

# A new in situ gas diffusion experiment: objectives, design and experimental protocol

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## Abstract

Over the last decade, SCK CEN obtained a large set of diffusion coefficients for different gases in different Members of the Boom Clay. These diffusion experiments were performed in the lab on Boom Clay samples with diameter 80 mm and length 30 mm. When performing safety calculations, diffusion of gas is assumed to take place within an approximate domain over 100's of meters. This led to the question: is the lab-scale diffusion coefficient also valid on a larger (meter) scale? In the past, a similar question was raised for more classical tracers such as HTO and iodide, and several *in situ* diffusion experiments lead to the confirmation of lab results. Hence, SCK CEN, ESV Euridice and ONDRAF/NIRAS will perform a new *in situ* diffusion experiment with dissolved neon in the HADES URL with as main objective to confirm/improve the current knowledge on diffusion of dissolved gas at a large scale.

The experiment preferably takes place under conditions that are relevant for the Boom Clay, thus not in an excavation damaged zone. Therefore, it was our aim to re-use an existing setup. All setups in HADES were screened against a list of criteria, and the MEGAS setup (drilled in 1992) was considered the best option.

Experiments with hydrogen are often complicated by the microbial conversion of hydrogen into methane, and therefore neon, considered to be the best proxy for hydrogen, will be used.

The experiment will consist of one injection circuit and three monitoring vessels. Dissolved neon will diffuse from the injection circuit/filter, through the clay, in the monitoring filters/vessels. The monitoring vessels are connected to a gas analyser which performs on line analysis of the neon concentration. By monitoring the concentration increase of neon in time in the monitoring vessels, the diffusion parameters can be determined.

This report describes both the technical aspects of the experiment, and the protocol to be followed.

## Keywords

Diffusion, gas, dissolved gas, Neon, MEGAS, HADES, gas transport, *in situ*

## 1 Objectives of the experiment

ONDRAF/NIRAS wishes to determine parameters associated to gas diffusion in Boom Clay. In addition to laboratory experimental programme, an in situ experiment aims to confirm and/or improve the current knowledge on diffusion of dissolved gas at a larger scale.

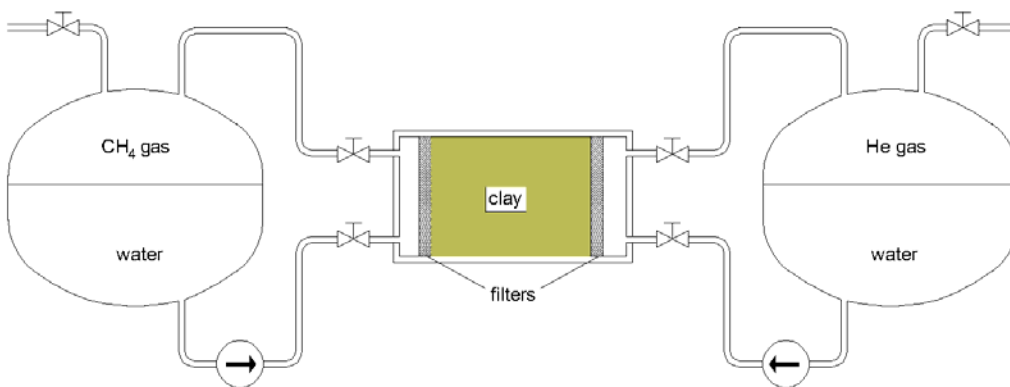
Based on the screening of in situ setups in HADES assessing the possibility to re-use an "old" setup for a gas diffusion experiment, MEGAS setup has been selected for this experiment in HADES URF (Jacops, Verstricht et al. 2018). An existing set up was preferred over a newly drilled set up because the clay in the excavation damaged zone will be more recovered and thus more representative for the Boom Clay Formation.

The MEGAS in situ setup will be adapted and improved by SCK•CEN to allow diffusion of dissolved gas from the source filter, through the clay, into the monitoring filters. When relevant, preliminary and complementary tests will be performed before the gas diffusion experiment starts (e.g. pore pressure measurements and permeability tests).

## 2 Equipment design

The main goal of the in-situ gas diffusion experiment is to compare the diffusion coefficient values measured in the lab on small samples (centimetre scale, samples have a diameter of 80 mm, height of 30 mm) to the values measured in situ (meter scale).

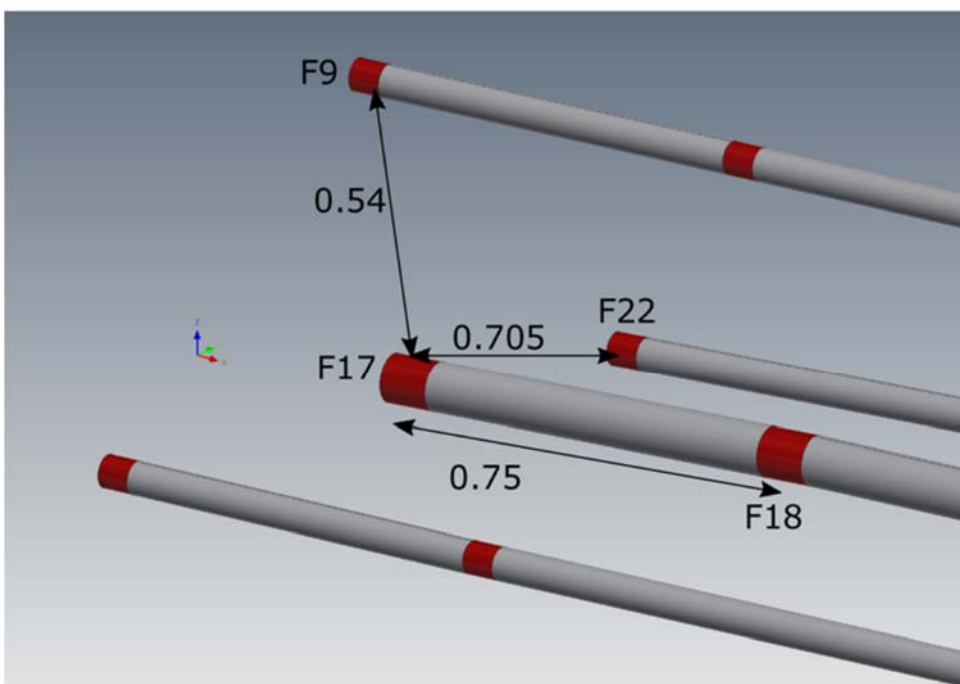
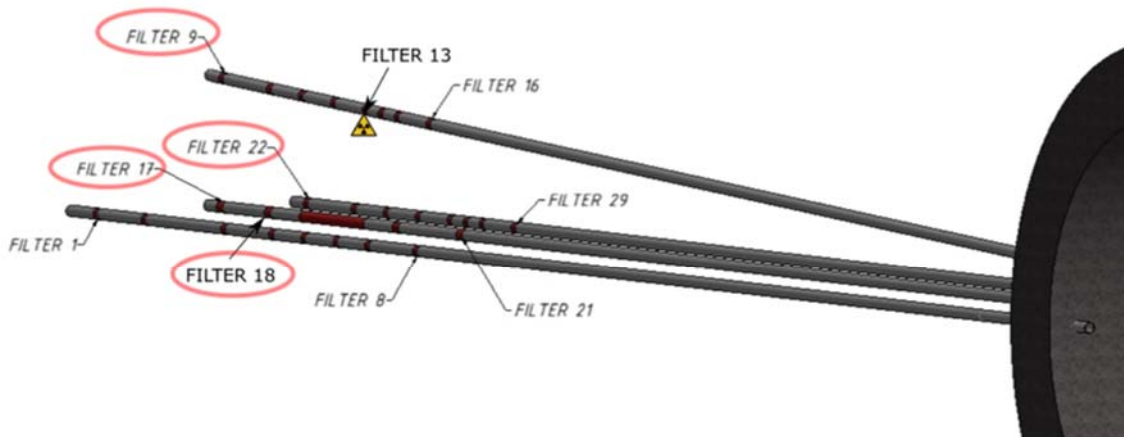
In the lab, a clay sample is loaded in a diffusion cell, and constant volume is obtained. Next, each side of the cell is connected to a 1 litre vessel, filled with half a litre of prepared synthetic Boom Clay water and half a litre of gas at 1 MPa. Gas dissolves in the water, and this dissolved gas will diffuse through the clay sample, to the downstream vessel. Figure 1 presents the lab diffusion set-up to measure diffusion parameters of Helium (He) and Methane (CH<sub>4</sub>) in saturated Boom Clay. In this set-up, He will diffuse into the CH<sub>4</sub> vessel, and CH<sub>4</sub> will diffuse into the He vessel. By monitoring the gas concentration evolutions in the respective downstream vessels, the diffusion coefficients of these two gases can be calculated (Jacops, Volckaert et al. 2013).



