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Abstract: A study carried out in the iron basin of Lorraine (North-East of France) highlighted the existence of under-oxygenated and noxious gas emissions (carbon dioxide, radon…) in vast built-up areas related to former mine workings.

Site investigations showed that the gas flow was mainly due to natural thermal air circulation between old mines and external atmosphere. The main hazards associated with this flow of gas and its potential accumulation in confined volumes in buildings are asphyxiation and intoxication.

A further study is in progress in order to better understand the origin of the gas generation within the old iron mine workings. Following the first hypothesis, the carbon dioxide emission is mainly due to the specific hydro-geo-chemical process associating the pyrite oxidation with the acid dissolution of carbonates.

KEY-WORDS : gas, emission, iron mine, pyrite.

1. INTRODUCTION

Some residential areas of the iron basin of Lorraine (in the North-East of France) are affected by noxious gas emissions (under-oxygenated and noxious gas loaded mixtures). The most spectacular phenomenon occurred in the built-up area of Moyeuvre-Grande in Moselle (figure 1), especially in the district of the town located very close to the former underground mine workings in which some inhabitants observed faulty working gas cookers and boilers.

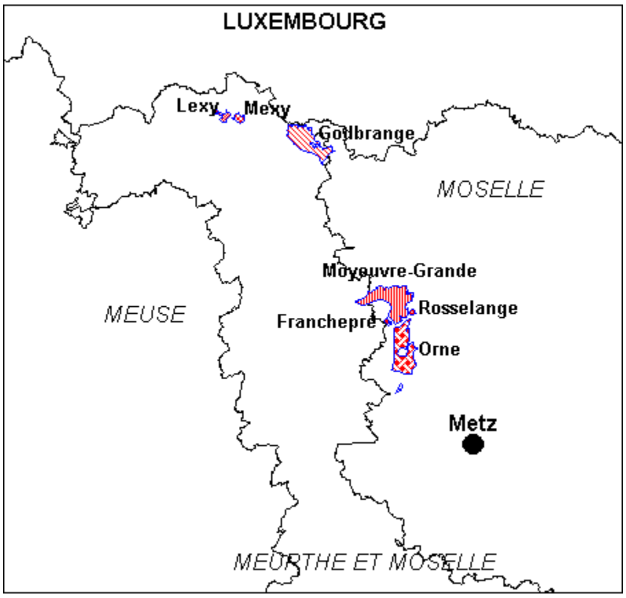


Figure 1. Localisation of the studied mining reservoirs

In 1999, following the Lorraine local office of the French Ministry of Industry, Research and Environment (DRIRE) request, INERIS (the French National Institute for Safety and Risks) started some investigations to assess the geographical extent of the problem. First of all, the research was focused on the underground old mine workings area of Moyeuvre-Grande and also some neighbouring closed mines (Rosselange, Franchepré and Orne) whose main features are given in table 1.

Table 1. Features of the studied reservoirs (old mine workings)

Name of the reservoir	**Moyeuvre Orne Franchepré**	**Rosselange**	**Lexy**	**Mexy**	**Godbrange**
Area (ha)	355,7	252			
Volumes (Mm^3)			1,3	0,7	>30
Operating periods	1873 - 1993	1890 - 1935	1867 - 1932	1871 - 1901	18 ? ? - 1978
Number of followed openings	18	3	3	2	5
Openings heights (m NGF)	170 à 180	170 à 180	260 à 270	280 à 295	330 à 360
Old workings heights (m NGF)	170 à 180	170 à 180	260 à 280	270 à 290	360 à 350
Plateau heights (m NGF)	300 à 330	310 à 325	300 à 350	350 à 375	450 à 410
Openings – plateau height differences (m)	120 à 160	130 à 150	30 à 90	55 à 95	50 à 100

These studies brought to the fore the existence of noxious gas emissions in relation with former mining workings that were not completely flooded. High level CO_2 content (up to 6 %) and low level O_2 content (down to 13 %) have been measured. During the same time, the Institute for Radio-protection and Nuclear Safety (IRSN) and the DASS (a government organisation under Ministry of Health) investigated and measured high level radon content (up to 15000 Bq/m^3) in the gaseous mixture.

