RADIATION MONITORING USING IMAGING PLATE TECHNOLOGY: A CASE STUDY OF LEAVES AFFECTED BY THE CHERNOBYL NUCLEAR POWER PLANT AND JCO CRITICALITY ACCIDENTS

by

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This paper describes the use of a photostimulable phosphor screen imaging technique to detect radioactive contamination in the leaves of wormwood (*Artemisia vulgaris L.*) and fern (*Dryopteris filix-max* CL. Schoff) plants affected by the Chernobyl nuclear power plant accident. The imaging plate technology is well known for many striking performances in two-dimensional radiation detection. Since imaging plate comprises an integrated detection system, it has been extensively applied to surface contamination distribution studies. In this study, plant samples were collected from high- and low-contaminated areas of Ukraine and Belarus, which were affected due to the Chernobyl accident and exposed to imaging technique. Samples from the highly contaminated areas revealed the highest photo-stimulated luminescence on the imaging plate. Moreover, the radionuclides detected in the leaves by gamma and beta ray spectroscopy were ^{137}Cs and ^{90}Sr, respectively. Additionally, in order to assess contamination, a comparison was also made with leaves of plants affected during the JCO criticality accident in Japan. Based on the results obtained, the importance of imaging plate technology in environmental radiation monitoring has been suggested.

Key words: CNPP accident, JCO criticality accident, imaging plate technique

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INTRODUCTION

The worst ever accident in the history of nuclear power plants occurred on April 26th, 1986, at Chernobyl, Ukraine, formerly a part of the USSR. It is referred to as the "Chernobyl nuclear power plant (CNPP) accident" [1, 2]. The accident followed a safety experiment in which the plant was operated outside of its designed parameters, at very low power and unfavorable cooling conditions. Radioactive particles that swept across the Ukraine, Belarus, and the western portion of Russia, eventually spread across Europe and the whole of the northern hemisphere. The CNPP accident exposed most of the population of the northern hemisphere to various degrees of radiation. Due to this, after 1986, the public perception of a nuclear risk was changed to a great extent. Other than the obvious and much studied health impact, the agricultural and environmental impacts, relatively unstudied, still pose a serious problem. At present, 20 years after the CNPP accident, contamination is still a major problem in Chernobyl and the surrounding areas originally included in the exclusion zone. Cae-

sium-137 (^{137}Cs, gamma- and beta-emitter), which has a half-life of 30.1 years, is the most important radionuclide from Chernobyl's catastrophic explosion, and is present at high concentrations in the 0-5 cm soil layer. Another damaging residual radionuclide, strontium-90 (^{90}Sr, beta-emitter), which has a half-life of 29.1 years, is also present in the soil layer. Both of these radionuclides pose a potential threat to plant life in the region.

The CNPP accident has been cited as a level 7 according to IAEA, whereas the JCO criticality accident has been evaluated as a level 4 incident. On September 30, 1999, a criticality accident (neutron accident and release) occurred in the precipitation tank at the uranium conversion building of the JCO Company Ltd. in Tokaimura, Ibaraki Prefecture, Japan [3]. A solution of enriched uranium (18.8% ^{235}U by mass) in an amount reportedly several times higher than the specified mass limit, had been poured directly into a precipitation tank, bypassing a dissolution tank and buffer column intended to avoid criticality. Although no serious contamination impact to the environment, neutron emission and fission products, including trace amounts of noble gases and gaseous iodine, escaped from the building [4, 5].

In general, when we monitor the effect of radioactivity released into the environment, the specific energy intensity of the radioactive nuclide contained in soil or plant is usually measured by gamma ray detection equipment or beta ray spectrometers. In order to explain this to the general public in a manner as simple as possible, specialized knowledge is necessary, and therein lies the aim/reason behind this study. By adopting the imaging plate (IP) technique, we hope to explain in simple, strictly visual terms, the presence of radioactivity in our surroundings, *e. g.* in/on plants. Additionally, this study examines the possibility of whether the IP technique can be applied to the decontamination of environmental radioactivity. We have collected plant samples from the Chernobyl exclusion zone (September, 2000/2001), as well as JCO site (Tokaimura), one week later and 27 days after the accident. The results obtained during this study reveal the usefulness of the IP technique in assessing the presence or absence (and quantification, where the contaminating radionuclides are known, as in the case of Chernobyl) of radioactivity in our immediate surroundings.

