NEW EVIDENCE OF THE PRESENCE OF RADON IN THE MARTIAN ATMOSPHERE AND PERSPECTIVE OF USE AS A GEOPHYSICAL TRACER.

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Introduction: Radon-222 is a radioactive gas belonging to the uranium-238 decay series (figure 1). A first indirect evidence of its presence in the Martian atmosphere has been provided by an analysis of the alpha spectra of *Opportunity*'s APXS, which revealed the signature of polonium-210, a long-lived decay product of radon-222 and alpha-emitter, on atmospheric dust collected by the magnets [1]. The radioactivity that was measured was translated into an estimate of the global average exhalation rate of radon, amounting to about 50 to 100 atom $m^{-2}\ s^{-1}$. As ~5 MeV alpha particles in the Martian atmosphere cannot reach space (their range is a few meters only), direct measurement of radon-222 or its decay products ^{218}Po, ^{214}Po and ^{210}Po require an *in situ* alpha detector [2]. Alternatively, the gamma-ray emission from bismuth-214, which is formed very shortly after radon disintegration (figure 1), can be measured by remote sensing. This radioelement is therefore routinely used to map uranium-238 (e.g., for airborne uranium exploration on Earth), assuming secular equilibrium between all the elements of its decay series in the soil. It is also being used by the Gamma Ray Spectrometer (GRS) on board *Mars Odyssey* to map ^{238}U in the martian subsurface [3,4], over a depth of the order of tens of cm. Thorium-232, another important radioisotope for geochemists, is measured through thalium-208 [3,4], which comes shortly after thoron, another radon isotope (radon-220) (figure 2). Both radon isotopes are the only gaseous species in their respective decay chains. However, in contrast to radon-220, which has a half-life of 55 seconds, radon-222 has a half-life of 3.8 days, making it more likely to escape its mineral host, eventually reach the atmosphere and be displaced from its production site. This can raise a problem not only for the U-Pb dating technique, but also for the mapping of uranium, as it can yield an overestimation of the uranium content of the surface. Indeed, if the diffusion length of radon in the soil is larger than the attenuation depth of ^{214}Bi gamma rays (~10 cm, R. Reedy, personal communication), the ^{214}Bi brought to the atmosphere by radon transport will lead to an excess of "apparent" uranium (or uranium-equivalent bismuth), which for a spectrometer placed in orbit corresponds to the atmospheric column density of ^{214}Bi (corrected for atmospheric attenuation). But one can also benefit from this phenomenon to look for the presence of radon in the atmosphere, derive its atmospheric column density and map its exhalation rate. We describe methods to carry out this study.

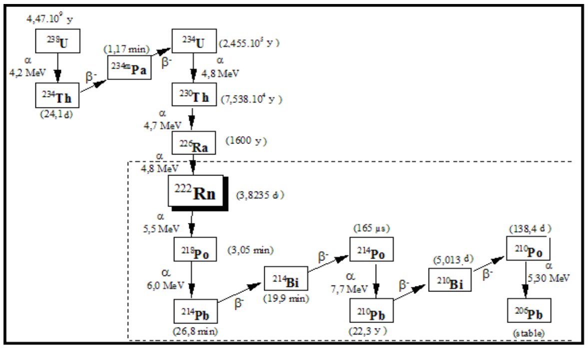

Figure 1. Uranium-238 decay series (with decay mode, energy and half-life).

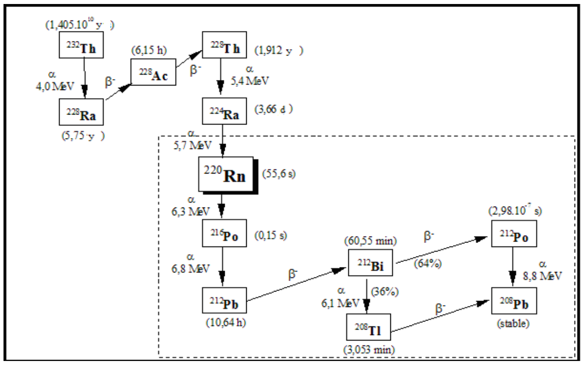

Figure 2. Thorium-232 decay series (with decay mode, energy and half-life).

Methods: If radon is present in the atmosphere, the main problem is to separate the atmospheric and the ground contributions to the total ^{214}Bi signal. Once the ^{214}Bi concentration in the atmosphere is known, it can safely be equated to the ^{222}Rn concentration, as ^{214}Bi is formed less than an hour after radon disintegration.

