QUANTIFICATION OF RADON MIGRATION FROM A URANIUM MINE THROUGH THE SOIL INTO BUILDINGS BY THE USE OF TRACER TECHNIQUES

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ABSTRACT:

This paper describes the results of a series of tracer gas tests performed in the mining community of Schlema in eastern Germany. The purpose of these tests was to determine the influence of various mechanisms and subterranean features on the radon levels in the ambient air and in the buildings of the community. Under the former Democratic Republic of Germany (the DDR regime), the mines in and near Schlema and in the ore mountains in Sachsen were an importance source of uranium. These mines have now been closed down and the area is currently under remediation. The remedial measures being applied are varied and consist of, for example: flooding the lower levels of the mine with water, filling the upper tunnels under the community with rock aggregate and blocking their entrances and applying impermeable layers and vegetation over the tilling dump piles near the community. The subterranean complex under the community is extensive due to the many centuries of mining activity. It is currently under depressurization due to the operation of huge mine exhaust fans whose primary function is to provide adequate ventilation air and environmental control for the miners working underground. The mining company WISMUT has undertaken several projects to study the effects of the remediation and the closing down of the mine on the nearby communities.

The project described here is one of these projects which was designed to determine the importance of air currents emulating from the mine complex on the radon levels in the community and to determine the influence of the flooding of the mine and the importance of the depressurization caused by the mine exhaust fans on the radon levels in the community. The purpose of this project was to quantify the component of the radon flux into the buildings from the uranium mine caused by the flow of air currents from the mine both when the mine was depressurized by the operation of the mine exhaust fans and when the mine exhaust fans were shut off.

Tracer gas measurements were carried out in two phases: with the main exhaust fan of the mine turned off and then with the exhaust fans on. By seeding the tunnels of the mine with a tracer gas, SF_6, the transport of air from tunnels in the mine through the soil above and then through the foundation of the buildings into the cellar was determined. Simultaneously the air change rate in the cellar was measured by the use of PDCB (Perfluorodicyclobutane) as a tracer gas to allow a complete mass balance of tracer for the cellar. A simple mass balance using the air flows calculated from the tracer gas measurements and the measured radon concentrations in ambient air and the mine was used to predict the radon level in the building due

to the air flows from the mine into the buildings. These predicted radon levels were compared to measured radon levels in the buildings. Fifteen (15) buildings in different areas of the community were examined. Some buildings exhibited almost immediate classical exponential tracer build-up response curves which indicated a strong communication with the mine tunnel complex. The calculated radon concentrations in the buildings based on the tracer measurements were in good agreement with the measured radon concentrations in the buildings, i.e. the buildings' radon concentration could be well predicted using the air flows from the mine into the buildings and the radon concentrations in the mine tunnels. Additional preliminary measurements of tracer migration from mine complex into radon dumps and more distant subterranean parts of the complex for which there was no direct flow paths indicated that the tracer technique used would be very well suited for also studying underground movement of contaminants including the determination of contaminant egress from radon dump piles in the area.

Introduction

The small mining town Schlema is located in the south of Saxonia, 15 km south-west of Chemnitz and 10 km north of the Czechoslovakian border.

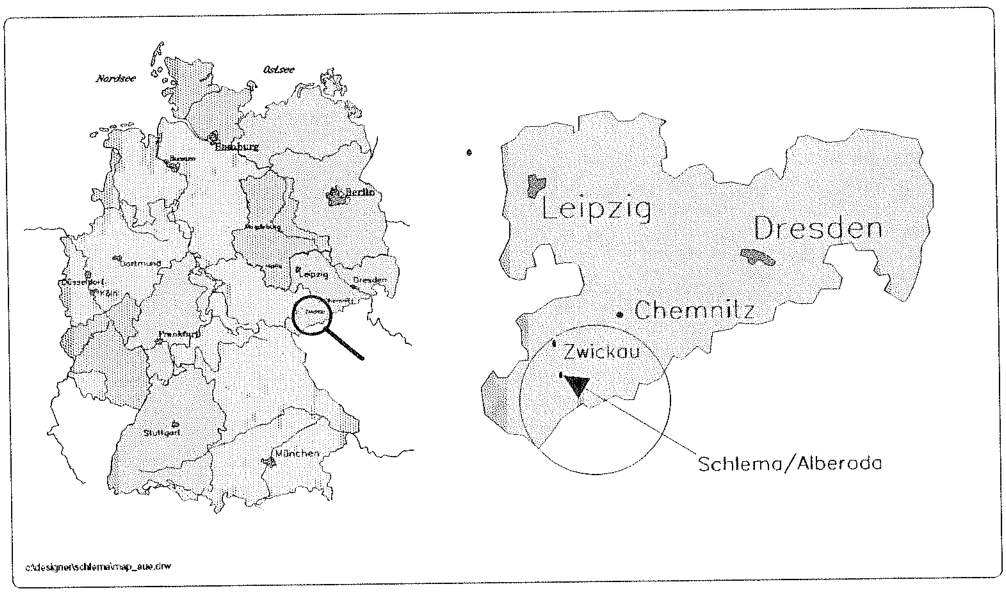

Figure 1. Map of Germany (left) and Saxonia (right) with the location of Schlema/Alberoda

After World War II the Russians started to extract uranium ore from the mines near Schlema. However, the region had been an active mining area for several centuries. The area around Schlema was very well known for the diversity of the ores and minerals found there. The neighboring village of Schneeberg was famous for its silver mines which were first exploited by the region's first silver miners in the 16th century. The silver miners from Schneeberg dug a tunnel, which is called the Markus-Semmler-Stollen (MSS tunnel) from Schneeberg through the Schlema valley. The tunnel ends at the river 'Mulde' in Schlema. The MSS tunnel was built to drain the silver mine in earlier times. It is 15 - 35 m beneath the surface, approximately 1 m wide and 2 m high. The uranium ore beneath the Schlema valley was extensively exploited during the last four decades. The mining activity created a complex of tunnels, cracks, fissures, cave-in areas and unknown flow paths. In the 1970's and 80's the mining activities moved deeper into areas below the Schlema valley, eventually reaching depths of more than 2000 m. Remedial flooding of the lower levels of the mine has filled the lower levels to a depth of about 1000 m below the surface. Since the radon exhalation is still very strong, it was necessary to keep the old mining area in the valley of Schlema depressurized to keep the radon concentration at tolerable levels.

Fig. 2 shows a map of Schlema. The thick line shows the underground mining area, the dashed line marks the MSS tunnel with its entrance at the river 'Mulde'. The main air supply and exhaust shaft of the mine are 3.5 km north east of Schlema. The reasons for elevated radon concentrations in this area are potentially multiple.