In some cases, these emissions affected directly inhabited buildings in connection with the mine entrances or with the old superficial mine workings.

Moreover, a more exhaustive series of measures has been taken in other non-flooded mining reservoirs of the North of the ferriferous basin: Mexy, Lexy and Godgrange (figure 1 and table 1). It showed that similar emissions were occurring in these areas. However the intensity of the phenomenon was far less spectacular than those observed in Moyeuvre-Grande area (Pokryszka and Grabowski, 2002).

INERIS is now carrying out research to better understand the phenomenon of gaseous emissions from iron mines. These works are performed in the framework of a program funded by the French Ministry in charge of Industry in collaboration with the Lorraine local administration (DRIRE).

2. THE RISKS CREATED BY THE GASEOUS EMISSIONS

In an average atmosphere, the volumetric oxygen content is 20.9 % and the carbon dioxide one is 0.03 %. The consequences of a CO_2 enrichment and an O_2 impoverishment on health are numerous. The significance depends on the contents of these two gases: it ranges from headaches for CO_2 contents between 3 and 4 % and O_2 content between 14 and 16 %, to death for O_2 content lower than 6 % and CO_2 higher than 10 % (Monomakhoff, 1978).

Fortunately, the composition of the air in inhabited buildings of the investigated area did not reach a level that could affect health. However in some cases, the measured contents were very close to the critical level for which harmful effects can appear by O_2 deficit and CO_2 excess.

Concerning the radon, it acts essentially on lung epithelium to further the development of cancer in case of long – lasting exposure with content higher than 500 – 1000 Bq/m^3 (Barrier and Lorentz, 2001).

Apart from these clinical effects, the observed dysfunctioning of combustion appliances (boiler, gas cooker) due to CO_2 polluted and under - oxygenated air are likely to increase the explosion and CO emission risks in concerned houses.

These observations induced INERIS to carry on its investigation, in order to better understand the mechanism of the gaseous emanations and to find reliable solutions to this problem.

3. GAS FLOW MECHANISM

In order to find the origin of the flow driving the gaseous exchanges between the old mine workings and the atmosphere, two measuring stations were installed at the entrance of two former galleries connected to the closed mine in Moyeuvre-Grande area.

These stations continuously monitored the intensity and the direction of the air flow, the temperature and the content of some components of the emitted gas (O_2, CO_2 and radon) as well as the outer temperature and the atmospheric pressure.

Figures 2 and 3 shows some interesting results obtained from the measures made from April 2000 to December 2002.

