

# SOIL GAS GEOCHEMISTRY AS PERMEABILITY TRACER OF THERMALLY ALTERED CLAYS AT ORCIATICO (Tuscany, Central Italy)

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**Introduction**  
 The Orciatico clay study was part of an EC project aimed to study natural analogues of the thermo-hydro-chemical and thermo-hydro-mechanical response of clay barriers. The main idea was to achieve information and experimental data regarding the long-term stability of permeability and plasticity of the clays affected by high temperatures. Natural analogues were identified in clay formations near contact with volcanic intrusions. Though available data do not allow exact evaluations of depth, many features of the Orciatico igneous body (widespread glass, highly vesicular peripheral facies etc.) point to a shallow emplacement, comparable with that reasonably forecast for a repository. Not even exact definitions of the temperature of magma at the moment of emplacement are feasible. Only some evaluations can be proposed: from its distinctly femic composition temperatures over 800°C may be assumed for the alkali-trachytic magma intrusion (Leoni et al., 1986; Hueckel and Pellegrini, 2002). These values are much higher than those expected around a radiowaste container (up to 300°C, according to Dayal and Wilke, 1982); therefore, as to the thermal aspects the Orciatico magmatic body and its metamorphic aureole should be regarded as an extreme condition model of a radiowaste repository and probably it can be mainly used to demonstrate a worst case.

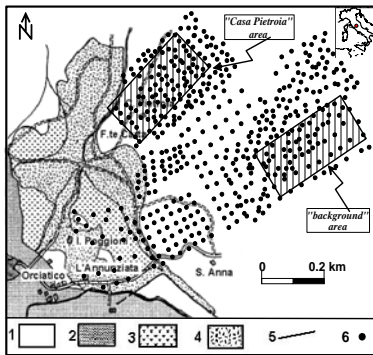


Figure 1. Geological sketch map of the Orciatico igneous body (from Canticelli et al., 1992) and soil-gas surveys. A first survey (July-September 1997) was performed along a regular grid with a sampling density of about 500 samples/km<sup>2</sup> (dark dots). The two dashed rectangles indicate two monitored zones (from April to September 1998) in order to study seasonal influence and/or local factors (i.e. seasonal vegetation) on soil-gas distributions. 1) Pliocene Quaternary sediments; 2) Miocene lacustrine and marine sediments; 3) thermally metamorphosed rocks (Termentite); 4) Orciatico andesite; 5) faults and inferred faults.

**Methodology**  
 A total of 1086 soil-gas samples was collected in the Orciatico area. A first survey was performed collecting 486 samples along a regular grid with a sampling density of about 500 samples/km<sup>2</sup>. A total of 600 samples (monthly surveys, from April to September 1998) were collected in two small zones within the studied area: the first one called "Casa Pietraia" area, characterized by altered clays and the second one, "background" area, situated in the unaltered clays (Fig. 1). The aim of the monthly surveys was to monitor possible variations of soil-gas concentration due to weather conditions. A total of 20 electrical soundings (VES) and 8 dipole electrical tomography (DET) profiles was carried out across the intrusive body and in the clays characterized by several thermal and mechanical alteration degrees.

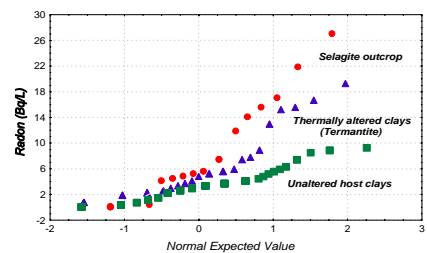


Figure 3. Merged probability plots of CO<sub>2</sub> and <sup>222</sup>Rn. The plots were elaborated in order to highlight the gas concentration differences on Selagite outcrop, metamorphosed clays and intact (un-metamorphosed) clays. There is a general decreasing trend of the measured concentrations in the three different sites, showing the lower values in the un-metamorphosed clays.

