# We are IntechOpen,
# the world's leading publisher of
# Open Access books
# Built by scientists, for scientists

## 4,400
Open access books available

## 118,000
International authors and editors

## 130M
Downloads

Our authors are among the

## 154
Countries delivered to

## TOP 1%
most cited scientists

## 12.2%
Contributors from top 500 universities

CLARIVATE ANALYTICS
BOOK CITATION INDEX
INDEXED

**WEB OF SCIENCE**™

Selection of our books indexed in the Book Citation Index
in Web of Science™ Core Collection (BKCI)

# Interested in publishing with us?
# Contact book.department@intechopen.com

Numbers displayed above are based on latest data collected.
For more information visit www.intechopen.com

# Natural Radioactive Isotopes in Water in Relation with Geology and Hydrological Investigations in the Territory of Luxembourg

Zornitza Tosheva, Harald Hofmann and Antoine Kies
*[1]University of Luxembourg,*
*[2]Monash University,*
*[1]Luxembourg*
*[2]Australia*

## 1. Introduction

Luxembourg is situated in the middle of Europe and is surrounded by Belgium in the north-west, France in the south-west and south, and Germany in the east (Figure 1). It is one of the smaller countries in Europe with a surface area of 2.600 km$^2$ and a population of approximately 500.000 inhabitants. The northern third, known as 'Eislek', has a plateau character with an average elevation of 450 m, whereas the southern two-third are know as the 'Gutland', good land in English. This region has a more varied topography and an average elevation of 240 m.

Groundwater is the main water source for drinking water supply in the Grand Duchy of Luxembourg. About 60% of the country's drinking water comes from so called "Luxemburg Sandstone", which forms the most important aquifer. Most of the groundwater captures are natural springs or shallow wells and represent 80 % of the volume of this aquifer. The majority of the springs are distributed along the main river valleys. The unconfined part of the sandstone is located in the centre of the country and covers about 300 km$^2$ with a surface to water table depths ranging from 0 to 100 meters (Colbach, 2006).

Except from some bores in the north of Luxembourg and some in the south, natural springs are mainly used for public drinking purpose.

The radionuclide content of more than 300 springs and bores was determined as part of 2 monitoring campaigns, in a first step to relate the physico-chemical parameters with local geological conditions and in a second to obtain data on groundwater pollution regarding agricultural contaminants, such as pesticides, nitrate etc. The second campaign allows a general quality assessment of the groundwater resources in the country, which is also required by the European Water Framework Directive (EU, 2000).

Radio-nuclides are of interest for two reasons. Firstly, a high activity poses a health risk. An example is dissolved radon ($222Rn$) in groundwater that degasses when the water leaves the aquifer. Radon concentrations increase in the air in buildings where the water is trapped, such as spring trappings, or when used like in households. The inhalation of radon and the deposition of its daughter products in the lungs can cause lung cancer. Secondly, radio-nuclides can be useful tools as tracers in the hydrological / hydro-geological cycle.

Radioactive carbon and tritium are widely known and utilized tracers for groundwater dating and follows the diffusion and migration processes. The radioactive carbon and tritium are cosmogenic. Most of the cosmogenically produced radio-nuclides in the atmosphere are oxidized and become attached to aerosol particles. These particles act as condensation nuclei for the formation of cloud droplets and eventually coagulate to form precipitation. Similar is the mechanism of naturally produced ^{210}Pb, a long-lived decay product of the noble gas radon from the uranium decay series, which reach the atmosphere and are transported with the atmospheric circulations in form of particles until deposition on the surface with the precipitations. Other radioactive elements, besides these isotopes, such those of the uranium and thorium decay series, can be used as groundwater flow and mixing tracers.



Fig. 1. Geological map of the main units in Luxembourg. The area of the Devonian formations in the North forms the 'Eislek', whereas the Southern 'Gutland' is represented by the Triassic and Jurassic units.

The uranium and thorium series isotopes derive from the release out of uranium / thorium bearing minerals in the soils and rocks. These naturally occurring radio-isotopes are supplementary tracers to conventional major ion chemistry and stable isotopes in hydrological applications, e.g. creates a particularly interest such as surface water/groundwater interactions.

There had been some research on the use of radio-nuclides in these fields but still only little was known about their distribution and the mobility over the country. If there presence is ubiquitous, there are physical and chemical aquifer characteristics, such as porosity,

Natural Radioactive Isotopes in Water in
Relation with Geology and Hydrological Investigations in the Territory of Luxembourg
379

hydraulic conductivity or organic matter content that play major roles in their occurrence and migration in the environment.

This study does not aim to solve all questions, which are related to the distribution and mobility of radio-nuclides in the groundwater but it helps to get closer to some solutions.

In this work we present two different kind of investigations performed with the natural radioactive isotopes in relation with geological and hydrological studies.

The aim of the first part is to deliver a high resolved spatial radioactive isotope distribution maps and an analysis for the causes of activity variations. We rely on the major ion chemistry data and physical parameters that have been measured in Luxembourg's drinking water supplies. The relations of the radionuclide concentrations with the major ion chemistry were studied. Comparison of the obtained results versus local geology was followed in this approach.

Besides a dense spatial variability and chemical behavior along the hydro-geological regions, the second part of this work aims analyses of temporal variations of radio-isotopes, especially in the case of radon. These results are crucial for the use of radio-isotope data as groundwater and surface water tracers.

## 2. Sampling and methodology

The samples were taken in 2 litres, airtight glass bottles directly from the spring. Where possible they were taken right at the emergence of the water from the host rock. Water from wells and bores was pumped with the preinstalled pumps from the water suppliers. Purging was necessary because the sampled bores are production bores that are constantly in use. The bottles were filled completely without air space and stored in insulated coolers to avoid degassing during the transport to the laboratory.

The electrical conductivity and the temperature of the water was measured in-situ using a WTW i340 multi probe. The electrical conductivity was temperature corrected to 20°C.

Major ion chemistry was done at the National Water Laboratory of Luxembourg by ion chromatography for the anions and induced-coupled mass spectrometry for the cation content and trace elements.

The radio-nuclides were measured at the Radio-physics Laboratory of the University of Luxembourg. Radon ($^{222}$Rn) was measured a few hours after sampling by liquid scintillation counting (LSC) on a Perkin-Elmer "Guardian" Liquid Scintillation Counter, using 10 ml of Perkin-Elmer Ultima Gold F scintillator and 12 ml of sample. For some choosen samples 3H were measured by LSC after purification with Eichrom tritium columns. The other isotopes ($^{238}$U, $^{235}$U, $^{234}$U, $^{232}$Th, $^{230}$Th, $^{228}$Th, $^{228}$Ra, $^{226}$Ra, $^{224}$Ra, $^{210}$Pb) were separated with sequential extraction (Tosheva et al. 2003, 2009) and $^{210}$Po by electro-deposition on silver discs. After separation they were measured on Canberra alpha detector or LSC respectively.

