



## Indication of the radioactive fallout in Riyadh, Saudi Arabia following the Fukushima nuclear accident



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### ABSTRACT

On March 2011, a severe damage has occurred to Fukushima Dai-ichi nuclear reactor complex in Japan following the huge earthquake and the resulting Tsunami. Consequently, vast amounts of radioactive fallout were released into the atmosphere and contaminated the environment in Japan. Soon after the accident, traces of anthropogenic radionuclides were detected in environmental samples collected in many parts in the northern hemisphere even very far away from Japan creating a global concern. There is no information about radioactive contamination in the Arabian Peninsula caused by the Japanese Fukushima nuclear accident. The first evidence of Fukushima radioactive fallout in Riyadh (24° 43' N, 46° 38' E), Saudi Arabia has been confirmed in April 8, 2011. The airborne fission products  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  were measured in air samples. The radionuclide concentrations were determined by identifying their characteristic gamma rays using a germanium detector. Their activity concentrations were studied as a function of time over a period of 20 days at the end of which they had mostly fallen below our limit of detection. The maximum activity concentration of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in air of, respectively,  $323.7 \pm 18.5$ ,  $17.2 \pm 1.0$  and  $26.0 \pm 1.8 \mu\text{Bq m}^{-3}$  were observed on April 10–11, 2011. The  $^{131}\text{I}/^{137}\text{Cs}$  and  $^{134}\text{Cs}/^{137}\text{Cs}$  activity ratio values in air were presented and discussed. Finally, the effective doses to the public of Riyadh city from inhalation of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  due to contribution from Fukushima incident was found far below levels of concern.

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### 1. Introduction

The Fukushima nuclear disaster, resulted from the 9.0 magnitude earthquake and the subsequent massive tsunami that hit off the Pacific coast of Japan on March 2011, was followed by radioactive releases of anthropogenic radionuclides into the atmosphere (Momoshima et al., 2012; Stohl et al., 2012; Butler, 2011; Chino et al., 2011). The large-scale radiation leak in Fukushima was considered the biggest source of global radioactive contamination after the catastrophe of Chernobyl in 1986. Both incidents were classified as level 7 in the International Nuclear and Radiological Event Scale (INES) (IAEA, 2011). Among the various radionuclides released in large quantities due to Fukushima accident, iodine isotope ( $^{131}\text{I}$ ) and cesium isotopes ( $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ) are of major interest for health impact assessments. In particular, the radioactive emissions of  $^{137}\text{Cs}$  and  $^{131}\text{I}$  into the atmosphere due to Fukushima accident were estimated to be 8.2 and 150 PBq, respectively,

according to the IAEA June 2012 Fukushima Daiichi Status report (IAEA, 2012). However, the estimated amounts of  $^{131}\text{I}$  and  $^{137}\text{Cs}$  emitted to the atmosphere due to Fukushima incident were approximated to 10% and 20%, respectively, of that released by Chernobyl accident (UNSCEAR, 2013). Most of these releases were deposited in the ocean and over Fukushima prefecture (National Research Council, 2014). Nevertheless, radiation released from the crippled reactors was started diffusing with air movement, transported long distances and ultimately fallout occurred over the northern hemisphere (Thakur et al., 2013) as detected far away in North America (Diaz Leon et al., 2011; Norman et al., 2011; Bowyer et al., 2011), Europe (Ioannidou et al., 2012; Kirchner et al., 2012; Kritidis et al., 2012; Lujanienė et al., 2012; Perrot et al., 2012; Pham et al., 2012; Piñero García and Ferro García, 2012; Manolopoulou et al., 2011; Pittauerová et al., 2011) and Asia (Enyuan et al., 2014; Kim et al., 2012; Long et al., 2012; Bolsunovskiy and Dementyev, 2011).