**Figure 1: set-up to measure diffusion parameters of gases in saturated porous media**

In order to measure in situ diffusion of dissolved gases, the MEGAS experiment setup has been selected. The MEGAS set-up was installed as a part of the EC MEGAS project (Modelling and Experiments on Gas Migration in Repository Host Rocks) (Volckaert, Ortiz et al. 1994, Volckaert, Ortiz et al. 1996). MEGAS E5 is located in the test drift (ring 17-24 W) and consists of 4 horizontally installed piezometers, which are placed in a 3D configuration (Figure 2). In total, 29 filters are available. The coordinates of the filters have been obtained from a borehole survey. Note that in the past, HTO has been injected in filter 13 (Ortiz, Volckaert et al. 2002). Filters 9, 10, 11, 12, 13, 14, 15 and 16 are known to be contaminated and their concentration of HTO is followed for more than 20 years. The HTO concentration in all other filters is also measured in preparation of this in situ gas diffusion experiment (annex I) and the modelling of the HTO diffusion was optimised and used to predict the HTO levels in the foreseen sampling filters (annex II).

To measure diffusion of dissolved gases, filter 17 will be used as source filter of dissolved gas and filters 9, 18 and 22 will be used for monitoring. Filter 23 could also be used for monitoring, but the tubing is clogged and no water is flowing out. The indicated distances refer to the distance between the surface of two filters. A general overview of the complete injection and monitoring system is shown in **Figure 3**.



**Figure 2: Location of the MEGAS piezometers as described in the initial design. Top: general view, the filters under consideration for a diffusion experiment of dissolved gases are indicated with a red circle. Bottom: detail of the involved filters. The indicated distance between the filters is the distance from the filter surfaces.**

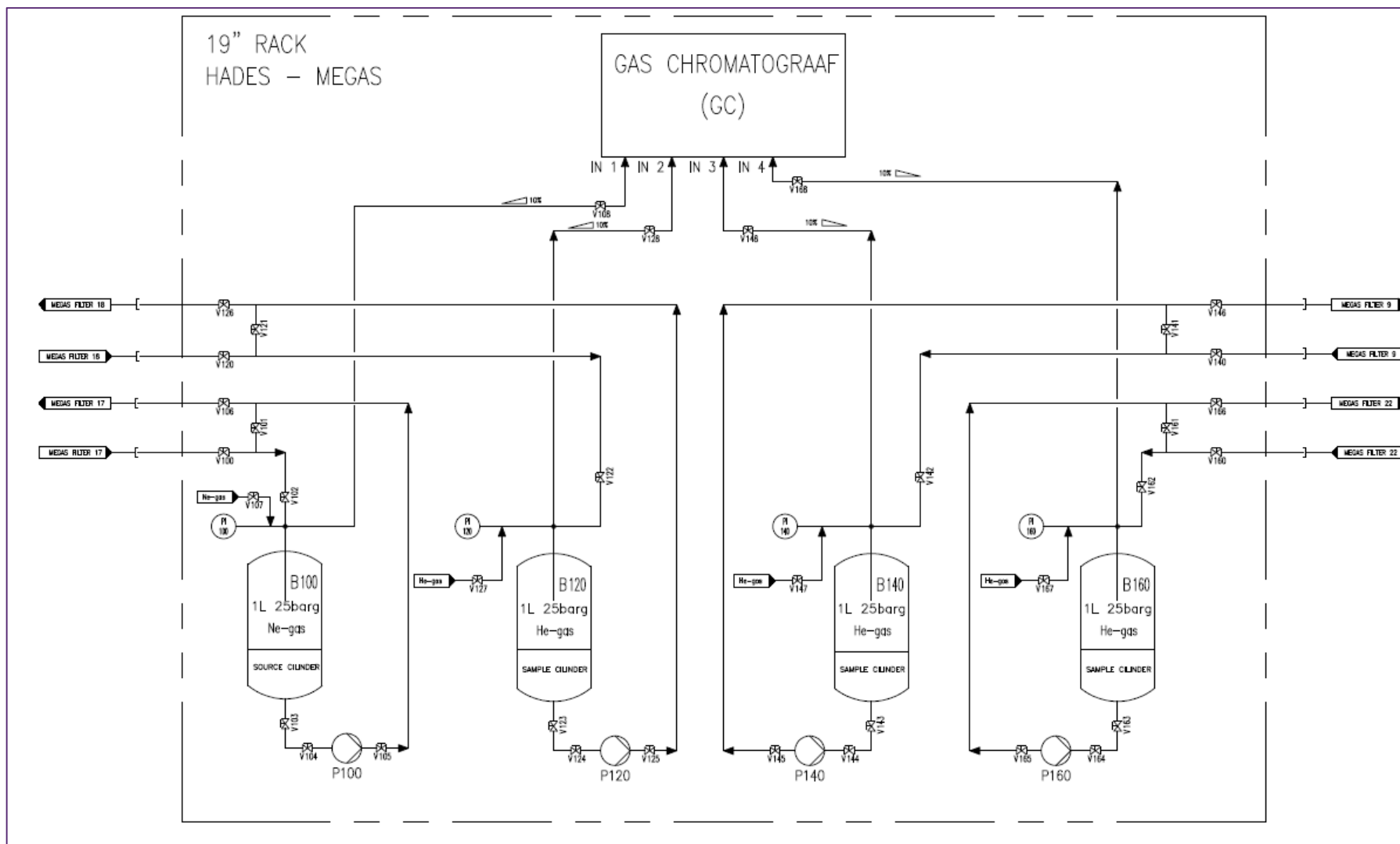


Figure 3: schematic overview of the setup to measure diffusion of dissolved gases. GC means gas chromatograph



Within a geological disposal facility, mainly hydrogen will be generated due to anaerobic corrosion and therefore, hydrogen is the most relevant gas. Given the experimental problems which might occur when performing experiment with hydrogen, this experiment will make use of neon which is considered to be the best proxy for hydrogen.