## 3. Local geology and hydrogeology

### 3.1 Geology

In the official web page of Luxembourg geological survey (geology.lu) is written that Luxembourg shows on its small area of about 2500 km² a dense geological diversity.

As mentioned above, Luxembourg is orographicaly and geologically divided in two major natural regions, which are geologically different. On the one hand, there is the 'Eislek' in the

North, which is built up from Lower Devonian formations in Siegen facies (Figure 1). It belongs to the rhenohercynian block of the Eifel Mountains. The outcropping rocks consist mostly of folded and slated medium grained sediments with actual thickness of several thousand meters, transformed into schist, meta-sandstones, and bluish-grey low-grade methamorphosed shists sediments. Inter-beddings of quartzite sandstone occur in some parts of the formation. The geological substrate is covered by a thin soil layer of 0.5 to 1.5 m thickness.

On the other hand, there is the 'Gutland' in the South, which encompasses Mesozoic units forms the Perm-Triassic boundary to the Middle Jurassic. They belong to the units of the Paris Basin and extend about 150 km from the southern Eifel in Germany, via Luxemburg towards Belgium (Berners, 1983). The sequence starts with the Lower Triassic units of the 'Buntsandstein' with fluvial red bed sand-, siltstones and clays, followed by marls of the Upper Keuper (Rhät). However, the unit, which bears most drinking water springs in the country, is the 'Luxembourg Sandstone'. It is carbonate bound sandstone from the Middle Liassic (li), which can be assigned to a shallow marine shelf environment with wave influencing reworking and re-sedimentation (Berners, 1983). Heterogeneous layering boundaries can be observed within and in between the sandstone formation and the Psilonotas marls. It consists of light yellow, fine to medium grained sandstones with a low to medium carbonate content (15-50 %), which origins from the spartic cement and bioclastic components (Colbach, 2006). Some layers contain very well rounded pebbles (ca. 10 cm).

The Luxembourg sandstone is covered by younger formations in the South-West of the country. They encompass the iron Minette ore formation carbonate units of the Middle Dogger and carbonates of particularly the Upper Dogger. These south-west part ironstones are present in the form of limonitic and pisolithic surface formations.

## 3.2 Hydrography and hydrogeology

Luxembourg has a temperate climate with an average rainfall of 800 mm per year and an average temperature of 9°C. The land-use distribution is as follows: forested 34%, pastures 26%, agricultural-cultivated 23%, other 17%.

The majority of the samples come from the Luxembourg sandstone (grès de Luxembourg) aquifer, which is the most important aquifer for the drinking water supply in the country. It is generally limited on its bottom and top by the limestone-shale alternations of the Lorraine facies, although interbeddings with these exist (Colbach, 2005). The porosity of the aquifer is closely related to the carbonate content with approximately 5% in well cemented sandy limestones to 35% in poorly cemented sections, whereas water flow occurs in the well developed open fracture network. It is a dual-porosity aquifer and shows very heterogeneous permeabilites across its occurrence. The general flow pattern is dominated by the dip to the south-west of the unit.

The other hydrogeological units, which were encompassed in the campaigns, are Devonian shists and sandstone in Siegen facies, red-bed sandstones (Buntsandstein), Muschelcalc sandstone, dolomitic Keuper limestones, carbonate-bound sandstones from the middle Lias and iron-bearing limestones form the Dogger.

The Devonian schist and sandstones in Siegen and Ems build up the northern part of the country and belong to the rheno-hercynian block of the Rhenish Slate Mountains and the Ardennes Mountains. They are folded and fractured schist and sandstones with a low hydraulic conductivity. Flow and storage happens mostly in the fracture network.

The Buntsandstein in Luxembourg shows a sandy facies of fluvial environments with conglomerate and silt/clay interbeddings. Groundwater flow and storage happens in the fracture network as well. It has mixture permeability of fissures and pores. The dolomitic limestones of the Keuper are characterized by a more developed fracture network than the older formations. Carbonate dissolution has created a reasonable specific storage and hydraulic conductivity. The same hold true for the middle Lias carbonate-bound sandstones. The Dogger formations consist of oolithic iron ores from the Minette-type. The hydrogeology of the Minette rocks is complicated due to the mining activities in the past but general characteristics of the aquifer encompass a medium to high hydraulic conductivity. This aquifer is a dual porosity aquifer with a well developed open fracture network. The pore space in the matrix is high in the weathered areas and low in the non-weathered areas.
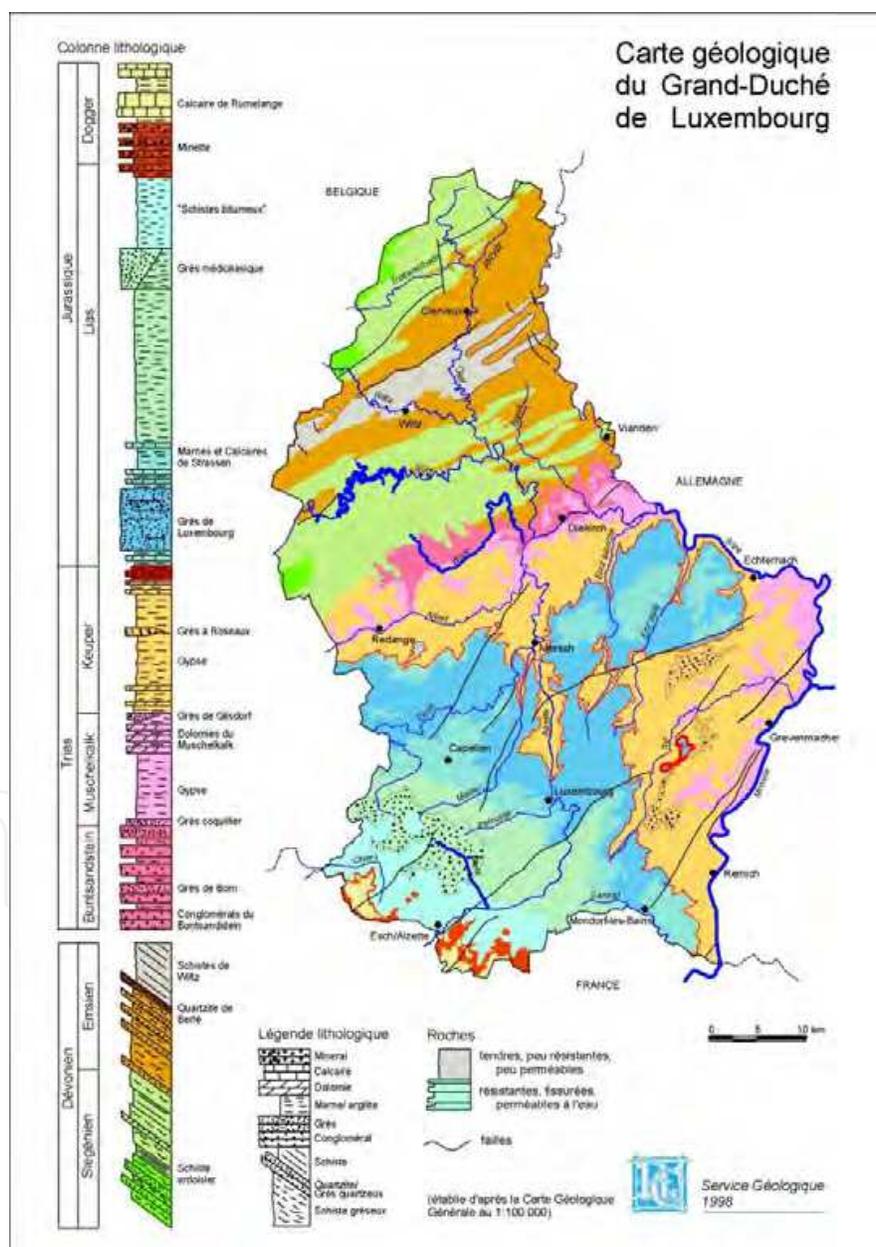


