Carpathian Journal of Earth and Environmental Sciences, May 2013, Vol. 8, No. 2, p. 163 - 168

RELATIONSHIP FROM GEOLOGY AND RADON IN OUTDOOR AIR IN MASSIF DITRĂU AREA, EASTERN CARPATHIANS – ROMANIA

Adriana ION
Geological Institute of Romania, Bucharest, 012270, Romania, adi75riana@yahoo.ro

Abstract: Radon activity concentration in outdoor air was measured using alpha radon monitors (Pylon AB-5 portable with Continuous Passive Radon Detector). Radon in outdoor air was measured in situ, in 5 points, for each lithological unit from the study area. The radon concentration was measured at a height of 15 cm above ground level. The radon exhalation rate was continuously measured for 24 hours with a counting time of 20 minute/interval in each site. The range is considerable: from 2.6 - 52 Bq m^{-3}. The release of radon from rocks and soil is controlled largely by the types of minerals in which uranium and thorium occur. In Ditrău Alkaline Massif, uranium (^{238}U), thorium (^{232}Th) and actinium (^{235}U) are concentrated in accessory minerals such as: zircon, monazite, titanite, allanite, apatite, xenotime, rutile, thorite, bastnäsite, parisite, pyrochlore.

Keywords: outdoor radon concentration, passive system, petrography, map

1. INTRODUCTION

Radon is a tasteless, odorless, invisible gas which is the single most important source of natural radiation to affect the human body (Cosma et al., 2009). Radon is a naturally occurring radioactive gas which has three isotopes ^{222}Rn, ^{220}Rn, and ^{219}Rn. These isotopes are originating from the natural radioactive decay of uranium (^{238}U), thorium (^{232}Th), and actinium (^{235}U). All radon isotopes have relatively short halflives ($T_{1/2}$) of 3.82 days, 51.5 seconds and 3.96 seconds. Regarding the radon level in the air, ^{222}Rn and ^{220}Rn are of higher interest, but because of its much shorter half-life ^{220}Rn can travel only very short distances in air and thus it is generally of less concern than ^{222}Rn, the most significant being ^{222}Rn (Dang Duc Nhan et al., 2012). Because (^{238}U), thorium (^{232}Th), and actinium (^{235}U) are constituents of the earth crust and radon is genetically associated with these isotopes, one can find radon everywhere in the environment.

Geology is the most important factor controlling the source and distribution of radon. High levels of radon emissions are associated with rock types present in the massif Ditrău area such as: syenite, nepheline syenite, granite, hornblendite, diorite, monzonite, monzodiorite, aplite, lamprophyre. The release of radon from rock-soil -air in Ditrău massif is given by the naturally occurring uranium, thorium in the mineral composition of rocks. Uranium and thorium are concentrated in accessory minerals (zircon, monazite, titanite, allanite, apatite, xenotime, thorite, bstnäsite, parisite, pyrochlore).

In this work, radon in outdoor air was measured in July, 2012 at 27 points distributed almost uniformly in each type of rock which occur in Ditrău massif area. Information on radon background levels is essential to assess anthropogenic contributions (UNESCEAR, 2000).

2. GEOLOGY OF THE AREA

The alkaline massif of Ditrău is singular in Romania by its size and petrographic variety. The massif is emplaced with in metamorphic basement rocks at the interior of the East Carpathians, it is an intermediate size massif (about 200 Km^2) and exhibits an eccentric ring structure in which the more basic rocks tend to lie to the west, with an arcuate zone of syenitic rocks, extending from the far north to the south-east, and a large area dominated by nepheline syenites on the eastern side (Constantinescu et al., 2010). The Ditrău Alkaline Massif is considered to represent an intrusion body with an internal zonal structure, which was emplaced into pre-Alpine metamorphic rocks of the Bucovinian nappe complex close the Neogene – Quaternary volcanic arc

of the Calimani-Gurghiu- Harghita Mountain chain (Kräutner & Bindea, 1995, 1998).

