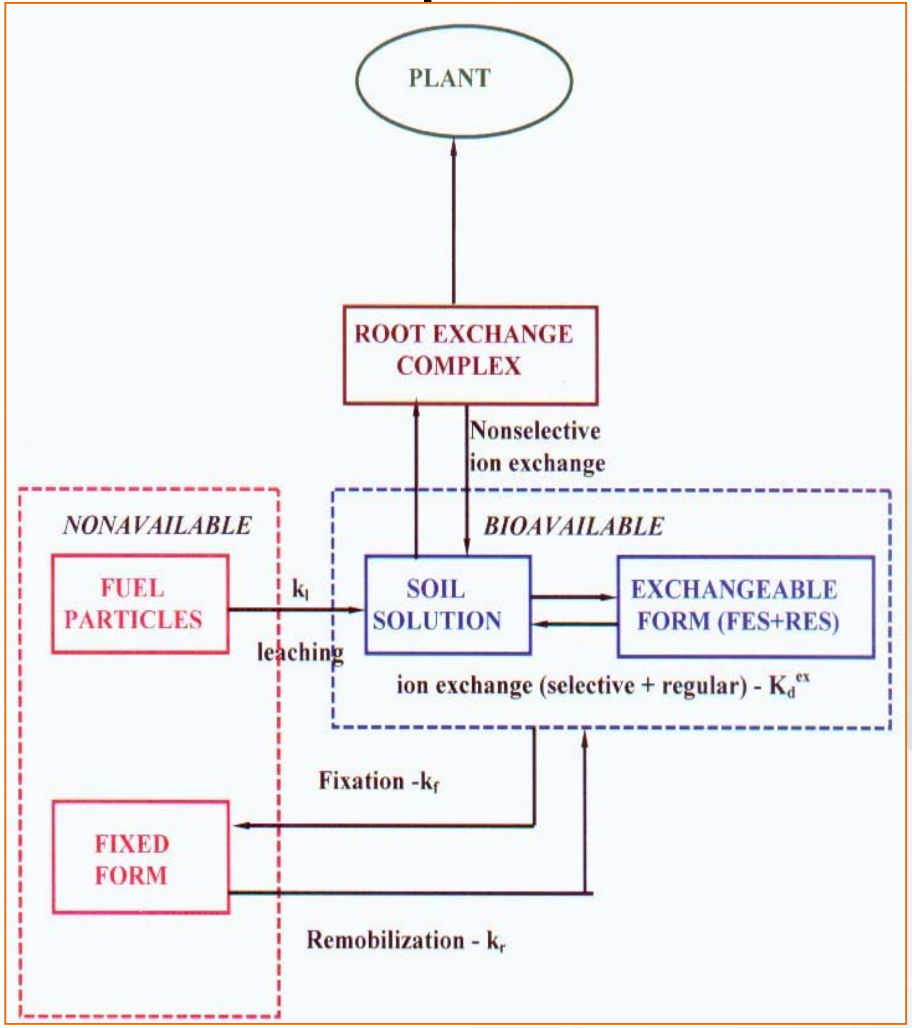
$$K_d(^{137}Cs) = \frac{RIP(K)}{([K^+]_W + K_c(N/K)[NH_4^{\ +}]_W)}$$

Clay-salt slimes increase RIP of soils.
(RIP – Radiocesium Interception Potential)

$$K_d(^{90}Sr) = \frac{CEC}{[Ca]_w + [Mg]_w}$$

Organic sapropels and hydrolized lignin increase CEC of soils.
(CEC –Cation Exchange Capacity)

Conceptual model of radionuclide soil-plant transfer



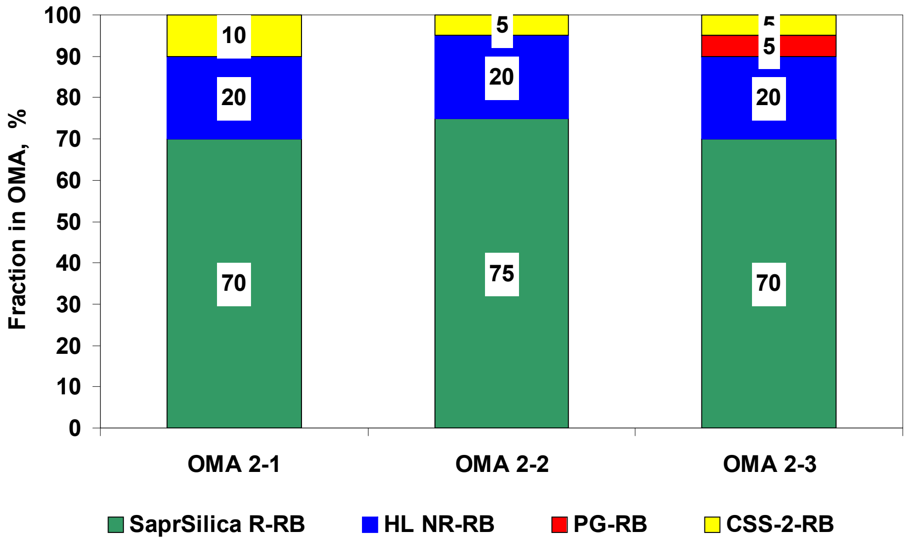
- Radiocesium Bioavailability ~ 1/RIP
- Radiostrontium bioavailability ~ 1/CEC

