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ARTICLE

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Tracking of Airborne Radionuclides from the Damaged Fukushima Dai-Ichi Nuclear Reactors by European Networks

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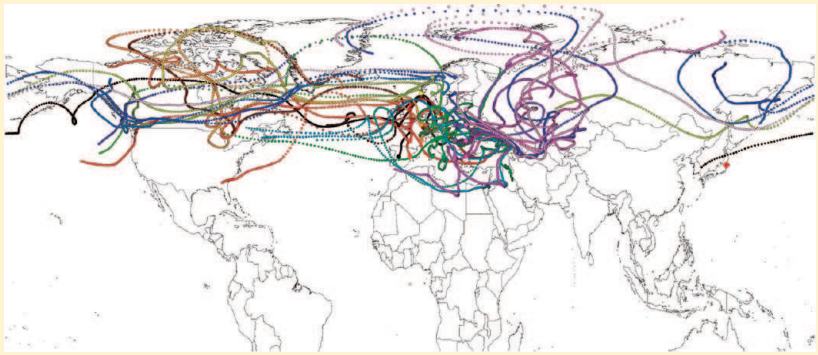
ABSTRACT: Radioactive emissions into the atmosphere from the damaged reactors of the Fukushima Dai-ichi nuclear power plant (NPP) started on March 12th, 2011. Among the various radionuclides released, iodine-131 (^{131}I) and cesium isotopes (^{137}Cs and ^{134}Cs) were transported across the Pacific toward the North American continent and reached Europe despite dispersion and washout along the route of the contaminated air masses. In Europe, the first signs of the releases were detected 7 days later while the first peak of activity level was observed between March 28th and March 30th. Time variations over a 20-day period and spatial variations across more than 150 sampling locations in Europe made it possible to characterize the contaminated air masses. After the Chernobyl accident, only a few measurements of the gaseous ^{131}I fraction were conducted compared to the number of measurements for the particulate fraction. Several studies had already pointed out the importance of the gaseous ^{131}I and the large underestimation of the total ^{131}I airborne activity level, and subsequent calculations of inhalation dose, if neglected. The measurements made across Europe following the releases from the Fukushima NPP reactors have provided a significant amount of new data on the ratio of the gaseous ^{131}I fraction to total ^{131}I, both on a spatial scale and its temporal variation. It can be pointed out that during the Fukushima event, the ^{134}Cs to ^{137}Cs ratio proved to be different from that observed after the Chernobyl accident. The data set provided in this paper is the most comprehensive survey of the main relevant airborne radionuclides from the Fukushima reactors, measured across Europe. A rough estimate of the total ^{131}I inventory that has passed over Europe during this period was <1% of the released amount. According to the measurements, airborne activity levels remain of no concern for public health in Europe.



INTRODUCTION

Radioactive emissions into the atmosphere from the damaged reactors of the Fukushima Dai-ichi nuclear power plant (NPP) started on March 12th. Xenon isotopes and especially Xe-133 (^{133}Xe, $T_{1/2}$ = 5.2 days), were the main contributors to the estimated source term (around 10^{18} Becquerels). However only five sophisticated sampling stations in Europe made it possible to quantify ^{133}Xe levels in air. Most of them are operated in the framework of the Comprehensive nuclear Test Ban Treaty Organization (CTBTO) and the results were not readily available outside of this organization. Among the various radionuclides released in large amounts, iodine-131 (^{131}I; $T_{1/2}$ = 8.0 days), cesium-134

(^{134}Cs; $T_{1/2}$ = 2.1 years) and cesium-137 (^{137}Cs; $T_{1/2}$ = 30.1 years) were easily detectable and of major interest for health impact assessments. Other short-lived radionuclides like tellurium-132 (^{132}Te; $T_{1/2}$ = 3.2 days) and iodine-132 (^{132}I; $T_{1/2}$ = 2.3 hours) were also measured at trace levels. All of them were transported across the Pacific toward the North American continent and reached Europe despite dispersion and deposition along the route of the contaminated air masses. After the Chernobyl accident, the importance of both the short and long-term effects of radioactive contamination due to ^{131}I were recognized [1,2] which is why special attention was dedicated to its determination. An understanding of the time and spatial variations of airborne concentrations levels will provide the basic data for further assessments of deposition mapping of those radionuclides onto the soil or to plants, their transfer to foodstuffs and finally dose estimates to humans. Additionally, the detailed knowledge of activity levels will offer a unique opportunity to test and enhance modeling of all sorts, thus providing worthwhile information for scientists in various fields.