EXPERIMENT

Sampling and exposure to IP

Plant leaves were prepared for exposure to an IP (BAS-MS2025, Fuji Photo Film Co., Ltd., To-

kyo, Japan) by pressing the leaves between pages of a notebook and subsequent drying in clean air. The exposure was carried out for 12, 24, or 72 h in a shield box, as mentioned in figure legends. The IP plate was analyzed using an IR Bio-Imaging Analyzer (BAS-2000, Fuji Photo Film Co., Ltd.). Photo-stimulated luminescence (PSL) intensity was also calculated and presented whenever necessary.

Gamma and beta ray measurements

Gamma ray measurement was performed before the processing of leaf samples for pressing and drying, using a Ge-detector (Ortec GMS-30185, 1.8 keV half bandwidth), coupled to a multi-channel analyzer (Seiko EG & G 7800). During measurement, one channel was set to 0.5 keV, so that measurements up to 2000 keV were possible. The spectrum of beta ray emission was measured using dried leaf samples. Approximately 0.4 g of dried leaves was put in an aluminum dish (25 mm in diameter) and placed inside a low background beta ray spectrometer (Fuji Electric Co., Pico-beta NPB00) coupled to a multi-channel analyzer (Laboratory Equipment Co., MCA/AT).

RESULTS AND DISCUSSION

Chernobyl (CNPP) samples

Sample collection points were at Gomel, Belarus, Masany ecology research center (10 km from the CNPP) in Ukraine, the three CNPP circumference areas (Kopachi, Prypyat, and Red Forest) and at the contrast (control) areas in Kiev and Kherson city off the black sea coast in Ukraine. Fern, wormwood, thistle, raspberry, mulberry, hop, pine, and moss plants were collected, and an index plant was selected based on the clarity of leaf structure, size and availability in all sampling areas mentioned above. Consequently, two kinds of index plants, namely wormwood and fern were selected. Wormwood is widely distributed from the subarctic to the subtropical zones, whereas the ferns are predominantly present in the dry sandy soils in and around Chernobyl.

The radioactive nuclide, which existed in these plants, was measured by a Ge-detector and a low background beta ray spectrometer. These radionuclides were identified as ^{137}Cs (half-life 30.07 year) and ^{90}Sr (half-life 28.78 year). This was compared with the same contaminated soil samples from the area and found to be 67.9   0.024 Bq and 3.18 0.33Bq for ^{137}Cs and ^{90}Sr, respectively. In this study, we have selected indicator plants such as wormwood and fern to check the IP technique. The influence of

^{40}K, which exists as a naturally occurring background radioactive source, was distinguished by checking the image in real time (data not shown). In order to investigate the distribution state of ^{137}Cs and ^{90}Sr, ^{137}Cs was separated using various shield materials, for example, since it was found that ^{90}Sr had reached radiation parallel with ^{90}Y, a radiation shield was used for ^{90}Y. Results presented in fig. 1 show ferns from Masany (both decontaminated and contaminated areas) exposed to the IP plate. It can be clearly seen that the leaves of the ferns from the contaminated areas show very high radioactivity, particularly in the veins (dark red color), strongly suggesting that these radionuclides had moved into the plants from the soil. In total contrast, control plants from the decontaminated area did not reveal any radioactivity. These initial data indicate the usefulness of IP in visually determining radioactivity.
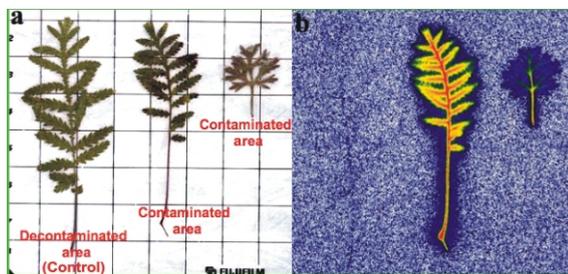

Figure 1. IP analysis of fern leaves
(a) photograph of fern before exposure to IP
(b) photograpgh after exposure to IP (exposure time was 24 h)

We examined the leaves of both wormwood and ferns from various locations around Chernobyl (figs. 2 and 3). The leaves of wormwood plants from Kopachi, Prypiat and Masany show presence of radioactivity. On the other hand, the ferns were strongly radioactive in comparison with the wormwood plants, and the leaves from the Red Forest, Kopachi and Masany showed high levels of contamination. Surprisingly, the leaves from the decontaminated area of Prypiat also showed radioactive contamination, though not at levels detected in the still-contaminated areas. These results, while clearly showing the efficiency of the IP technique, disturbingly reveal that some of the decontaminated areas have a certain amount of radio-contamination still left in the plants, which is most likely to originate from the soil. This makes us very anxious about in-the-body contamination, *i. e*. the shifting of the radioactive nuclide from contaminated soil to agricultural products.

