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An approach to define potential radon emission level maps using indoor radon concentration measurements and radiogeochemical data positive proportion relationships

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A B S T R A C T

The aim of this paper is to present the first step of a new approach to make a map of radonprone areas showing different potential radon emission levels in the Quebec province. This map is a tool intended to assist the Quebec government in identifying populations with a higher risk of indoor radon gas exposure. This map of radon-prone areas used available radiogeochemical information for the province of Quebec: (1) Equivalent uranium (eU) concentration from airborne surface gamma-ray surveys; (2) uranium concentration measurements in sediments; and (3) bedrock and surficial geology. Positive proportion relationships (PPR) between each individual criterion and the 1417 available basement radon concentrations were demonstrated. It was also shown that those criteria were reliable indicators of radon-prone areas. The three criteria were discretized into 3, 2 and 2 statistically significant different classes respectively. For each class, statistical heterogeneity was validated by Kruskal–Wallis one way analyses of variance on ranks. Maps of radon-prone areas were traced down for each criterion.

Based on this statistical study and on the maps of radon-prone areas in Quebec, 18% of the dwellings located in areas with an equivalent uranium (eU) concentration from airborne surface gamma-ray surveys under 0.75 ppm showed indoor radon concentrations above 150 Bq/m3. This percentage increases to 33% when eU concentrations are between 0.75 ppm and 1.25 ppm and exceeds 40% when eU concentrations are above 1.25 ppm. A uranium concentration in sediments above 20 ppm showed an indoor radon concentration geometric mean of 215 Bq/m3 with more than 69% of the dwellings exceeding 150 Bq/m3 or more than 50% of dwellings exceeding the Canadian radon guideline of 200 Bq/m3. It is also shown that the radon emission potential is higher where a uranium-rich bedrock unit is not covered by a low permeability (silt/clay) surficial deposit.

1. Introduction

Radon is a naturally occurring noble gas which is colourless, odourless and tasteless. It is found in soil and rocks and drifts upward from the ground to the outdoor air. As a highly volatile gas, radon is quickly diluted to harmless levels in exterior environments. However, it can also infiltrate dwellings through cracks or holes in the foundations and can reach high indoor concentration levels. Because radon gas has a higher density than air, it tends to accumulate in basements, the lowest dwelling level and also the least ventilated (Dessau et al., 2004).

There are three natural radon isotopes, but radon-222 (^{222}Rn) is the most stable and decays with a half-life of 3.823 days. ^{222}Rn is produced by the radioactive decay of radium-226 (^{226}Ra), both daughter elements of uranium-238 (^{238}U). ^{220}Rn and ^{219}Rn (decay products of thorium-234 and uranium-235 respectively) are of no concern from an air quality point of view because their half-lives are short ($^{219}Rn = 3.96$ s and $^{220}Rn = 55.6$ s) (Nambi and Aitken, 1986) and as they cannot move very far. Because of its longer half-life, ^{222}Rn can relocate from soil to indoor air before emitting a

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high energy alpha particle of 5.49 MeV (Nero et al., 1990). Daughter products of ^{222}Rn are solid elements and may attach to dust and aerosol particles.

This radioactive dust may be deposited on epithelial tissue of the lungs when inhaled. Radon progeny, especially polonium-218 and polonium-214, also emit high energy alpha particles (6.00 MeV for ^{218}Po and 7.69 MeV for ^{214}Po) and decay with short half-lives (^{218}Po = 3.04 min and ^{214}Po = 164 μs) (Nero et al., 1990). Epidemiologic studies concluded that alpha particles emitted from radon gas and from radon progeny can alter DNA of the cells lining lungs and may lead to lung cancer. Radon is considered to be the second leading cause of lung cancer after tobacco smoking (WHO, 2009).

The World Health Organization estimates that 3%–14% of lung cancers are attributable to radon (WHO, 2009) which result in 21,000 radon related deaths per year in the United States (USEPA, 2003) and proportionally to more than 600 deaths per year in Quebec (CCSSC, 2009). The United States Environmental Protection Agency (USEPA) classified radon as a "A class" cancer-causing substance (Dessau et al., 2004) and the International Agency for Research on Cancer (IARC), as a "group 1" substance (IARC, 1988). Those agencies concluded that "radon and its decay products are carcinogenic to humans" (IARC, 1988).

In agreement with recent epidemiological studies, Health Canada lowered the Canadian guideline from 800 to 200 Bq/m^3 in 2007 (Health Canada, 2007) and Quebec intersectorial radon committee prepared an *Action plan about radon*, effective in 2008. One of the main objectives of the *Action plan* was to develop a map of radon-prone areas for the entire province to help public health authorities identify radon potential zones and inform populations living in these regions.

The objective of this paper is to show that radiogeochemical data and indoor radon concentration measurements PPR can be used to define different radon potential levels. Such a study was initiated in the 90's by Martel (1991) and Lévesque et al. (1995). Recent indoor radon measurements and radiogeochemical data enable us to confirm the relationships between radiogeochemical data and indoor radon concentrations and to map different levels of indoor radon exposure.

Therefore, the paper presents the conclusions from a previous literature survey of the mostly used criteria for mapping radon-prone areas (Drolet, 2011), the available dataset used in Quebec to make such a map, a PPR and a statistical analysis between indoor radon concentrations and radiogeochemical information and, finally, a map for each selected criterion showing different levels of radon emission potential.

2. Survey of criteria used for mapping radon-prone areas

A redundancy analysis of radon mapping criteria (Drolet, 2011) (Table 1) shows that 89% of 62 studies published during the last 25 years used indoor radon concentrations to map radon-prone areas.

The presence of a radon source is an essential condition for having high indoor radon concentrations in dwellings. Uranium rich rocks (and sediments derived from them) are generally the main source of radon in the Quebec area (Martel, 1991). Many mapping projects used geology as a primary criterion for locating radon-prone areas (Alexander and Devocelle, 1997; Apte et al., 1999; Gundersen and Schumann, 1996; Kemski et al., 2001, 2008; Miles and Ball, 1996; Zhu et al., 2001) and 66% of the surveyed international works employed this approach.

Uranium-rich sedimentary rocks are commonly associated with high organic matter content (Harrell et al., 1991; Martel, 1991; Poirson and Pagel, 1990). Uranium adsorption by organic matter is effective when carbonaceous materials are present in a reducing environment (Fertl and Chilingar, 1988). Such conditions are especially met during the formation of black shales in marine environments. Organic-rich black shales have uranium concentrations ranging from 8 ppm to 168 ppm (aluminium-rich shale) (Wedepohl, 1969). Fig. 1 shows the correlation between uranium concentrations and total organic carbon (TOC) in the Ohio Shale.