A first way to analyze the data consists of looking at the ^{214}Bi/^{232}Th ratio, or apparent ^{238}U/^{232}Th ratio, on a global scale. Indeed, ^{238}U and ^{232}Th are both incompatible elements that are usually strongly correlated in the crust in the absence of aqueous alteration or mechanical erosion [5,6]. The value of the ^{238}U/^{232}Th ratio is typically between 0.24 and 0.28 (ppm/ppm) in chondrites, in the Earth's upper continental crust and in SNC meteorites [e.g.,7,8,9]. Any significant departure from this standard value on a global scale (towards higher values) is a first evidence of the presence of radon in the atmosphere.

On a local scale, the ^{238}U/^{232}Th ratio can vary from this standard value because of geochemical fractionation. However, several studies have shown that fractionation of incompatible elements has been probably very limited in the martian soil [6,10,11,12]. Therefore, to a first order, it is reasonable to assume that the ^{238}U/^{232}Th ratio is constant throughout the surface of the planet. Spatial variations of the apparent ^{238}U/^{232}Th ratio could then stem from differences in the exhalation rate of radon or from atmospheric circulation of this gas. Prediction of these effects requires proper modeling of radon production and diffusion in the soil and modeling of its atmospheric transport. This has been achieved by implementing a coupled subsurface and atmospheric transport model of radon in a Martian General Circulation Model (LMDZ-Mars) [13], whose input parameters were derived either experimentally (emanation and adsorption coefficients) or from a realistic model of porous media (diffusion coefficient) [14]. The source term was inferred from the ^{232}Th map measured by *Mars Odyssey* GRS, assuming uniform ^{238}U/^{232}Th and ^{226}Ra/^{238}U ratios. Besides soil structural parameters such as grain size and specific surface area, the only free parameter in the model is the subsurface pore water content (adsorbed + free pore water), which controls the radon emanation factor.

If radon is present in the atmosphere, one would also expect to observe temporal variations of the apparent ^{238}U/^{232}Th ratio on two different timescales : on a diurnal timescale owing to the dynamics of the boundary layer, which affect the vertical profile of radon and thus the attenuation pattern of gamma ray lines produced in the atmosphere; on a seasonal timescale owing to changes in atmospheric circulation patterns as well as changes of soil temperature, which controls the adsorption coefficient of radon in the soil and thus its exhalation rate (see below).

Lastly, ^{214}Bi emits gamma rays at different energies, which are thus more or less attenuated before reaching the detector. If radon is confined in the ground and assumed homogeneously distributed with depth, proper modeling of the relative attenuation of these rays should yield a ^{214}Bi (and thus ^{238}U) content that is independent of the photons energy. However, application of this same attenuation model to the atmospheric ^{214}Bi fraction, if it exists, will induce a dependence upon energy of the apparent uranium content, namely a decrease of apparent ^{238}U as the energy increases. Analysis of this dependency can yield both the atmospheric column density and the real ^{238}U content of the soil. For this purpose, analysis of two gamma ray lines is theoretically enough if the atmospheric profile is known or derived from modeling. Any other lines can be used to improve these estimates or to constrain the atmospheric profile.

Preliminary results: Analysis of GRS spectra is complex in some energy ranges, as many peaks can interfere and must be deconvolved first [4]. None of the ^{214}Bi lines is easily extracted. Therefore, uranium maps have not been published yet. However, preliminary results tend to confirm our predictions and support the presence of radon in the atmosphere. First of all, the global average apparent ^{238}U content derived from ^{214}Bi measurement is significantly larger than would be expected from a geochemical standpoint, given the global average ^{232}Th content of the crust that has precisely been measured. A global average exhalation rate of 200 to 300 atom m^{-2} s^{-1} of radon could account for this anomaly, which is consistent with the value derived from Opportunity's APXS. If this preliminary exhalation rate could be confirmed, it would place the magnitude of Mars' radon out gassing intermediate between that of the Earth and Moon. This tentative conclusion is most likely due to differences in soil humidity (see below). Secondly, simulations with the GCM predict an accumulation of radon in some locations, which to first order match the GRS data quite well (see figure 3), in particular around Valles Marineris, over Hellas Basin and over Margaritifer Terra. Anomalies of the apparent U/Th ratio seem to be correlated with regions of low thorium (and thus, probably low uranium if U and Th have not fractionated from each other) content, which is also expected if radon is transported by winds over these regions. Finally, preliminary results indicate an energy dependence of the uranium content (after correcting for the attenuation), as expected. Other lines of evidence (temporal variations for instance) will be studied in the future.