During the experiment, filter 17 will be connected to the source circuit. One tube of the filter will be connected to the inlet of the source gas system (V100). The inlet is connected via V102 to the upper connector of a vessel of 1 litre (B100) of which half the volume is filled with water and half with Neon (pressurized at the same pressure as the in-situ measured pore water pressure). The lower connector of the vessel (V103) is directly connected to the inlet (V104) of a magnetically coupled gear pump (P100), which will pump around the water and speed up the homogenization of the dissolved gas in the water in the source gas system and the water in the filter, filter chamber and two microtubes connected to the filter. The outlet of the pump (V105) is connected to the outlet of the source circuit (V106), and the latter is connected to the other tube of the filter which allows the water to return to the filter. Also a bypass between the inlet and the outlet of the source gas system is foreseen (V101). This bypass allows the source gas system to be internally flushed, pressurised, etc. without influencing the filter itself and thus the in situ conditions. Via an optional connection (V108), this circuit can be connected to the gas analyser. Filters 9, 18 and 22 will be connected to the monitoring systems. The monitoring systems are identical to the source circuit, but are directly connected to the gas analyser. Gas can be sampled directly from the top part of the vessel as a sample line is going directly to the gas analyser (via V128/148/168).

Gas from all three monitoring systems can be analysed automatically, using a CGC4 (Compact Gas Chromatograph 4) gas analyser from Interscience. The gas analyser is equipped with a multi position valve which allows automated sampling.

### 3 Expected results: scoping calculations

Updated scoping calculations (Figure 4 and Figure 5) show that, as expected, filters 9, 18 and 22 are most interesting to monitor. As diffusion is a slow process, the detection limit of 100 ppm Ne will be passed only after 3 years.

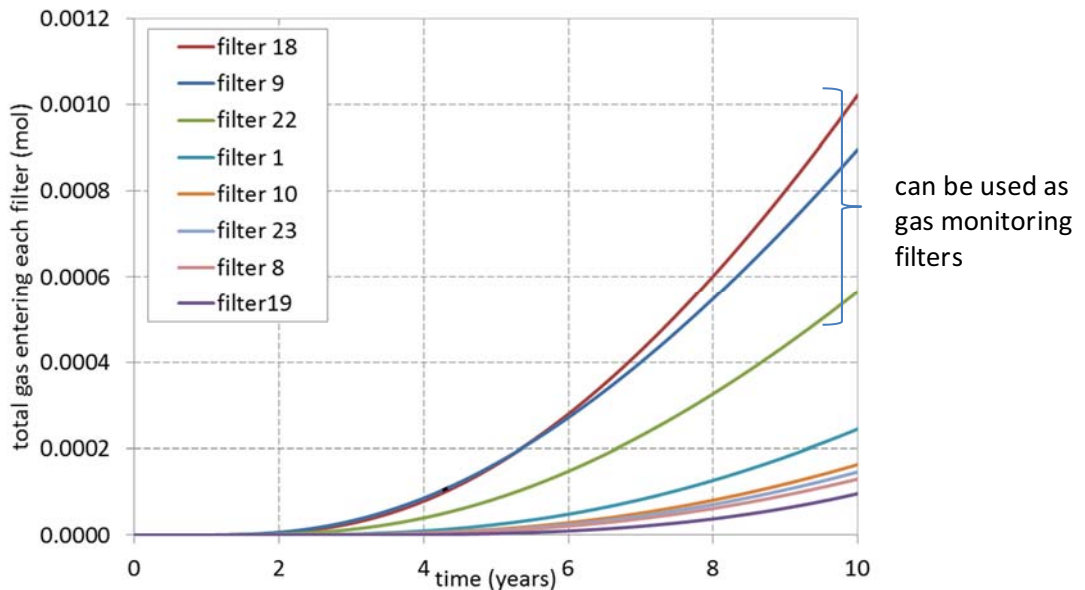


Figure 4: cumulative gas diffused into neighbouring filters around the gas injection filter

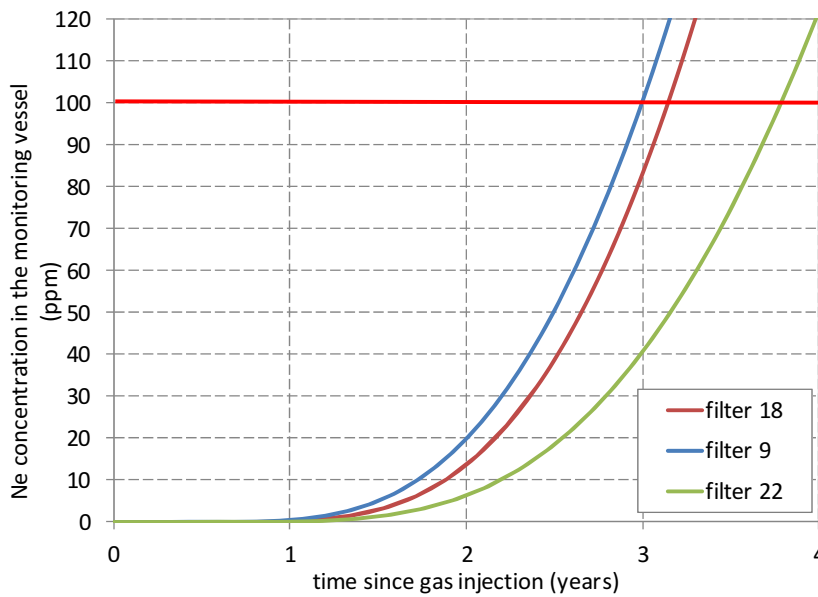


Figure 5 Zoom of Ne concentration in the gas phase (in ppm) to indicate when the detection limit of 100ppm is passed considering a total gas pressure of 15.8 bar in the monitoring vessel

## 4 Equipment

The set-up will be built, using stainless steel tubing of 6 mm outer diameter. Stainless steel is selected because it is inert (not reacting with gas), has a high corrosion resistance and is gas tight. The size is selected in order to minimize the resistance caused by the walls of capillary tubing of e.g. 1/8 inch. All connectors, valves and vessels are ordered from Swagelok, a brand which has proven its high quality and high degree of leak tightness.