For igneous and metamorphic rocks, uranium concentration is usually associated with silica (SiO_2) content. Felsic rocks (silica content greater than 65% by weight) such as granites have generally higher uranium concentrations than mafic and ultramafic rocks (Wedepohl, 1969). Felsic rocks have the highest uranium content, but intermediate rocks (silica content between than 52% and 65% by weight) can also have high values. With uranium content one or two orders of magnitude higher than that of mafic and ultramafic rocks, felsic and intermediate volcanic rocks are considered as likely radon sources.

There is no direct link between uranium-rich bedrock and elevated indoor radon concentrations. In fact, interactions are numerous and have been identified previously in the literature (Nazaroff, 1992; Nazaroff et al., 1987; Nguyen et al., 2011). Radon emanation and migration depends on porosity, fractures, permeability of geological units, depth of the water table and saturation,

Table 1
Redundancy of the most used indoor radon mapping criteria surveyed from 62 international studies (surveyed studies are listed in Drolet, 2011).

Radon mapping criteria	Redundancies: proportion of studies using the criteria (%)
Indoor radon concentrations	89%
Bedrock units	66%
Surficial deposits	35%
Equivalent uranium (eU) concentration from surface gamma-ray measurements	34%
Soil gas radon concentration	31%
Building characteristics	26%
Geochemistry (uranium concentration in sediment samples)	23%
Temperature	6%
Precipitation	2%

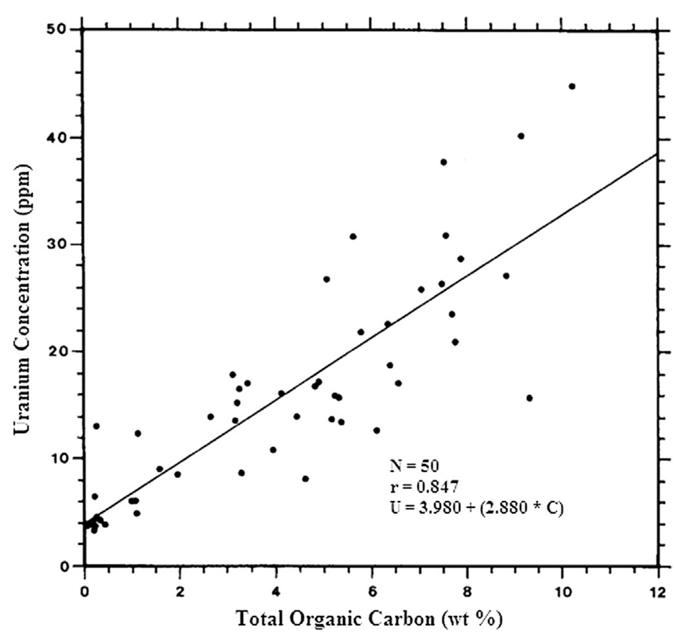

Fig. 1. Scatter plot and linear regression for uranium concentration and total organic carbon (TOC) in the Ohio Shale (adapted from Harrell et al., 1991).

dissolution/migration/precipitation of uranium and radium. Radon gas emitted from the underlying bedrock must be able to migrate through bedrock to near-surface soil before reaching house basements and decaying into its daughter product 218Po. Therefore, soil permeability is the main surficial deposit characteristic reported in 35% of the surveyed studies. Sediment permeability controls radon mobility and hence the distance that radon can travel upward (Alexander and Devocelle, 1997). Coarse grained surficial sediments like sand or gravel have high permeabilities and provide excellent pathways for upward radon migration. Finer grained materials such as clay and silt, with significantly lower permeabilities, can form near-impermeable barriers to radon migration (Alexander and Devocelle, 1997; Miles and Ball, 1996). The silt/clay barriers are effective only when they are thick enough to avoid being penetrated by buildings foundations (Miles and Ball, 1996). While some marine silt and clay sediments may be derived from uranium-rich rocks or glacial sediments and may thus constitute radon sources themselves, the scarcity of data on uranium concentrations in Late Quaternary marine sediments of southern Quebec does not allow us to further investigate this possibility. However, only low uranium concentrations in glacial and glaciomarine sediments have been reported in geochemical surveys from uranium-rich Shield terrains north of Gatineau (Kettles and Shilts, 1989), which suggests that generally low uranium concentrations may be expected in southern Quebec fine-grained marine sediments.

One third of the surveyed international studies used airborne surface gamma-ray surveys to predict radon-prone areas. In the literature, this criterion is considered effective to identify radon-prone areas (Akerblom, 1995; Ball et al., 1995; Chen, 2009; Christensen and Rigby, 1995; Doyle et al., 1990; Drolet, 2011; Duval, 1989; Ford et al., 2001; Heincke et al., 2008; Jackson, 1992; Kline et al., 1989; Lévesque et al., 1995; Martel, 1991; Mose et al., 1990; Pellerin et al., 1999; Savard et al., 1998; Smethurst et al., 2008). Earlier studies carried out by Martel (1991) and Lévesque et al. (1995) in Quebec set 2 ppm in eU as the threshold for regions with a high indoor radon potential.

Also, uranium concentration in sediments (geochemical data) has been used as a proxy for radon potential in 23% of the surveyed international studies. Martel (1991) and Lévesque et al. (1995) used a uranium concentration in sediments above 4 ppm to identify radon-prone areas.

Even if they are often used in international radon studies, soil gas radon concentrations and building characteristics were not included in our study because of the overall lack of reliable data. Other criteria such as temperature and precipitation were also ignored (Table 1).

The next section shows the available radiogeochemical data that were used to define the radon-prone areas in Quebec.

3. Available datasets and selection of criteria for the Quebec study

In our study, five datasets were used to map radon-prone areas: (1) indoor radon concentrations, (2) equivalent uranium (eU) concentrations from surface gamma-ray measurements, (3) uranium concentrations in sediments, (4) bedrock units and (5) surficial deposits.