Fig. 2. Geo-morphological map of the main units in Luxembourg.

## 4. Results and discussion

The major ion chemistry of the water in Luxembourg is dominated by $Ca^{2+}$, $Mg^{2+}$, $HCO_3^-$ and $SO_4^{2-}$ and therefore the waters can be assigned to a calcium-bicarbonate type (figure 3). However $Na^+$, $K^+$ and $Cl^-$ play a significant role, especially in places with higher permeability and fractures. Another not negligible contribution comes from $NO_3^-$ ions, especially on the plateaus of Luxembourg Sandstones when agriculture affects the water table or organic matter is present in the rocks. The carbonate species are dominant in the carbonate units of the Luxembourg Sandstones and the younger Jurassic lime stones formations and carbonate units if Luxembourg's sandstones with carbonate cements and bioclastic components. Calcium values range from a few mg/l to over 200 mg/l, where values of the main aquifers, such as the Luxemburg sandstone, range from 80 to 200 mg/l. The bicarbonate follows the same trend with concentrations in between 1 to 40 mg/l.
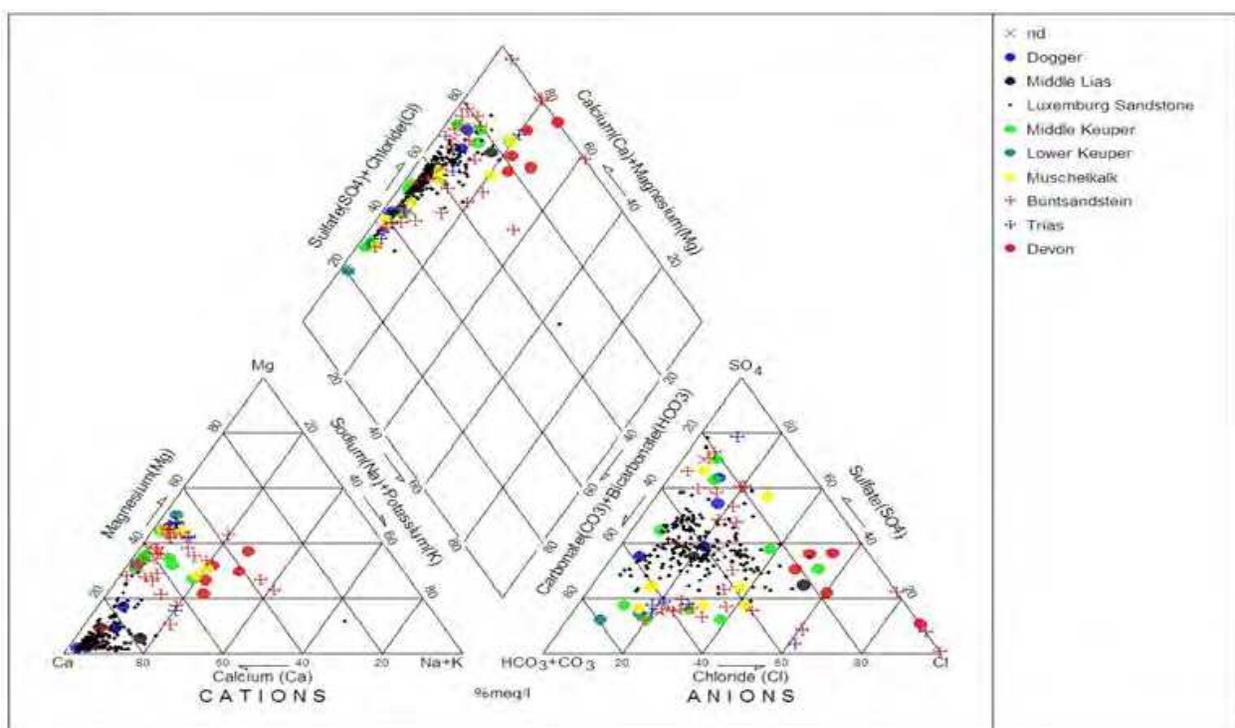


Fig. 3. Piper-diagram of the sampled water in the Spatial Monitoring campaign.

Radionuclide concentrations mainly vary with the local geology and in many cases are supported by different chemical mecanismus. Values for activity concentration of uranium vary from 0.2 to 74 mBq/L with predominant 10–30 mBq/L. They correspond to aquifers with similar mineralogy described by[Porceli et al., 2003].

Radium in natural water is derived mainly by interaction between the water and radium bearing materials, such as rock, soil and deposits due to its good solubility. As it forms sulfates, carbonates, nitrates and chlorides at different degrees and as such is a good indicator of the geochemical situation. In the analyzed samples the concentration varied from 3 to 35 mBq/L. Only five samples show significantly increased results, which can be explained with the different geological material in the aquifers (Devonian and Trias formations). Another data set with higher than median values was from a spring where the waters have a quite different chemistry and high salinity [Oliveira et al, 2006].

Natural Radioactive Isotopes in Water in
Relation with Geology and Hydrological Investigations in the Territory of Luxembourg
383

Lead mobility in water depends on the pH, hardness, sulfate and carbonate ion concentration and the presence of organic complexion agents [Oliveira et al, 2006]. In our study the spatial distribution of $^{210}$Pb follows the same pattern as for $^{238}$U and $^{230}$Th (figure 4). These data prove that the investigated sources have almost no contact with the surface, mostly there has been no exchange between water horizons [Gonzalez-Labajo et al,2001] and the radio-nuclides originate from the surrounding rocks and soils.