The selected pumps are magnetically coupled gear pumps (type Reglo Z) from Ismatec. The pump heads (from Micropump) are equipped with teflon gears (type GA-T23-JFS-B) and allow a flow rate of 0.092 ml/rev, which corresponds – when considering maximum 5500 rev/min - to maximum 506 ml/min ([http://www.ismatec.com/int\\_e/pumps/g\\_gearpumps/reglo\\_z.htm](http://www.ismatec.com/int_e/pumps/g_gearpumps/reglo_z.htm)). As the lifetime of the pump heads is limited to 1 or 2 years, valves have been foreseen at both sides of the pump heads which allows the replacement of the pump heads without disturbing the experiment.

The selected gas analyser is a compact gas chromatograph version 4 (CGC4) from Interscience (<https://gassite.com/compactgc4-0/>) and is operated using Chromeleon software. The gas analyser is equipped with carboxen 1010 and a molsieve 5A column which will allow for the separation of neon, larger gases will be backflushed to avoid blocking of the molsieve column. The detector is a thermal conductivity (TCD) detector. The gas analyser is equipped with a multi position valve, which allows automated gas sampling from the three monitoring systems. The schema of this analyser is shown in Figure 6. Based on our experience with our current CGC4 gas analyser, the calibration for neon is expected to be stable for more than 12 months. Therefore, only verification is recommended prior to each measurement. Gas pressure will be measured by using Unik5000 pressure transducers from General Electric. The selected pressure transducers have a range of 0 to 18 bar (absolute pressure), with a precision of 0.2% full scale (which corresponds to 0.04 bar). The membrane of the sensor is welded with a laser, which avoids gas leakage through the sensor. (<https://www.dimed.eu/products/pressure-measurement/sensors-probes-transducers/unik-5000-pressure-transducer.html>)

\* High pressure compatible valve and vacuum compatible

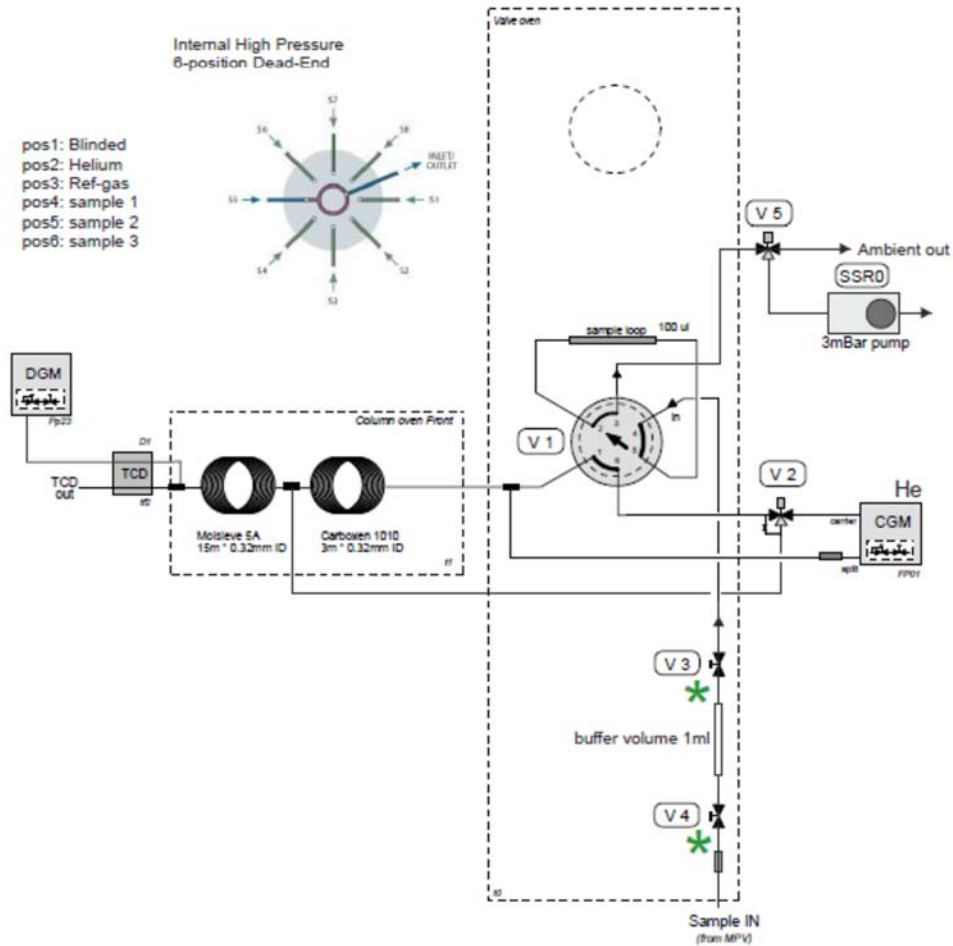


Figure 6: Schema of the selected gas analyser (Compact gas chromatograph 4 – CGC4 from Interscience (The Netherlands))

## 5 Experimental protocol

### 5.1 Pore water pressure measurement

During the preparation phase, the pore water pressure will be monitored in the relevant filters, using the existing pressure transducers. The currently available data recorder is no longer functional, but the pressure transducers still work and the output signal (4 – 20 mA) can be measured. Once the data acquisition system has been renewed (continuous monitoring is possible).

### 5.2 Hydraulic conductivity measurement

The Boom Clay hydraulic conductivity around the different filters will then be measured during the pore water sampling of all filters which is carried out in the framework of the monitoring of the MEGAS HTO injection experiment. From a single filters, several water samples will be taken and the analyses of the water outflow over a certain period allows to assess the hydraulic conductivity.

### 5.3 Leak test of the set up

Prior to the installation in HADES URL, the experimental setup will be tested extensively in the lab in several steps. Immediately after construction, a leak test with helium will be performed by following the pressure over time.

### 5.4 Validation of the measurement protocol for neon on the gas analyser

In a first step, the measurement protocol on the gas analyser will be validated. Measurements obtained with the new apparatus will be compared to those of an existing gas analyser which is used for lab-scale diffusion experiments.

### 5.5 Accuracy of the gas measurements and impact of sampling on the pressure of the set up

In a second step, the correctness of the gas concentration and the gas consumption due to sampling will be assessed. The vessels of the setup will be filled with synthetic Boom Clay water and will be pressurised at the exact in-situ pore water pressure of the corresponding filter. For the source gas system, pure neon will be used, for the monitoring system mixtures of Ne in He will be used, varying between 100 and 10000 ppm. Water circulation will occur over the bypass, and the pump rate and frequency are set to replace the volume of 500 ml at least once per day. On a regular base, gas samples will be taken and the measured concentration will be compared to the known concentration in the vessel and of course both values should be equal. Also the pressure drop due to sampling will be monitored.