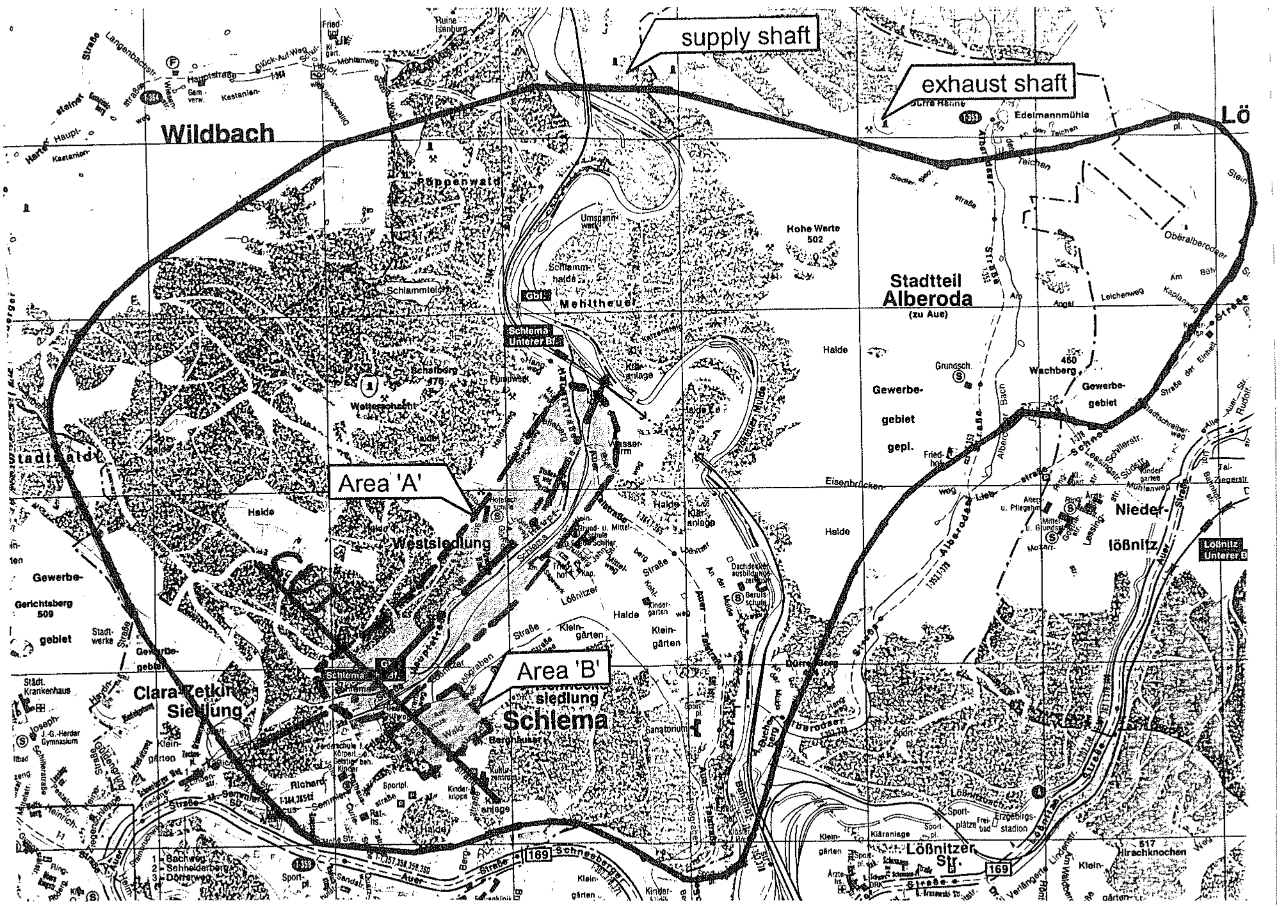

Figure 2. Map of the Schlema valley with the MSS tunnel, Area 'A' and 'B' and main exhaust and supply shafts

Fig. 3 shows the most important potential radon paths. These possible paths include: the traditional near field flow of ambient air into the soil and then into the building due to the natural driving forces which the building causes by depressurization produced to the "stack" and wind effects; the movement of air from the mine directly into the buildings; the movement of air from the mine into the exterior ambient through fissures and cracks in the soil; the exhalation of air from the mine shafts into the ambient; the exhalation of air from the tilling dump piles which could be a combination of air movements caused by wind and thermal variations of the ambient or by air currents from the mine through the dump piles; and also the possible contamination of the valleys near the exhaust fans from the mine.

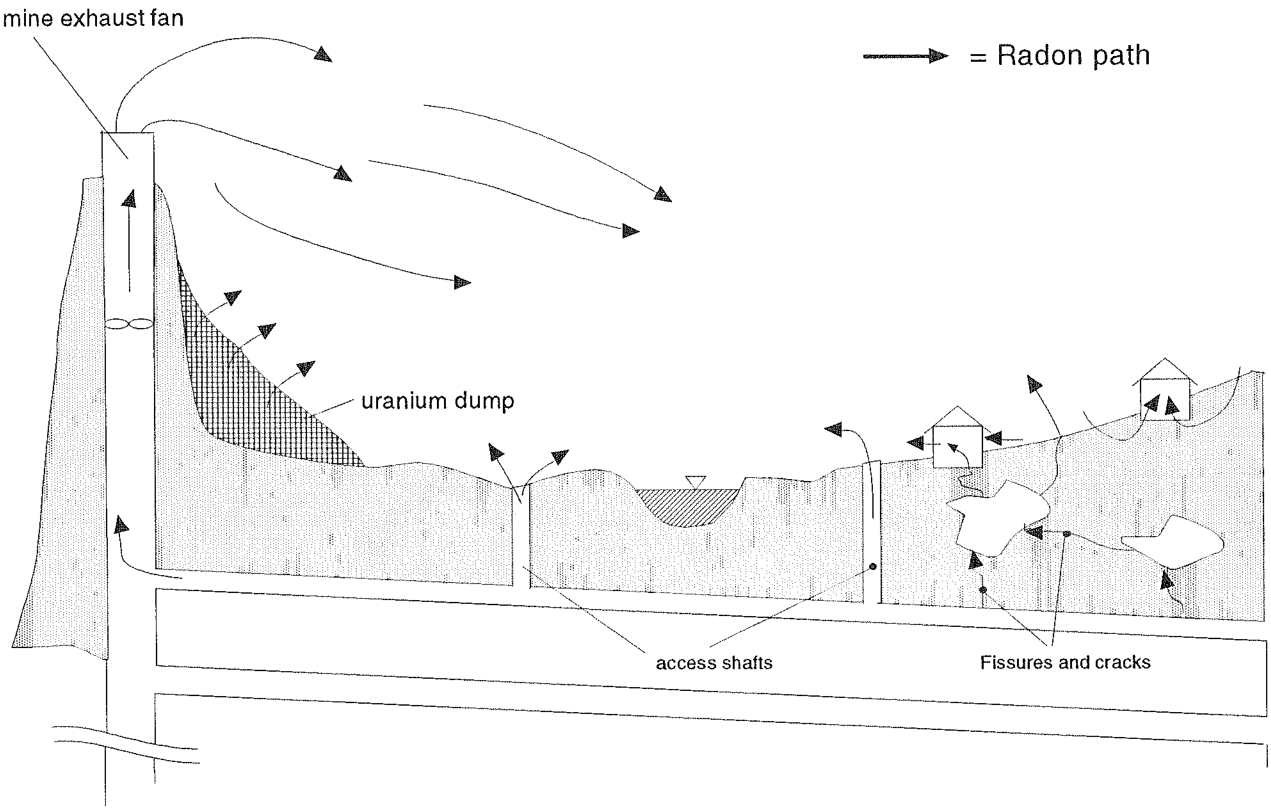

Figure 3. Sketch of the potential radon sources and paths in the Schlema valley

Since the radon levels in the dwellings could be caused by different sources and flow paths and each influence the others in a very complex manner, it is currently not possible

- to quantify the radon emissions through the soil by measuring the radon concentration alone
- to predict the radon exposure to the people due to a change in the mine ventilation.

One way of approach to answer these questions is the use of tracer technology in which separate parts of the subterranean complex are seeded with a tracer gas and the migration of the gas monitored over a period of time.

3. THE EFFECT OF THE VENTILATION OF THE MINE

3.1 Forced ventilation with exhaust fans in operation

Fig. 4 shows a schematic of the main airflow paths and directions of the mine. It can be seen, that the mine exhaust fans draw one part of air through the main supply shaft and the other part through the MSS tunnel and from other diffuse openings and paths to the surface. This depressurizes the underground of the Schlema valley and keeps radon concentrations in the houses low.