Most European countries have developed monitoring programmes of airborne radionuclides since the 1950s. More than 150 sampling locations, most of them belonging to national networks, are spread out over Europe. They are equipped with high volume samplers for particulate species and some of them with activated charcoal traps for gaseous iodine. They are operated continuously within the framework of networks devoted to trace levels of airborne radioactivity. Sampling periods were reduced to provide an overall analysis of daily changes of, for example, ^{131}I especially in its particulate form and to a lesser extent in its gaseous form.

Most of the data were reported from laboratories participating in a network called "Ring of Five (Ro5)" which is an informal information group started in 1983 for the purpose of rapidly exchanging data on occasional enhanced concentrations of man-made radionuclides at trace levels (from micro to 10 of micro-Becquerels per cubic meter) in the atmosphere. In addition to the "measuring laboratories", there are also partners participating such as national meteorological services or staff members of universities or of the International Atomic Energy Agency (IAEA).

EXPERIMENTAL SECTION

The sampling stations in Europe have various types of aerosol samplers and operate at different flow rates. In all cases, the particulate fraction is filtered through conventional filters, mainly glass fibers or polypropylene fibers. For sampling of gaseous radioiodine, activated charcoal traps were also used. After sampling, activity levels were measured by gamma ray-spectrometry (usually High Purity Germanium detectors). The unique energies of the γ-rays (364.5 keV for ^{131}I; 604.7 and 795.9 keV for ^{134}Cs; 661.7 keV for ^{137}Cs) allows both identification and quantification of the radionuclides. During the studied period (March 20th to April 13rd), radioisotopes of cesium and iodine were detected above their limits of detection. Summation effect correction was applied to the determination of ^{134}Cs in