### 5.6 Testing the set up on lab scale

In a next step, the system will be tested in a lab scale diffusion experiment. The main purpose is to assess whether the gas analyser correctly captures the concentration change in time, and thus whether the gas that goes to the multi position valve corresponds to the gas in the vessel. A clay sample is loaded in a constant volume diffusion cell and connected to the injection system and one monitoring system. Both vessels are filled with synthetic pore water and gas (same gas pressure to avoid breakthrough): the injection vessels is filled with pure Ne, the monitoring vessel is filled with pure He. Once the valves between the sample and the injection and monitoring system are opened, diffusion starts. Gas sampling will take place regularly (at least once per week), and the concentration increase in time is followed. The obtained concentration increase and time, and hence diffusion coefficient can be compared to those of previous lab scale experiments. Afterwards, the system is emptied and cleaned.

### 5.7 Transport to HADES URL and leak test

In a next step, the system is moved to HADES URL and is connected, but the valves at the inlet of the cabinet are closed. After transport, a leak test is performed again by measuring the pressure evolution over time.

### 5.8 Preliminary in situ test of the set up

Once the system is considered to be leak tight, the set up will be tested under in situ conditions. All vessels will be filled again with artificial pore water and are pressurised to the exact in-situ pore water pressure of the corresponding filter. The gas, used to pressurise both vessels should not interfere with the later diffusion experiment (with neon and eventually ethane), therefore He or N<sub>2</sub> are proposed. Now the influence of the pumping rate on the pore water pressure will be

investigated because pumping should not cause major pressure changes ( $> 0.5$  bar). The pump rate and frequency are set at the values which were determined in the lab in previous steps and the pressure is monitored during several days. Should the impact be too large (more than 0.5 bar pressure fluctuation), the frequency or flow rate have to be adapted. At the same time, also the impact of gas sampling and analysis on the full system will be assessed because the pressure drop due to sampling should be as small as possible to avoid disturbance of the in situ conditions. Gas samples are taken regularly (e.g. daily) and their impact is assessed. Too much pressure loss might force us to decrease the sampling frequency.

## 5.9 Starting the diffusion experiment

After finishing all preliminary tests, the source vessel is pressurised with pure neon, the monitoring vessels are pressurised with pure helium – all at the corresponding pore water pressure. Afterwards, the valves between the tubing and the experimental setup are opened and the experiment can start.

Once the experiment is running, the pressure is monitored carefully, only minor deviation from the in-situ undisturbed pore pressure are allowed ( $< 0.5$  bar). During the experiment, sampling will occur every 4 weeks. Based on the outcome of the gas analyses and the caused pressure drop, the sampling frequency can be adapted during the course of the experiment.

## 6 Conclusion

The main objective of the new diffusion experiment with dissolved gases is to determine parameters associated to gas diffusion in Boom Clay at larger scale. In addition to laboratory experimental programme, an in situ experiment aims to confirm and/or improve the current knowledge on diffusion of dissolved gas at a larger scale.

Based on the screening of in situ setups in HADES assessing the possibility to re-use an "old" setup for a gas diffusion experiment, MEGAS setup has been chosen for this experiment in HADES URF.

The MEGAS in situ setup will be adapted and improved to allow in-diffusion of dissolved gas from one filter into the clay. The experiment sept-up will consist of one injection/source vessel and three monitoring vessels with associated injection/monitoring lines connected to the filters. The monitoring vessels are connected to a gas analyser which performs on line analysis of the neon concentration. By monitoring the concentration change of Ne in time in the "injection" and the "observation" vessels, the diffusion parameters can be determined.

This report describes both the technical aspects of the experiment, and the protocol to be followed.

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## Annex I: screening of HTO activity in all filters of the MEGAS set-up

The MEGAS experiment has been used for a large-scale long-term migration experiment with HTO starting in 1998. The HTO diffusion has been monitored since then but only in 1 direction, more particularly in the neighbouring filters located on the same piezometer.

In order to assess the radiological impact of the HTO diffusion test on all filters, but with special emphasis for those considered for the gas diffusion experiment, all filters were sampled for HTO.

A first quick screening from filters 9/17/18/22 was done in November 2018 as part of the screening study of in-situ set-ups that may be used for a new gas diffusion test (Jacops et al. (2018) Screening of experiment set-ups in HADES for evaluating the possibilities for a new gas in situ experiment, SCK•CEN-ER-0512). Note that during the sampling, the dead volume of the tubing was not removed. This first screening showed possible, but low contamination.

In a second screening, all filters in the set-up are sampled in a more rigorous way and the data are used to compare to an updated modelling of the HTO experiment (see annex II). This updated modelling will be the base for the modelling of the gas test. In order to have representative samples, the dead volume of the tubing should be rinsed/changed by water that is present within the filter chamber and representative for the water in contact with the filter (which was not done during the first screening).

In the table below, the dead volumes are tabulated in order to estimate sampling time.

**Table 1: overview of the calculated dead volume for each filter and the estimated sampling time, required to remove the dead volume**

Filter No.	Filter length	Filter external diameter	Filter volume	Chamber volume	micro tube volume	Total dead volume	flow rate estimate d (ml/day)	time for 1 rinse (day)
	cm	cm	ml	ml	ml	ml		
A-17	9	8.9	35.63	22.05	53.56	164.81	28	1.9
A-18	9	8.9	35.63	22.05	51.21	160.10	28	1.8
A-19	100	8.9	395.84	245.04	48.07	737.02	120	0.4
A-20	9	8.9	35.63	22.05	44.92	147.53	28	1.6
A-21	9	8.9	35.63	22.05	41.78	141.25	28	1.5
B-22	6	5.56	8.36	9.24	53.41	124.41	18	3.0
B-23	6	5.56	8.36	9.24	51.05	119.70	18	2.8
B-24	6	5.56	8.36	9.24	49.48	116.56	18	2.7
B-25	6	5.56	8.36	9.24	47.91	113.42	18	2.7
B-26	6	5.56	8.36	9.24	46.34	110.27	18	2.6
B-27	6	5.56	8.36	9.24	45.55	108.70	18	2.5
B-28	6	5.56	8.36	9.24	44.77	107.13	18	2.5
B-29	6	5.56	8.36	9.24	43.20	103.99	18	2.4
C-9	6	5.56	8.36	9.24	54.90	127.40	18	3.0
C-10	6	5.56	8.36	9.24	52.54	122.68	18	2.9
C-11	6	5.56	8.36	9.24	50.97	119.54	18	2.8
C-12	6	5.56	8.36	9.24	49.40	116.40	18	2.7
C-13	6	5.56	8.36	9.24	47.83	113.26	18	2.7
C-14	6	5.56	8.36	9.24	47.05	111.69	18	2.6
C-15	6	5.56	8.36	9.24	46.26	110.12	18	2.6
C-16	6	5.56	8.36	9.24	44.69	106.97	18	2.5
D-1	6	5.56	8.36	9.24	57.49	132.58	18	3.2
D-8	6	5.56	8.36	9.24	55.13	127.87	18	3.1
D-2	6	5.56	8.36	9.24	51.21	120.01	18	2.8
D-3	6	5.56	8.36	9.24	48.85	115.30	18	2.7
D-4	6	5.56	8.36	9.24	47.28	112.16	18	2.6
D-5	6	5.56	8.36	9.24	45.71	109.02	18	2.5
D-6	6	5.56	8.36	9.24	44.14	105.88	18	2.5
D-7	6	5.56	8.36	9.24	41.78	101.16	18	2.3

Septum bottles of 100 ml were used for collecting samples. Sampling was done in consecutive mode for about 2-3 days/sampling. A first sample was the "rinse" sample, the second one was considered the representative sample.