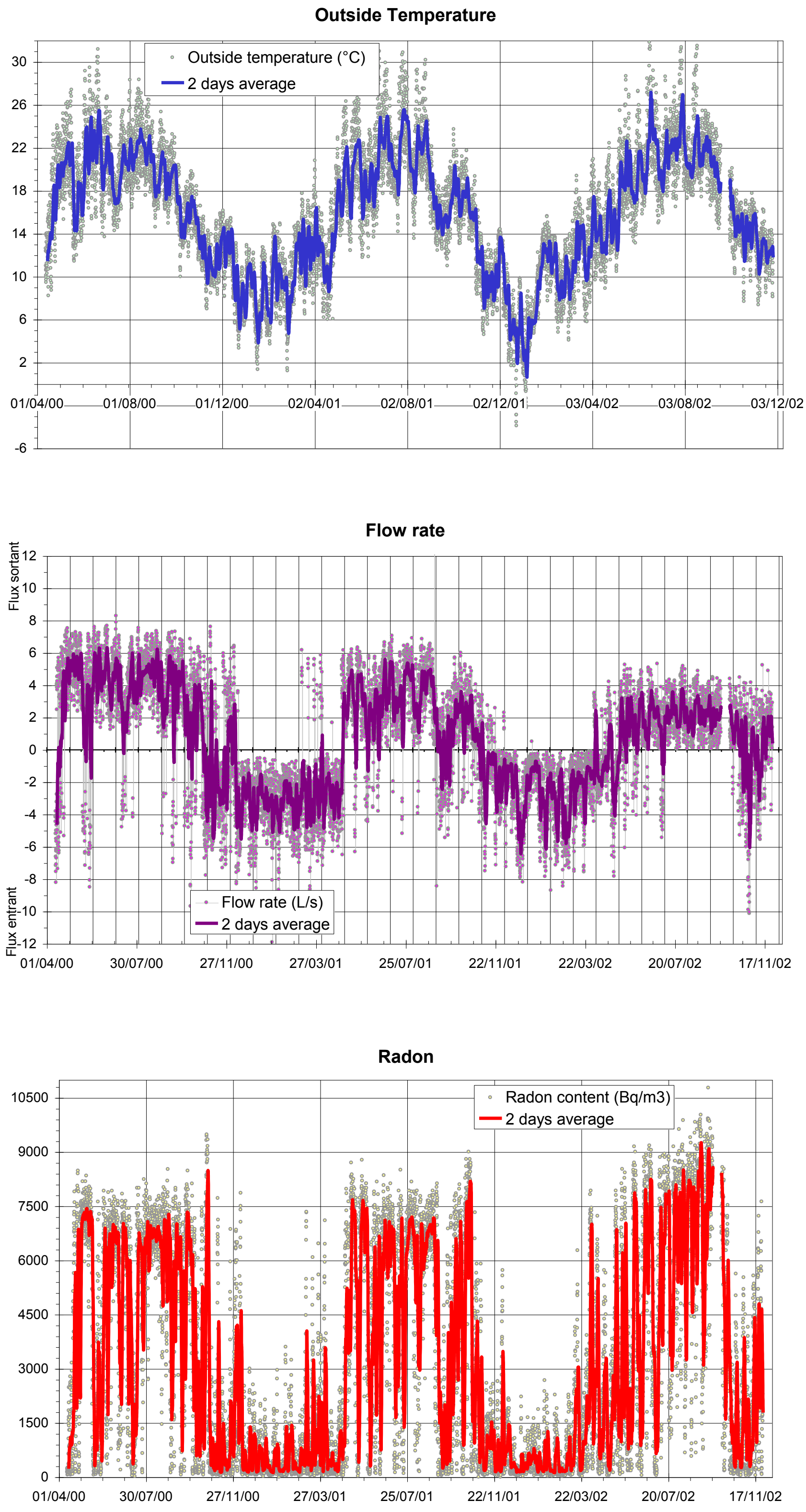


Figure 2. Outside temperature, air flow rate and radon content evolutions observed in a gallery connected to the Moyeuvre-Grande reservoir.