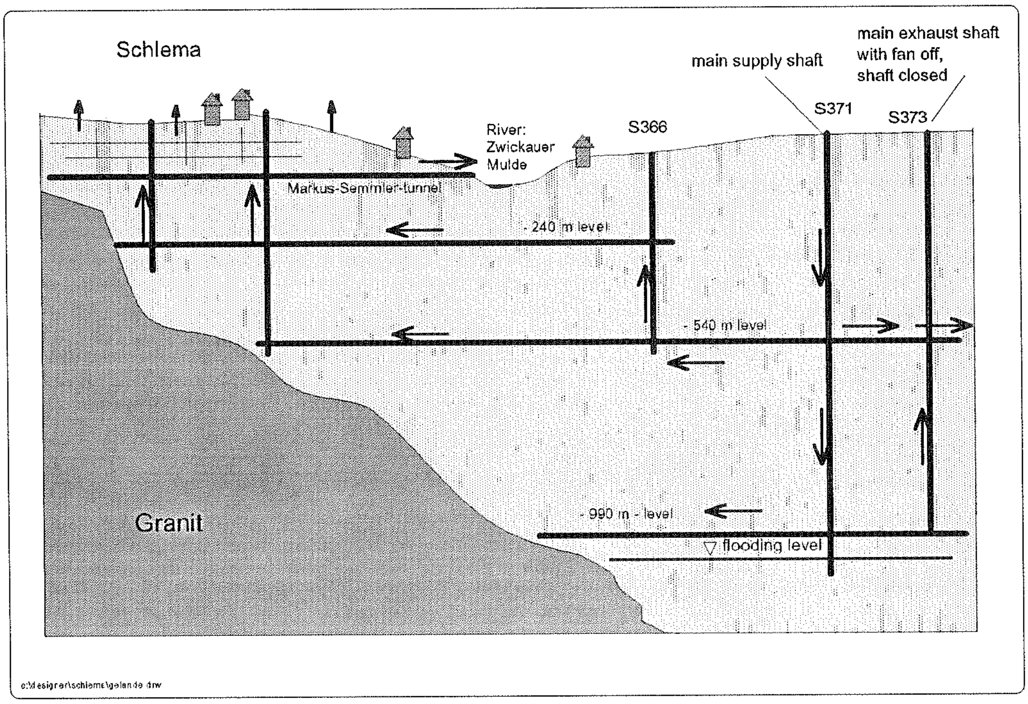

Figure 5. Schematic of airflow paths in the mine with the main fan off

5. TEST PROCEDURE

5.1 Tracer injection into the mine

There were two test phases: phase I under natural ventilating conditions and phase II under normal mechanical ventilation. It was planned to inject a constant rate of SF_6 tracer into the MSS tunnel to achieve an almost constant concentration in the mine. The target concentration was ~ 10 ppm, though it was felt that, given the sensitivity of the tracer monitors, a level of 100 ppb would suffice. The injection flow rate was set at a value considered sufficient to produce 10 ppm due to the lack of precise information on the extent of the underground complex and the porosity and permeability of the soil.

All injection and sampling tubes were installed two weeks before the tests since the mine was closed with the main fan off due to the high radon concentrations. Polyethylene tubing (6 mm inner diameter for sampling and 4 mm i. d. for dosing) was used. Air samples were drawn from various locations in the MSS using dual-head pumps. Syringes were drawn from the pump exhausts for later analyses.

Pure SF_6 was injected via a pressure regulator and a mass flow meter through dosing tubes into the MSS tunnel.

5.2 Sample locations and concentration measurements

From all the buildings, air samples were sequentially drawn at approximately 4 hour intervals and analyzed for the arrival of tracer molecules. Additionally, ambient air samples up wind of each building were also taken using syringes and the syringes were analyzed for SF_6. The radon concentration in the mine, inside the buildings and outside the buildings were measured. To allow a complete mass balance for each building, air exchange rate measurements were

made using the tracer-decay method with a second tracer gas, Perfluorodicyclobutane(PDCB)
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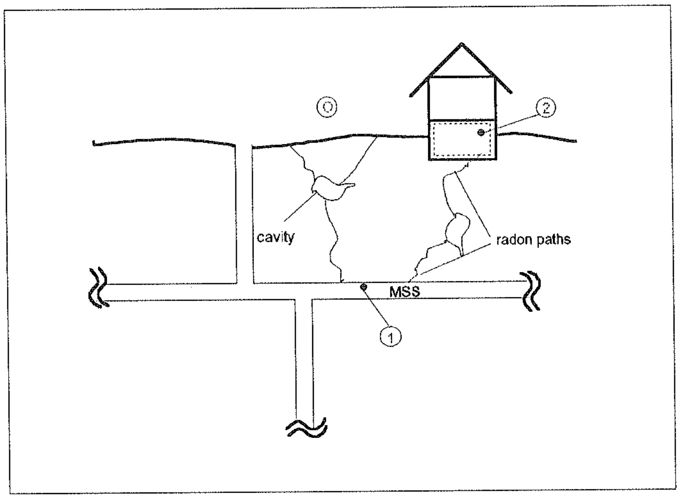
Fig. 6 visualizes the mass balance of SF_6 for a cellar.



$$Q_{12} = Q_{20} \frac{C_{S,2} - C_{S,0}}{C_{S,1} - C_{S,0}} \quad (1)$$

$$C^*_{Ra,2} = \frac{Q_{12}}{Q_{20}} (C_{Ra,1} - C_{Ra,0}) + C_{Ra,0} \quad (2)$$

Figure 6. Schematic of SF_6 mass balance in a cellar of a building

Q_{12} = volume flow rate of air from the MSS tunnel (zone 1) through the soil into the cellar of a building (zone 2)

Q_{20} = volume flow rate of air from the cellar (zone 2) into the ambient (zone 0)

C_{SO}, C_{S1}, C_{S2} = SF_6 concentration by volume for zones 0, 1 and 2

$C^*_{Ra,2}$ = predicted Radon concentration in the cellar of the house due to airflow from the MSS tunnel.

$C_{Ra,1}$; $C_{Ra,0}$ = measured Radon concentration in the MSS tunnel (zone 1) and in the ambient air outside the house (zone 0)

To calculate the airflow Q_{12} from the MSS tunnel through the soil into the cellar, the exfiltration airflow Q_{20} of each house must be known. Q_{20} was obtained by an air exchange rate measurement (see section 5.4) using the decay of the PDCB tracer.

5.3 Tracer gas equipment

SF_6 and PDCB were detected by gas chromatography with an electron capture detector (GC-ECD). The detection limit, range and accuracy of the equipment used are shown in table 1.

	Tracer Gas	
	SF_6	PDCB
Accuracy	3% of reading	3% of reading
Detection Limit	< 50 ppt	< 50 ppt
Detection Range	0.050-50ppb	0.1-40 ppb

Table 2. Technical Specifications of ECD gas chromatograph AUTOTRAC Used for the Measurements. Samples above the upper detection limit were diluted with a tracer free gas before analysis.