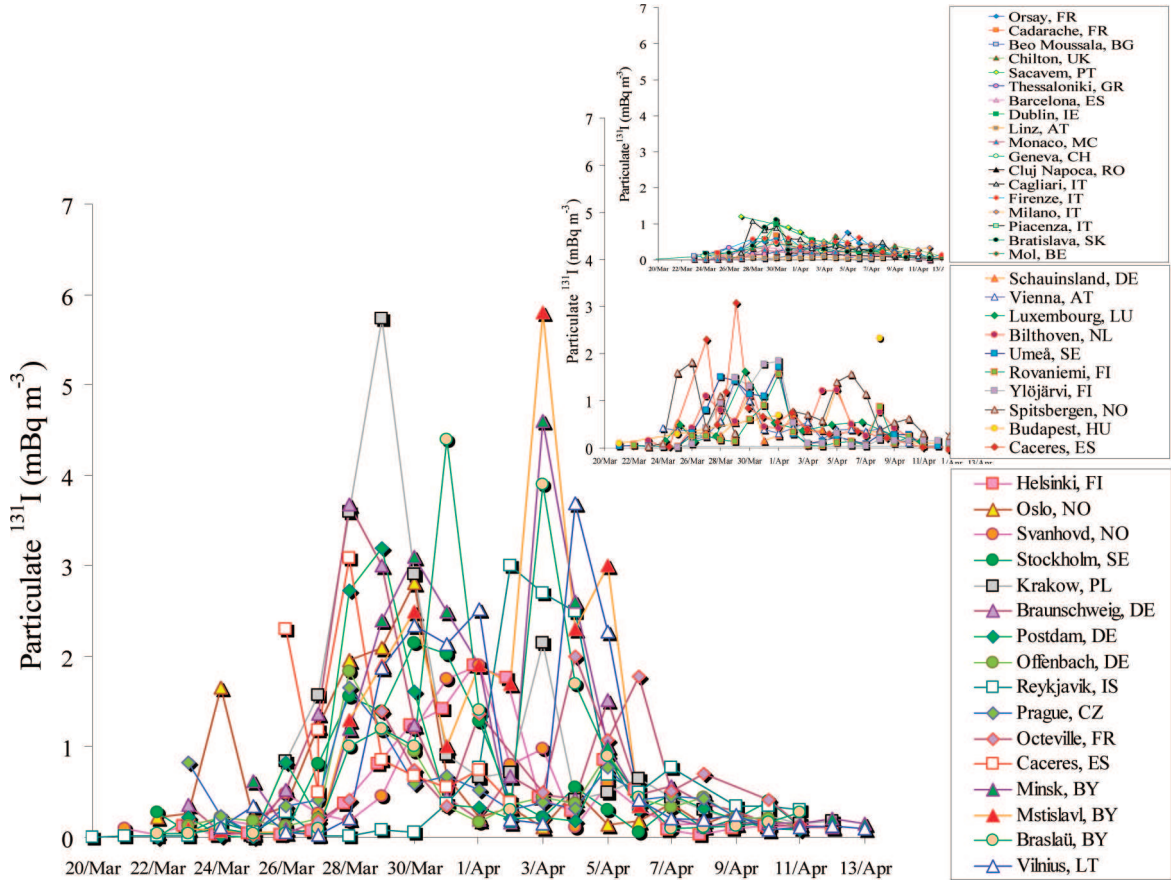


Figure 1. Time series of particulate ^{131}I (mBq m^{-3}) in northern and central Europe (bottom), western and southern Europe (middle and top) due to the Fukushima releases.

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order to get the true $^{134}Cs/^{137}Cs$ ratio. After the Chernobyl accident considerable variation was noticed regarding this ratio because such corrections were probably not commonly used at that time. The middle of the sampling period was chosen as the reference time for all stations. For the short-lived ^{131}I, decay correction was applied according to this reference date. For quality control, the level of naturally occurring ^{7}Be was used as an internal standard. The natural range of the specific activities of ^{7}Be is well-known for each station, which makes it possible to validate the sampling and measurement procedures.

RESULTS AND DISCUSSION

According to the source term declared by the Nuclear and Industrial Safety Agency (NISA) of Japan,[3] around 150 PBq (1 PBq = 10^{15} Bq) of ^{131}I and 6 to 15 PBq of ^{137}Cs, were released into the atmosphere. Releases started on March, 12th. The contaminated air masses entered North America from March 17th.[4] The first European detection of ^{131}I from Fukushima occurred between March 19th and March 20th in Iceland, between March 19th and March 21st in the northern part of Scandinavia and between March 23rd and 24th for most of the other European countries. Increasing levels were generally noticed during the next 10−12 days until March 28th to March 30th for western and central Europe and until April 3rd for the Republic of Belarus, as shown in Figure 1. A second peak with a similar magnitude was also detected between April 3rd and April 5th. The decreasing values between these peaks coincided with the arrival of a pronounced rainfall event over most part of Europe and may be also explained by precipitation scavenging experienced during transport, dry deposition, decrease in the atmospheric source term as well as the short half-life of ^{131}I. For the entire period, peak values ranged from less than 1 mBq m^{-3} up to 6 mBq m^{-3} for the particulate ^{131}I, and up to 11 mBq m^{-3} for the gaseous ^{131}I fraction. These values are 3−4 orders of magnitude lower than activity levels encountered in Western Europe after the Chernobyl accident in April 1986. On the European scale, the average ^{132}Te and ^{132}I activity levels were 0.032 and 0.043 mBq m^{-3}, respectively and the maximum value for both was 0.12 mBq m^{-3}. For a given location, the levels of ^{132}Te and ^{132}I were approximately in equilibrium. They were not measurable above detection limits from the second week of April.

During the last weeks of March meteorological conditions led to a corridor of higher activity levels through Europe (Figure.2). It extended along a NW to SE axis from Scandinavia, across eastern Germany, Poland, the Czech Republic and Belarus. Peak values of particulate ^{131}I of around 6 mBq m^{-3} were registered in