The massif lies at the innert border of the Mesozoic crystalline zone, within the Tulghes Group (Tulghes Terrane, according to Balintoni et al., 2009). The alkaline massif of Ditrău has an intrusive character and its trend of enrootment has been by petrologic and geophysical arguments, too. It constitutes a multistage magmatic intrusion in a high level of the Earth's crust (Atanasiu et al., 1994). Parts of the Ditrău massif are uncomformably overlain by andesitic piroclastics with some interbedded basalt-andesite lava flows from the Neogene Harghita – Calimani and by Pliocene to Pleistocene lignite-bearing lacustrine deposits of the Jolotca basin (Rădulescu, 1973).

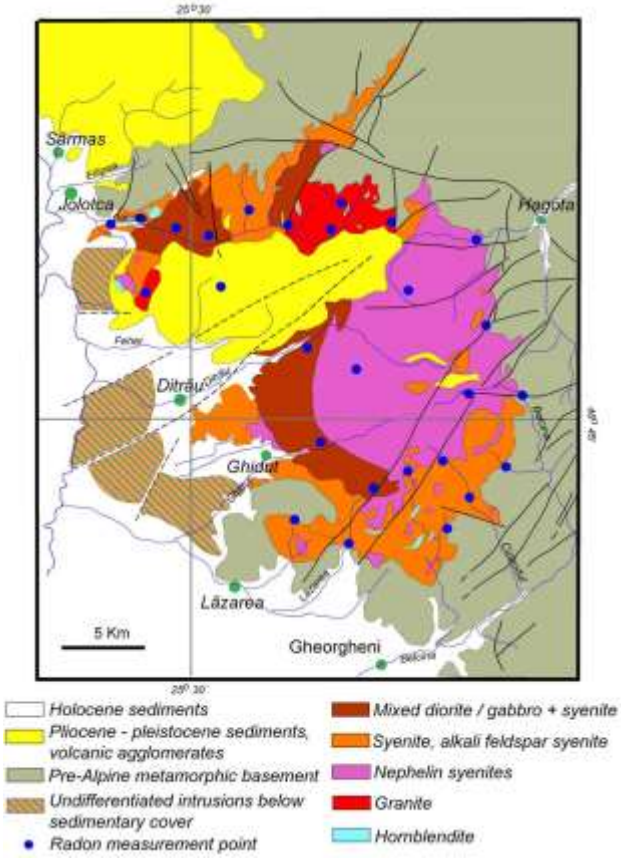

Figure 1. Geological map of Ditrău Alkaline Massif and radon measurement point (simplified from Kräutner and Bindea, 1995).

The center of the Ditrău massif was formed by nepheline syenite, which is surrounded by syenite and monzonite. The north–western and north–eastern marginal sectors are composed of hornblende gabbro/hornblendite, alkali diorite, monzodiorite, monzosyenite and alkali granite. Small discrete ultramafic bodies (kaersutite-bearing peridotite, olivine, pyroxenite and hornblendite) and alkali gabbros occur in the Jolotca area (Morogan et al., 2000). Hornblende gaabro/hornblendite and diorite represent the earliest intrusive phase, and are embedded within younger syenite and granite Dallmeyer et al., 1997; Morogan et al., 2000). All these rocks are cut by late-stage dykes with a large variety of composition including tinguaite, phonolite, nepheline syenite, microsyenite, and aplite (Codarcea et al., 1958; Streckeisein & Hunziker, 1974; Atanasiu & Constantinescu, 1984 Atanasiu et al., 1994).

3. MEASUREMENT METHODS

Radon activity concentration in outdoor air was measured using a portable alpha radon monitor AB-5 (Pylon Electronic Development Company, Ltd. Canada) with Continuous Passive Radon Detector. The Pylon AB-5 equipment includes a detector, which was connected to a photomultiplier and a system of data acquisition based on a microprocessor.

The system uses for alpha detection a passive scintillation cell Silver Activated Zinc Sulfide (ZnS(Ag)) detector which uses the principle of passive diffusion as the sampling method with data acquisition and storage system for radon levels as low as 0.3pCi/l. (Călin et al., 2012).

