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NOXIOUS GAS EMISSIONS FROM THE CLOSED IRON MINES TO THE BUILT-UP AREAS ON THE SURFACE

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ABSTRACT: Some residential areas of the iron basin of Lorraine (in the North-East of France) are affected by noxious gas emissions (under oxygenated, carbon dioxide loaded and radon loaded gas mixtures).

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

1. Introduction

A measurements campaign carried out in the iron basin of Lorraine (North-east of France) put the spot on the existence of under-oxygenated and noxious gas emissions in vast built-up areas related to former mining works. The most spectacular phenomenon occurred in the built-up area of Moyeuvre-Grande in Moselle (figure1), especially in the district of the town located very close to the former underground mine workings, in which some inhabitants observed the dysfunctioning of gas cookers and boilers.



Figure 1. Location of the studied mining reservoirs (north-east of France)

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 extend of the problem. These studies brought to the fore the existence of noxious gas emissions in relation with former mining workings that were not completely submerged. High level CO_2 content (up to 6 %) and low level O_2 content (down to 13 %) have been measured. At the same time, the Institute for Radio-protection and Nuclear Safety (IRSN) and the DASS (a government organisation responsible to the national Ministry of Health) investigated and measured high level radon content (up to 15000 Bq/m³) in the gaseous mixture.

In some cases, these emissions affected directly inhabited buildings that were shown to have a link with the mine entrances or with the old superficial mining works.

The consequences of a CO_2 enrichment and an O_2 impoverishment on health are numerous. The seriousness 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 content in inhabited buildings of the investigated areas didn't look directly risky. 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 $500 - 1000 \text{ Bq/m}^3$ (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.

2. Gas flow mechanism:

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

These stations enable to continuously monitor 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.

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



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Figure 2. Outside temperature, air flow rate and CO₂ 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 influence on the flow (like the atmospheric pressure for instance) 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 temperature difference existing between the outer atmosphere and the old workings. The temperature in the latter is usually almost constant (in that case, it was appraised to be comprised between 12 and 14 °C), unlike the atmospheric temperature, the level of which varies with 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 situated mainly 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 3. 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 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 during which the flow is globally coming into the old mine workings. This 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 contents measured in an opening of the Moyeuvre-Grande reservoir as a function of temperature. It clearly shows that the atmosphere poor in O_2 and rich in CO_2 appears for temperatures higher than 12 °C to 14 °C, corresponding to the outflow stage.



Correlation between the outside temperature and the CO2 content

Figure 4. Correlation between the outside temperature and the O₂ and CO₂ 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 temperature change of 10 °C. Such a charge seems to be large enough to create the gas circulation in this context.

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Figure 5. Estimation of the aeraulic load thanks to the hydrostatic method (with non modified dry air).

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 inverse them at long term.

3. Atmosphere modification mechanism in the mining 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 consisted in a O_2 consumption accompanied by a less than proportional production of CO_2 .

This result, in accordance with a literature review into the 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:

$$FeS_2 + 3,75 O_2 + CaCO_3 + 3,5 H_2O \Leftrightarrow FeOH_3 + 2CO_2 + 2SO_4^{2-} + 2Ca^{2+}$$

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).

A further study is in progress in order to better understand the origin of the gas generation within the old iron mine workings. This study is performed in real site conditions (Tressange pilot site) and consists in a controlled flooding of a small part of the old mine workings (GISOS, 2003).

4. Conclusions

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The non or partly flooded old mines in the iron basin of Lorraine (North-East of France) constitute a CO_2 laden polluted and under-oxygenated air source. In some cases, these noxious emissions affect directly inhabited buildings that were shown to have a link with the mine entrances or with the old superficial mining works. The main hazards associated with this flow of gas and its potential accumulation in confined volumes in buildings are asphyxiation and intoxication.

The gas flows are generated by natural thermal draught. A numerical simulation showed that the aeraulic charge due to this phenomenon was estimated, in the local conditions, at 20 to 30 Pa for a temperature change of 10 °C. Such a charge seems to be large enough to create the gas circulation in the studied mine workings.

The initial atmosphere composition modification is very likely due to a set of chemical reactions involving marly interstratifications, pyrite and calcite, in contact with water and air.

A further study is in progress in order to better understand the origin of the gas generation within the old iron mine workings. This research is performed in real site conditions (Tressange pilot site) and consists in a controlled flooding of a small part of the old mining workings.

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