The correlations among radio-nuclides and major ions were not significant, suggesting independence among the variables. However the analyzed waters were from a bicarbonate type; the main influence on the radionuclide distribution is due to chloride ions. Some explanation is related to the surprising fact that most of the waters contain non negligible iron and manganese concentrations.
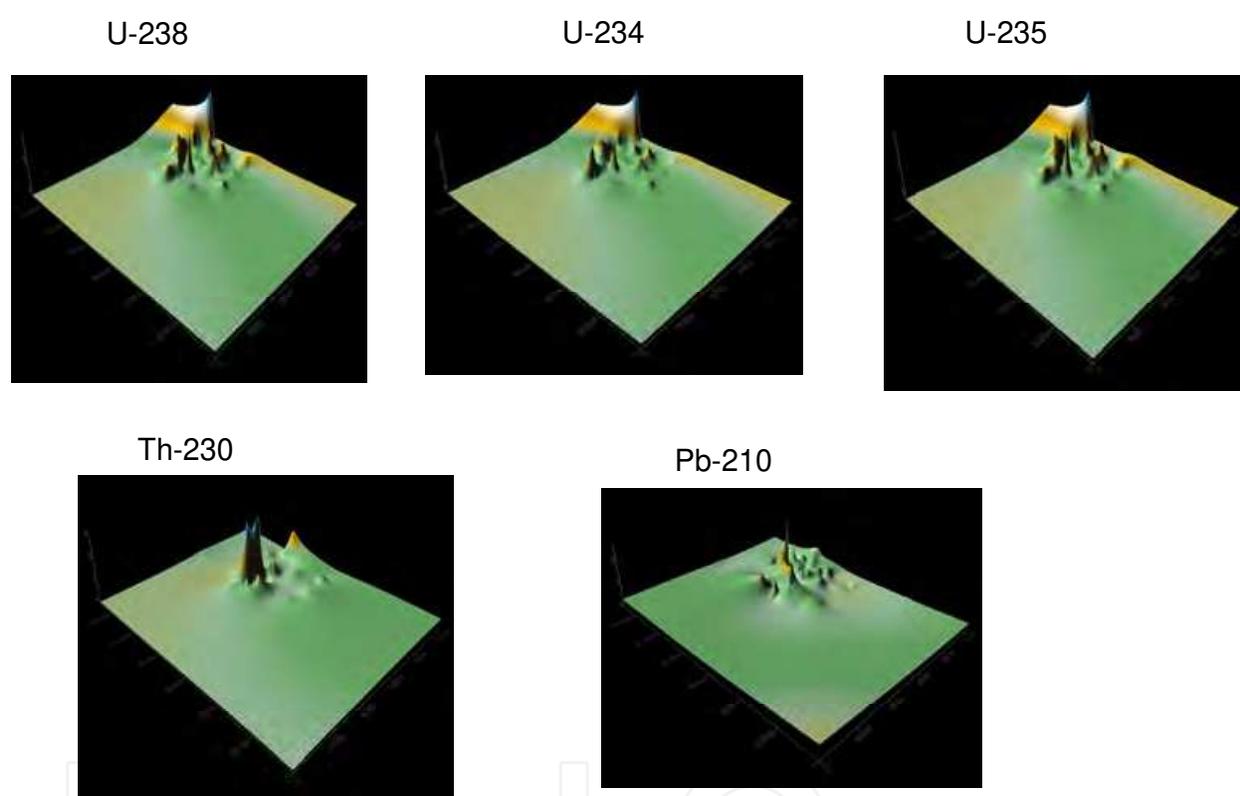
U-238

U-234

U-235



Th-230

Pb-210



Fig. 4. Spatial distribution of uranium series isotopes over Luxembourg country. Activity concentration, represented on axe 'z' is in mBq/L

Correlation analysis show also no clear relation between calcium and magnesium content, however calcium is substituted well by strontium in almost all geological formations (figure 5). We obtain good correlation of strontium and radium radio-nuclides in Buntsandstein and Luxembourg sandstones formation where these form sulfates. In both formations reducing conditions exist in the deep wells.

In figure 6 are represented average values of main ions influencing radio-nuclide distribution over geological formations as well as the mean of different radioisotopes distributions over the same formations. The values are in 'ppm', except where for representation facility, they are converted in 'mg'. Radioactive elements are in 'mBq/L except radon that is in Bq/L. In Figure 6a are represented main cations and anions that may affect the solution of natural radio-nuclides, following the fact that generally Luxembourg

waters are bicarbonate. In Figure 6b, after detailed analysis of the data are represented the main influencing radioactive distribution ions. Strontium and sulfates affect the radium distribution, as discussed previously; whereas iron and its supported copper and manganese are responsible for uranium and thorium distributions.
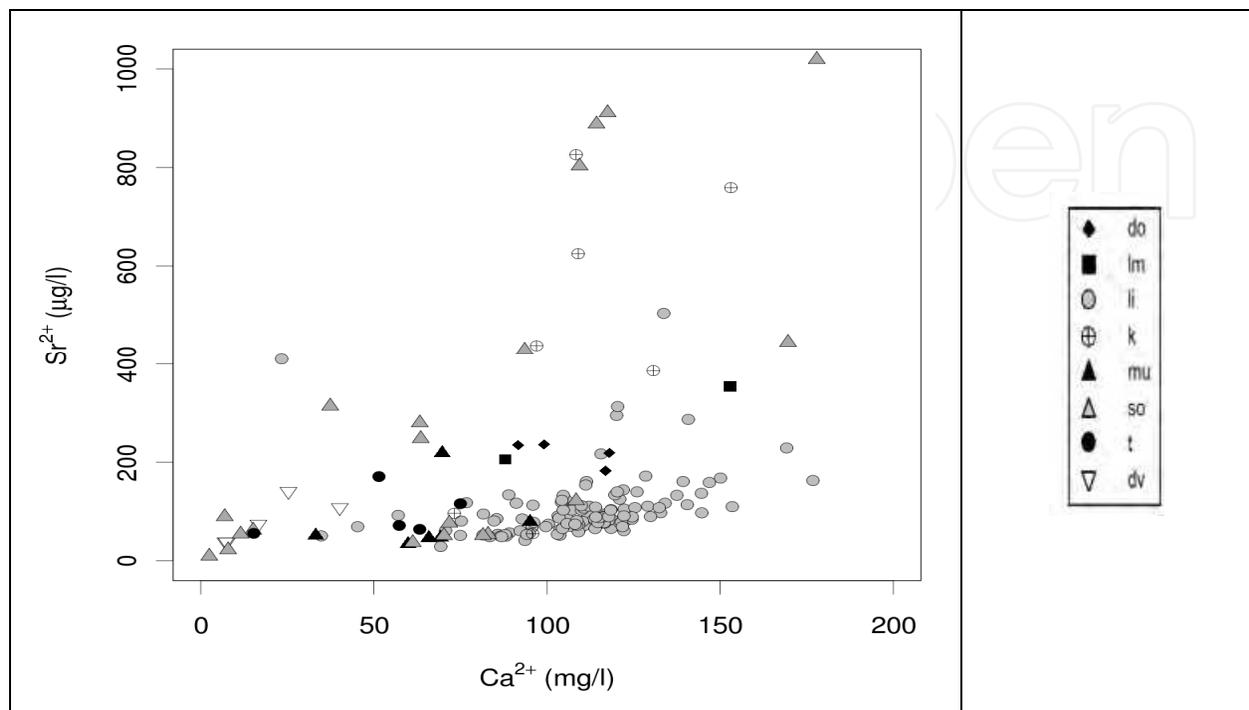