The sampling campaign was split in 2 parts. In a first part, the filters of piezometer C with filters 9-16 were sampled within the sampling campaign of the in-situ diffusion experiments under the NIRAS/ONDRAF RP Migration. These samples were provided to the laboratory of low activity measurements (LRM) at SCK•CEN for ISO17025 certified HTO measurement.

The other filters were sampled in similar way but the activity was determined in our own laboratory (not certified).

From the representative samples, 1 ml subsamples were taken and transferred into a 20 ml LSC vial and mixed with 18 ml of scintillation cocktail. Samples were analysed in a Packard TRI-CARB 2100TR liquid scintillation counter. The samples were 3 times measured in the 0-18.6 keV range for 15 minutes. The average of 3 measurements was corrected for the background and a counting efficiency of 65% was used to convert counts to Bq.

The results can be found in Table 2.

**Table 2: overview of the measured activity in each of the MEGAS filters**

Filter No.	Activity [Bq/ml]
BOREHOLE A	
A-17	0.91
A-18	6.96
A-19	21.59
A-20	27.20
A-21	5.58
BOREHOLE B	
B-22	0.43
B-23	no water
B-24	3.90
B-25	7.83
B-26	8.94
B-27	10.11
B-28	5.55
B-29	6.18
BOREHOLE C	
C-9	1.42
C-10	16.50
C-11	54.00
C-12	106.00
C-13 (injection filter)	192.00
C-14	165.00
C-15	no water
C-16	55.40
BOREHOLE D	

D-1	0.40
D-8	3.94
D-2	10.29
D-3	7.80
D-4	3.86
D-5	1.40
D-6	1.20
D-7	1.15
<i>Detection limit</i>	<i>0.4</i>

As can be observed, HTO can be detected in all filters of the MEGAS set-up. The filters foreseen for the gas in-situ diffusion experiment do exhibit HTO activity but are at low level because of the distances to the HTO injection point. However, this does not pose any problem (safety or otherwise) to the planned experiment as we will do on-line gas measurements in closed circulation loops. But it should be kept in mind that manipulations to equipment in contact with these filters will lead to contact with low HTO contaminated water.

## Annex II: Updated modelling of the HTO migration experiment

A total amount of  $7.61 \times 10^8$  Bq of HTO was injected into the filter 13 of MEGAS piezometers in April of 1998 (Fig. 2). Concentrations at filters along the piezometer C have been continuously measured until 2015 Nov. In 2019, concentrations were measured at all filters in MEGAS piezometer network, and results are presented in Table 2 of Annex I.

Before the HTO migration experiment, an *in situ* gas breakthrough test was carried out between filters 13 and 14. Gas breakthrough was triggered with an excess gas pressure of 0.3 MPa. After the breakthrough, a steady-state gas flow-rate of  $2.1 \times 10^{-2}$  ml/s STP was maintained for one year under a constant gas pressure gradient. Forty days after the end of the gas test, HTO migration experiment started (Ortiz et al., 2002).

The HTO migration test was simulated using diffusion of a point source in infinite domain and results were published in Ortiz et al. (2002), named as previous modelling in the following text. The objectives of updating the modelling of HTO migration experiment are to:

- improve the simulation results by considering filter dead volumes and more precise boundary conditions.
- screen the possible HTO contaminated filters so as to assist the design of the new *in situ* gas diffusion experiment.
- help interpreting the future gas diffusion data by estimating filters coordinate deviations (if any).

The excavation of the HADES URL galleries in 1982 is taken as the start of the drainage period. In 1998, after 16 years of drainage and when the HTO migration experiment started, a large drainage field, in the order of tens of meters, was formed around the MEGAS piezometer network. Initially, diffusion of HTO happened only within a limited local region around the injection filter 13. For such a flow-diffusion coupling problem, a sequential workflow is adopted to enhance the numerical calculation efficiency, with a larger domain for the hydraulic modelling (a block of 30 m X 30 m X 20 m) and a local domain for the diffusion modelling (a block of 10 m x 10 m x 10 m).

Detailed information about hydraulic modelling can be found in Jacops et al. (2018). Figure 7 presents the evolution of the water velocity at filter 13. Results indicate that the water flow reaches steady state after 16 years of drainage with an order of  $10^{-4}$  (m/year). The steady-state water velocity field is extracted from the hydraulic modelling and serves as input in the subsequent diffusion modelling. Actually, such a small velocity has negligible effects on the subsequent diffusion modelling.

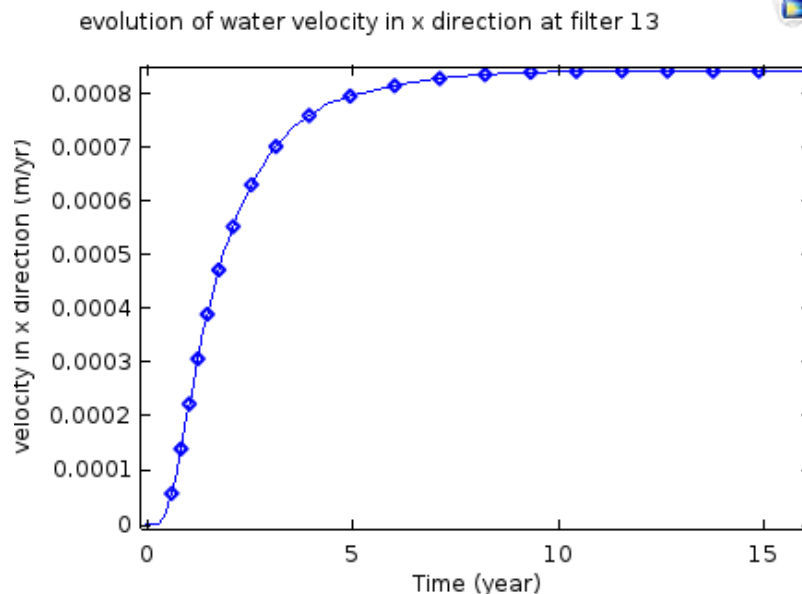
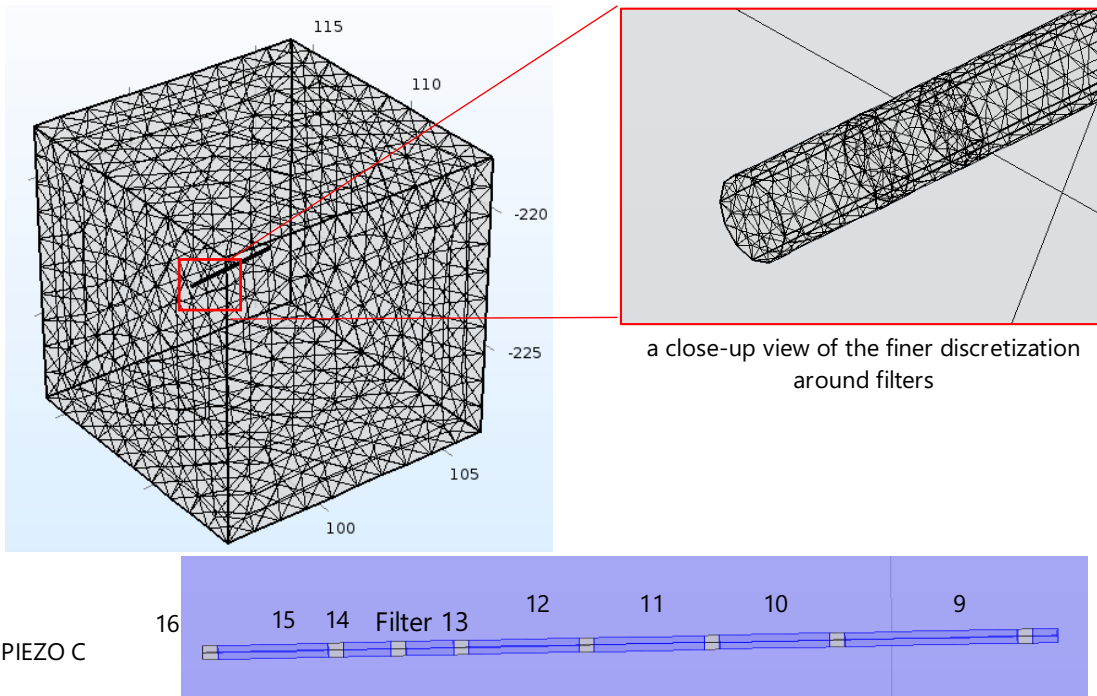