Tokaimura (JCO) samples

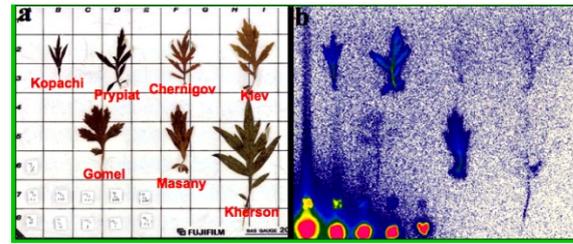As a comparison, and in order to test the IP technique, we conducted certain experiments on

Figure 2. IP analysis of wormwood leaves
(a) photograph before exposure to IP
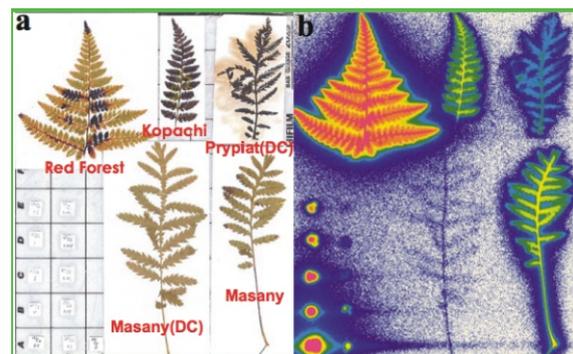**(b) photograph after exposure to IP (exposure time was 72 h)**

Figure 3. IP analysis of fern leaves from the Chernobyl exclusion zone
(a) photograph of wormwood before exposure to IP (DC, decontaminated area)
(b) photograph after exposure to IP (exposure time was 12 h)

plants from the Tokaimura accident site in Japan (JCO criticality accident site). At first, in order to select an IP index plant around the JCO site, ivy, wormwood, mulberry and fern plants were collected and examined. The sampling points are shown in fig. 4 (S1-S14). Consequently, wormwood, whose detection sensitivity of the radioactivity by IP was good (fig. 5), was adopted as the index plant. This was based on its presence at almost every point where it was easy to form a plant community, and a suitable height from the ground in the range of 10 to 30 cm. This result further provided support to the use of the IP technique for measuring radioactivity in plants. However, as seen in fig. 5, S7 and S8 samples showed higher radioactivity than S9, which is closest to the precipitation tank (conversion building), fig. 6). Hence, these initial results revealed that it was not advisable to depend on the distances of the wormwood plants from the precipitation tank, the main generating source of neutrons. Moreover, in order to investigate the contribution of surface contamination by the radioactive noble gas as another cause of the contamination, the distance of a sampling point was measured from the surface of a wall near the exhaust duct. As a result,
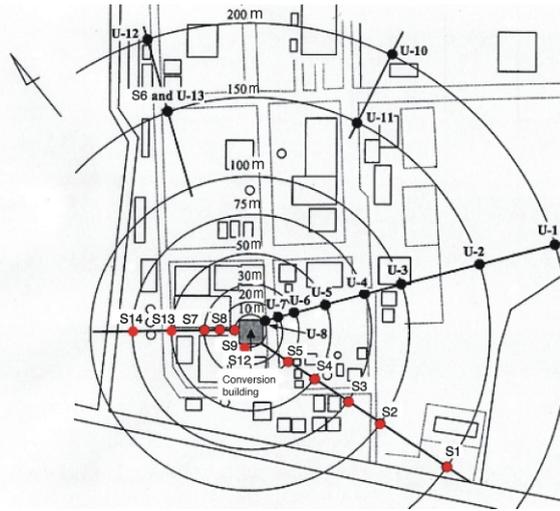
Figure 4. Sampling point of the Tokaimura JCO site Distance from the uranium conversion building was 2.1 m (S9), 23.8 m (S8), 37.1 m (S7), 49.1 m (S13), and 63.5 m (S14)

i. e. the radioactive noble gas that leaked from the exhaust duct.