5.4 Air exchange rate measurement

To clearly distinguish between ambient air and soil gas entry into the cellar of the house, a SF_6 mass balance of the cellar volume had to be made. Fig. 7 shows the flow paths of air

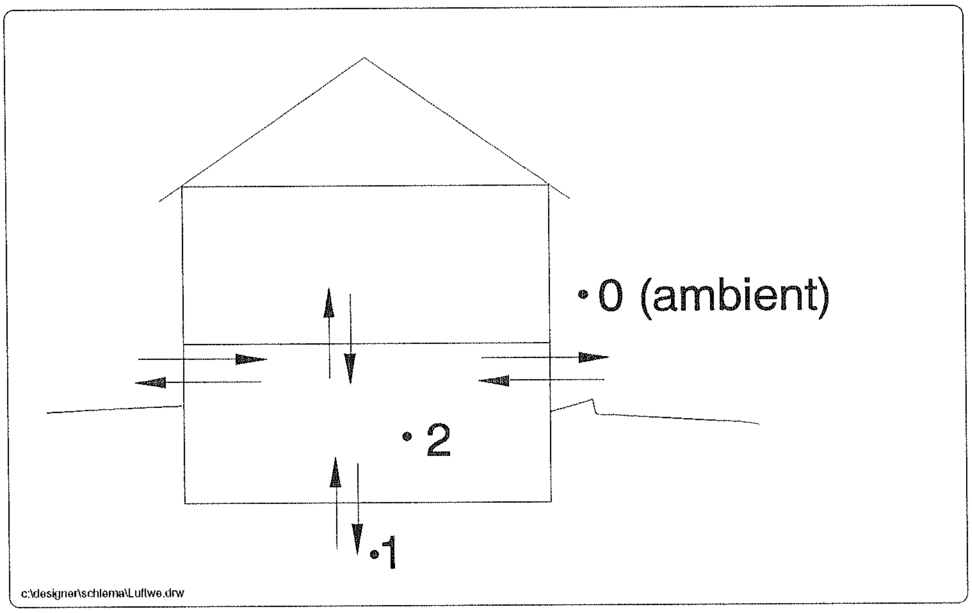


Figure 7. Flow paths into and out of the cellar zone

and SF_6 inside and outside of the cellar. To measure these airflows, the air in the cellar was tagged with PDCB. The decay of the tracer with time after stopping injection is a measure of the air change rate. If the tracer concentration is plotted on a logarithmic scale versus time, the slope of this curve is equivalent to the air change rate, n. For this part of the tests, the tracer monitor AUTOTRAC automatically analyzed the tracer data from each building and calculated the air change rate using a regression method. The data were also entered into a specially designed spread sheet template which checked the analysis.

A 2 l bottle of 0.2 % PDCB in N_2 was used for injection by releasing the mixture for 5 to 10 minutes injected at a flow rate of 0.2 l/min. The advantage of the small bottle was that one could walk with it through the cellar area slowly injecting the tracer in all parts of the cellar. This improved the mixing process of PDCB with the room air considerably. The initial concentrations were between 100 to 300 ppb. Fifteen (15) minutes after the end of injection, the first air sample was drawn by a 50 ml syringe. The advantage of a syringe is also that you can walk through the cellar rooms while slowly drawing air into the syringe to obtain a local mean concentration. At intervals of approximately 15 minutes, 4 additional air samples were taken. Fig. 8 gives an example of an air change rate tracer decay in building B2.

If the room volume V_R of the cellar is known, the exfiltration rate, *n* (air change rate) can be obtained from the equation:

$$Q_{20} = n \cdot V_R$$

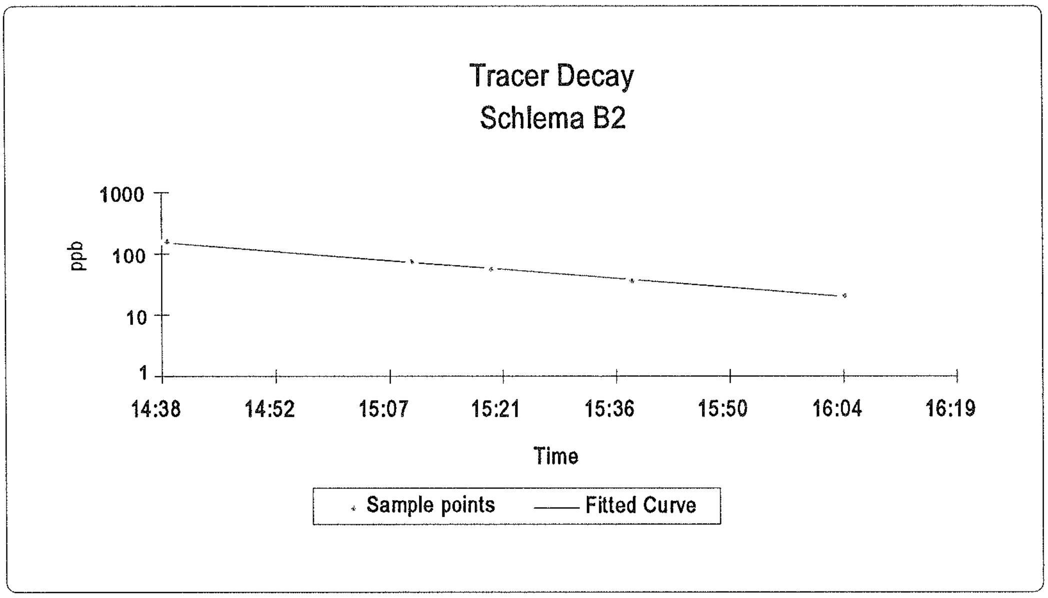


Figure 8. PDCB tracer decay curve used to determine the air change rate in building, B2

6. MEASUREMENT RESULTS OF PHASE I - MINE EXHAUST FAN OFF

6.1 Buildings in the Schlema valley, area A

The tracer test phase I was performed between Christmas and new year 1993. Ambient temperatures varied between -2 to - 12°C at night with weak winds from west parallel to the Schlema valley. A constant tracer gas flow was injected at diverse underground locations to insure an almost constant concentration of SF_6 in the mine. Fig. 9 gives a detailed sketch on the flow in the main tunnels and shafts. UG 113 and UG 91 were 2 x 2 m shafts for the miners to have direct access to the MSS tunnel to do remediation work.

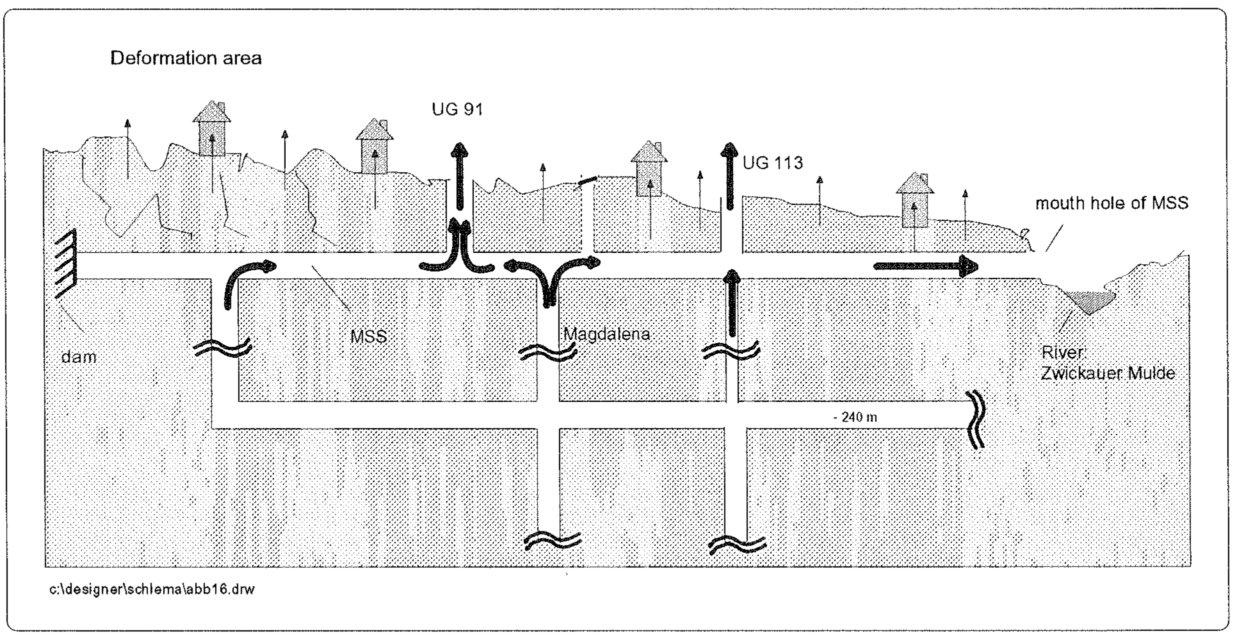


Figure 9. Main flow paths around the MSS under natural ventilation in the mine

Fig. 10 displays the measurement results for building B 2. On Dec. 28 at 14:36 the dosing in the MSS tunnel section below B 2 was started. At 16:45 the equilibrium concentration of ≈ 7 ppm war reached. The first SF_6 molecules in the cellar of B2 were