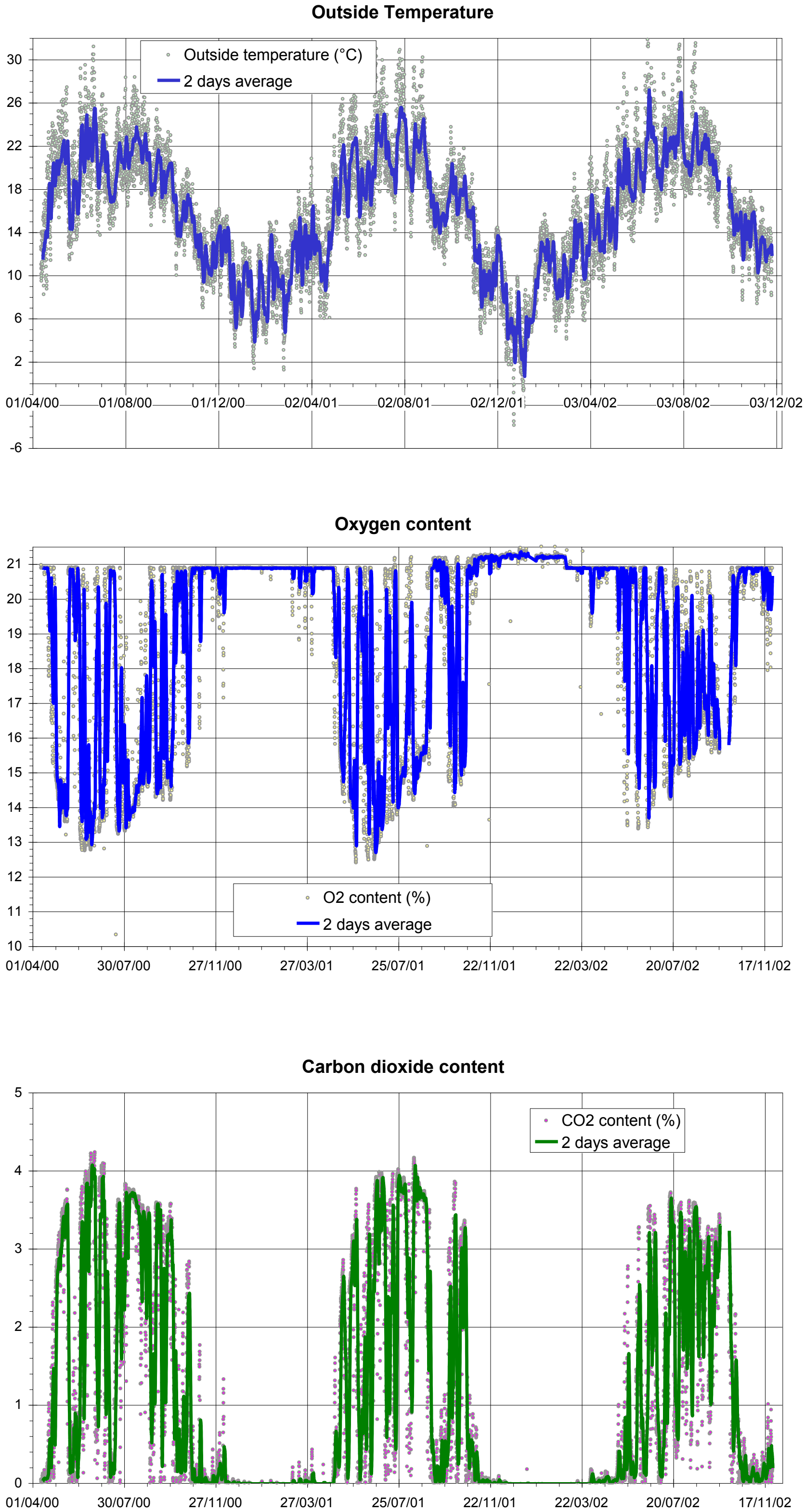


Figure 3. Outside temperature, CO_2 and O_2 content evolutions observed in a gallery connected to the Moyeuvre-Grande reservoir.

These results, in agreement with the numerous measurements done during the preliminary reconnaissance phase between 1999 and 2000 (Pokryszka and Grabowski, 2002), clearly show a link between the direction and the air flow rate, and the outer temperature. No other parameter having a significant effect on the flow (like the atmospheric pressure) has been identified.

This fact led us to consider the natural thermal convection as the phenomenon creating the gaseous flow between the mining reservoir and the surface. This phenomenon is due to the thermal differential existing between the outer atmosphere and the old workings. The temperature in the latter is almost constant (in that case, it was between 12 and 14 °C), unlike the atmospheric temperature whose level varies with the seasons.

In the same way, the surface topography above the mining reservoir of Moyeuvre-Grande and other concerned reservoirs seems to favour the natural thermal convection.

Indeed, the investigated areas are globally characterised by a very contrasted topography with rather large plateau distinctly intersected by an irregular network of valleys. The ferriferous deposit is located at the base of the plateau. Outcrops can be frequently seen on the hillsides.

During summer, a part of the atmospheric air in contact with the upper old workings, by unsealed openings or cracks, is subject to a progressive cooling. An increase in the bulk density due to the cooling causes the gas migration to the lower parts of the old emerged mine workings. Then the gas can leave the reservoir by the various openings and apertures mainly situated along the deposit outcrop on the hillsides. An air circulation becomes established in this way the principle of which is presented on figure 4. The flow direction is shown for the summer period.