In order to prove this, an identification of the radionuclide was attempted by using a Ge-detector and a low background beta ray spectrometer. Radionuclides such as ^{140}Ba (0.537 MeV), ^{140}La (1.349 MeV), ^{137}Cs (0.662 MeV), ^{131}I (0.365 MeV) and ^{133}I (0.529 MeV) were detected by a Ge-detector (fig. 9a). According to the report of Ban-nai *et al*. [5], the level of ^{131}I was about 120 Bq/kg at the JCO site. This was less than the index (pine leaf: 2000 Bq/kg) ingestion restrictions imposed on food and beverages for ^{131}I. As an identification of the -ray nuclide by low background beta ray spectrometer, the existence of ^{89}Sr (1.497 MeV) from the fission product origin ^{89}Kr was found in the leaves [6] (fig. 9b-d) and confirmed by purifying Sr from the S9 leaf sample (fig. 9e). The detection of ^{89}Sr, which is a beta ray emitter, is a new observation, clarified due to the emission into the environment. These results were also in good accord with the PSL intensity of IP (fig. 9).
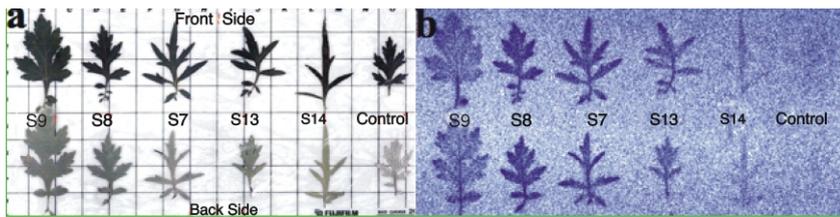

Figure 5. IP image of the collected wormwood leaves (a) photograph before exposure of wormwood to IP (b) photograph after exposure to IP (exposure time was 72 h)
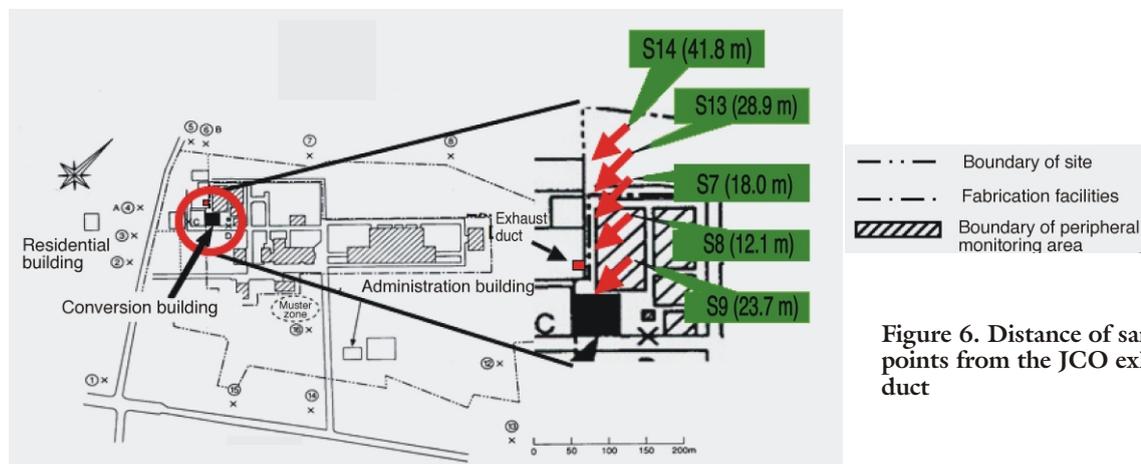

Figure 6. Distance of sampling points from the JCO exhaust duct

S9 (23.7 m), S8 (12.1 m), S7 (18.0 m), S13 (28.9m), and S14 (41.8 m) sampling points were selected, which proved to be in good agreement with the photo stimulated luminescence (PSL) density of IP (fig. 7). We also checked the PSL density of the same samples (S9-S14) in a time-dependent manner. It can be clearly seen that the PSL density decreased with time (day), fig. 8. Therefore, it can be suggested that the PSL density was strongly influenced by the neutron-induced fission product,