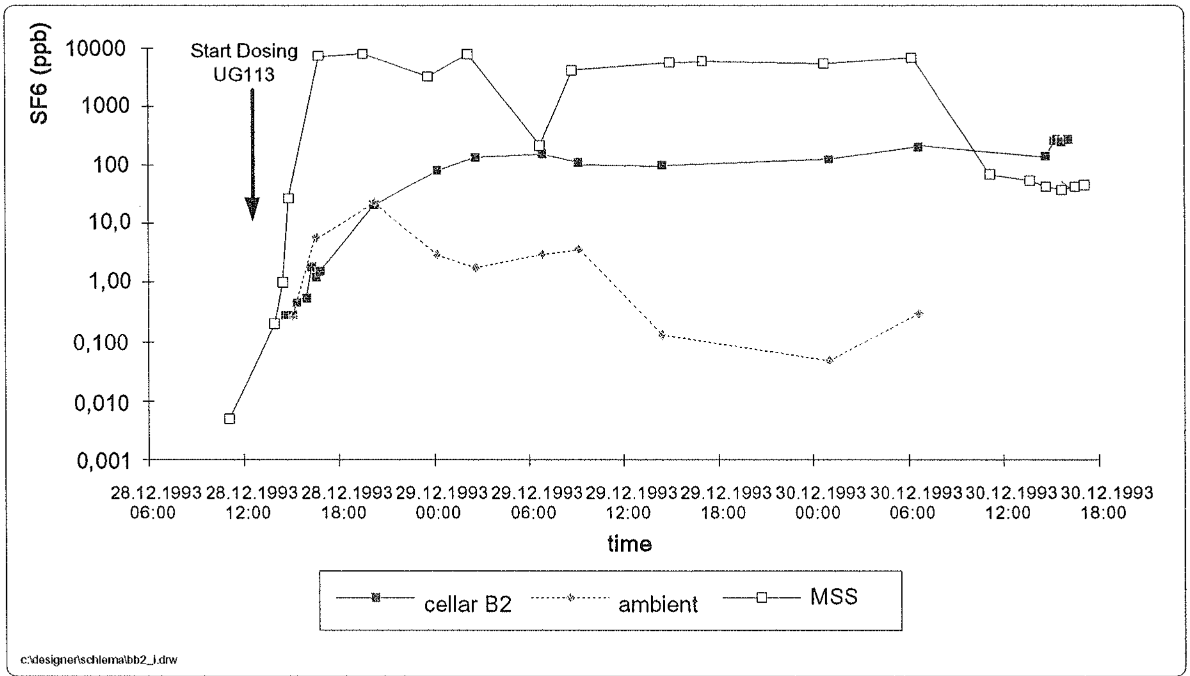


Figure 10. Tracer Response for Building B2.

detected only 45 minutes after dosing start at 15:20 with concentration of 450 ppt. With a direct path length of approximately 15 m through the soil to the building foundation, the data indicate that the mean velocity of the flow from the tunnel to the building was 0.33 m/min.

The air change rate of the building B2 was measured as $n = 1.43h^{-1} \pm 0.04$, see Fig. 8. Applying equation 1, the airflow through the soil into the cellar was calculated to be 11 m^3/h. In building B2 the radon concentration was measured twice, once as a 3 day mean concentration with passive E-Perm samplers and also continuously with Alpha GUARD devices. Fig. 11 shows the measured and predicted radon concentration in B 2 during a time span of 3 hours. Due to random oscillations in the mine it was not possible to keep the underground tracer concentration constant. Therefore data uncertainties were in some cases significant.

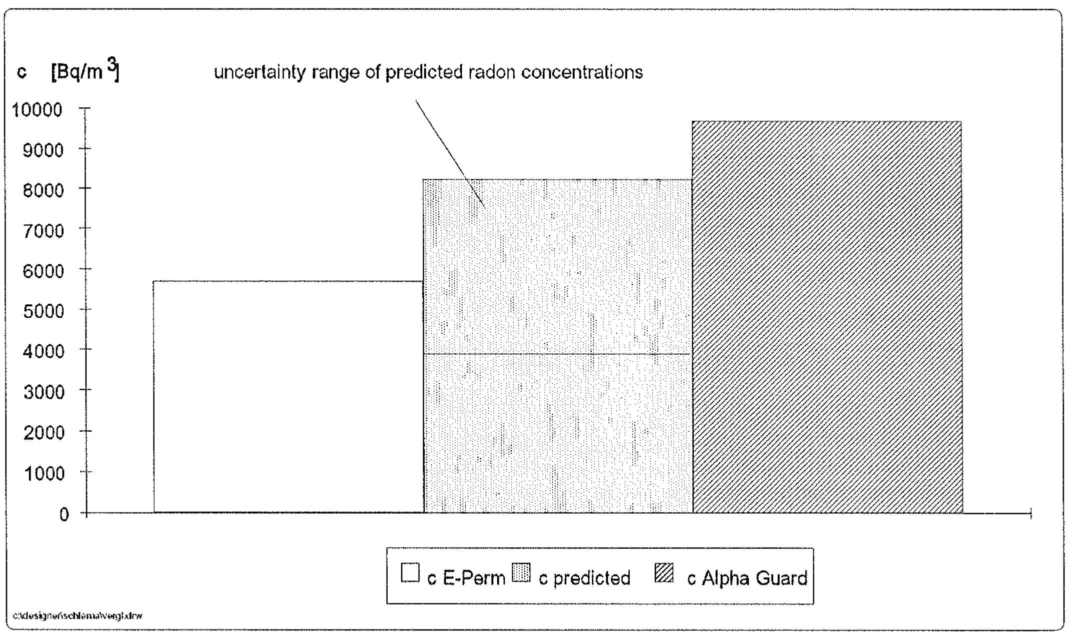


Figure 11. Measured and predicted radon concentrations in building B2

The soil gas flows of six buildings out of ten in the area Schlema valley are summarized in Fig. 12. Fig. 13 shows the measured and predicted radon concentrations according to equation 2. Only in B4 was the agreement between measured and calculated radon concentration poor.

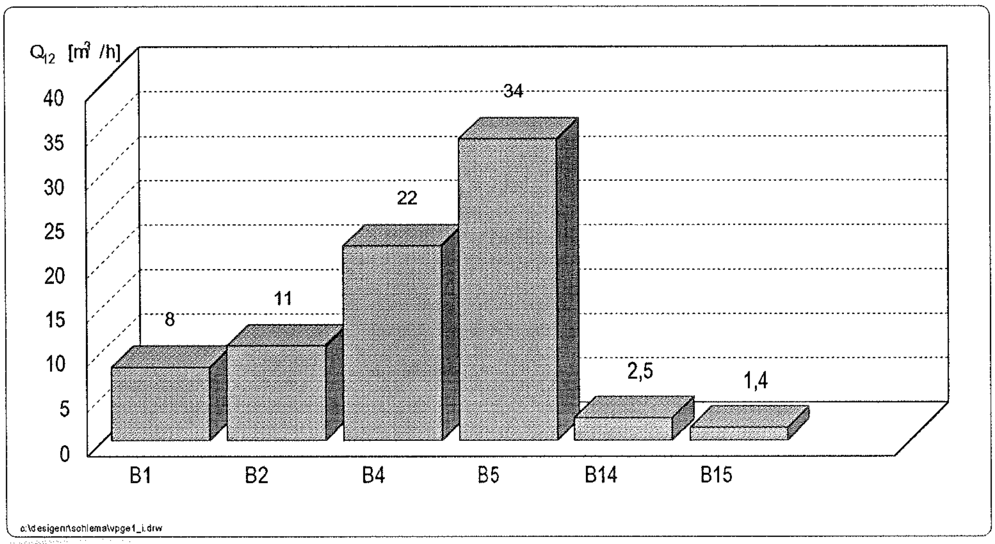


Figure 12. Measured airflows from MSS into buildings of the Schlema valley during measurement phase I, main fan off