Fig. 5. Correlation between calcium and strontium content in analysed water vs. geological formation. The abbreviations are: do (dogger), km (keuper), li (liasic), mu (muschelcalc), so (buntsandstein 1), t (buntsandstein 2), dv (devonien)

In Figure 6c and 6d are represented the uranium and thorium series. The geological formations are also separated as for the Buntsandstein, where we observe a mixture of fissures and pores and permeability; besides an increasing mineralization from north to north-east. In the first ones we distinguish more sulfate and bicarbonate, while in the second more chlorides. This explains the low radium radio-isotopes and polonium contents in the bi-carbonate clay-sand and gypsum part, as well the presence of lead sulfate, and its dissolutions in mobile chlorides. Noticed is also the clear influence of iron and strontium in reducing conditions for the uranium and thorium presence. For similar reasons the sandstones of Luxembourg are presented into two different sub-formations. In the widely represented Luxembourg sandstones water circulation is favoured along vertically formed diaclases; the water table is charged continuously. Fractures are enlarged by water passages and by carbonate dissolutions; whereas the dissolved residue fraction in the sands constitutes a natural filter. As a consequence the reducing/oxidizing conditions change and induce the formation of sulfates where stable and radioactive thorium (80%) correlates with sodium, copper and manganese.

The $^{226}$Ra/$^{238}$U ratios over the country range from the unique very low value of 0.3 to the values of 20, having a mean value not significantly different from 1. Similar is the range of ratios between $^{210}$Po and $^{210}$Pb. The highest observed values can be explained with the high solubility of radium and with polonium mobility in anoxic water.
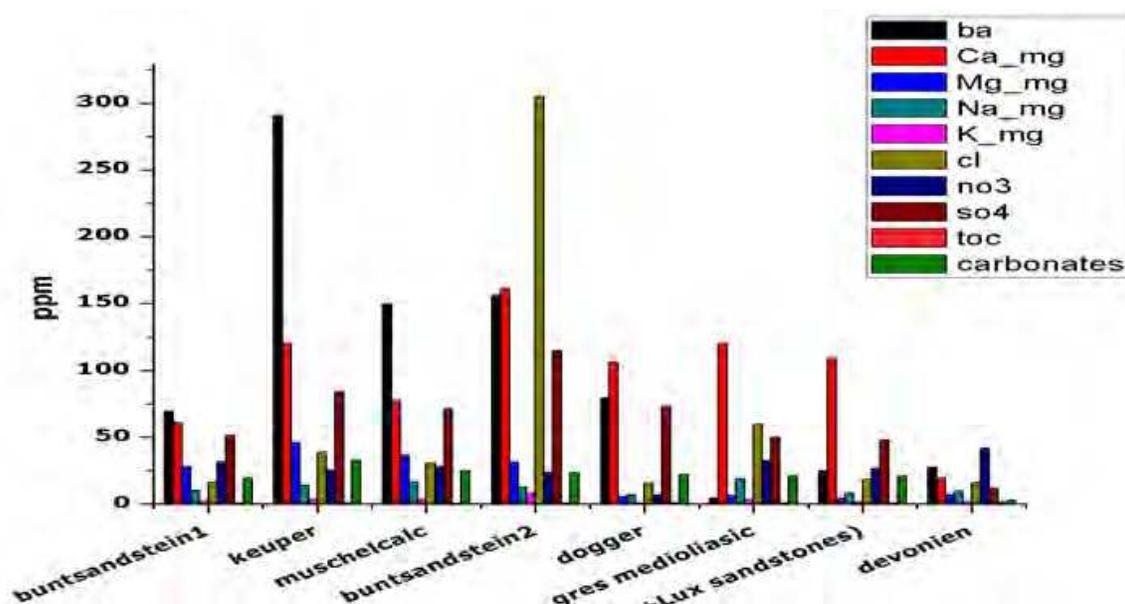
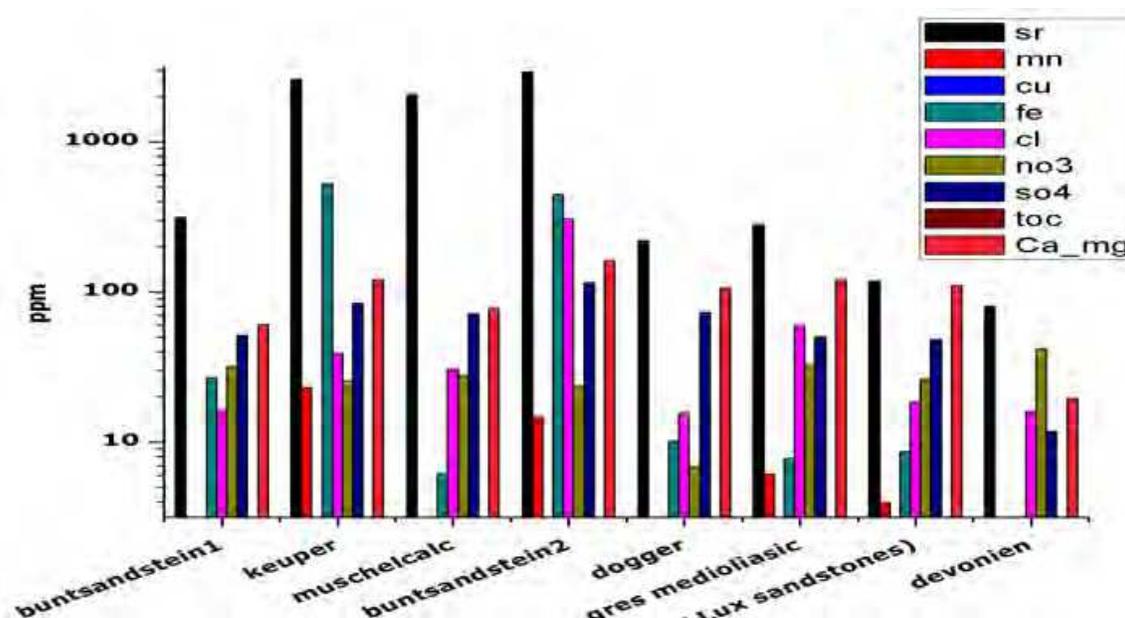Fig. 6a. Distribution of major ions versus geological formation



Fig. 6b. Spatial distribution of some influencing radio-nuclides occurrence ions versus geological formations

Analyzing the cross ratios $^{238}U/^{232}Th$ and $^{230}Th/^{232}Th$, it is observed that the first ones are higher than the second ones, but follow the same surface distribution. This fact again is explained with the specific chemistry of these radio-nuclides and should be attributed to the very low thorium solubility.