3.1. *Indoor radon concentration measurements*

1417 basement radon measurements were collected by different organizations between 2008 and 2011 (Fig. 2). 90 measurements were collected by Radioprotection inc. (a private company) and Health Canada in one specific area in north-western Quebec in 2008. 585 alpha-track detectors sampled indoor radon concentrations in 65 schools from eastern and central-western Quebec during winter 2010. These measurements were made on different floors, (Poulin and Leclerc, 2010) but only the 48 basement radon measurements were included in the dataset. Quebec Lung Association (QLA) provided the remaining 1279 basement radon concentrations measured with alpha-track detectors. Basement radon concentration measurements are the only ones used in this study. Basement radon concentrations are generally higher than values typical of the first floor of the same building. The goal of the exclusion of the first floor data was to generate the worst case scenario. Radon measurements were scattered all over Quebec from volunteers who had chosen to test radon concentrations in their dwellings. Naturally, higher populated regions had more radon tested dwellings. For the purpose of mapping, the

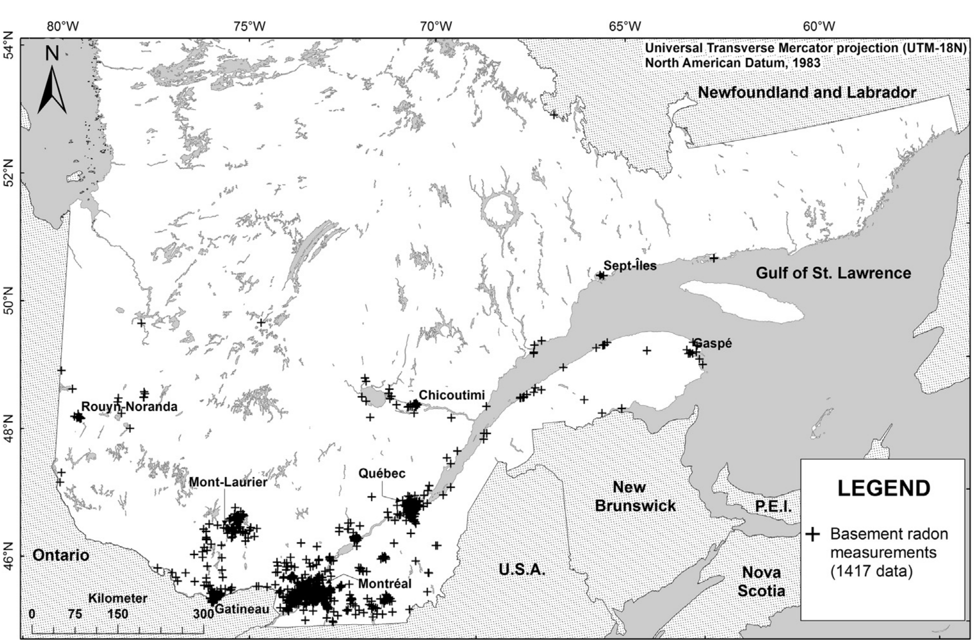

Fig. 2. Locations of the 1417 basement radon concentration measurements across Quebec.

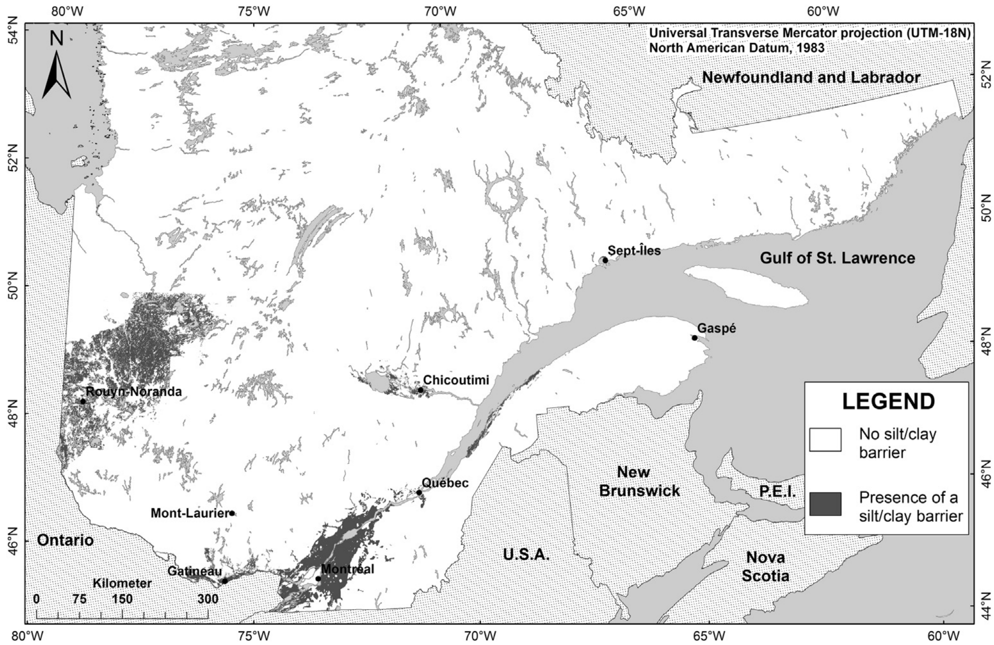

Fig. 3. Location of the low permeability silt/clay barrier across Quebec.

civic addresses of tested dwellings were systematically converted into latitude/longitude coordinates to be included into a geo-referenced database. Web mapping services such as Google Earth, Yahoo! Maps and MapQuest were used to locate the dwellings. When two or more concentrations were measured for a specific address, the maximum measurement was kept in the dataset. All the radon measurements are owned by the Ministère de la Santé et des Services sociaux (MSSS) and were obtained with the homeowner's consent.

3.2. Equivalent uranium (eU) concentrations from airborne surface gamma-ray measurements

Airborne surface gamma-ray surveys provide concentrations of natural radioactive elements (uranium, thorium and potassium) present in rocks or sediments from the analysis of the energy spectrum. Airborne surface gamma-ray surveys were downloaded from Natural Resources Canada web site. Airborne surface gamma-ray measurements mainly cover the southern reach of Quebec (Fig. 7A). Typically, airborne surface gamma-ray surveys are flown with flight line spacing ranging from 200 to 5000 m, at a 120 m terrain clearance. Higher density surveys (200–500 m spacing) were done for detailed mapping purposes while lower density surveys (5000 m) were flown for reconnaissance scale purposes (NRCan, 2007).

Uranium concentrations are generally low and measured in parts per million (ppm). Airborne surface gamma-ray measurements do not measure uranium-238 concentrations directly. The term *equivalent uranium concentration* (eU in ppm) is used because bismuth-214, a ^{238}U daughter product, is measured by the gamma surveys and it is assumed to be in equilibrium with its parent isotope ^{238}U. By measuring a radon-daughter element (bismuth-214) emitted few minutes after the radon-222 decay (Nero et al., 1990), the airborne surface gamma-ray measurements are a fairly direct method of establishing the amount of radon in the upper part of the ground.