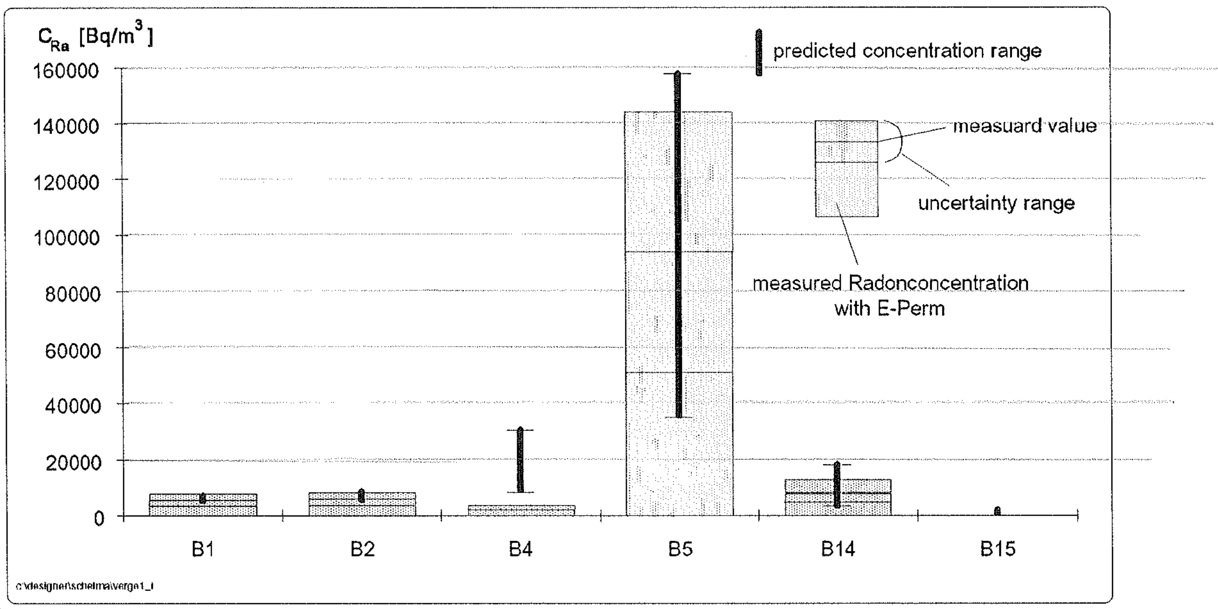


Figure 13. Measured versus predicted radon concentrations in buildings of the Schlema valley during measurement phase I, main fan off

Looking at building B 5, the highest mean radon concentration of 98000 Bq/m^3 was measured. Although the uncertainty is high, the predicted mean value is in excellent agreement with the measured value. In this case, the airflow through the soil contributed to 21 % to the infiltrated air in the cellar. The data from this phase indicate that the air flow from the mine into the buildings was the principal mechanism which caused the excessive radon levels in the buildings when the mine exhaust fans were shut down.

6.2 Buildings in area 'B', 60 m uphill from the MSS tunnel.

As mentioned above, there was no direct connection known between the main mine and the small tunnel (LL2-ST3) for reconstruction work below the 5 selected buildings of area 'B'.

Fig. 14 shows a vertical cut through MSS area 'A' and area 'B'.

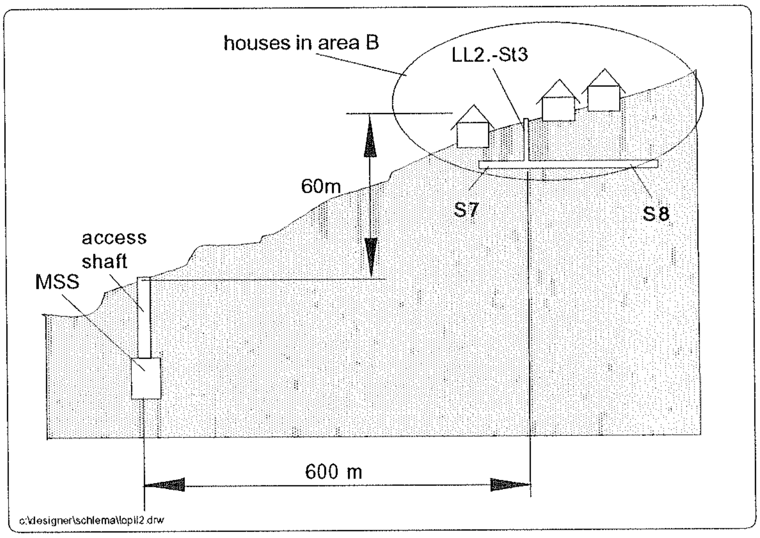


Figure 14. Topography between area 'A' and 'B'

The dosing of SF_6 into LL2-ST3 started one day later than in the MSS tunnel. This was to check if SF_6 tracer molecules arrive in area 'B' from the tracer injection in area 'A'. Fig. 15 clearly indicates that 6 hours after injection start in MSS the first SF_6 molecules arrived in the left part (S 7) of tunnel LL2-ST3.

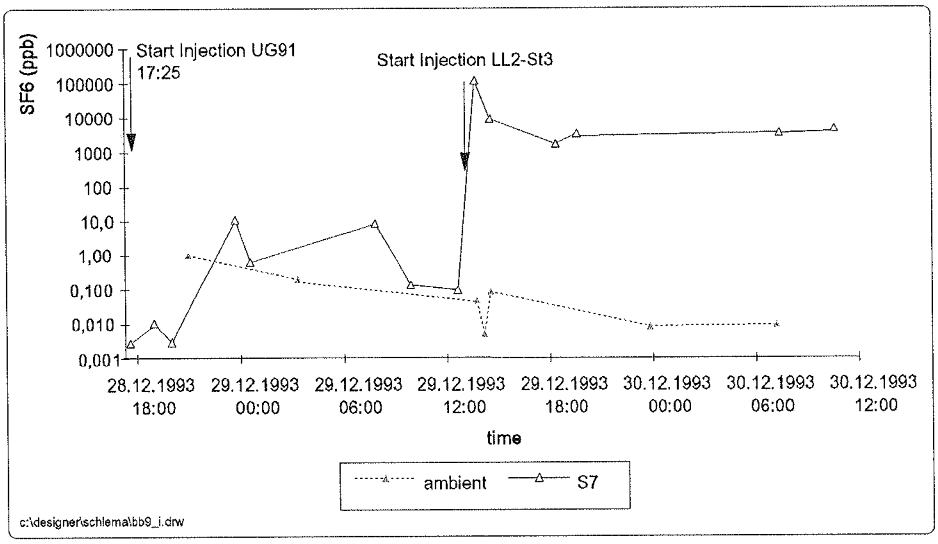


Figure 15. SF_6 arrival from MSS in tunnel LL2-ST3 at sample point S7

It is assumed that there exists an uphill convection flow through fissures, porous soil and cracks to the 60 m higher area 'B'. The very low ambient temperatures ≈ - 10°C enhanced the stack effect and this convection flow.