Perspectives: It can be shown that for purely diffusive transport, the exhalation rate of radon is given by:

$$\Phi = E\ \rho_b\ C_{Ra} \sqrt{\frac{D_b}{\lambda\ \beta}} = E\ \rho_b\ C_{Ra}\ L_{diff}$$

where E is the emanation factor, ρ_b the soil bulk density, C_{Ra} the radium concentration, D_b the bulk diffusion coefficient, λ the decay constant of radon and β the so-called partition-corrected porosity, which depends on the adsorption coefficient and thus strongly on temperature and on the specific surface area of the grains. E increases with increasing moisture content, while D_b and β decrease. L_{diff} is called the diffusion length. Hence, the exhalation rate is given by the product of a source term and a transport term, both of which depend on the soil moisture content and structural properties. The previous equation shows that mapping the radon exhalation rate can provide valuable information on subsurface-atmosphere gaseous exchange. This also implies that the ^{214}Bi signal, once corrected for atmospheric transport away from its production site and for differences in uranium content, should be correlated with latitude, albedo, thermal inertia (which control the soil temperature) and possibly hydrogen maps, if this hydrogen corresponds to pore water (adsorbed or free pore water). Correction for atmospheric transport through modeling with a GCM is made possible owing to the limited half-life of radon, which cannot move very far from its emission zone.

A more detailed analysis of GRS ^{214}Bi data should thus be conducted in the future and can potentially lead to innovative results on surface-atmosphere interactions (including the water cycle), as well as on the dynamics of the boundary layer and the global circulation. It is probably necessary to take into account the transport of radon to derive real uranium maps, which can help understanding the history of Mars surface alteration [6]. GRS ^{214}Bi data could also bring new information on the dust cycle [15]. Lastly, it may be used to survey recent outgassing activity, or at least to constrain it, as it has been done for the Moon since the Apollo missions.

Nonetheless, the spatial and temporal resolution of the GRS data will present a large challenge to the feasibility of these applications [4,11,16,17].

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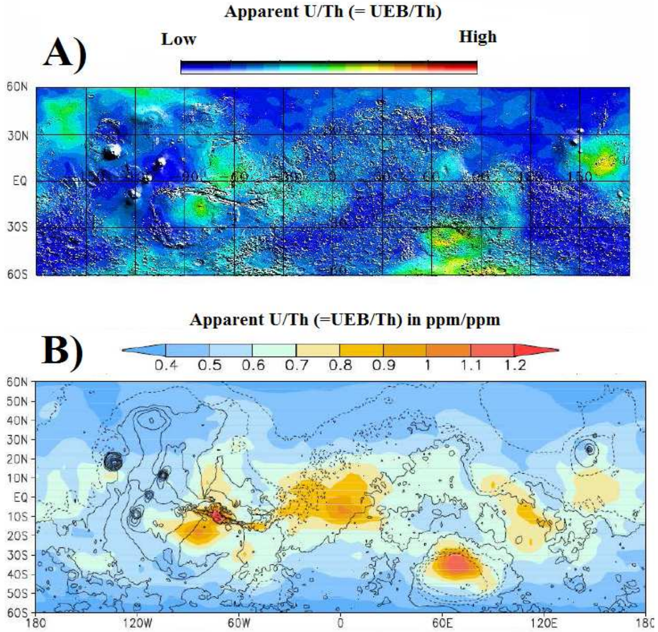

Figure 3. Comparison between GRS data and GCM simulation: A) Apparent U/Th ratio (i.e., uranium-equivalent bismuth/Th, or UEB/Th) measured by the GRS between 60°S and 60°N. B) Apparent U/Th ratio (in ppm/ppm) simulated with the LMDZ Global Circulation Model , coupled to a radon subsurface transport model: the ^{238}U signal is obtained by summing the radon atmospheric column density and the radon concentration in the soil integrated over a depth of 10 cm, average over one Martian year. The source term is derived from the thorium map, so that the anomalies are either due to enhanced exhalation rate or subsequent atmospheric transport from the source regions.