One can see that in many cases over the country, an opposite distribution between radon and radium is present. An analysis of the relation between radium and oxygen content shows higher radium associated with lower oxygen content. The initially anoxic water, when enriched in oxygen, deposits radium; this radium will produce elevated concentrations of radon in the source.
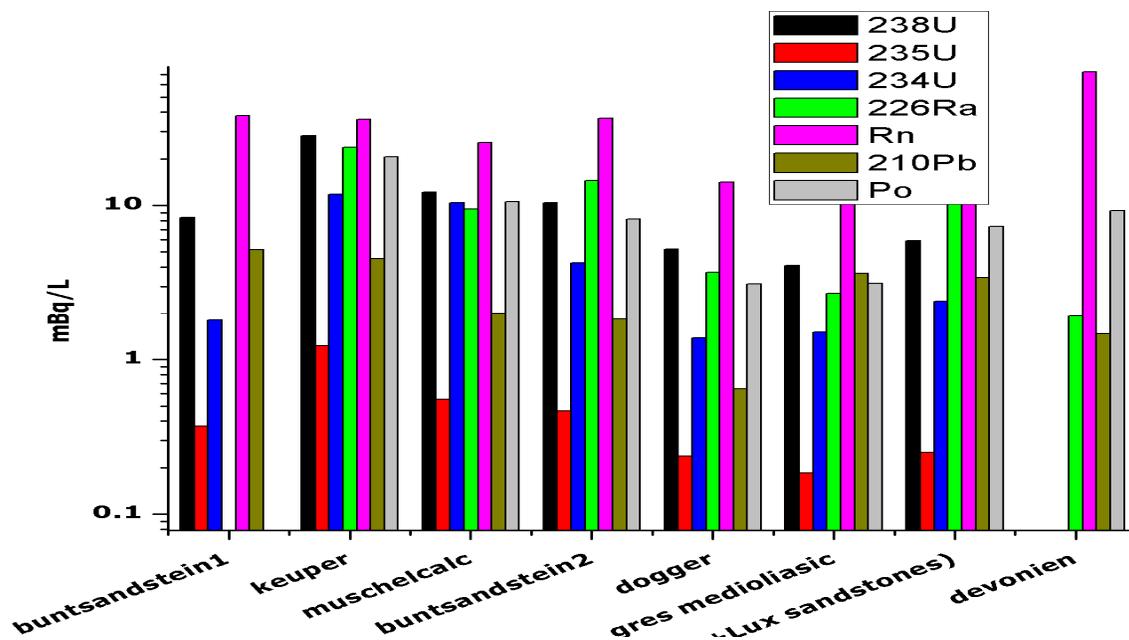
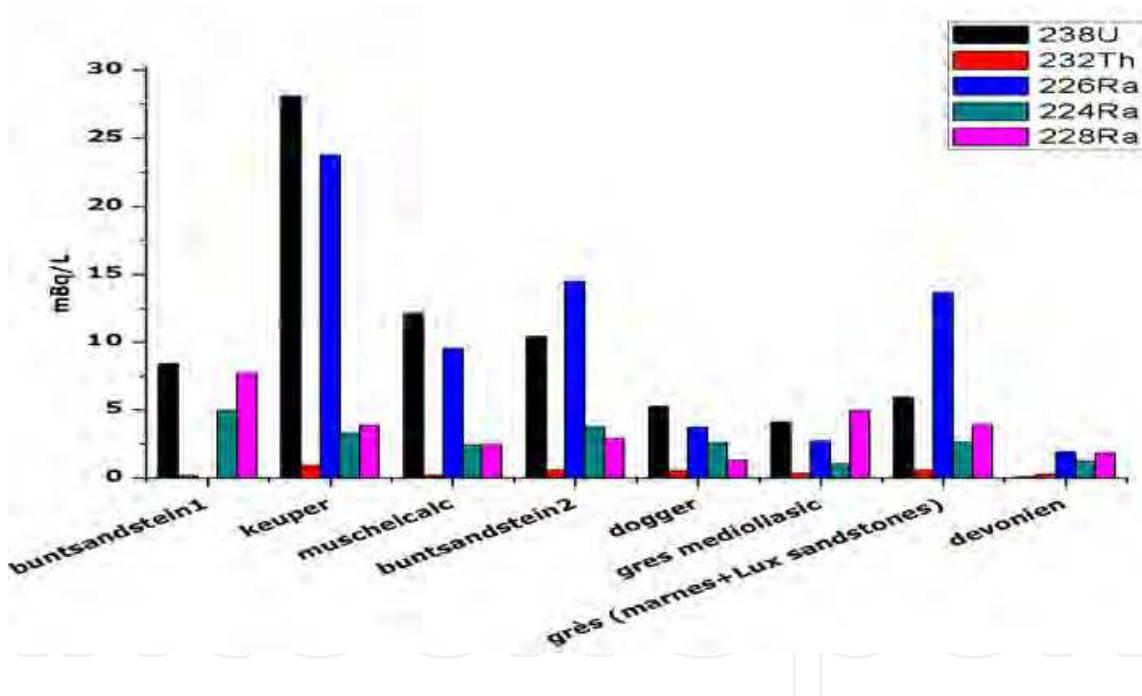Fig. 6c. Uranium series isotopes distribution versus geology



Fig. 6d. Distribution of main radio-nuclides versus geology

Another feature is the relatively high concentrations of [210]Po compared to the parent [226]Ra in the middle part of the country. One explanation is that, polonium product of ^{210}Pb is supplied to body waters from atmospheric input, run-off and the ''in situ'' decay of [226]Ra, its precursor in the water column. As the lead distribution follows the uranium one and it is 10 times lower than polonium the dependence of the [210]Po/[226]Ra ratio with the radon should be investigated in location with high values. Naturally produced 210Pb can exhale from the soils and can get transported through the lower atmosphere. It is a long-lived decay product of the noble gas radon from the uranium decay series, which reaches the atmosphere and is

transported with the atmospheric circulations in form of particles until deposition on the surface with the precipitations. Generally in keuper, muschelcalc and sandstone formations we observed some equilibrium between radon and radium. In these cases the 226Ra/210Po ratio is upper or near to 1. In the escape cases we had some spots of stable lead and it is related to Ba/Ca sulfates formations. Polonium values were high due to re-deposition of aerosols too.

In the devonien formation radon concentrations were higher than radium ones and 226Ra/210Po ratios lower than 1. In this case possible explanation of disequilibrium between polonium and lead were the exhalation to atmosphere processes.