3.3. Geochemistry

Uranium concentrations in sediment samples were available online via SIGEOM (GEOMining Information System), a web search engine of the Quebec Ministère des Ressources naturelles et de la Faune (MRNF). SIGEOM is a complete database that contains up-to-date information on mining rights, geology, geochronology, geophysics, geochemistry, deposits, drillings, Quaternary geology and hydrogeology in Quebec. There are 278,841 uranium point samples that provide uranium concentrations from analysed stream and lake sediments, till or soil. Samples were analysed by different analytical methods such as inductively coupled plasma mass spectrometry, fluorometry, neutron activation, atomic absorption, paper chromatography and plasma emission. Uranium concentrations from geochemical surveys were interpolated to a regular grid using the inverse distance weighted (IDW) method implemented in ESRI's ArcGIS (ESRI, 2012; Drolet, 2011). This interpolation technique was used with success by Lamothe (2009) and Trépanier (2009) to identify uranium geochemical anomalies (uranium-bearing sediments) for mineral exploration purposes. The grid dataset covers 80% of the studied territory (Fig. 7B), with the exception of the highly populated Saint-Lawrence Lowlands region of southern Quebec. Also, north-western Quebec is not covered by grid dataset. However, this area is largely uninhabited.

3.4. Bedrock units

The bedrock geology of Quebec is relatively well known. Uranium-rich sedimentary, igneous and metamorphic rocks were tentatively identified on geological maps available online via SIGEOM. Those were maps available at 1:20,000 or 1:50,000 scales and cover the entire province. It is the only radon-prone mapping dataset that covers the entire province of Quebec (Fig. 7C).

In southeastern Quebec (Saint-Lawrence Lowlands and the Appalachians), most bedrock units are grouped into formations based on their lithological characteristics. Sedimentary black shales and intermediate/silicic volcanic rock units in those two geological and physiographic domains were identified from descriptions found in Globensky et al. (1993).

In northern and north-western Quebec (Canadian Shield), rock units are rarely grouped into formations. Radon-prone igneous and metamorphic rocks were identified from their mineralogy as documented in Moureau and Brace (2000). A Streckeisen double triangle was also used to identify intermediate/silicic rocks (Streckeisen, 1976). Potentially uranium-rich bedrock units across Quebec were more specifically identified in Drolet (2011).

3.5. Surficial deposits

Silt/clay surficial sediments were identified on nine Quaternary geological maps (Dredge, 1983; Lamarche, 2011; Lasalle and Tremblay, 1978; Parent, 2012; St-Onge, 2009; Veillette, 1996; Veillette and Cloutier, 1993, 2012; Veillette et al., 2003). These cover only 30% of the studied territory. However, close to 85% of these maps are from areas invaded by postglacial seas (Occhietti et al., 2001); the marine fine grained sedimentation likely resulted in deposition of irregular silt/clay barriers to radon migration. Paper maps were digitized prior to identifying deep water marine, glaciomarine and glaciolacustrine sediments. Surficial sediments inferred to be thinner than 3 m were not included in the silt/clay barrier because most dwelling foundations break through it when the soil is excavated. The silt/clay barrier is shown in Fig. 3.

4. Methodology for PPR and statistical analysis

4.1. PPR between basement radon concentrations and other selected criteria

The PPR (positive proportion relationship) between equivalent uranium concentration from airborne surface gamma-ray measurements and indoor radon concentration measurements was evaluated to verify the capacity of the eU concentration dataset to predict radon-prone areas in Quebec. The 1417 basement radon concentrations were compared to eU concentrations from airborne surface gamma-ray measurements, but only 1077 of the radon measurements coincide with the eU dataset. The 1077 basement radon concentrations were linked to eU concentrations between 0 and 7 ppm. Equivalent uranium concentration from airborne surface gamma-ray measurements were discretized into five classes. The discretization process was carried out because the relationship between indoor radon concentrations and eU concentrations (and the other two criteria) are not straight forward. Factors such as building structure and indoor/outdoor air exchanges also control indoor radon concentrations. Therefore, a small increase in eU concentration is not necessarily associated with a proportional increase in indoor radon concentration. Moreover, all three criteria are obtained from interpolated data and are thus approximated. These errors justify working with classes instead of continuous data. Each proposed class had to respect three conditions: (1) classes cover approximately an equal range of eU concentrations, (2) classes should encompass approximately the same number of radon measurements and (3) there are at least 50 radon measurements per class.

The same process was performed with geochemical data. The 1417 geo-referenced dwellings were superimposed on the layer of uranium concentration interpolated from geochemical surveys. Uranium concentrations ranging from 0 to 76 ppm were extracted for the 448 basement radon concentration measurements covered by the interpolated geochemical layer. Uranium concentrations interpolated from geochemical surveys generated six classes that respect the previously stated three conditions.

Before superimposing basement radon concentrations on *bedrock units* and *surficial deposits*, both geological datasets were merged to create a new variable herein identified as the *geology* criterion. Radon-prone areas based on geology consist of zones where there is an assumed radon source in the underlying bedrock unit (presence of a uranium-rich rock or unconsolidated unit) that is not covered by a silt/clay barrier. The potential to measure a high indoor radon concentration associated to the geology criterion is low when there is no radon source in the subsurface. It is also low when a potentially uranium-rich bedrock unit is overlain by a silt/clay barrier which prevents radon gas migration from deeper in the ground upwards into dwelling basements.

The 1417 geo-referenced dwellings were superimposed on the geology criterion. Two output classes were created from this criterion because there are only two situations considered in this paper: 1) potential for high radon emissions based on geology (uncapped uranium-rich bedrock unit) and 2) potential for low radon emissions based on geology (all other combinations of bedrock units and surficial deposits).

PPR between basement radon concentration measurements and each of the criteria were evaluated by calculating, for each previously proposed class:

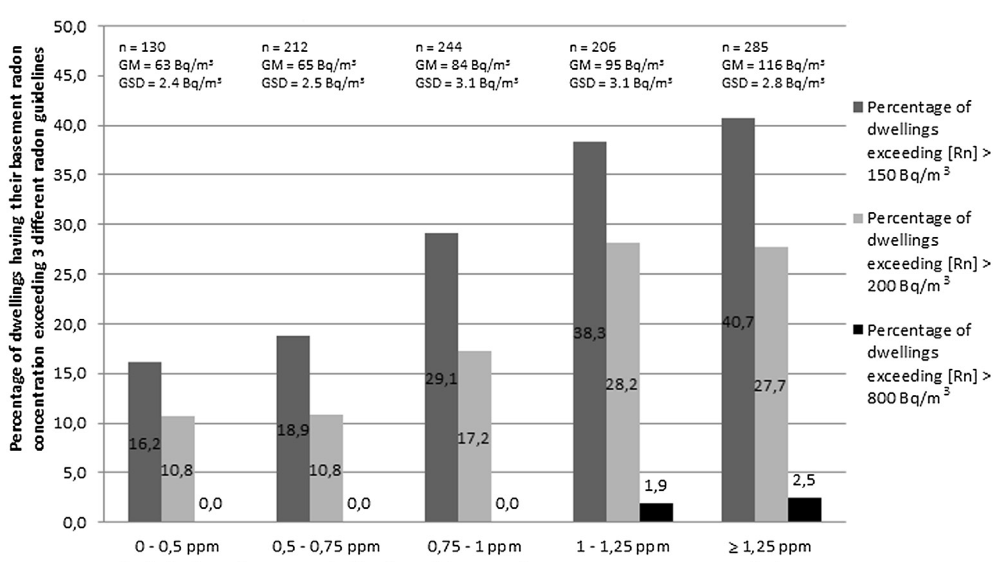

Fig. 4. Graph of equivalent uranium concentration measured by airborne surface gamma-ray surveys discretized into 5 classes as a function of percentage of dwellings having indoor radon concentrations above three existing radon guidelines (made from 1077 indoor radon measurements in basements).