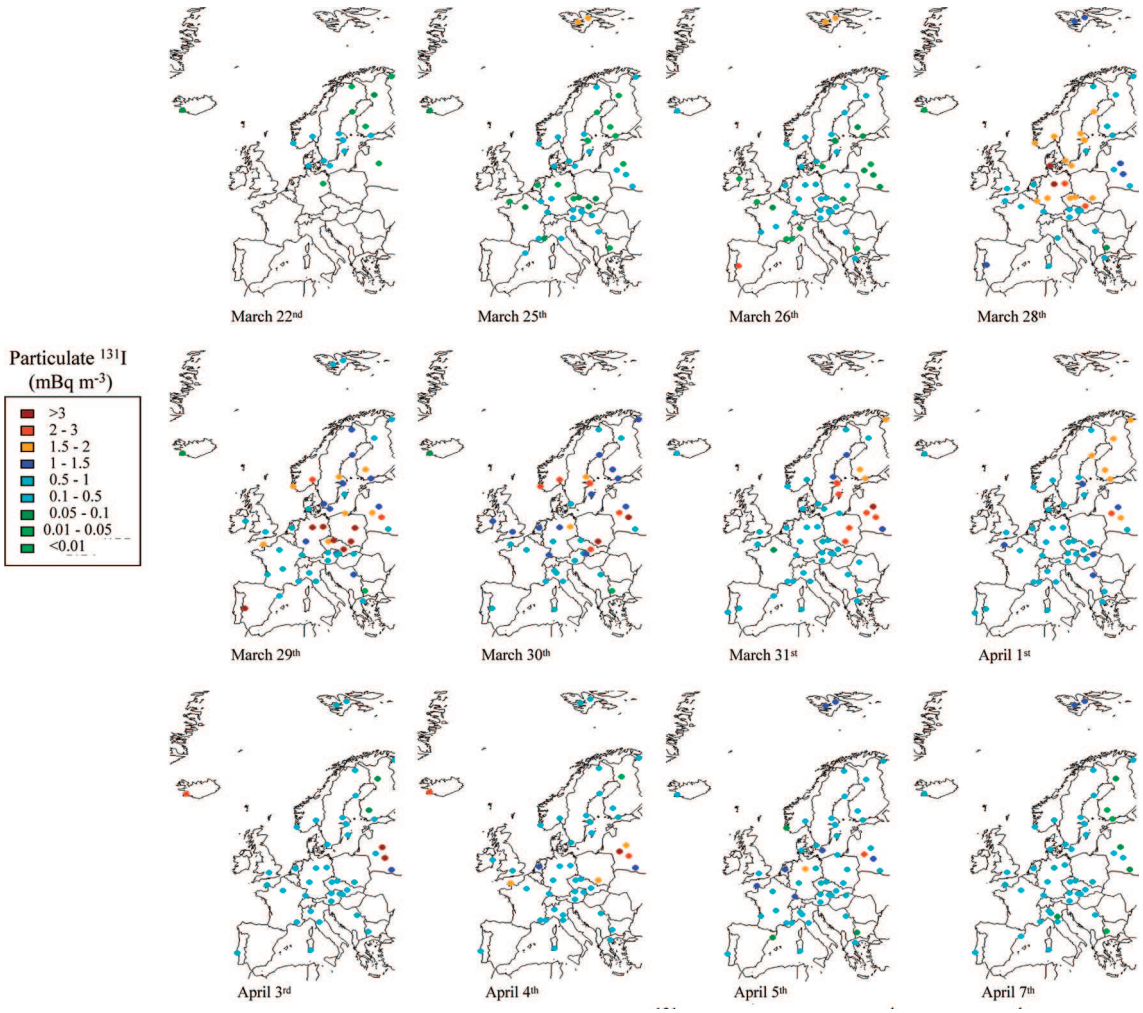

Figure 2. Measurements of daily activity levels of particulate ^{131}I between March 22nd and April 7th 2011.