The system was set to operate in *continuous mode* with the calibration factors of 0.060 ± 3.58% cpm/(Bq/m^3) (1.664±4%). The radon concentration in air was calculated with the following equation:

$$CRn = \frac{CR/\Delta T - CB}{K} [Bq/m^3] \qquad (1)$$

where CRn is the radon concentration expressed in Bq/m^3, CR is the integral counts of alpha-radiation of radon expressed in counts per minute (cpm) for the time interval ΔT (min), CB is the counts of the background (cpm), K is the sensitivity. The output data of the radon monitor AB-5 were processed with the specialized software *Transfer Utility 1.1* (DTU - Data Transfer Utility)

Radon in outdoor air was measured in situ, in 5 points, for each rock type within the study area (Fig. 1). The radon concentration was measured at a height of 15 cm above ground level. The radon exhalation rate was continuously measured for 24 hours with a counting time of 20 minute/interval in each site. The data recorded in the first 3 hours were not taken into account because this time is necessary for the system to reach its equilibrium point (Călin et al., 2011). After radon concentration measurements in the field, a spatial point grid was built (using the geographic coordinates of each measuring point). With each point were associated the measured values of radon

concentration. *Topo to raster* is the interpolation method that was chosen to obtain the spatial distribution of radon concentration in the massif area. The software used for graphical representation and interpolation of the data is Arc GIS 9.3 from ESRI.

4. RESULTS AND DISCUSSION

The results of the measured values of the radon concentrations are shown in table 1.The results of measurements for different locations in the area under study for ^{222}Rn in the range of 2.6 to 52 Bq m^{-3}, with a median value of 20.35 Bq m^{-3}. These values are not group plotted within the investigated area, thus reducing the degree of precision in interpretation (Fig. 2).

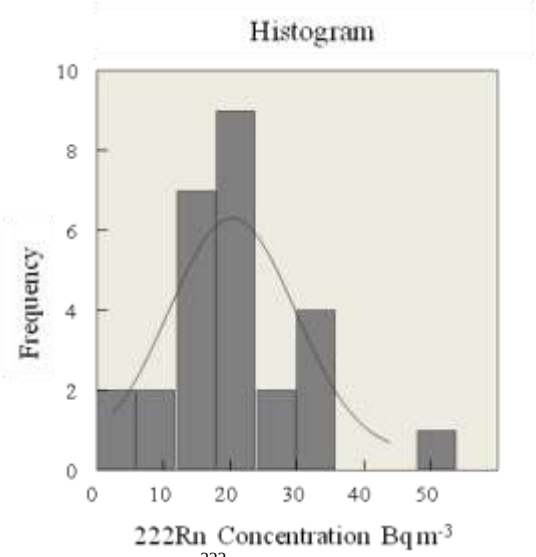

Figure 2. Outdoor ^{222}Rn - Frequency distribution

The spatial distribution of radon in the case analyzed indicates values higher than normal for most of the investigated area (fig.3). US EPA (Environmental Protection Agency) indicated the natural outdoor level of radon gas (0.4 pCi/L) as the target radon level for indoor radon levels (US EPA, 1999; Zeeb & Shannoun, 2009).

The variation of the radon concentration and release of radon from rocks is directly controlled by the minerals in which uranium and thorium occur. In igneous rocks uranium appears in the valence state U^{4+} with crystallochemical properties close to Th^{4+} and the Rare Earth Elements (REE), which explains the coherent geochemistry of U, Th and REE in igneous rocks (IAEA - TECDOC - 1363, 2003). U^{4+} and Th^{4+} becomes concentrated in late stage of magmatic differentiates in accessory minerals: zicon - $ZrSiO_4$, monazite – $(Ce,La)PO_4$, titanite $(Ca,TiSiO_5)$, allanite - $(Ce,Ca, Y,La)_2 (Al, Fe^{3+})_3 (SiO_4)_3(OH)$,

Table 1. Location of the ^{222}Rn outdoor air measurement point and ^{222}Rn concentration in the Massif Ditrău area.