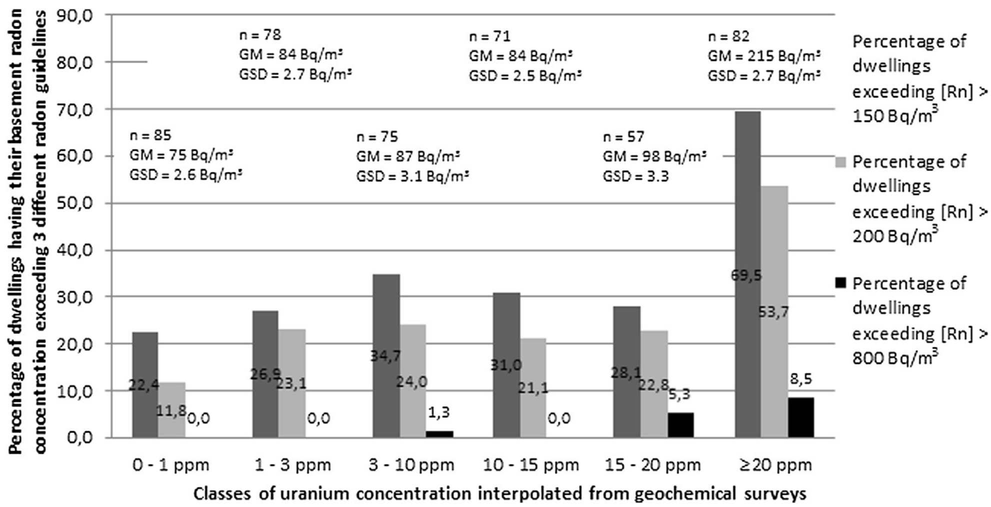

Fig. 5. Graph of uranium concentration measured in sediments discretized into 6 classes as a function of percentage of dwellings having indoor radon concentrations above three existing radon guidelines (made from 448 indoor radon measurements in basements).

- the percentage of dwellings exceeding the United States radon guideline (150 Bq/m^3);
- the percentage of dwellings exceeding the actual Canadian radon guideline (200 Bq/m^3);
- the percentage of dwellings exceeding the previous Canadian radon guideline (800 Bq/m^3);
- the geometric mean (GM);
- the geometric standard deviation (GSD).

Geometric mean (GM) and geometric standard deviation (GSD) allowed us to interpret classes in terms of log-normal distributions and our basement radon concentrations dataset is log-normally distributed.

4.2. Statistical study of relationships between basement radon concentrations and the selected criteria

The Kruskal–Wallis one way analysis of variance on ranks (ANOVA) were performed on indoor radon concentrations for each predetermined class of the three selected criteria (equivalent uranium concentration from surface airborne gamma-ray measurements, uranium concentration in sediments and geology). This was done in order to test if there is a statistically significant difference between each class. A *p*-value, calculated by the ANOVA, lower than 0.05 was set as the threshold for a statistically significant difference. A *p*-value above 0.05 allowed us to merge classes based on statistical similarities. This analysis was carried out for each of the three

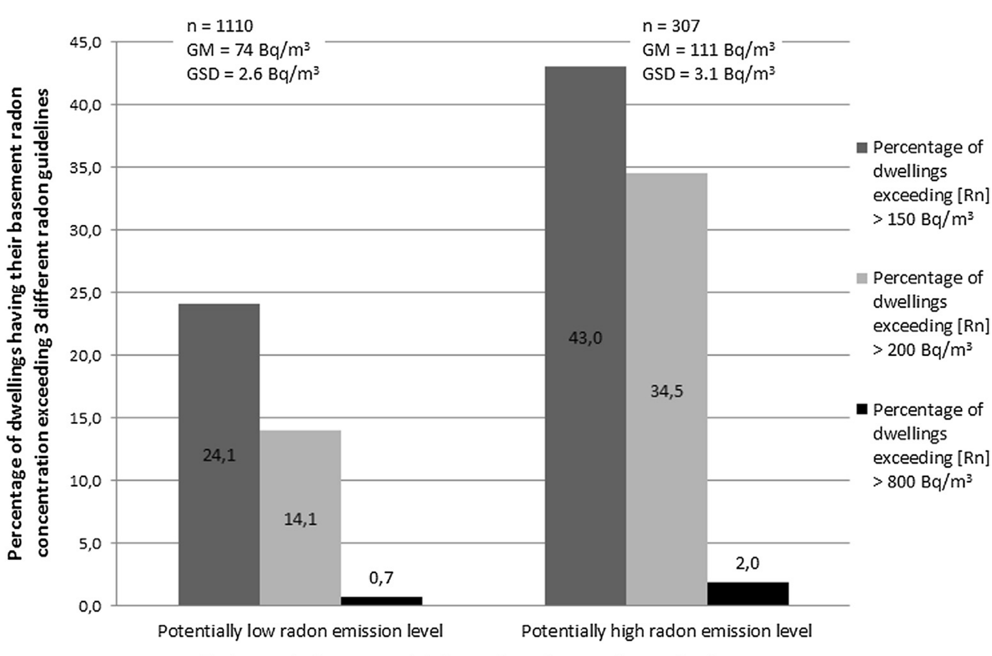

Fig. 6. Graph of the geology criterion discretized into 2 classes as a function of percentage of dwellings having indoor radon concentrations above three existing radon guidelines (made from 1417 indoor radon measurements in basements).

Table 2
Kruskal–Wallis one way ANOVA on five classes of equivalent uranium concentration from airborne surface gamma-ray measurements. A *p*-value >0.05 means that the hypothesis that they share a common mean cannot be rejected at the 95% confidence level (bold characters). The median of indoor radon concentration in basement, in Bq/m^3, is included in parenthesis for each class.