These observations of fallout of short and long lived anthropogenic radionuclides, including  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , from Fukushima promoted conducting measurements and subsequent dose

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assessments across the globe. For example, the radioactive fallout has been detected in environmental samples collected during the period of April–May 2011 in the USA and Greece which are at a far distant from Japan (Norman et al., 2011; Manolopoulou et al., 2011). High levels of  $^{131}\text{I}$  and  $^{137}\text{Cs}/^{134}\text{Cs}$  and  $^{131}\text{I}/^{137}\text{Cs}$  ratios were observed in water samples collected in Russia and Greece (Norman et al., 2011; Manolopoulou et al., 2011). Samples of water, soil, grass and cow-milk collected from Northwest Germany during the period from March to May 2011, revealed the presence of traces of  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  (Pittauerová et al., 2011). Contaminated air masses from Fukushima were also detected in air filter membrane in Russia by (Bolsunovsky and Dementyev, 2011). In addition, relatively high releases of radiation from Fukushima have been observed in states of the United State of America (Diaz Leon et al., 2011; Pittauerová et al., 2011; Manolopoulou et al., 2011; Norman et al., 2011).

All of these reasons together with the global wind circulations, suggesting high-velocity movement of the radioactive contaminants from the Fukushima Nuclear Accident to other parts of the world. To date no study has been dedicated to examine the airborne fission products from Fukushima Dai-ichi nuclear accident in the Arabian Peninsula. Therefore, we conducted this study with the aim to measure the radiological impact of Fukushima accident to air samples collected at Riyadh, Saudi Arabia and determine the dose contributions from Fukushima fallout to inhalation and submersion effective doses of the public in Riyadh.

## 2. Materials and methods

The air samples were collected on the roof of the building of the Atomic Energy Research Institute building at the King Abdulaziz City for Science and Technology (KACST) in Riyadh (24° 42' 57" N, 46° 38' 20.7" E). The system for air filtration was obtained from Senya, Finland (JL-900 SNOW WHITE) which is designed for continuous outdoor use. It is a high volume air sampler, with a fixable capacity from 300 to 900 m<sup>3</sup>/h that can be controlled with frequency converter, thus providing possibility for changing volume flow. The filters used were glass fiber filters with a size of 570 × 460 mm from Whatman GF/AMacherey-Nagel MN 85/90 Camfil Media. These types of filters have a high retention capacity even for accumulation-mode aerosol particles to about 98% (Mattsson et al., 1965).

Air samples were collected for 24 h on the roof of the building in Riyadh from 08 April to 28 April 2011 using the above mentioned air filter system. After 24 h, the air filter was changed and sample collection was started again on the next morning. Air particles together with airborne radionuclides were collected on the surface of the filter paper and filter paper was taken off for counting and detection of the radioactive fission products.

The radioactivity of air (filter radioactivity) was analyzed using a high-resolution, p-type high purity coaxial germanium detector (HPGe) supplied by ORTEC, USA. The system had a relative efficiency of 80% and a resolution of 1.98 keV at 1332 keV. The HPGe detector was housed in shielding with 104.8 cm thickness lead covered with 0.12 cm, 0.15 cm and 0.06 cm of steel, copper and plastic respectively to reduce the effect of the background from the building and cosmic rays. The HPGe detector was calibrated using the Amersham point source of different radionuclides of  $^{22}\text{Na}$ ,  $^{57}\text{Co}$ ,  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^{133}\text{Ba}$  and  $^{241}\text{Am}$ . The efficiency of 500 mL geometry calibration of the detector was performed using a mix standard radionuclide source of  $^{210}\text{Pb}$  (46.5 keV),  $^{241}\text{Am}$  (59.5 keV),  $^{109}\text{Cd}$  (88.0 keV),  $^{57}\text{Co}$  (122.1 keV),  $^{139}\text{Ce}$  (165.9 keV),  $^{203}\text{Hg}$  (279.2 keV),  $^{113}\text{Sn}$  (391.7 keV),  $^{85}\text{Sr}$  (514.0 keV),  $^{137}\text{Cs}$  (661.7 keV),  $^{60}\text{Co}$  (1173.2 keV and 1332.5 keV) and  $^{88}\text{Y}$  (898.0 keV and 1836.1 keV). Each sample was counted for 86,400 s. There set of each material