For every analysed water sample the activity ratios $^{238}U/^{232}Th$ and $^{230}Th/^{232}Th$ are calculated. From the results is we obtain that these ratios may have significant values. In most cases, they can be attributed to the very low $^{232}Th$ concentrations and not to high $^{238}U$. The mean ratio value of $^{238}U/^{232}Th$ for surface and soil infiltrated water is 2.4, whereas it exceeds 12 for underground water. It should be mentioned that in the investigated underground water with high $^{238}U/^{232}Th$ ratios, we remarked that the $^{210}Pb$ content is very low, and the $^{230}Th$ concentrations are twice as high as those of $^{232}Th$ [5]. This is associated with the higher solubility of uranium isotopes compared with thorium isotopes.

The obtained data also help to study the radioactive equilibrium disturbance within the uranium series. For each sample, the ratios $^{210}Pb/^{238}U$, $^{234}U/^{238}U$ are calculated. As expected for surface waters, the $^{238}U/^{234}U$ ratio is close to 1 and reveals a secular equilibrium (normal background). In the same samples, the lead content exceeds by more than 30 times the uranium content related to the normal hydrological cycle and atmospheric precipitations.

In all drilled wells the $^{234}U/^{238}U$ ratios range from the very low value of 1.3 to a value of 5, with a mean value of 1.8. A correlation with increasing depth or fault zones is presumed, which causes the irregularities.

As above -mentioned, the majority of the natural springs occurs in the outcropping region of the Luxembourg sandstone in the central part of the country. The normal background radon concentrations in water vary in the range of 2–60 Bq/L with an average value of 17 Bq/L. The distribution of the obtained data follows a positively skewed log- -normal dependence with the maximum at 15 Bq/L that is characteristic for deeper groundwater in this area. Only 3% of the obtained results are close to 100 Bq/L and are well explained with the high mineralization of these sources.

The comparison between indoor radon concentrations over the country and water radon concentrations show no direct relation. The reasons for this lack of relationship are not clear because soil air radon and, therefore, indoor radon concentrations are dependent on the geological substrates and their U-bearing minerals in general. It is supposed that the radon propagation in the water is more dependent of the local hydrology processes than from the geological structures.

The geographical distribution of radon shows two axes of elevated concentrations. One is located in the north-western region, followed by the second one in the central East of the country. They both are mainly situated on Lower Jurassic and Triassic formations. A radiation profile map of Luxembourg, prepared with aero-gamma measurements in 1995 [Geo-service of Luxembourg, 1997], also reveals that these parts of the country have increased radiation levels as a consequence of high geochemical distribution of 238U in the rocks of these formations. The test of radon dependency on main physical parameters, such as EC, temperature and pH, did not show any correlation for all sampled springs. The temperature of the waters varied in-between 8–16°C with average values in the range of 10–12°C. An anti-

correlation of the radon and temperature in 65% of the cases is known from the literature, but this could not be observed in this case due to the small temperature fluctuations of the groundwater [Huyadi et al, 1999]. The same situation was found for the EC.

Radon concentrations in the north-central part show some differences in the 2003 and the 2007 campaigns. The latter campaign coincides with the recent construction of the northern highway in the time of this investigation. A hydro-geological and hydro-geochemical study of the Geological Survey of Luxembourg showed that the construction works influenced the flow regime and the hydro-geochemistry. A decreasing radon concentration could be observed in the same period. It is assumed that drilling and blasting works changed the flow path within the aquifer and, therefore, also changed the water chemistry. This was correlated with degreasing discharges in most of the monitored springs.

The spatial variations can be led back to the different geological materials and structural/tectonic features. Figure 6c shows that the radon activity changes with different geological units where the sources are located. Most aquifers in the region are double porosity aquifers, with proportion of matrix storage and fracture flow. Radon concentrations are influenced by the contact time between the water and the aquifer rock. Water from fast fracture flow is usually in contact with the rock matrix only for a short period of time and, therefore, cannot accumulate so much radon. On the other hand, radon with its short half-life reaches secular equilibrium after approximately 30 days. Hence, the groundwater flow should not influence its concentrations because groundwater flow is usually by magnitudes slower. Nevertheless, hydraulic conductivities of up to 100 m/d have been found along open fracture zones through tracer tests [Pistre et al, 2005], which support the results of this study.

The relation between radon and radium is of particular interest. Radon is a tracer, which can be used in a variety of groundwater studies, such as hydrograph separation, or any other type of groundwater/surface water interaction [Schmidt et al, 2009; Wu et al, 2004]. It is a daughter isotope of radium. Reasonable amounts of radium get into the hydrological cycle mainly through the dissolution of silicates and carbonates. It is not very mobile and can precipitate with other bivalent cation, such as $Ba^{2+}$ or $Ca^{2+}$, as sulfates or carbonates. Nevertheless, considerable amounts of radium were measured in the sampled sources.

It is interesting to notice that the gamma airborne survey has shown a strong anti-correlation of radon and radium over Luxembourg, especially in the central-eastern part of the country. The radium concentrations in water are negatively correlated with radon. High levels of radium are associated with low levels of radon and vice versa. Radon activity in groundwater is controlled by the concentration of radium in the aquifer matrix [Vinson et al, 2009]. However, the amount of radon that can enter the groundwater depends on the emanating coefficient of the radium bearing material in the aquifer. This means that the radon emanation potential of the rocks is in some cases very low, especially for low permeable rocks. Then again, low radium content rock can nevertheless emit higher concentrations of radon, when their porosity and interconnected pore space is large. Both interpretations do not explain the negatively correlated behaviour in Luxembourg. The waters from the Devonian rocks, low porosity, show high concentrations in radon (Figure 6c), but low concentrations in radium, whereas the waters from the Lias and Buntsandstein, higher porosity, formations are low in radon, but high in radium. Hence, a possible interpretation is that in the first case the radon derives directly from emanating form the rock. The Devonian shits and sandstones are rich in uranium and radium, whereas the radon comes form dissolved radium in the water in the second case. The relation between radium and oxygen content shows higher radium concentrations associated with lower

oxygen content. The initially anoxic water, poor in oxygen, mobilizes only a little amount of radium. If this water is deposited on the surface layer, it will produce elevated concentrations of radon in the springs.