Compared class 1 (median)	Compared class 2 (median)	*p*-value
0–0.5 ppm (63)	**0.5–0.75 ppm (59)**	**0.814**
0–0.5 ppm (63)	0.75–1.00 ppm (81)	0.007
0–0.5 ppm (63)	1.00–1.25 ppm (85)	0.001
0–0.5 ppm (63)	≥1.25 ppm (122)	<0.001
0.5–0.75 ppm (59)	0.75–1.00 ppm (81)	0.004
0.5–0.75 ppm (59)	1.00–1.25 ppm (85)	<0.001
0.5–0.75 ppm (59)	≥1.25 ppm (122)	<0.001
0.75–1.00 ppm (81)	**1.00–1.25 ppm (85)**	**0.233**
0.75–1.00 ppm (81)	≥1.25 ppm (122)	<0.001
1.00–1.25 ppm	**≥1.25 ppm (122)**	**0.065**

criteria individually with SigmaStat's functions featured into SigmaPlot 12 (from Systat Software Inc.).

5. Results

5.1. PPR between basement radon concentrations and other selected criteria

An increase in the percentage of dwellings exceeding three North American radon guidelines is observed from lower to higher classes of equivalent uranium concentration from airborne surface gamma-ray measurements (Fig. 4). The geometric mean increase of indoor radon concentration in the basement of dwellings is in accordance with increasing eU concentrations classes, thus strongly suggesting a statistically significant PPR between those two parameters.

The increase in the percentage of dwellings that exceed North American radon guidelines from low uranium concentrations interpolated from geochemical surveys to higher levels (Fig. 5) is not as obvious as it is for the gamma-ray spectrometry data. The highest class (>20 ppm; Fig. 5*)* shows higher percentages of dwellings exceeding the three guidelines than for the highest class of eU (Fig. 4). Moreover, a higher geometric mean of indoor radon concentration is associated with this class compared to that of the highest class of eU. The first five classes might be statistically homogeneous and a statistical study on the similarities of classes is needed (see section 5.2).

The PPR between indoor radon concentrations in basements and the geology criterion is shown on Fig. 6. The percentage of dwellings exceeding all three North American radon guidelines and the geometric mean of indoor radon concentration are higher for the potentially high radon emission level class compared with the potentially low radon emission level class.

5.2. Statistical study

Statistically significant differences and similarities between each class for all three above selected criteria were validated by

Table 4
Kruskal–Wallis one way ANOVA on six classes of uranium concentration interpolated from geochemical surveys. A *p*-value >0.05 means statistically significant similarities between the tested classes (bold characters). The median of indoor radon concentration in basement, in Bq/m^3, is included in parenthesis for each class.

Compared class 1 (median)	Compared class 2 (median)	*p*-value
0–1 ppm (81)	**1–3 ppm (78)**	**0.602**
0–1 ppm (81)	**3–10 ppm (78)**	**0.479**
0–1 ppm (81)	**10–15 ppm (81)**	**0.470**
0–1 ppm (81)	**15–20 ppm (80)**	**0.422**
0–1 ppm (81)	≥20 ppm (212)	<0.001
1–3 ppm (78)	**3–10 ppm (78)**	**0.987**
1–3 ppm (78)	**10–15 ppm (81)**	**0.839**
1–3 ppm (78)	**15–20 ppm (80)**	**0.642**
1–3 ppm (78)	≥20 ppm (212)	<0.001
3–10 ppm (78)	**10–15 ppm (81)**	**0.945**
3–10 ppm (78)	**15–20 ppm (80)**	**0.765**
3–10 ppm (78)	≥20 ppm (212)	<0.001
10–15 ppm (81)	**15–20 ppm (81)**	**0.863**
10–15 ppm (81)	≥20 ppm (212)	<0.001
15–20 ppm (80)	≥20 ppm (212)	<0.001

Kruskal–Wallis one way analyses of variance on ranks (ANOVA). Table 2 shows *p*-values calculated for all combinations of compared classes (compared class 1 *versus* compared class 2 in Table 2) for the gamma-ray spectrometry criterion.

The "0–0.5 ppm" and "0.5–0.75 ppm" classes are merged because they are not statistically different. Also, statistical similarities exist between "0.75–1.00 ppm" and "1.00–1.25 ppm" classes, and also between "1.00–1.25 ppm" and "≥1.25 ppm" classes. The choice to regroup the "1.00–1.25 ppm" class with the "0.75–1.00 ppm" class instead of grouping with the "≥1.25 ppm" class was based on higher *p*-values (0.233 and 0.065 respectively).

Kruskal–Wallis one way ANOVA analyses were performed on the three new groups ("0–0.75 ppm", "0.75–1.25 ppm" and "≥1.25 ppm") of equivalent uranium (eU) concentration from airborne surface gamma-ray measurements to ensure statistical significance (Table 3). Percentages of dwellings having their indoor radon concentrations in basements exceeding the three North American radon guidelines of 150, 200 and 800 Bq/m^3 are shown as a function of three eU concentration groups.

Statistics presented in Table 3 show that the probability of encountering a higher proportion of elevated radon concentrations increases when eU concentration from airborne surveys is above 0.75 ppm. Regions located in areas with an eU concentration above 0.75 ppm were considered radon-prone areas because Duval (1989) showed that there is a high health risk when 30% of the dwellings in a region exceed 150 Bq/m^3. When the eU concentration is above 1.25 ppm, indoor radon emission potential increases and over 40% of dwellings exceed the 150 Bq/m^3 guideline.

The same validation procedure of classes was made for the geochemistry criterion (Table 4). The classes "0–1 ppm", "1–3 ppm", "3–10 ppm", "10–15 ppm" and "15–20 ppm" were merged because it was impossible to reject the hypothesis that they share a common mean at the 95% confidence level (a *p*-value above 0.05). Two new groups were created: "0–20 ppm" and "≥20 ppm". The statistical dissimilitude between the "0–20 ppm" group and the

Table 3
Kruskal–Wallis one way ANOVA on three groups of equivalent uranium concentration from airborne surface gamma-ray measurements.

Group (x in ppm of eU)	n	Median (Bq/m^3)	25th percentile (Bq/m^3)	75th percentile (Bq/m^3)	GM (Bq/m^3)	GSD (Bq/m^3)	% [Rn] ≥ 150 Bq/m^3	% [Rn] ≥ 200 Bq/m^3	% [Rn] ≥ 800 Bq/m^3
$0 < x < 0.75$	342	59	33	115	64	2.5	17.833.3	10.8	0
$0.75 \le x < 1.25$	450	81	41	185	89	2.8	33.3	22.2	0.9
$x \ge 1.25$	285	122	56	219	116	2.8	40.7	27.7	2.5

$\chi^2 = 53{,}375$ with 2 degrees of freedom. (*p*-value between three groups ≤ 0.001; statistically significant difference).

Table 5
Kruskal–Wallis one way ANOVA on one group and one class of uranium concentration interpolated from geochemical surveys.