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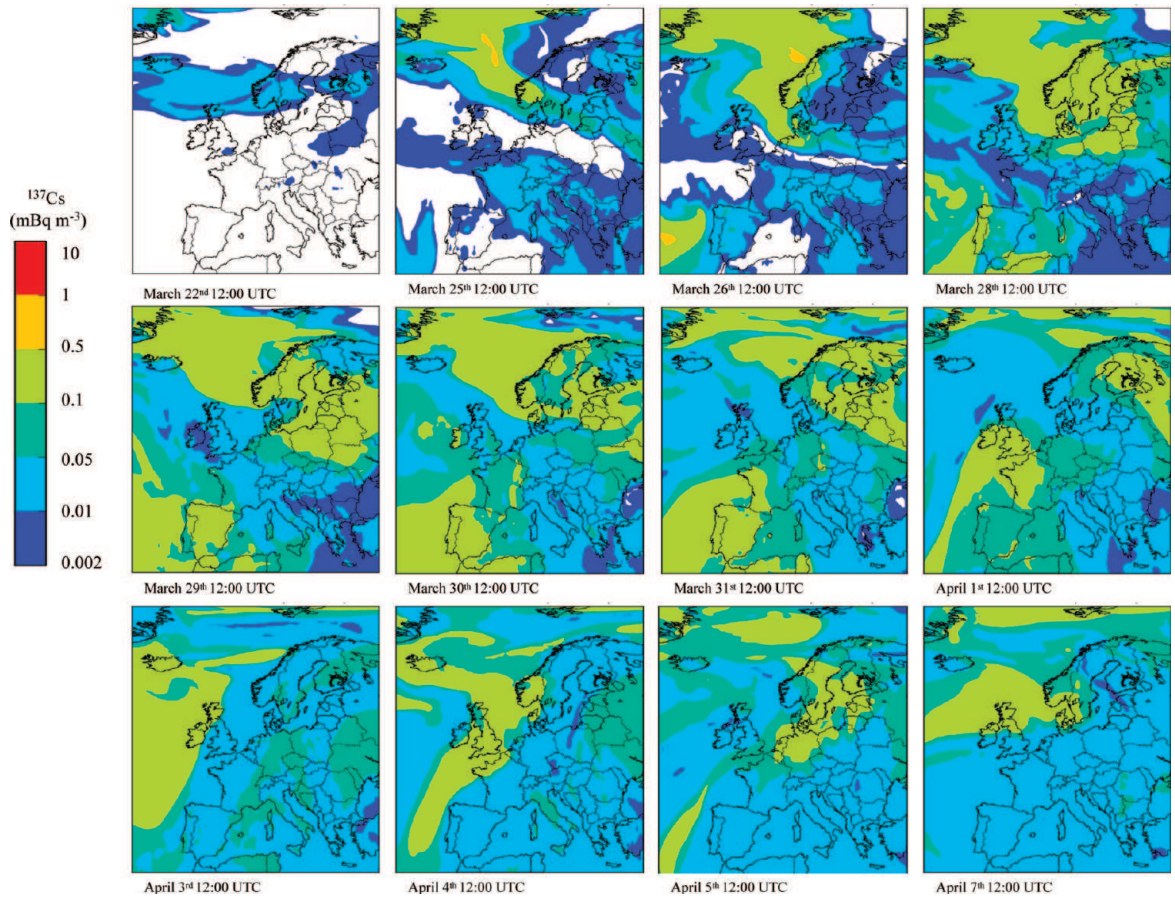

Figure 3. Simulations of ^{137}Cs activity levels in the atmosphere between March 22nd and April 7th 2011.

Krakow (Poland), Braslaü and Mstislavl (northern Belarus) and in Braunschweig (northeastern Germany). Countries located on the western and southern edges (Ireland, UK, Belgium, Switzerland, France, Italy, Portugal and Greece) measured maximum levels of typically <1.5 mBq m^{-3}. The dates that the contaminated air masses arrived and the so-called corridor of higher values were also well-predicted by simulations using the Eulerian LdX dispersion model, developed by IRSN and part of the Polyphemus platform[5] and are consistent with what was measured. Figure 3 shows the simulations for ^{137}Cs, arbitrarily taken at 12:00 UTC for convenient purpose. The pattern of contamination is broadly similar for ^{131}I as the ratio between the levels of ^{131}I and ^{137}Cs was observed to be rather constant as discussed later in this paper. The dry deposition of ^{137}Cs was modeled using a simple scheme with a constant deposition velocity ($V_d = 2 \times 10^{-3}$ cm s^{-1}). For wet scavenging, the parametrization used in this study was of the form $\Lambda = ap_0^b$; with $a = 5 \times 10^{-5}$ h mm^{-1} s^{-1}, b = 1, and the rain intensity (p_0) in mm h^{-1}. This model used meteorological data provided by the Arpege model from Meteo-France with a 0.5° resolution.