7. MEASUREMENT RESULTS OF PHASE II - MINE EXHAUST FANS ON

During Jan. 24 - 28, 1994, with the mine exhaust fans with a capacity of 226 m^3/s operating (note: the operation of these fan can require up to 4 megawatts of electrical generating capacity), the second measurement phase was conducted. The ambient temperatures were with + 2 to + 7°C about 10 K higher than during phase I. The wind speed was a little higher, but had the same direction west-east through the Schlema valley.

The results of phase II are summarized in Fig. 16 and 17.

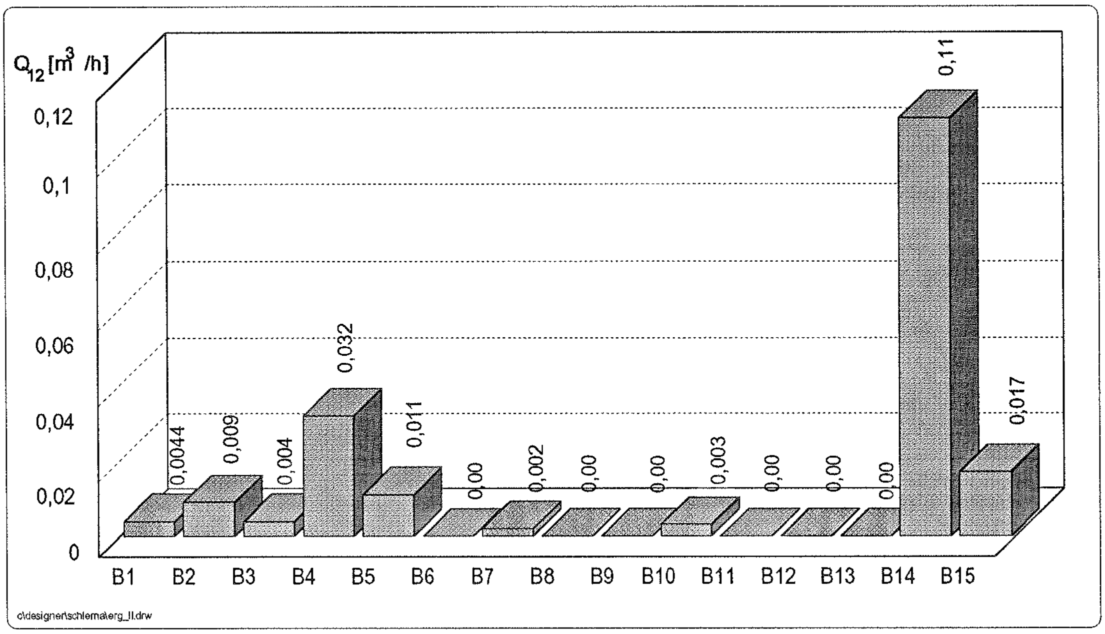


Figure 16. Measured airflows from MSS into buildings of the Schlema valley during measurement phase II, when mine exhaust fans were in operation

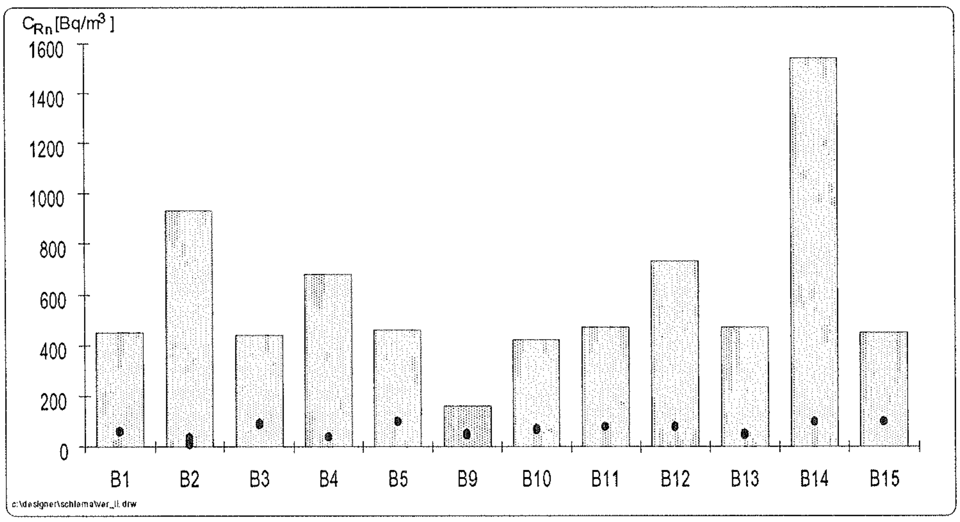


Figure 17. Measured (bars) versus predicted (points) radon concentrations in buildings of the Schlema valley during measurement phase II, when mine exhaust fan was in operation. The bars indicate the predicted concentrations.

If we compare the airflows through the soil of phase I and II, a reduction by a factor of 700 to 3000 had taken place. In Fig. 17 we see that the measured radon concentration is underestimated by concentration predicted by using only the radon transported by air movement from the MSS tunnel. This would indicate that in contrast to the situation with the fan off the airflow from the mine is not the main source of radon in the dwellings when the main mine exhaust fan is operating. Fig. 18 tries to explain the reason for this discrepancy.

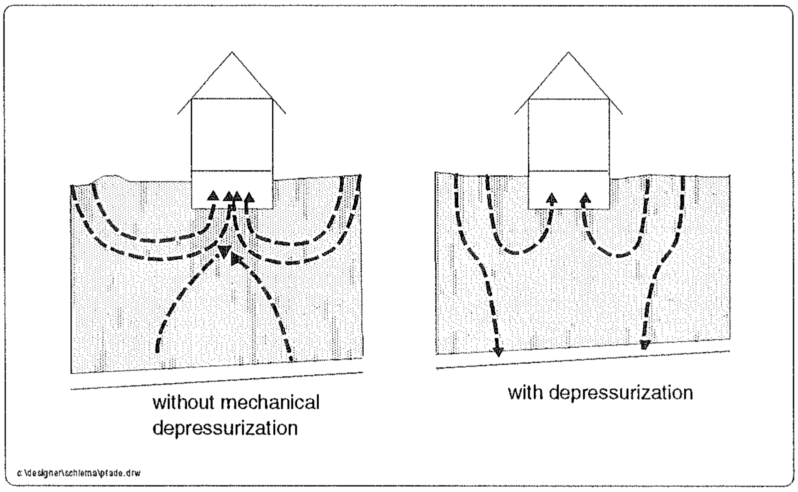


Figure 18. Flow paths of radon and air with and without mechanical ventilation

With no mechanical depressurization of the mine, air with around 450 kBq/m^3 moves from the very deep tunnels in the mine into the MSS tunnel and a small portion of this flow enters the buildings through cracks in the soil. There may be a further enrichment with radon of the air from MSS into the building as the uranium rocks near the surface also emit radon. This effect is not accounted for in the calculation and mass balance in equations 1 & 2. For phase I the results prove that a possible enrichment of the airflow from MSS into the cellar was negligibly small. The typical building induced natural radon path, where ambient air moves into the soil around the building and enters the building due to pressure differences caused by the stack effect of the building, had also a very small impact during phase I. The good agreement of the results supports this assumption.

But with mechanical depressurization, the flow from the MSS tunnel into the buildings was very effectively reduced. What still remains is the natural pathway of air from the surface around the building, into the soil, and then into the cellar. The magnitude of these volume flows may be much smaller - but with a continuous generation rate of radon in the soil, the radon concentration of the soil air is higher and produces still significant high radon concentrations in the buildings.

The depressurization of the mine and the change of the pressure field around the building may also effect the natural pathways of radon in such a way, that air from the surface, which, without depressurization would enter the building, would now be directed into the mine (see Fig. 18). This would cause an even lower radon level in the buildings then if there were a not depressurized mine beneath the buildings.