Group (x in ppm)	n	Median (Bq/m^3)	25th percentile (Bq/m^3)	75th percentile (Bq/m^3)	GM (Bq/m^3)	GSD (Bq/m^3)	% [Rn] ≥ 150 Bq/m^3	% [Rn] ≥ 200 Bq/m^3	% [Rn] ≥ 800 Bq/m^3
$0 < x < 20$	366	80	39	167	84	2.8	28.4	20.2	1.1
$x \geq 20$	82	212	136	358	215	2.7	69.5	53.7	8.5

$\chi^2 = 52{,}384$ with 1 degree of freedom. (p-value between two groups ≤ 0.001; statistically significant difference).

Table 6
Kruskal–Wallis one way ANOVA on two potential radon emission levels based on the geology criterion.

Group	n	Median (Bq/m^3)	25th percentile (Bq/m^3)	75th percentile (Bq/m^3)	GM (Bq/m^3)	GSD (Bq/m^3)	% [Rn] ≥ 150 Bq/m^3	% [Rn] ≥ 200 Bq/m^3	% [Rn] ≥ 800 Bq/m^3
Potentially low radon emission level based on geology	1110	74	37	144	74	2.6	24.1	14.1	0.7
Potentially high radon emission level based on geology	307	115	44	281	111	3.1	43.0	34.5	2.0

$\chi^2 = 33{,}097$ with 1 degree of freedom. (p-value between two groups ≤ 0.001; statistically significant difference).

"≥20 ppm" class was validated by another Kruskal–Wallis one way ANOVA test (Table 5).

The "≥20 ppm" class of uranium in sediments has 70% of the dwellings exceeding the 150 Bq/m^3 guideline and is considered to represent a high health risk as suggested by Duval (1989). Also, the geometric mean of indoor radon concentration of this class is 215 Bq/m^3 which is higher than the actual Canadian indoor radon guideline. The presence of such uranium concentrations interpolated from geochemical surveys in a region indicates that it is a radon-prone zone.

The last Kruskal–Wallis one way ANOVA on ranks was carried out in order to determine if there is a significant statistical difference between measured basement radon concentrations with potentially low and high radon emission levels as defined by the geology criterion (Table 6). A p-value lower than 0.001 confirms that the populations of radon values assigned to the low and high radon potential classes are statistically different and that the high radon potential class can be considered as a radon-prone area. This class has 43% of dwellings exceeding an indoor radon concentration of 150 Bq/m^3.

5.3. Maps

Fig. 7 shows the three proposed criteria used in Quebec for radon mapping (A: equivalent uranium concentration from airborne surface gamma-ray measurements with three classes; B: uranium concentration in sediment geochemistry with two classes; C: geology with two classes). Based on the statistical study with the three criteria and by combining radiogeochemical information from Figs. 7A, 7B and 7C, it is possible to determine a radon emission potential map. Regions where each criterion indicates a high potential radon emission level (Map A: "≥1.25 ppm"; B: "≥20 ppm"; C: "high potential of measuring an elevated indoor radon concentration") were considered as radon-prone areas. Dwellings built in south-western Quebec could potentially have high basement radon concentration measurements based on radiogeochemical information. Fig. 7A and C show high potential radon emission levels for south-eastern Quebec not covered by the geochemistry criterion (see Fig. 7B) and is considered as radon-prone. If a region has only one criterion with a high radon emission potential, the zone only has a medium potential for high indoor radon concentrations. Identification of potential radon emission level is less accurate in zones only covered by the geology criterion. The Montréal region and the region north of Montréal are the only highly populated zones in this situation; where an airborne surface gamma-ray survey is needed to increase radon mapping accuracy.

The synthesized provincial map of radon-prone areas is not presented herein because a more complicated approach than the simple addition of potential radon emission level maps has been used. The approach used (Kruskal–Wallis one way ANOVA on ranks to combine radon emission potential maps of radiogeochemical information) is explained in details in another paper (Drolet et al., 2013). This map shows four different levels of indoor radon exposure for the whole province of Quebec.

Fig. 8 shows a map of radon potential based on basement radon measurements only. This is a direct way of estimating the radon potential. It is useful to evaluate the radon model predictions presented in Fig. 7A, B and C. This radon potential map based on direct indoor radon measurements was generated by calculating the proportion of dwellings above the action level of 200 Bq/m^3 for each municipality. A minimum of 5 basement radon measurements was required to assign a radon potential to the municipalities.

6. Discussion

Indoor radon concentration sampling has not been done randomly. It is plausible that a significant proportion of the measurements were performed because homeowners had some reason to suspect their home might have a high radon concentration. This could result in an overrepresentation of high values in the dataset (Burke and Murphy, 2011). To illustrate this effect, it is interesting to note that the proportion of homes above 200 Bq/m^3 is 10.8% in areas where eU concentration is between 0 and 0.75 ppm (Table 3), which is comparable to the proportion recently published by Health Canada for the entire province (10.1%; Health Canada, 2012). These considerations emphasize the need to obtain new data from all regions of the province, if possible randomly, to improve the external validity of the model.

Also all three criteria contain uncertainties and may create variability in the results obtained in the PPR and the statistical studies. Uranium concentrations from geochemical surveys were interpolated by the inverse distance weighted technique on ArcGIS creating variability in the model. Equivalent uranium concentrations from gamma-ray surveys and the bedrock units were interpolated by kriging also showing variability. Also, bedrock units come from many local geological maps that were all merged together. Some non-geological boundaries can be seen in Fig. 7C because the ways rock types have been identified by different geologists. The less precise variable was the silt/clay barrier. The