Many uncertainties resulted from the source term assessment, meteorological conditions along the route, parametrization of the scavenging efficiency and the long-range transport from Japan to Europe. The comparison between measured values and the simulation cannot therefore be used to check absolute values but shows the capability of both measurements and simulations to highlight areas where levels are elevated and possible hotspots. For example the relatively higher value

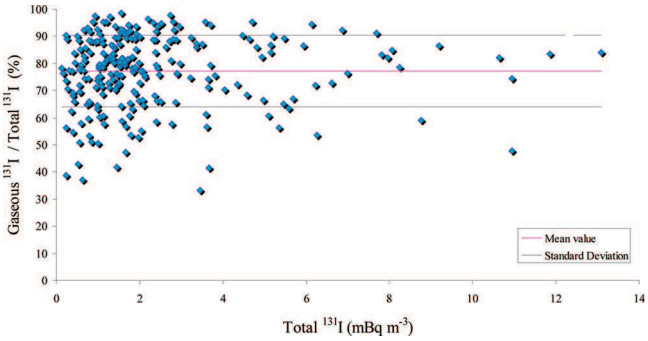

Figure 4. Gaseous/Total ^{131}I ratio in the atmosphere in Europe following the Fukushima NPP accident.

measured in the Iberian Peninsula, on March 29th was also predicted using the Ldx model.

Characterization of airborne iodine levels is complicated as it coexists in both particulate and gaseous forms, which requires different kinds of air samplers. At the time of the Chernobyl accident, few measurements of the gaseous ^{131}I were performed. In the following years, many sampling locations were equipped with activated charcoal to collect the gaseous fraction. This means it is now possible to provide more information on this fraction and for the gaseous to particulate ratio, both on a spatial scale and regarding its temporal variation. Based on about 210 pairs of values acquired up to April 12th, the average gaseous/

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total ^{131}I ratio is 77.2% ± 13.6% (Figure 4). This is the same average ratio as the one found after Chernobyl.[6] The higher variability between measured ratios can be explained by higher measurement uncertainties for low levels of total iodine. The ratio also varies with time as the particulate fraction alone will be subject to wet deposition. For example, the gaseous/total ratio at one site varies between 0.79 and 0.90 and even up to 0.96 for a few days. Another source of uncertainty can come from the properties of the adsorption media used and especially its collection efficiency in humid air conditions. Apart from the specifications of the gaseous trap itself, global uncertainties probably reflect different washout/rainout histories and aerosol chemistry-related effects of the two fractions during transport. Thus the predictive estimate of the gaseous fraction from the aerosol must be considered carefully.

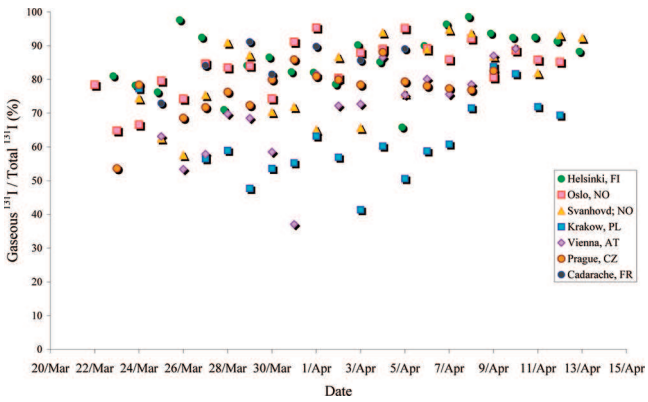

Figure 5. Time variation of the gaseous/total ^{131}I ratio in Europe following the Fukushima NPP accident.

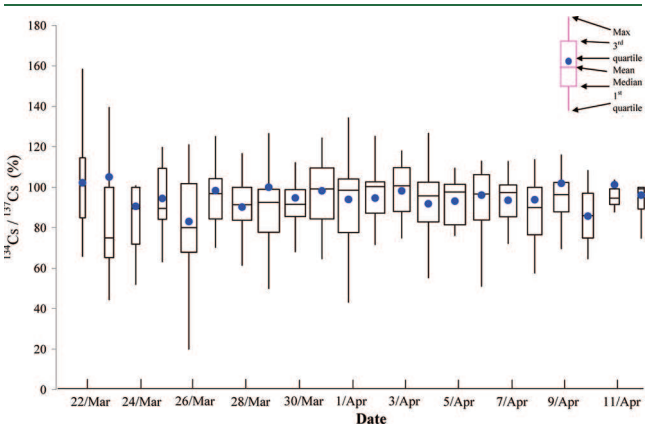

Figure 6. Time variation of ^{134}Cs/^{137}Cs in Europe following the Fukushima NPP accident.