8. ADDITIONAL SUPPLEMENTARY TEST RESULTS

One day before the phase I tracer test in the mine was conducted, a pulse test into the main supply shaft of the mine was made to

- estimate the air velocity from the supply shaft to MSS
- check if there is an air path between the mine and a uranium dump, which was above the mine.

8.1 Air velocity in the mine

Fig. 5 shows the airflows in the mine under natural ventilating conditions. At Dec. 27, 1993 at 14:20 a 10 l bottle of pure SF_6 was emptied in 5 minutes into the main supply shaft. The shortest way of the SF_6-tagged supply air to reach the MSS tunnel was to go down to the -540 m level, then 3.5 km horizontally, then go up again to the -240 m level and then arrive at MSS. As shown in Fig. 19, at measurement point S6 in the MSS tunnel

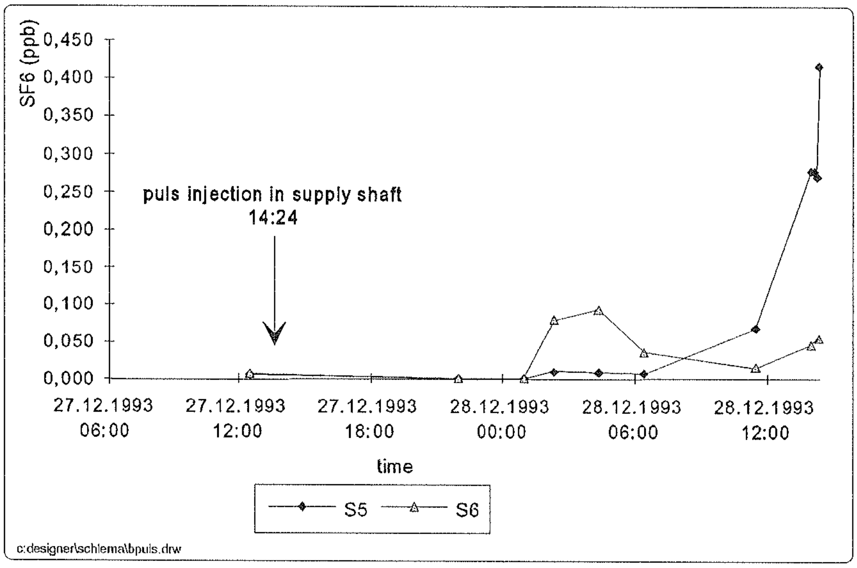


Figure 19. Concentration histories in MSS tunnel after the pulse injection

the arrival of the first SF_6 molecules was on Dec. 28 at 2:20 (12 hours after injection). At sample point S5, the pulse arrived on Dec., 28, at 11:28 (21 hours after injection). With a total flow path length of approximately 4 km, the maximum air velocity was estimated at 2.8 m/min.

8.2 Check of communication between the mine and an uranium dump

Fig. 20 shows a scheme of the mine and an uranium dump on top of it.

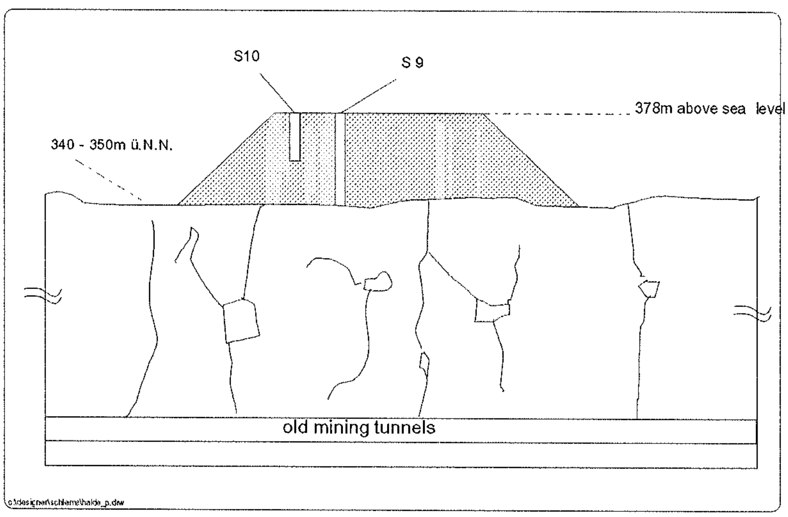


Figure 20. Schematic of mine and uranium dump with air paths

The dump has an oval form with a length of ~ 900 m, a width of 300 m with a top height of ~ 30 m above the ground level. There was one bore hole which passed down to the dump sole (S 9) and the other bore hole which was only 6 m deep (S 10). After the pulse injection, air samples were also taken at these two sampling locations. It should be noted, that the ambient temperature was -12°C at night, very low. This led to a strong stack flow from the bottom of the dump to the top surface. On Dec. 29, at 0:22 the first SF_6 molecules arrived on top of the dump as can be seen in Fig. 21

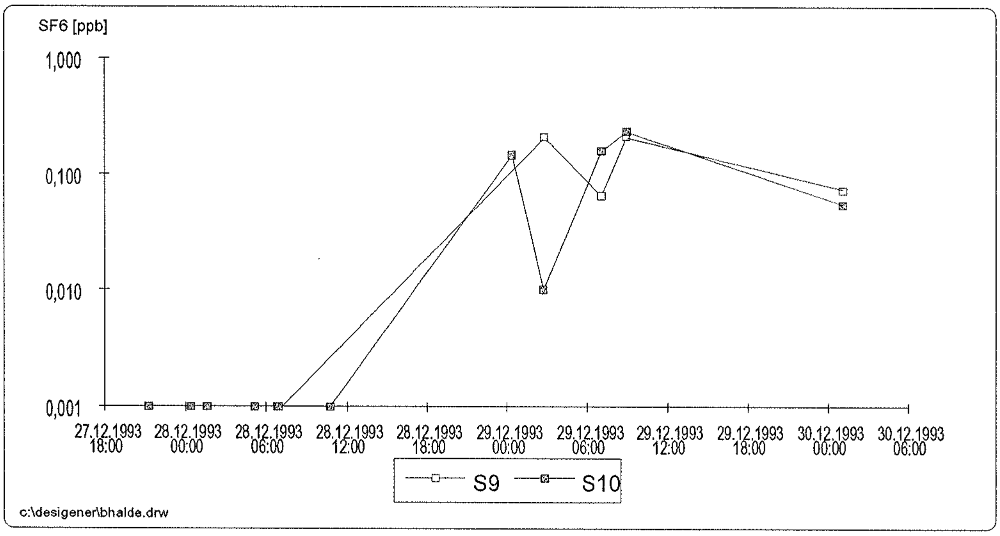


Figure 21. Measured SF_6 concentrations on top of the uranium dump after the pulse test

9. CONCLUSIONS

The tests reported here showed that tracer technology is capable of precisely measuring air-flows in mines and through the soil and can be a powerful tool for determining the importance of various mechanism on the radon levels in buildings (and potentially, the movement of other contaminants through the soil). The tests clearly demonstrated the effect of the operation of the mine exhaust fans on the radon levels in the ambient atmosphere and in the dwellings in the community of Schlema located over the mining complex. The tests also showed that air for the mine flows through the soil even if direct tunnel connections do not exist and can cause exhalation of radon, for example, at the top of the tilling dump piles over the mine. The importance of these flows on the ambient radon levels in the valleys near the dumps will require further testing. Essential for these kind of measurements is the choice of a good tracer and sensitive analyzing equipment (ppt level sensitivity). SF_6 proved to be a very good and inexpensive tracer for this purpose. Due to the high sensitivity and automated features of the AUTOTRAC GC-ECD used, it was possible to analyze and process a large number of samples and obtain reliable results even in the ppt range, minimizing the quantity of tracer required.