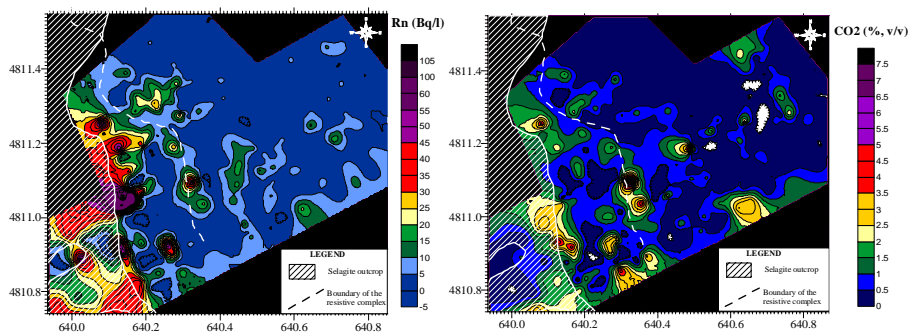


Figure 2. Radon and carbon dioxide distributions in soil-gases. Anomalous values (CO<sub>2</sub> > 2 % v/v, Rn > 25 Bq/l) are in correspondence of the boundary of the resistive complex supposed on geoelectrical results.

**Results and discussion**

The radon, as well as the CO<sub>2</sub> contour line maps, Fig. 2, show that highest values (<sup>222</sup>Rn > 25 Bq/l, CO<sub>2</sub> > 2 % v/v) occur in the south-western part of the studied area (characterized by the presence of Selagite outcrop) and along a narrow belt, with direction NNW-SSE, where metamorphosed clays (named "Termentite") are present. Furthermore, anomalous values occur in unaltered clays especially in correspondence of the boundary of the resistive complex supposed on geoelectrical results. All over the north-eastern sector, in non metamorphosed clays, radon and carbon dioxide values are very similar to background values reported in literature (Rn: 10-15 Bq/l; CO<sub>2</sub>: 0.5 % v/v). Figure 3 shows two merged probability plots elaborated to highlight different concentrations on Selagite outcrop, Termentite (metamorphosed clays) and intact (unaltered) clays: it is well evident a general decreasing trend of the measured concentrations in the three different sites, showing the lowest values in the un-metamorphosed clays. Results from monthly surveys (from April to September 1998) showed (Fig. 4) that soil-gas concentrations remain constant confirming that they are directly correlated with clay permeability. Geoelectrical surveys were performed in order to identify the geometry of the trachite-alkaline intrusive body (Selagite) outcropping a few hundred meters NE of the village of Orciatico. A total of 20 vertical electrical soundings (VES) and 8 dipole electrical tomography (DET) profiles have been performed across the intrusive body. The VES defined three electrical layers: the first and third layers have a resistivity between 5 and 10 ohm·m and are attributed to Pliocene clays, while the intermediate layer has a resistivity between 15 and 60 ohm·m and is correlated with the Selagite intrusion, near the outcrop, and with the clays that underwent modification because of the intrusion effects. The metamorphosis degree of clays is function of their distance from the intrusion, and, clearly, highest metamorphosed clays are near Selagite outcrop.

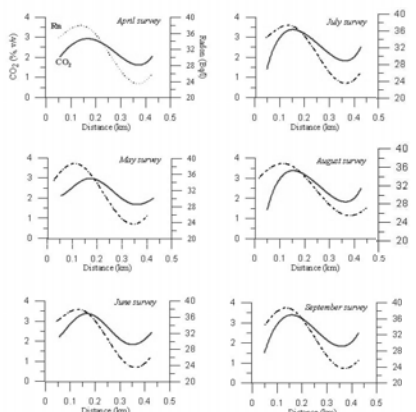


Figure 4. Polynomial regression (5° degree) of radon and carbon dioxide concentrations at the "Casa Pietraia" area during six month monitoring (April-September 1998). The left axis represents carbon dioxide values (straight line) whilst on the right axis there are radon values (dashed line). Graphs highlight a general decreasing trend of soil gas values towards NE. Results from these monthly surveys showed that in spite of seasonal variations, soil gas concentrations remain constant.