As a first estimate, it could be supposed that a transfer of iodine gradually takes place from gaseous to particulate form, as if gaseous ^{131}I acts as a reservoir for the particulate form. According to the measurements taken on the Fukushima Dai-ichi NPP site from March 22nd to April 4th, the average particulate/gaseous ^{131}I ratio was 0.46 ± 0.17, i.e. a gaseous/total ratio of 71% ± 11%. This is very similar to the value observed in Europe (Figure 4) leading to the assumption that ^{131}I remains mainly in its gaseous form during transport. Thus, the transfer from gaseous to particulate form, if it exists, was not sufficient within the two-week interval to counterbalance the decrease of particulate ^{131}I due to its deposition, mainly by rain. Actually, the gas-to-particle conversion time for ^{131}I typically requires about 2−3 weeks to occur.[7] However, the behavior of the gaseous and particulate fractions of iodine and related transfer processes are not yet fully understood. Further detailed investigations are in progress.

The time variation of the gaseous/total ^{131}I ratio shows on average a slight increase with time (Figure 5). This corresponds to the faster decrease in the particulate form.

As in the case of the Chernobyl accident, both ^{134}Cs and ^{137}Cs were detectable. During the monitoring of air masses from Fukushima, cesium isotopes were typically detected with a time-lag of 3 to 4 days after ^{131}I due to the fact that cesium isotopes exceeded the detection criterion later than iodine. In Europe, ^{134}Cs had not been measured in the atmosphere since the middle of the 1990s due to its fairly short half-life of 2.06 years. On the contrary, ^{137}Cs was still measurable before the Fukushima accident at trace levels; below 1 μBq m^{-3} in Central Europe [8,9] and lower values (<0.3 μBq m^{-3}) were typically observed in northern and southern Europe.[10,11] Its persistence in the atmosphere can be explained by a longer half-life (30.1 years) and regular inputs through resuspension of soil particles containing traces from former deposition (Chernobyl fallout and/or global fallout from nuclear weapon tests) or through the emission of flying ashes during occasional biomass burnings.[12] Contrary to iodine which is mainly found in gaseous form, cesium is rapidly bound to aerosols and thus highly subject to washout removal by rain from the contaminated air masses. The time and spatial average values from March 20th to

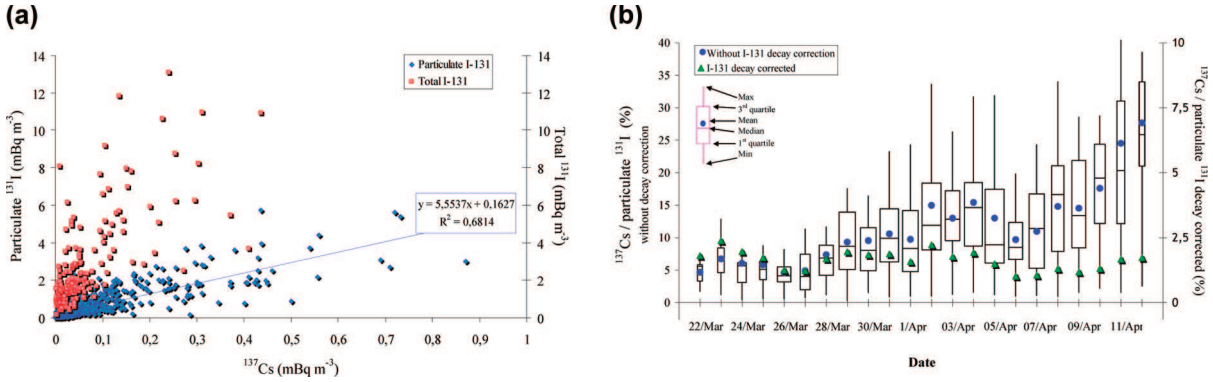

Figure 7. (a)Particulate ^{131}I and total ^{131}I, versus ^{137}Cs in Europe after the Fukushima accident. (b) Time variation of the ^{137}Cs/ ^{131}I average ratio in Europe after the Fukushima accident.