No. measurement point	GPS position: longitude latitude	^{222}Rn concentration Bq m^{-3}
1	25^{0}33'25"E 46^{0}44'57"N	3.9
2	25^{0}31'40"E 46^{0}45'44"N	2.6
3	25^{0}33'38"E 46^{0}45'59"N	32.54
4	25^{0}34'52"E 46^{0}45'27"N	19.16
5	25^{0}35'21"E 46^{0}46'21"N	19.71
6	25^{0}35'22"E 46^{0}46'00"N	26.32
7	25^{0}36'05"E 46^{0}45'34"N	17.84
8	25^{0}37'38"E 46^{0}45'25"N	18.77
9	25^{0}38'49"E 46^{0}46'58"N	19.12
10	25^{0}38'02"E 46^{0}47'40"N	30.49
11	25^{0}40'56"E 46^{0}47'50"N	19.87
12	25^{0}39'34"E 46^{0}48'55"N	52.00
13	25^{0}38'05"E 46^{0}48'36"N	24.15
14	25^{0}36'59"E 46^{0}49'20"N	17.14
15	25^{0}35'23"E 46^{0}50'02"N	18.16
16	25^{0}32'57"E 46^{0}49'36"N	17.88
17	25^{0}33'32"E 46^{0}49'35"N	9.78
18	25^{0}32'10"E 46^{0}51'56"N	14.75
19	25^{0}33'05"E 46^{0}52'22"N	20.15
20	25^{0}30'55"E 46^{0}51'48"N	15.04
21	25^{0}34'23"E 46^{0}51'49"N	18.41
22	25^{0}30'42"E 46^{0}51'56"N	14.12
23	25^{0}29'23"E 46^{0}52'08"N	17.05
24	25^{0}35'11"E 46^{0}52'37"N	35.12
25	25^{0}28'35"E 46^{0}52'00"N	35.47
26	25^{0}29'23"E 46^{0}52'03"N	20.14
27	25^{0}29'23"E 46^{0}52'08"N	10.02