Figure 7: evolution of water velocity at filter 13

HTO migration test is modelled using 3D COMSOL advection-diffusion module with the Boom Clay (BC) block centered around the injection filter 13 (Figure 8). The domain is discretized into a total of 113978 quadratic tetrahedral elements. In order to mimic the field measurements, dilution of the tracer in the filter dead volume (casing + tube) must be considered. In the modelling, filters are treated as porous cylinders with an equivalent porosity calculated from the dead volume. The external surface of the rest part of the piezometers is considered impermeable. Detailed information of MEGAS piezometers and filters are listed in Table Annex II-1 for reference.



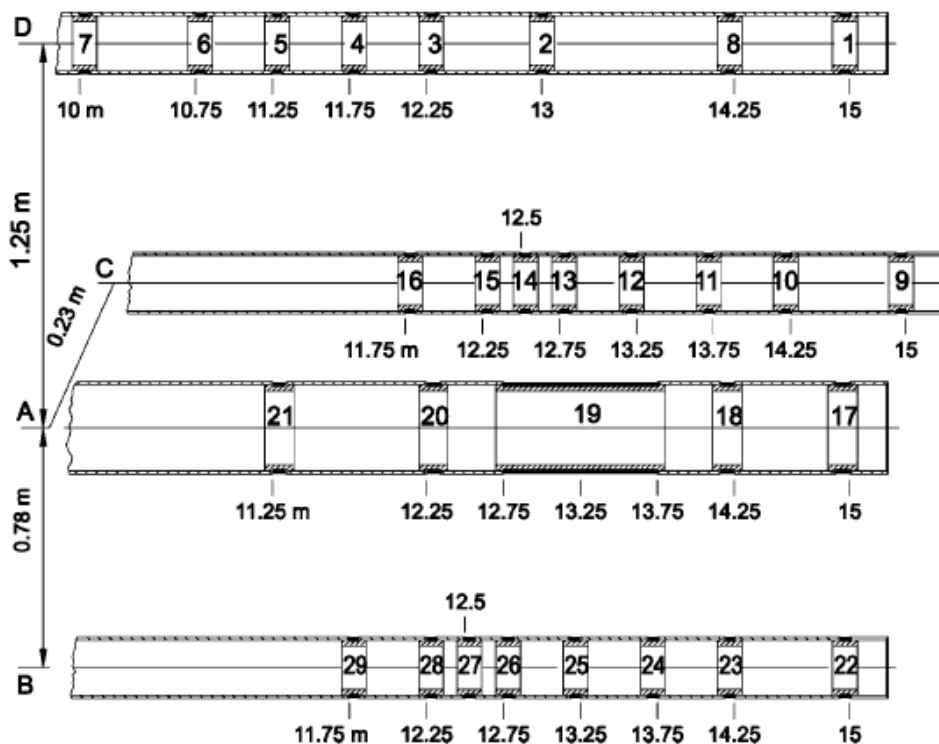
**Figure 8: geometry for diffusion modelling (above) ;geometry of piezometer C where filters are treated as porous cylinder and the piezometer tube between filters is impermeable (below)**

Parameters used in the advection-diffusion modelling include:

- Porosity of the Boom Clay: 0.38;
- Anisotropic apparent diffusion coefficients of HTO in BC are taken from Aertsens et al., (2005), Bruggeman et al., (2009), Weetjens et al., (2011):  $D_H=4.1 \times 10^{-10}$  and  $D_V=2.05 \times 10^{-10}$  ( $m^2/s$ ), with an anisotropic ratio of 2. Apparent diffusion coefficient of HTO within filters is set  $2.2 \times 10^{-9}$  ( $m^2/s$ );
- The half-life of  $H^3$  is 12.33 years.

**Table Annex II-1: information of the MEGAS piezometers and filters**

MEGAS piezometer layout



**PIEZO A:**

- The length of filter 17, 18, 20, 21 is 9 cm and 1 m for filter 19;
- Diameters of filter external surface, filter inner surface and inner surface of casing are 9/79/77 mm respectively.
- Porosity of filters=0.3
- The dead volume of filters is calculated as the sum of
  - ✓ Filter and Casing volume:  $(\pi/4 \times (89^2-79^2) \times 0.3 + \pi/4 \times (79^2-77^2)) \times \text{length of filter} = 57680 \text{ mm}^3$  for filter 17, 18, 20, 21 and  $640884 \text{ mm}^3$  for filter 19;
  - ✓ tube volume: each filter contains two tubes with a tube diameter of 2 mm. The average length of one tube is: depth of filter+casing entrance to lining extrados+tube outside the casing= depth of filter+0.7 m+1.35 m for PIEZO A.