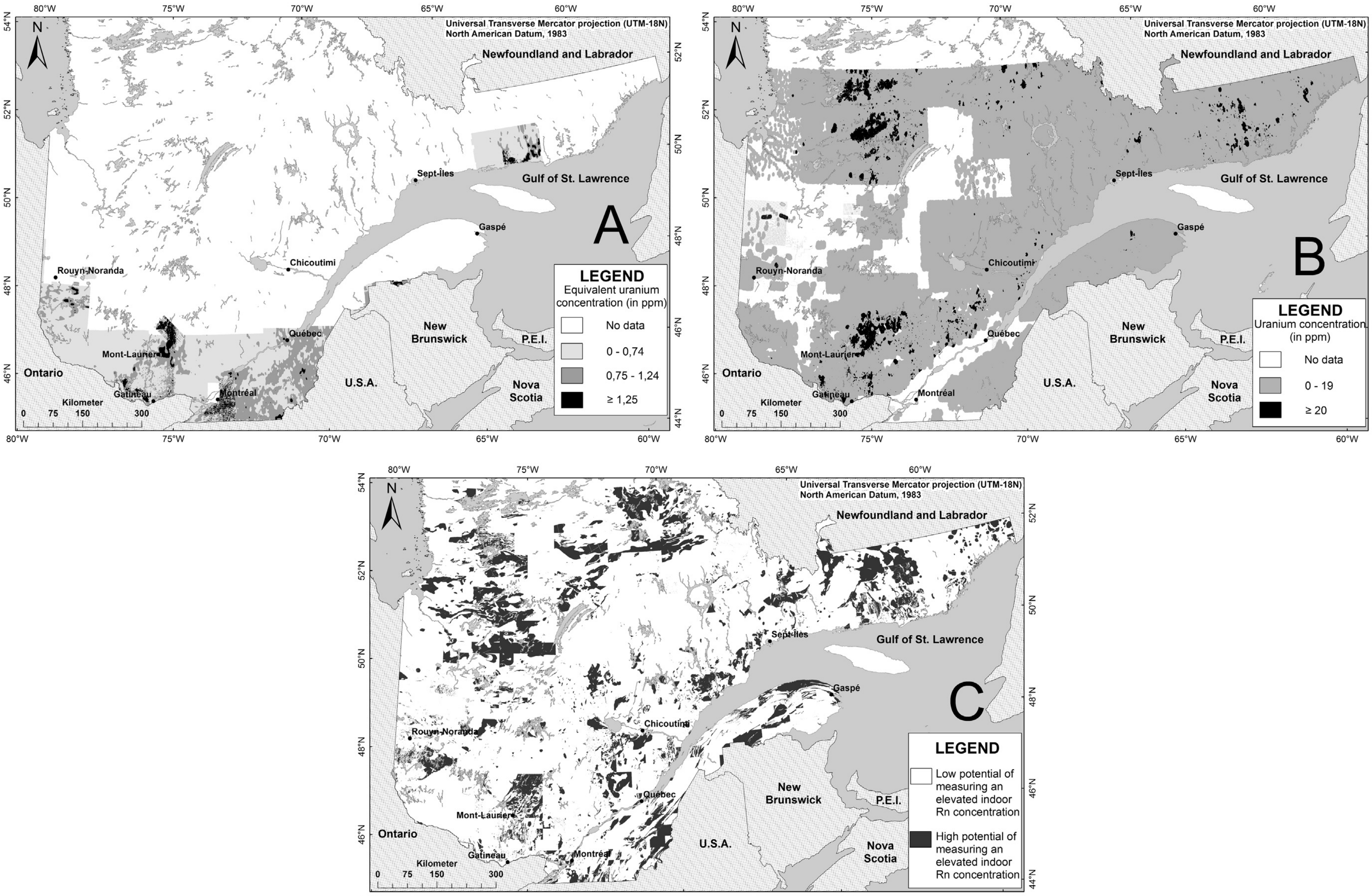

Fig. 7. Discretized criteria across Quebec based on the statistical study to define radon emission potential maps. Map A: Indoor radon emission potential based on equivalent uranium concentration from airborne surface gamma-ray measurements; Map B: Indoor radon emission potential based on uranium concentration in sediments from geochemistry surveys; Map C: Indoor radon emission potential based on bedrock geology combined with low permeability silt/clay surficial deposits.

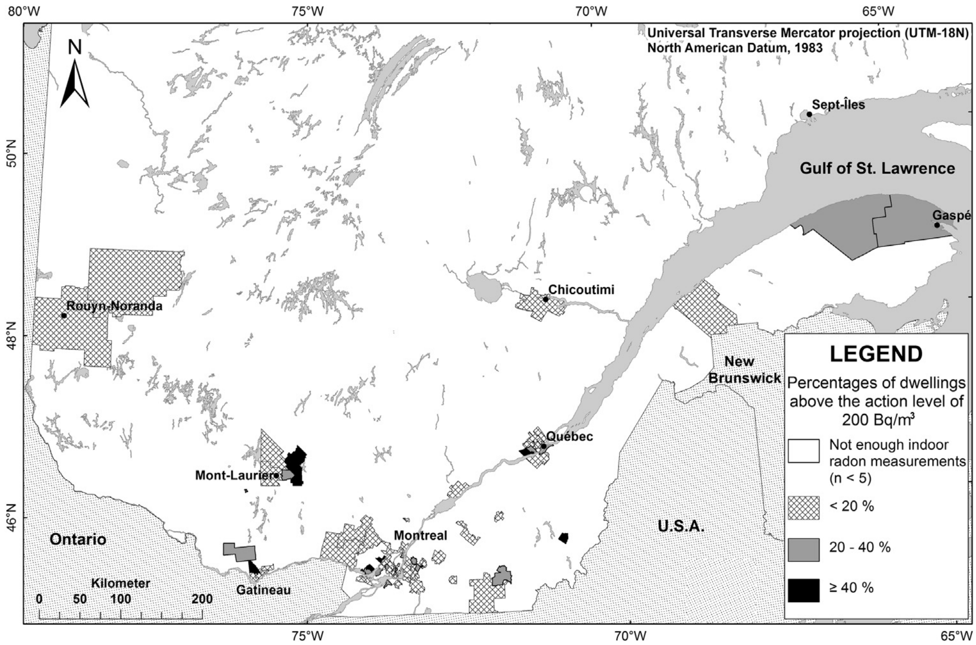

Fig. 8. Radon potential map based on direct basement radon measurements.

geology of surficial sediments source datasets were preliminarily interpolated by kriging causing some noise on the output datasets. Also, the thickness and the mineralogy of the surficial deposits were not detailed in the datasets for such a huge territory causing uncertainties on the areal extent of the "thick enough (3 m) non radon emitting silt/clay barrier". Nonetheless, the three criteria are reliable enough to identify high indoor radon concentrations and show that variability in the model is not large enough to invalidate the PPR.

7. Conclusion

Radiogeochemical information is useful to generate maps of radon-prone areas. PPR (positive proportion relationships) between three selected criteria and radon concentrations in basements demonstrate that these criteria are statistically reliable predictors for potential radon emission levels. A statistical study proposes thresholds where potential radon emission levels increase for each criteria. This methodology allows mapping radon-prone areas in regions where no actual indoor radon measurements are available. While radon potential is based on radiogeochemical information, accuracy of the potential radon emission level increases with the number of available criteria. New gamma-ray surveys and/or uranium concentration measurements in sediments would improve the model accuracy. There is always at least one criterion that can be used to determine radon potential because the geology criterion is available over the entire study area. A combined map of radon-prone areas in the province of Quebec based on the three criteria (airborne gamma-ray spectrometry, uranium concentrations in sediment samples and geology) and on indoor radon measurements still has to be made. Putting the three criteria together into one total radon potential requires more indoor radon measurements to determine the model efficiency. Public health authorities may use such a combined map of radon-prone areas to inform populations living in potentially high radon concentrations regions. Indoor radon measurements and mitigation measures need to be promoted and conducted in such areas to help lowering the occurrence of radon-related lung cancers.

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