We further examined whether other radio--nuclides, like the beta ray emitters ^{32}P, ^{35}S, and ^{45}Ca, neutron-induced, were present in the leaves. We therefore selected the S9 leaf sample, and consequently, ^{32}P, which is the neutron-induced radionuclide of phosphorus, identifying a sulfurous origin (fig. 9f). In general, based on all these results, it can be suggested that, in the JCO criticality accident, the influence of fission products was more pronounced than the effect of the
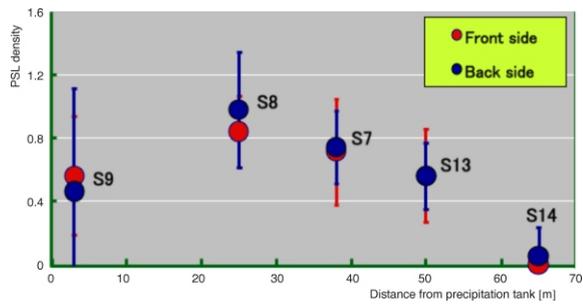
Figure 7. Relation between sampling point and the photo-stimulated luminescence (PSL) density by IP
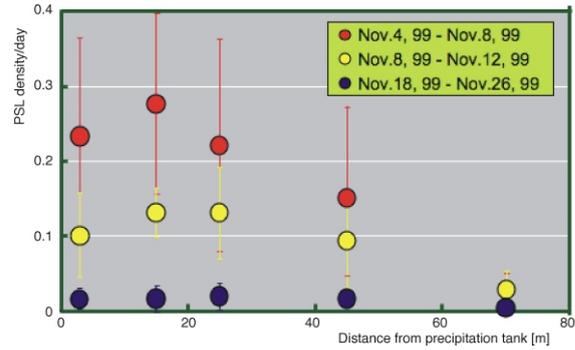

Figure 8. Decrease of radioactivity in collected wormwood samples during the progress of time (days) after the JCO criticality accident
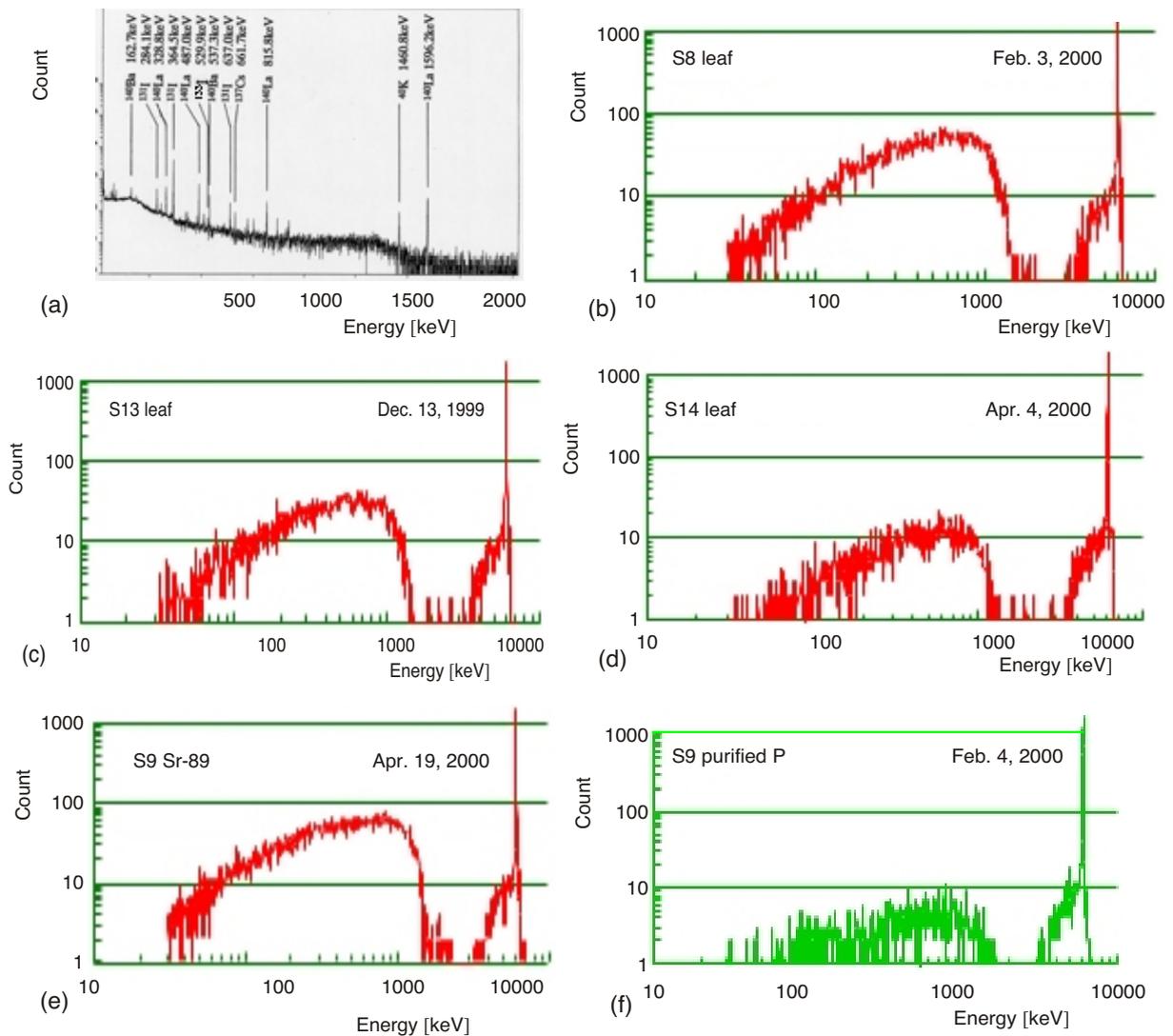