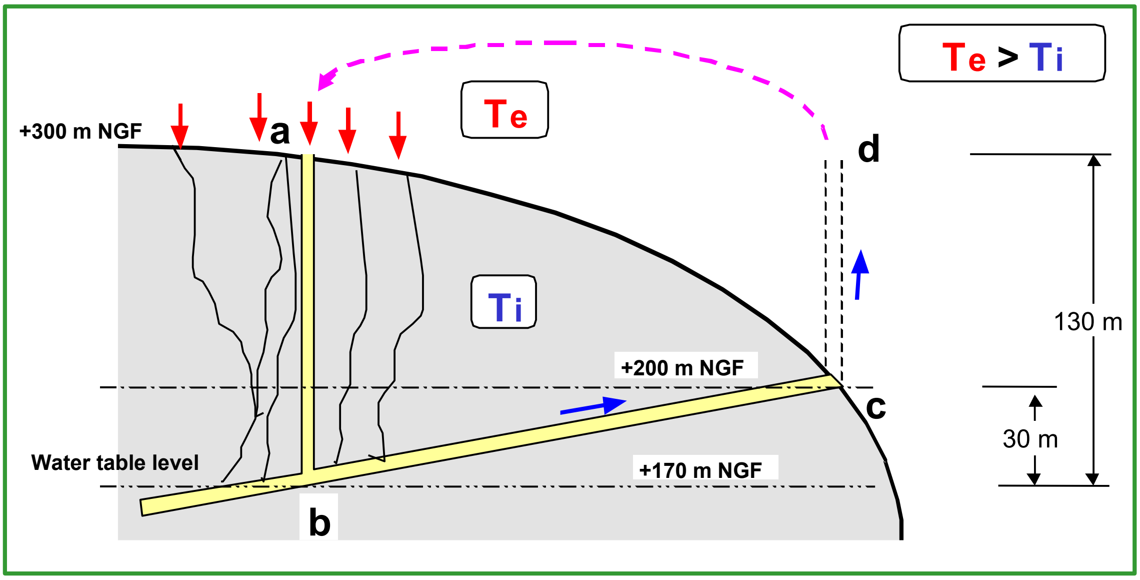


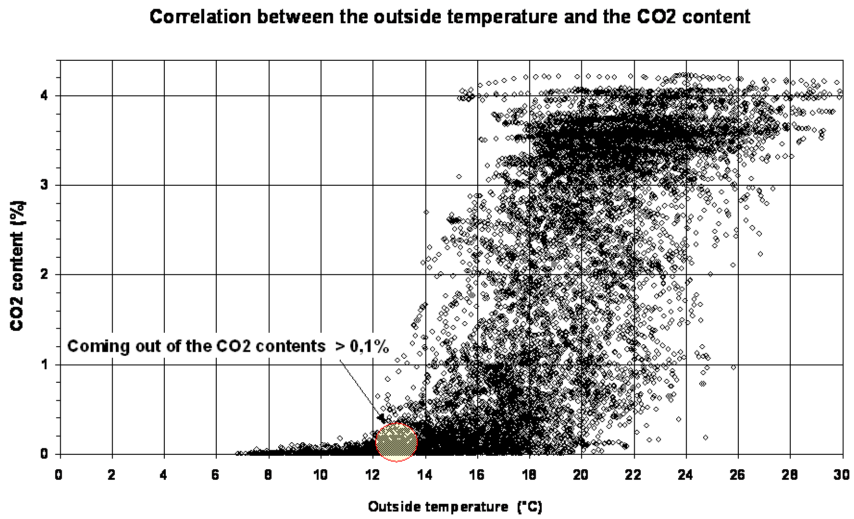
Figure 4. Schematic diagram of the natural thermal draught during the summer period (Moyeuvre-Grande reservoir case)

According to this model, the reservoir working can be divided in two distinct stages:

- the summer stage during which the flow is globally coming out of the massif. The outer temperature is significantly higher than the inside one (between 12 °C and 14 °C);
- the winter stage during which the flow is globally coming into the old mine workings. The flow rate is clearly setting when the outer temperature is lower than the old mine workings temperature.

These two stages are separated by transitional periods during which the flow is fluctuating.

Figure 5 represents the O_2 and CO_2 rates measured in an opening of the Moyeuvre-Grande reservoir as a function of temperature. It clearly shows that the O_2 impoverished and CO_2 enriched atmosphere appears for temperatures higher than 12 °C to 14 °C, that corresponds to the coming out flow stage.