Chemical characteristics of source components and soils

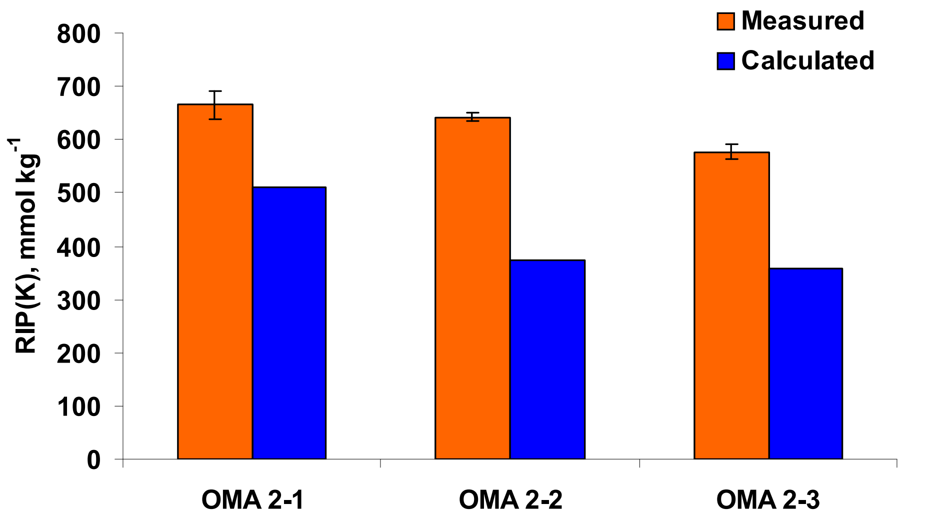
Sample code	C_{org}, %	pH_{KCl}	CEC, $cmol_c\ kg^{-1}$	RIP(K), $mmol\ kg^{-1}$
CSS-1-RB	1,50±0,12	7,7	14.2±1.0	6343±1120
CSS-2-RB	1,96±0,29	7,3	162.±1.0	3041±334
PG-RB	0,05±0,01	4,9	-	17.6±1.6
HL AR-RB	34,6±1,7	3,0	100±3	7,2±0,8
HL NR-RB	47,8±2,4	6,3	64.3±0.8	23,3±1,8
HL DR-RB	39,8±1,9	2,8	72.4±2.0	32,2±1,2
SaprSilica R-RB	14,3±0,6	4,7	69.6±5.0	596,7±0,3
SPS-1- RB	0,30±0,05	4,2	8.7±1.6	35.1 ±1.2
SPS-RF	0,62±0,03	3,6	5.7±0.3	440 ±70
HGS-RF	8,6±0,6	3,2	33.9±0.4	1200 ±70

RIP in ternary and quaternary OMAs

Composition of OMAs (organic mineral additives)