Besides, a dense spatial resolution we analyze temporal variations of radon as well. The 316 springs from 2007-2008 campaign were sampled twice, once during the winter month, and once during the summer month and therefore during base flow conditions. Furthermore, five of the selected springs have been equipped with continuous flow and electrical conductivity (EC) devices and have been sampled for radon on monthly basis over a whole year (figure 7). Data from a previous campaign in 2003 are also used for comparison.
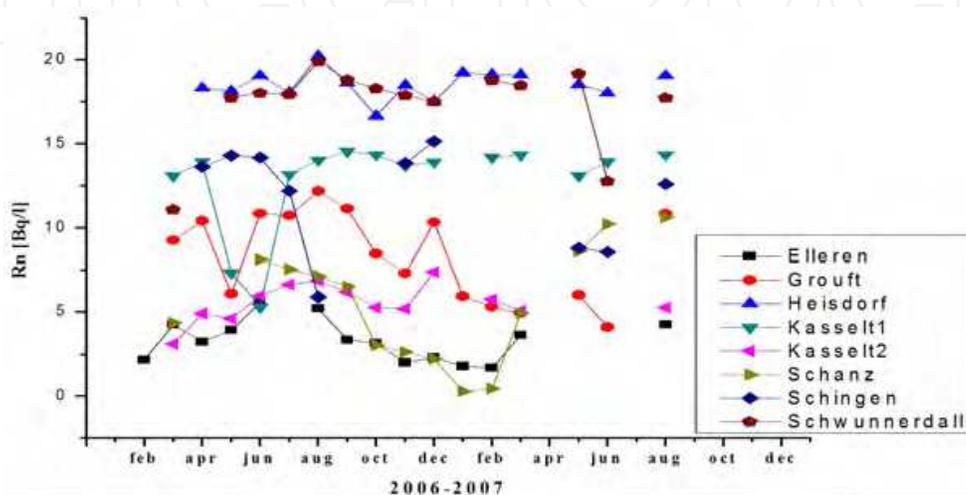


Fig. 7. Temporal variations of radon in sources of drinking water

Spatial heterogeneity was assumed in the rocks, with different correlation lengths. Long-scale effects were dominant for most of the sources.

The possible application of tritium is an essential tool for hydrological investigations. The most important applications are identifications of recharges in aquifers, estimation of hydraulic parameters related to pollutant transfer and determination of turnover time of groundwater. In the present work are analyzed for this isotope only 83 samples mainly for pollution investigation purposes this does not allow us to make significant conclusions. In all of them the concentrations are near to the capability of registration of used equipment.

Analyses of all obtained data over these two campaigns are in progress of elaboration.

## 5. Conclusions

The spatial distributions of the radio-nuclides as well the major ions are driven by two kinds of forces, anthropogenic (agricultural land use, iron mining etc.) and geogenic Uranium / thorium series isotopes are integrated in the crystal lattice of minerals, e.g. as it is the case with uranium in zircons, or substitute positively charged major ions, e.g. the substitution of calcium and magnesium by radium. Depending of local site geo-morphology this substitution is guided by carbonates (trias, buntsandstein), sulphates and chlorides (deep water in keuper, muschelcalc, lias) or nitrates and iron oxide / hydroxides (Luxembourg sandstones diaclases, land-used trias part).

Radon (in this case $^{222}Rn$) constitutes a particular case, as it is a noble gas and therefore inert. It is the daughter nuclide of Radium ($^{226}Ra$ decays to $^{222}Rn$) and accumulates in open crevasses in the rock matrix or gets dissolved in the groundwater.

As the distribution of the analysed sources is much more concentrated in the part of Luxembourg sandstones and alluvial formation, more of the results are reported to trias and inferior lias formations.

In medio-liasic sandstones and supra-liasic formations (Minette), due to iron mining, showed a marked uranium/thorium disequilibrium.

In Muschelcalc formation radon concentrations are low due to low permeability, increased levels are measured in sources due to the existence of fractures. In Luxembourg sandstones due to changing oxidizing/reducing conditions variable radon concentrations are observed.

Seasonal heterogeneities of radon are mainly relied to runoff production processes.

The response of radio-nuclides as tracers is more direct in sandstones compared to the schist in rain periods.

The investigation on spatial distribution dependency shows that the distribution in uranium series radio-nuclides, except of radium and polonium, are in correlation with local hydrogeology features.

Comparisons between two measurement campaigns are coherent and give us important information for the mechanism of distributions of the natural radio-nuclides in ground and surface water.

## 6. Acknowledgment

We want to thank the Water Service Department of Ministry of Interior and the Great Region of Luxembourg andespecially Dr. Denis Pittois form the Public Research Center Henri Tudor - CRTE for the chemical data and support. We would like to acknowledge the Service of Geology of Luxembourg for the guidelines and help during the analysis and whole project.

## 7. References

Colbach R (2005) Overview of the geology of the Luxem- bourg Sandstones. Scientific work of Museum of natural history of Luxembourg vol.44, pp. 155–160

EU Directive of drinking water (2000)

Tosheva Z., Taskaeva I., Kies A. (2006). International Journal of Environmental Analytical Chemistry, vol.86 No 9, pp.657-661

Tosheva Z., Hofmann H., Kies A. (2009). Journal of Radioanalytical Nuclear Chemistry, vol.282, pp.501-505

www.geology.lu

Berners (1983) Annales de la Société Geologique de Belgique 106:87-102

Porceli D., Swarzenski P (2003). Mineral Geochemistry, vol.52, pp.317-321

Oliveira J., Carvalho F (2006). Czechoslovak Journal of Physics, vol.56, No1, D545

Gonzales-Labajo J., Bolvar J.P., García-Tenorio R (2001). Radiation Physical Chemistry, vol.61, No3, pp.643-652

Geochemical map of G-D Luxembourg (1997). Geological Service Division of Luxembourg

Huyadi I., Csige I., Haki J., Baradacs E. (1999). Radiation and Measurements, vol.31, No116, pp.301-316

Pistre S., Marliak S., Joudre A., Bidoux P. (2005). Ground Watater, vol.40, pp.232-241

Schmidt A., Stringer Ch., Haferkorn U., Schubert M. (2009). Environmental Geology, vol. 56, No5, pp.855-863

Wu Y., Wen X., Zhang Y (2004). Environmental Geology, vol.45, No5, pp.647-653

Vinson D., Vengosh A., Hirschfeld D., Dwyer G. (2009). Chemical Geology, vol.260, No3/14, pp.159-171

**Earth and Environmental Sciences**

Edited by Dr. Imran Ahmad Dar

We are increasingly faced with environmental problems and required to make important decisions. In many cases an understanding of one or more geologic processes is essential to finding the appropriate solution. Earth and Environmental Sciences are by their very nature a dynamic field in which new issues continue to arise and old ones often evolve. The principal aim of this book is to present the reader with a broad overview of Earth and Environmental Sciences. Hopefully, this recent research will provide the reader with a useful foundation for discussing and evaluating specific environmental issues, as well as for developing ideas for problem solving. The book has been divided into nine sections; Geology, Geochemistry, Seismology, Hydrology, Hydrogeology, Mineralogy, Soil, Remote Sensing and Environmental Sciences.

**How to reference**

In order to correctly reference this scholarly work, feel free to copy and paste the following:

Zornitza Tosheva, Harald Hofmann and Antoine Kies (2011). Natural Radioactive Isotopes in Water in Relation with Geology and Hydrological Investigations in the Territory of Luxembourg, Earth and Environmental Sciences, Dr. Imran Ahmad Dar (Ed.), ISBN: 978-953-307-468-9, InTech, Available from: http://www.intechopen.com/books/earth-and-environmental-sciences/natural-radioactive-isotopes-in-water-in-relation-with-geology-and-hydrological-investigations-in-th

# INTECH
open science | open minds