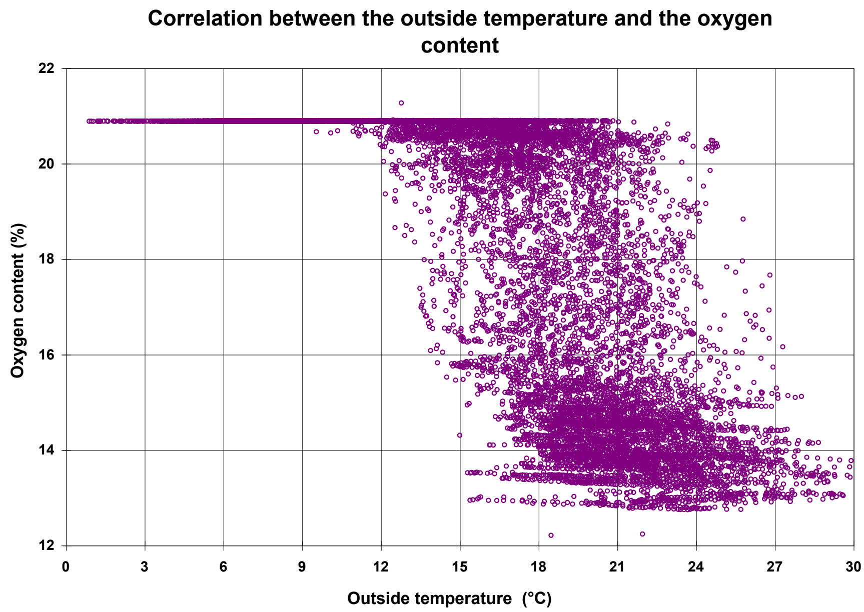


Figure 5. Correlation between the outside temperature and the O_2 and CO_2 contents observed in an opening of the Moyeuvre-Grande reservoir

Furthermore, a simulation based on the simplified model of Budryk (Budryk, 1932) showed that the aeraulic charge due to this phenomenon was estimated in the region of 20 to 30 Pa for a thermal differential of 10 °C. Such a charge seems to be large enough to create the gas circulation in this context.

It is also important to underline that other natural events (such as the atmospheric pressure variations, the wind effect, the aquifer fluttering …) add to the natural thermal draught phenomenon and can disturb temporarily the two main stages characterised previously, without being enough to reverse them on a long term basis.

4. ATMOSPHERE MODIFICATION MECHANISM IN THE MINING RESERVOIRS

4.1. Atmosphere modification in the studied reservoirs

The old mine workings gas composition, monitored continuously in the Moyeuvre-Grande reservoir and punctually in other reservoirs of the North of the ferriferous basin, have shown that the atmosphere changes within the old mine workings were consisting in a O_2 consumption accompanied by a less than proportional production of CO_2.

This result, in accordance with a literature review on geochemical reactions observed in similar mining development cases (Feuga, 2000), led us to consider the pyrite oxidation (found in the ferriferous deposit) coupled with the dissolving of calcite by the produced sulphuric acid as the most convincing hypothesis to explain the CO_2 emission and the O_2 consumption.

The reaction balance can be written:

$$FeS_2 + 3{,}75\ O_2 + 2\ CaCO_3 + 1{,}5\ H_2O \Leftrightarrow Fe(OH)_3 + 2\ CO_2 + 2\ SO_4^{2-} + 2\ Ca^{2+}$$

Moreover, an other study revealed the presence of reactive pyrite in the rich in calcite marly interstratifications of the Lorraine ferriferous field (Collon and al., 2002).

Furthermore, another way to check the assumption about the pointed out geochemical mechanism occurred with the opportunity of the Tressange pilot site experimentation, which consists in a controlled submergence of a small part of the old mine workings.

4.2. Atmosphere modification measured in the massif during the submergence

In order to estimate the flooding effect on the abandoned mining structures and on the water and atmosphere quality, a specific experiment has been carried out within the framework of the GISOS research program (GISOS, 2003). It is realised in real and in situ conditions, in the old mine of Tressange.

For the needs of that experimentation, a part of a former mine stripped by the room and pillar method used in two geological levels has been isolated, fitted out and prepared to be flooded in a controlled way.

Only the deeper layer and the base of the marly interstratifications have been flooded. A simplified diagram of the experimental site is presented on figure 6.

INERIS took advantage of this opportunity to check the effect of the ferriferous massif flooding on the possible gas production and on the atmosphere composition within the rock.