Figure 9. Gamma ray and beta ray spectrum from wormwood collected within the JCO site
(a) gamma ray spectrum from wormwood,
(b-d) beta ray spectrum from wormwood leaves,
(e-f) identification of the beta ray emission nuclide by the beta ray spectrometer after the purification of Sr (e), and of P (f)

neutrons themselves, which may turn out to be data of great interest to researchers.

CONCLUSION

Sample selection of plants is very simple at any accident site. It does not require any special treatments or sample preparation. Results can be obtained within a few hours (0.2~1 h), ascertaining the nature of the contamination due to radiation. For example, Chernobyl exclusion zone samples can be detected within 10 minutes. Measurement of environmental radioactivity using an IP can serve as an inexpensive and rapid method/technique to check surface contamination in case of an emergency. Moreover, it can also be used for monitoring the long-term effects caused by a nuclear accident.

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РАДИЈАЦИОНИ МОНИТОРИНГ ПОСРЕДСТВОМ ФОТО ПЛОЧЕ СЛУЧАЈ ПРОУЧАВАЊА ЛИШЋА ПОДВРГНУТОГ ЧЕРНОБИЉСКОМ АКЦИДЕНТУ И JSO АКЦИДЕНТУ КРИТИЧНОСТИ

Описано је коришћење технике сликања фотостимулисаним фосфорним екраном за детекцију радиоактивне контаминације лишћа биљака пелена (*Artemisia vulgaris L.*) и папрати (*Dryopteris filix-max* CL Schoff) подвргнутих акциденту чернобиљске нуклеарне електране. Технологија фото плоче добро је знана у дводимензионалној радијационој детекцији по многим упадљивим особинама. Пошто укључује интегрисани систем за детекцију, широко је примењивана за проучавање расподеле површинске контаминације. У овом раду проучавају се узорци биљака, сакупљених са подручја високе и ниске контаминације Украјине и Белорусије, које су претрпеле чернобиљски акцидент и потом обрађене технологијом фото плоче. Примерци са високо контаминираних области испољили су најјачу фотостимулисану луминисценцију у поступку фото плоче. У лишћу су гама и бета спектрометријом детектовани радионуклиди ^{137}Cs и ^{90}Sr. У циљу оцене контаминације, извршено је поређење са лишћем биљака оболелог током JSO акцидента критичности у Јапану. На основу добијених резултата, указано је на значај технологије фото плоче за радијациони мониторинг околине.

Кључне речи: чернобиљски акциденш, JCO акциденш кришичносши, шехника фошо плоче