**Comparison between geochemical and geoelectrical results**  
 As Rn and CO<sub>2</sub> values seem to decrease gradually from Selagite outcrop towards un-metamorphosed clays, soil-gas data set was projected along two longitudinal lines coinciding with one geoelectrical profile that highlights a quick resistivity decrease both in the vertical and in the horizontal way. Figure 5 shows polynomial regression (3° degree) of Rn and CO<sub>2</sub> values plotted against the distance from a reference point. Graphs highlight a slight decreasing trend of radon (continuous line) and carbon dioxide (dashed line) soil-gas values towards the NE, from Selagite outcrop until un-metamorphosed clays. Highest CO<sub>2</sub> and Rn values overlap and were found between Selagite outcrop and the first resistive limit, in a narrow belt characterized by a high alteration degree and, probably, by an intense shallow fracturing.

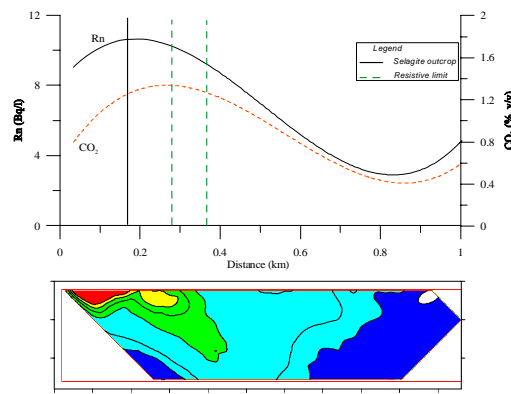


Figure 5. Comparison between polynomial regression (3° degree) map and geoelectrical profile n°4. Radon graph (continuous line) highlights a general slightly decreasing trend of soil-gas values towards the NE, from Selagite outcrop until un-metamorphosed clays. The same behaviour is well evident also for CO<sub>2</sub> polynomial regression (dashed line).

**Conclusions**  
 The results of this study provided specific information about soil-gas permeability on the Orciatico clay units characterised by different degrees of thermal alteration. This research represents the first study performed in thermally and mechanically altered clays and results demonstrated that the method gives interesting information also in clays that apparently did not undergo to mineral and geotechnical variations. Radon and carbon dioxide soil-gas anomalies seem to be concentrated in zones where Selagite outcrop and thermally altered clays may be inferred: regional trend (that excludes possible local effects on soil-gas concentrations) of soil-gas distribution highlights the strong difference of radon and carbon dioxide concentrations over Selagite and over unaltered clays. These distributions are interpreted as being due to intense shallow fracturing of clays along the inferred laccolith boundary: the intrusion of the laccolith caused thermo-hydro-chemical and thermo-hydro-mechanical stress and contact metamorphism in the clay. Soil-gas results in the Orciatico area were supported by detailed geoelectrical survey: the main conductive lineaments representing highly permeable zones within the clays, were outlined in an area where highest Rn and CO<sub>2</sub> soil-gas anomalies occur. This study allowed to highlight the role of soil-gas technique for the identification of secondary permeability in a clay sequence. In particular, at Orciatico site, this secondary permeability was found along the boundary of the resistive complex forming a lesser impermeable barrier for naturally migrating gas. It means that a clay sequence, if affected by thermal alteration, can strongly modify its characteristics and the effect is not always visible through traditional investigative methods.

**References**  
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**Acknowledgments**  
 This research was funded by EC project (Contract F14W/CT95/0014, "Natural Analogue of the Thermo-hydro-chemical and Thermo-hydro-mechanical Response of Clay Barriers"):the authors would like to thank all partners available for exchanging information in the framework of the project.