An experiment has been set to follow the atmosphere evolution (O_2 and CO_2 contents) in the isolated enclosure, during the flooding. The atmosphere has been monitored thanks to two boreholes drilled through the marly interstratifications and emerging into the mine workings of the brown layer to be flooded.

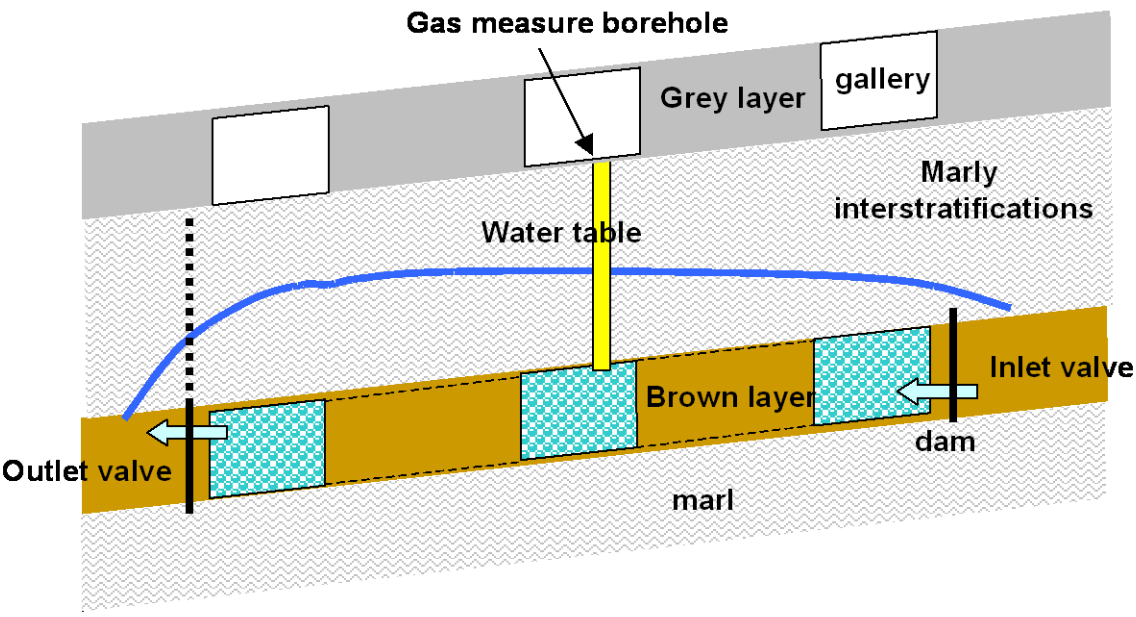


Figure 6. Schematic section of the atmosphere watch over system during the flooding of the Tressange experimental site

Figure 7 shows the evolution of the O_2 and CO_2 contents measured within the boreholes concurrently with the rising of the submergence water.