April 12th in Europe were 0.076 and 0.072 mBq m^{-3} for ^{137}Cs and ^{134}Cs, respectively with a first maximum of 0.75 mBq m^{-3} in Lodz, Poland between March 28th and 30th and a second maximum of 1.0 mBq m^{-3} in Vilnius, Lithuania between April 3rd and April 4th. Compared to the situation after the Chernobyl accident, ^{137}Cs airborne activity levels reported in this study were at least 10 000 to 100 000 times lower. Whatever the radionuclide considered, airborne activity levels remained sufficiently low that they were of no concern to public health in Europe.

The $^{134}Cs/^{137}Cs$ ratio in the Chernobyl fallout in 1986 was in the range 0.5 - 0.6,[13,14] whereas it was close to 1 for the Fukushima fallout (Figure 6). Variation of this ratio was fairly constant with time.

Figure 7a shows the relation between ^{131}I (particulate fraction and total fraction) and ^{137}Cs values. Dry deposition of the gaseous fraction of ^{131}I is known to be important and significantly higher than the dry deposition of particulate cesium.[15] The $^{137}Cs/^{131}I$ ratio range found in this study is consistent with deposition values observed in the UK [16] after the Chernobyl accident. The variation was attributed to the amount of rain from place to place. This parameter must still be checked and compared with the other parameters previously mentioned.

Between March 19th and April fourth measurements taken close to the NPP by the operator show that the particulate $^{137}Cs/^{131}I$ ratio was increasing from 0.35% to 11%, that is, the radioactive cloud contained more iodine at the beginning of the accident as it is more volatile than cesium. In Europe, as ^{137}Cs started to be detected (March 22nd), the overall average ratio ranged between 4.6% and 6.5% until March 27th before it increased due to the arrival of the most concentrated air masses (Figure 7b). This may indicate adsorption of gaseous ^{131}I onto uncontaminated aerosols at the edge of the air masses. This interpretation would be in line with significantly lower $^{137}Cs/^{131}I$ ratios observed at the Jungfraujoch station (Switzerland, 3450 m a.s.l), at an altitude which corresponds approximately to the upper limit of the planetary boundary layer.[17]

To check if other parameters than the radioactive decay of ^{131}I could explain this increase with time, average values were decay-corrected (triangles on Figure 7b) for the half-life of ^{131}I to March 11th, the date of the automatic shutdown of the reactors after the earthquake detection. It can be clearly shown that the ^{131}I decay is mainly responsible for the ratio increase. Consequently, it can be assumed that the washout coefficient for cesium also stands for particulate ^{131}I and that transfer of particulate iodine from the gaseous fraction, if it exists, remains of minor importance after a long passage in the atmosphere from Fukushima.

The highest ^{131}I concentration over Europe measured at ground stations persisted for approximately 10 days. The average concentration in the corridor of highest activity levels was approximately 1 mBq m^{-3} for particulate ^{131}I or 4 mBq m^{-3} for both fractions. Less is known about the ^{131}I vertical distribution in the troposphere but activity levels seem to be rather homogeneous in the vertical plane over Europe. Indeed, the altitude station at Jungfraujoch (3450 m a.s.l) showed ^{131}I activity levels up to 0.9 mBq m^{-3} and high altitude samples taken with airplanes on March 30th above Switzerland (up to 8000 m) measured 2.3 mBq m^{-3} as well as a few mBq m^{-3} above northern Germany (up to 10 000 m). Thus it seems reasonable to assume that ^{131}I was well mixed up to the upper troposphere. Assuming that the above-mentioned average concentration was valid for a 10-day period (240 h); that the air masses had moved at an average speed of 50 km h^{-1}; that the corridor with the highest concentrations was 3,000 km wide with a mixing layer height of 5.5 km, results in an estimated volume of air of $3000 \times (240 \times 50) \times 5.5$ km^3. Using this volume and an average concentration of 4 mBq m^{-3}; roughly 1 PBq of ^{131}I passed over Europe, thus less than 1% of the released amount.

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