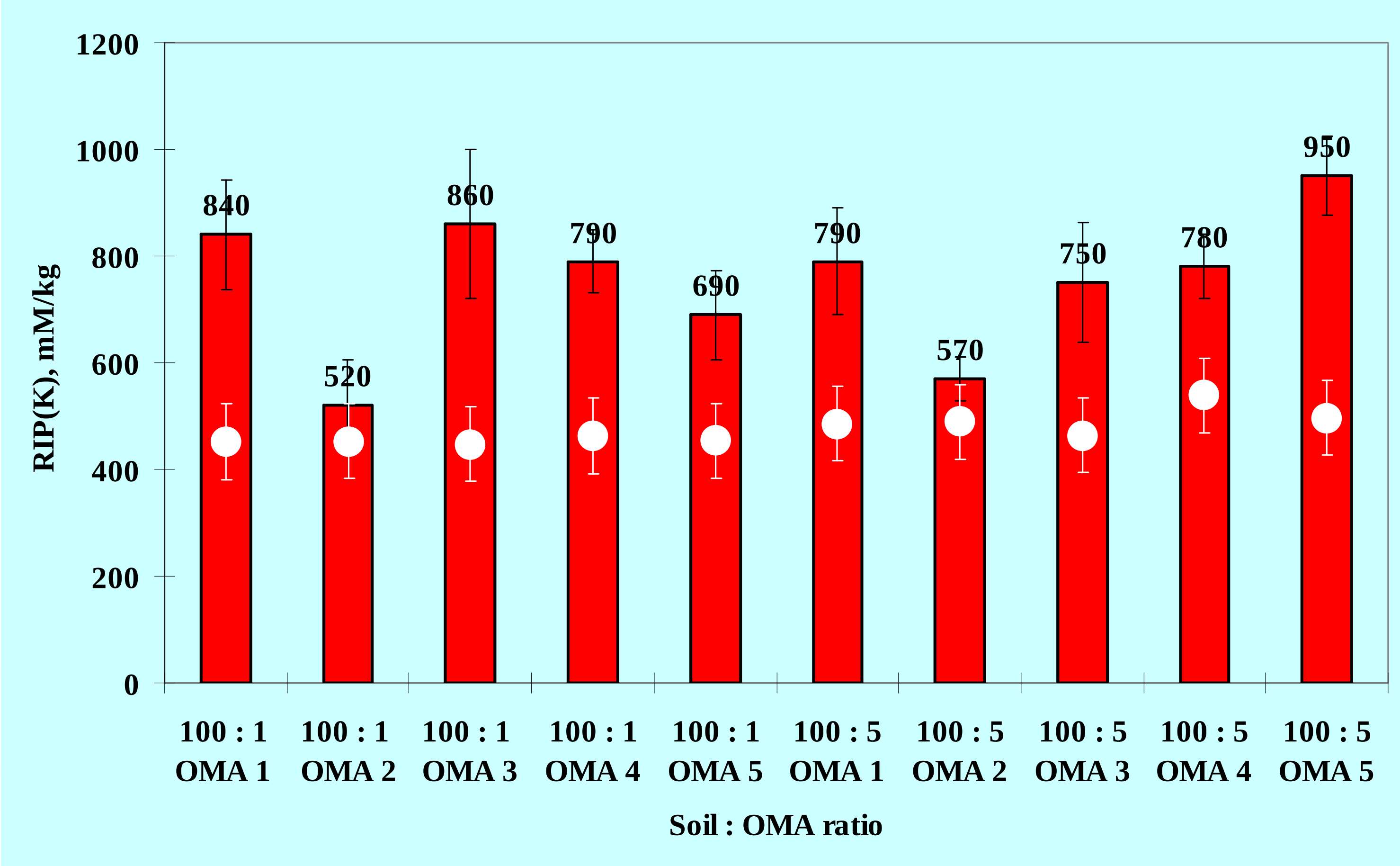


RIP(K)



$$\frac{RIP(K)_{Exper}}{RIP(K)_{Calcul}} = 1.3 - 1.7$$

RIP(K) of SPS-RF enriched with OMAs



Assessment of the efficiency of OMA application

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Soil samples	K_d, ml/g	Efficiency (for ^{90}Sr)	RIP(K), mmol/kg	Efficiency (for ^{137}Cs)
Soddy-podzolic sandy soil (SPS-1)	8,6		35,1	
SPS-1 + 2% OMA 1	14,3	1,7	120,1	**3,4**
SPS-1 + 4% OMA 1	25,5	2,9	145,3	**4,1**
SPS-1 + 8% OMA 1	31,8	3,7	178,1	**5,1**
SPS-1 + 2% OMA 2	18,3	**2,1**	-	-
SPS-1 + 4% OMA 2	31,6	**3,7**	-	-
SPS-1 + 8% OMA 2	35,2	**4,1**	-	-

Taking into account that a specific density of a soils sample (SPS-1) is roughly 1300 kg/m^3, the rate of the OMA application on soddy-podzolic soils is 50-100 t/ha and for a soil sample (SPS-3) with a specific density of 1500 kg/m^3, is 60-120 t/ha respectively.

Efficiency of organo-mineral additives

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- Mixing of organic and mineral components to produce OMAs leads results in an increase of up to 2 times of the sorption of Cs
- Application of OMAs can reduce radiocesium soil-plant transfer up to 10 times as compared to original soils;
- The reduction of radiostrontium transfer to plants in poor sandy soils can be as high as 500-600 times.
- Application of these sorbents is therefore most effective on poor sandy soils.

Conclusions

- Organic source materials (sapropels and hydrolyzed lignin) and phosphogypsum are promising components of additives for reduction of radiostrontium uptake by plants;
- Additives containing clay-salt slimes are most promising to reduce radiocesium and radiostrontium uptake by plants;

Conclusions

- The optimum proportions of the components in the organo-mineral additives for the concurrent 137Cs and 90Sr sorption and fixation are 75-70% - sapropel, 20% - hydrolized lignin, 5-10% - clay-salt slimes and phosphogypsum – 5%;
- For sod podzolic soils the recommended dose of additives application is 2-4 %;
- OMA application rate is 50-120 t/hectare depending on soil type.

Proposals for Fukushima site remediation

- Characterization of Japanese soils on Fukushima site in terms of radiocesium mobility and soil-plant transfer;
- Production of additives on the basis of natural raw materials and industrial waste (CIS and Japan origin) and their application for rehabilitation of soils in Fukushima site;
- Assessment of additives efficiency to reduce radionuclide mobility and soil-plant transfer at soils on the Fukushima site.

THANK YOU VERY MUCH FOR YOUR ATTENTION!

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QUESTIONS?

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