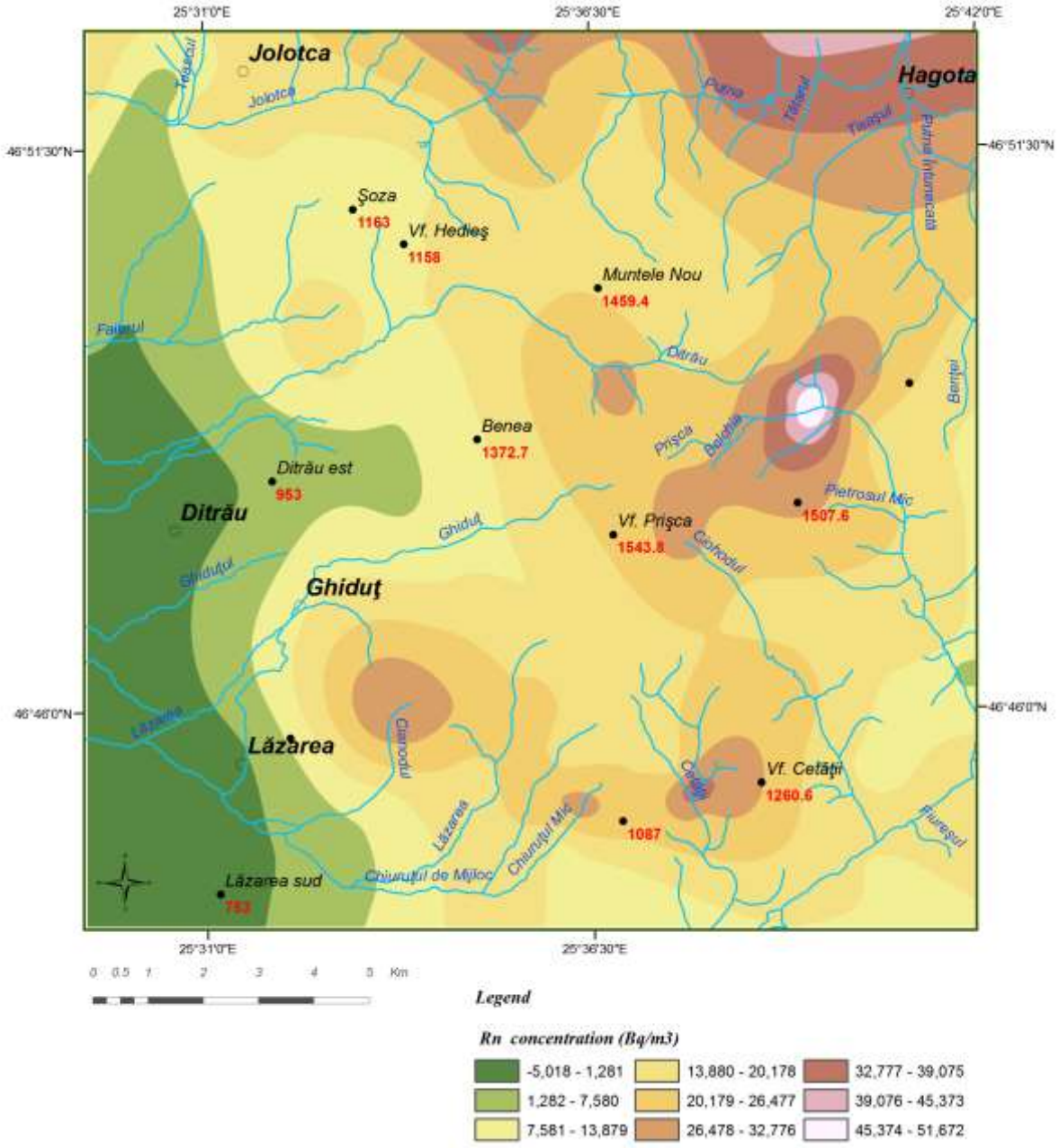

Fig. 3. Spatial distribution of Radon in Ditrău massif area.

apatite - $Ca_5(PO_4)_3(F,Cl,OH)$, xenotime - YPO_4, thorite - ThO_2, pyroclore - $Na,Ca)_2Nb_2O_6(OH,F)$, bastnäsite - $Ce,La,Y)CO_3F$, parisite - $Ca(Ce,La)_2(CO_3)_3F_2$.

The lowest values of radon content 2.6 Bq m^{-3} respectively 3.9 Bq m^{-3} were measured in Lăzarea zone and are geologically associated with crystalline limestone. The highest radon potential radon are associated with marginal syenitic rocks (red syenite ± liebnerite) which occur in the eastern part of the massif (52 Bq m^{-3}), Aurora area (Belcina Valley). The Pb, Zn + REE + Mo + Th vein mineralization from Belcina play an important role in generating increased levels of radon in the area. Elevated radon potential is also associated in the center of the massif with syenite, nephelin syenite and aplitic rocks from Prişca zone.

The relative increase of radon concentration in the air measured in Cianod -Hereb area could be explained by a contribution of ^{222}Rn exhalation area generated by the disseminated mineralization of the Mo, REE, Zr and Th which occurs in red syenites.
Slightly elevated values of radon content are genetically associated with granite which occur in north and north–east of massif area. In hornblendite, gabbro/hornblendite, alkali diorite, monzodiorite, monzosyenite and alkali granite which composed the marginal sector from north - western of massif,

radon outdoor concentration shows normal values for basic rocks.

5. CONCLUSION

The study shows that average radon in the air in Ditrău massif area is higher than recommended values. For all types of rocks the radon measurements confirm that radon activity increases from basic to acidic rocks and depends on the uranium and thorium concentration of rocks. Another aspect of the geochemistry of radon is the spatial variation on the some types of rocks. This difference in the distribution of radon is consistent with the concentration of both uranium and thorium mainly in accessory minerals (zircon, monazite, allanite, apatite, xenotime). The irregular distribution of the accessory minerals probably directly influences the oscillation range of radon values.

All these, clearly indicate that radon concentration depend on the lithology which is the primary source of radon indoor. This study also will make possible a risk assessment given by radon emanation in Dritău massif inhabited area.

REFERENCES

Atanasiu N. & Constantinescu E., 1984. *Contributions á la connaissance pétrologique et structurale du massif alcalin de Ditrău.*Analele Universitătii. Bucuresti, XXXIII, 43 - 49.

Atanasiu N., Garbaşevschi N., Jakab G & Vlad S., 1994. *Mesozoic rift related magmatism / metallogeny at Ditrău.* In Borcos M. & Vlad, S. (eds.). Plate tectonics and metallogeny in the East Charpatians and Apuseni Mts.,. Field trip guide, IGCP Project No. 356 Bucharest, Geological Institute of Romania , pp 14 – 16.