was counted an average was taken. The air filters were then subjected to gamma-ray spectrometric analysis with a counting time of 86,400 s (i.e., 24 h). The radioactivity concentration of  $^{131}\text{I}$ ,  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  were measured using their gamma-ray photons at 364 keV (81.2%), 662 keV (85.2%) and 605 keV (97.6%), respectively. Genie 2000 Basic Spectroscopy Software is used to data acquisition and analyze the photo peaks. The energy and efficiency of the detector have been calibrated using a mixed standard reference filter source with gamma lines covering the range between 59 to 1500 keV. Fig. 1 shows the spectrum of air-filter radioactivity level using gamma-ray spectrometry equipped with HPGe detector.

The activity concentrations given in the text, tables and figures are all decay-corrected at the mean-time of the sampling. The overall uncertainties in the measured activities, with no more than 5%, are mainly attributed to the statistical counting and sampling uncertainties.

## 3. Results and discussion

### 3.1. $^{131}\text{I}$ , $^{134}\text{Cs}$ and $^{137}\text{Cs}$ activity concentration in air samples

In Riyadh, Saudi Arabia the first evidence of Fukushima radioactive fallout was confirmed with  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  measured in air filter samples collected on April 08, 2011. We could measure traces of fission products until April 28 at which there was no more detectable presence of any of these artificial radionuclides. Thus, radioiodine and radiocaesium isotopes were not present in air in Riyadh before April 08 or after April 28. However, the visible gamma lines detected on the air filters before April 08 and after April 28 were attributed to known background radioactivity from natural and/or cosmogenic origin. The widespread emissions from Fukushima were transferred by wind and cloud and caused detectable radioactive contamination in air over Riyadh. The filters were measured and the results of airborne radionuclide concentration during the 20-day period are presented in Table 1.

Shown in Table 1,  $^{131}\text{I}$ ,  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  were first detected at concentrations of, respectively,  $281.3 \pm 16.1$ ,  $15.6 \pm 0.9$  and  $7.3 \pm 0.4$   $\mu\text{Bq m}^{-3}$  on an air filter taken on April 08. On April 10,  $^{131}\text{I}$  and  $^{137}\text{Cs}$  had their maximum activity concentrations of, respectively,  $323.7 \pm 18.5$  and  $26.0 \pm 1.8$   $\mu\text{Bq m}^{-3}$  while the activity of ( $7.3 \pm 0.5$   $\mu\text{Bq m}^{-3}$ ) was recorded for  $^{134}\text{Cs}$ . After one day, on April 11,  $^{134}\text{Cs}$  in air peaked to ( $17.2 \pm 1.2$   $\mu\text{Bq m}^{-3}$ ) while  $^{131}\text{I}$  and  $^{137}\text{Cs}$  concentrations were decreased about 25% of their maximum initial values.

The time evolution of the three radioisotopes during the period of study was elucidated in Fig. 2. It is clear that similar maximum

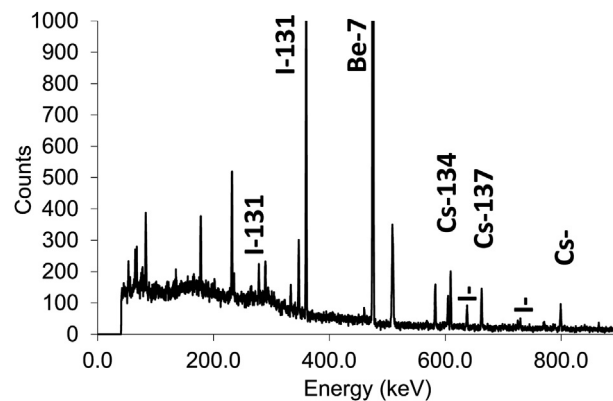


Fig. 1. Spectrum of air-filter showing the radioactivity level by using gamma-ray spectrometry (HPGe detector).

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