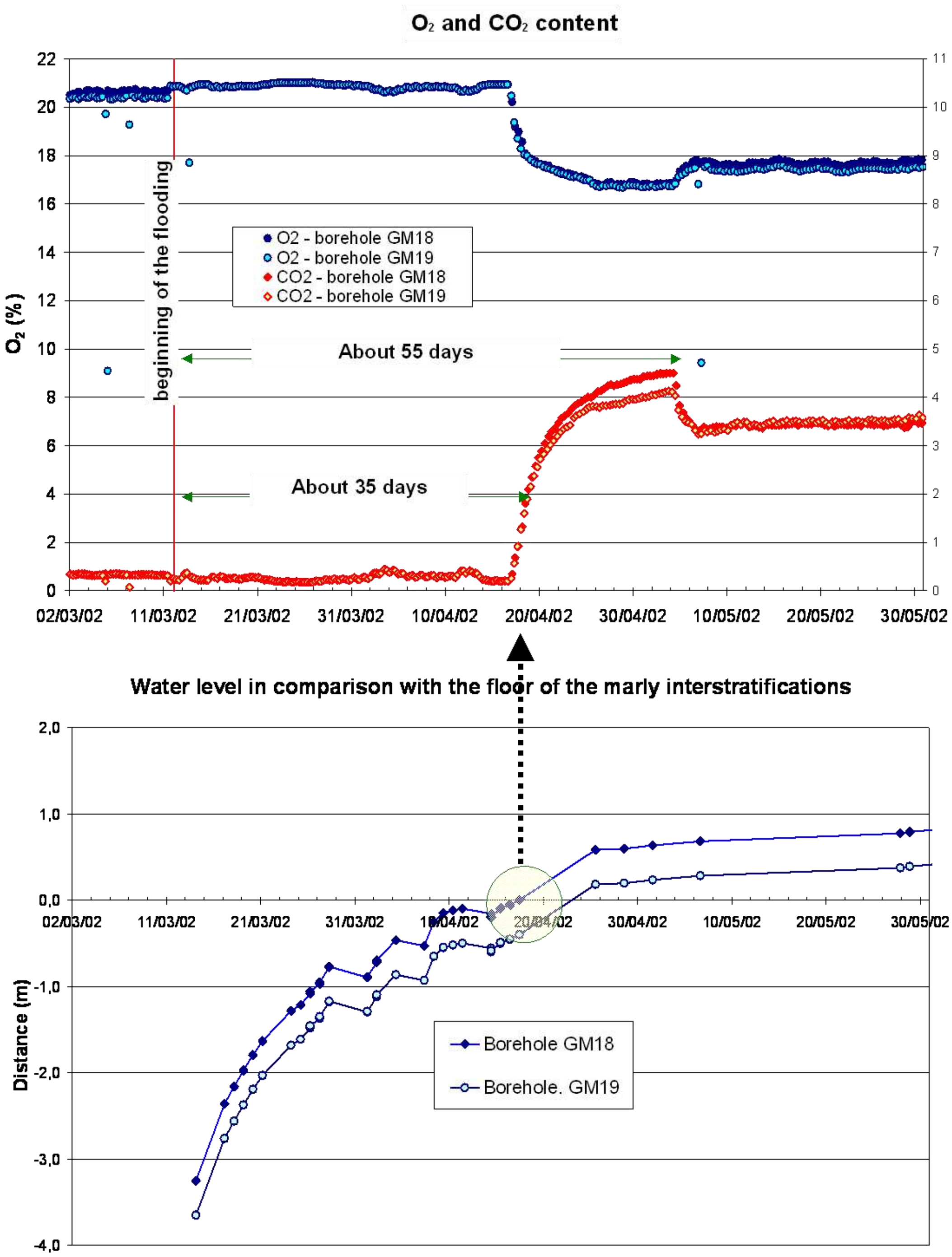


Figure 7. Evolution of the massif gaseous composition during the flooding of the Tressange pilot site.

Therefore, three main stages can be distinguished:

- Before water reaches the marly interstratifications level (35 first days), the atmosphere composition measured at the base of the boreholes does not evolve;
- As soon as the water level reaches the marly interstratifications base and progresses inside this layer (end of April 2002), the air contained in the borehole is becoming poorer in O_2 and richer in CO_2;
- Once the water level is almost stabilised in the marly interstratifications, the atmosphere composition does not evolve any more.

Such results clearly show an obvious link between the atmosphere composition modification (O_2 consumption and CO_2 production) and the marly interstratifications flooding on the experimental site.

The atmosphere modification would be resulting from a chemical reaction between the pyritic marl and the flooding water, which tends to confirm our hypothesis according to which the CO_2 emission would be due to a geochemical reaction combining pyrite from marl with calcite dissolved by produced acids.

Hydrogeochemical analysis being carried out in laboratory are in progress in the framework of the GISOS scientific program to provide additional information about the mineralogical massif composition and so to confirm this in-situ obtained result.

5. CONCLUSIONS

The non or partly flooded mine workings constitute a CO_2 laden polluted air source. The flows are generated by natural thermal draught and the initial atmosphere composition modification is very likely due to a chemical reactions set involving marly interstratifications, pyrite and calcite, in contact with water and air.

This hypothesis seems to be confirmed by the investigations carried out in real mine conditions (Tressange experimental site) where the evolution of the atmosphere in contact with the marly interstratifications has been observed.

All these results indicate that the flooding of old iron mines can lead to a production of gas (CO_2,…) and noxious emissions outside the mine.

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