Balintoni I., Balica C., Ducea M.N., Fikun Chen, Hann H.P.& Şablovschi V., 2009. *Late Cambrian-Early Ordovician Gondwanan terranes in the Romanian Charpatianas: A zircon U-Pb provenance study.* Gondwana Research, 119 – 133.

Călin M. R., Zoran M. & Călin M. A., 2012. *Radon levels assessment in some Northern Romanian salt mines.* Journal of Radioanalytical and Nuclear Chemistry, 293: 565 - 572.

Călin M. R., Simion, C.A., Simionică, Gh. I, Călin M.A. & DrukerA. E., 2011. *The Characterization of the radioactivity in the Cacica salt mine.* Romanian Reports in Physics, 63, 2, 483 - 502.

Codarcea A, Codarcea-Dessila, M.& Ianovici, V., 1958. *Structure géologique du massif des roches alcalines de Ditrău.* Révue Roumaine de Géologie, Géophysique et Géographie, Géologie, 1, 1-135.

Constantinescu E., Atanasiu N. & Iancu O.G., 2010. *The Ditrău Alkaline Massif.* Acta Mineralogica-Petrographica, Field Guid Series, 19, 22 - 25.

Cosma C., Szacsvai K., Dinu A., Ciorba D., Dicu T. & Suciu L., 2009, Preliminary integrated indoor radon measurements in Transylvania (Romania), Isotopes Environ Health Stud.,45(3):259-68.

Dallmeyer R.D., Kräutner H.G. & Neubauer F., 1997. *Middle - late Triassic $^{40}Ar/^{39}Ar$ hornblende ages for early intrusion within the Ditrău Alkaline Massif, Romania: implication for Alpine rifting in the Carpathian orogen.* Geologica Charpatica, 48, 347 - 352.

Dang Duc Nhan, Carvalho P. Fernando, Nguyen Thi Thu Ha, Nguyen Quang Long, Dao Dinh Thuan & Heloisa Fonseca, 2012. *Radon (^{222}Rn) concentration in indoor air near the coal mining area of Nui Beo, North of Vietnam.* Journal of Environmental Radioactivity, 110, 98 - 103.

IAEA - TECDOC - 1363, 2003. *Guidelines for radioelement mapping using gamma ray spectrometry data.* International Atomic Energy Agency, Viena, 115 – 122.

Kräutner H.G. & Bindea G., 1995. *The Ditrău alkaline intrusive complex and its geological environment.* Romanian Journal of Mineralogy , In Guidebook to excursion E, (3rd Symposium on Mineralogy, Bucharest), 77, 3, 18.

Kräutner H.G. & Bindea G., 1998 *Timing of the Ditrău Alkaline Intrusive Complex (Eastern Charpatians, Romana).* Slovak Geological Magazine, 4, 213 – 221.

Morogan V., Upton, B.G.J. & Fitton J.G., 2000. *The petrology of the Ditrău alkaline complex, Eastern Carpathian.,* Mineralogy and Petrology, 69, 227 – 275.

Rădulescu D., 1973. *Considerations on the origin of magmas of the Neozoic subsequent volcanism in the East Charpatians.* Annuala Report , Geological Institute of Romania 41, 67 – 76,

Streckeisen, A. & Hunziker, J.C., 1974. *On the origin and age of the nepheline-syenite complex of Ditró (Transylvania, Rumania).* Schweizer Mineralogische Mitteilunge, 54, 1, 59 - 77.

UNSCEAR Report, 2000. *Sources and effects of ionizing radiation.* Report to the general assembly with scientific annexes, United Nation, New York , 89 - 105.

US EPA, 1999. *Indoor Air Quality, Objectives - Radon.* US EPA Publication.

Zeeb H. & Shannoun F. (Eds), 2009. *WHO Handbook on Indoor Radon: A Public Health Perspective. World Health Organization press, 91.*

Received at: 11. 01. 2013

Revised at: 08. 04. 2013
Accepted for publication at: 22. 04. 2013
Published online at: 26. 04. 2013