depth	Filter number	x	y	z	Tube length (m)	dead volume (m <sup>3</sup> )	Equivalent porosity (%)
11.25	21	102.599	111.650	-223.051	14.65	1.41E-04	25.23
12.25	20	101.605	111.539	-223.039	15.65	1.48E-04	26.35
12.75	19 (front)	101.109	111.477	-223.046	16.65	7.37E-04	11.85
13.25	19 (middle)	100.612	111.420	-223.051			
13.75	19 (bottom)	100.116	111.360	-223.057			
14.25	18	99.618	111.308	-223.058	17.65	1.60E-04	28.59
15.00	17	98.876	111.198	-223.067	18.4	1.65E-04	29.44

**PIEZO B:**

- Length of filter=6 cm ;
- Diameters of filter: 55.6/50/ 48mm , therefore filter and casing volume is:  
 $(\pi/4 \times (55.6^2-50^2) \times 0.3 + \pi/4 \times (50^2-48^2)) \times 60 = 17596 \text{ mm}^3$ ;
- Each filter contains two tubes and tube diameter=2 mm. The total length of one tube is depth of filter+casing entrance to lining extrados+tube outside the casing= depth of filter+0.7 m+1.3 m

depth	filter number	x	y	z	Tube length (m)	dead volume (m <sup>3</sup> )	Equivalent porosity (%)
11.75	29	101.961	112.346	-223.201	15.05	1.04E-04	71.38
12.25	28	101.465	112.286	-223.211	15.55	1.07E-04	73.54
12.5	27	101.217	112.255	-223.215	15.8	1.09E-04	74.62
12.75	26	100.969	112.223	-223.218	16.05	1.10E-04	75.70
13.25	25	100.473	112.163	-223.229	16.55	1.13E-04	77.85
13.75	24	99.977	112.101	-223.236	17.05	1.17E-04	80.01
14.25	23	99.481	112.038	-223.239	17.55	1.20E-04	82.17
15	22	98.737	111.944	-223.256	18.3	1.24E-04	85.40

**PIEZO C** (same as PIEZO B) except the tube length

The total length of one tube is depth of filter+casing entrance to lining extrados+tube outside the casing= depth of filter+0.7 m+1.775 m

depth	filter number	x	y	z	Tube length (m)	dead volume (m <sup>3</sup> )	Equivalent porosity (%)
11.750	16	102.117	111.495	-222.496	16	1.07E-04	73.43
12.250	15	101.622	111.428	-222.491	16.5	1.10E-04	75.59
12.500	14	101.374	111.394	-222.489	16.75	1.12E-04	76.67
12.750	13	101.126	111.361	-222.487	17	1.13E-04	77.75

13.250	12	100.630	111.294	-222.483	17.5	1.16E-04	79.90
13.750	11	100.134	111.227	-222.479	18	1.20E-04	82.06
14.250	10	99.639	111.160	-222.474	18.5	1.23E-04	84.22
15.00	9	98.895	111.059	-222.468	19.25	1.27E-04	87.45

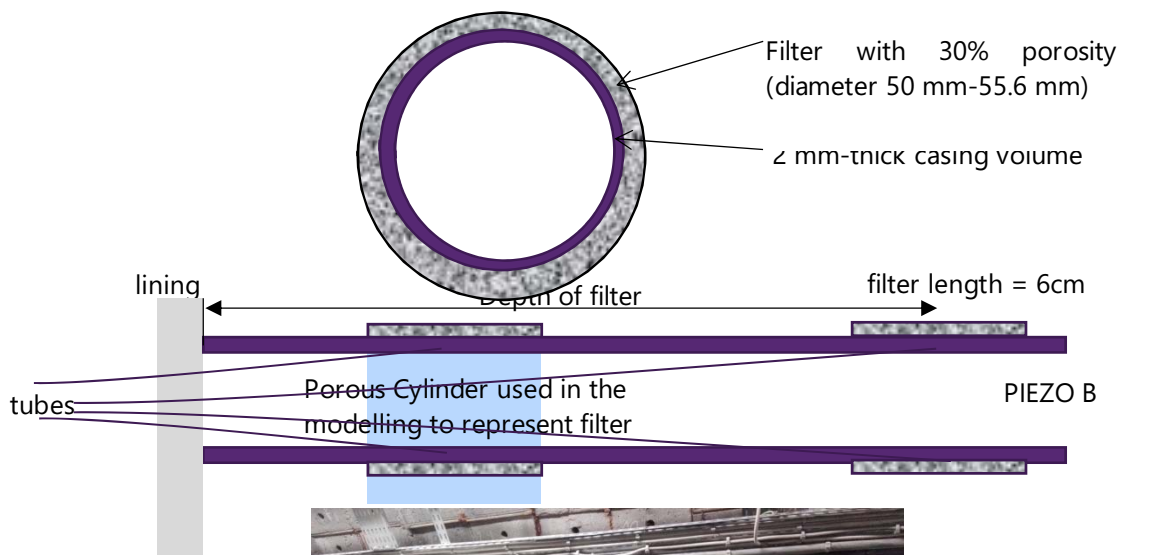
**PIEZO D** (same as PIEZO B) except the tube length

The total length of one tube is depth of filter+casing entrance to lining extrados+tube outside the casing= depth of filter+0.7 m+0.21 m+2.6 m

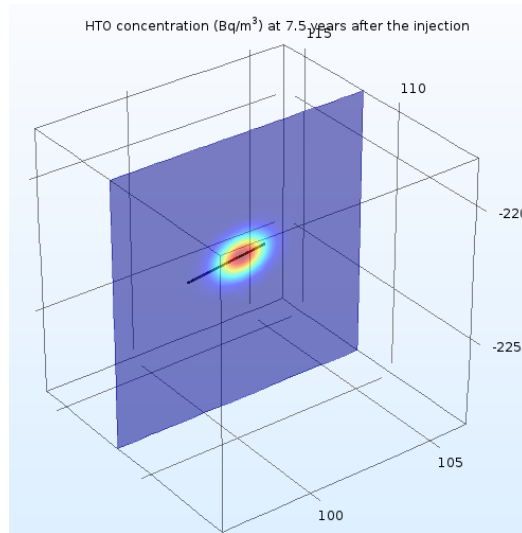
depth	filter number	x	y	z	Tube length (m)	dead volume (m <sup>3</sup> )	Equivalent porosity (%)
9.790	7	103.742	110.762	-222.944	15.69	1.01E-04	69.44
10.540	6	102.995	110.688	-222.947	16.44	1.06E-04	72.68
11.040	5	102.498	110.642	-222.949	16.94	1.09E-04	74.83
11.540	4	102.000	110.591	-222.954	17.44	1.12E-04	76.99
12.040	3	101.503	110.535	-222.951	17.94	1.15E-04	79.15
12.790	2	100.756	110.466	-222.943	18.69	1.20E-04	82.38
14.040	8	99.513	110.340	-222.952	19.94	1.28E-04	87.77
14.790	1	98.793	110.265	-222.946	20.69	1.33E-04	91.01

Note:

- Filter coordinate (X,Y,Z) is supposed referring to the filter section marked by the arrows in the MEGAS piezometer layout;
- The depth refers to the distance of the filter to the extrados of the lining;
- The following gives an illustration of a filter in PIEZO B, where dead volume includes two tube volumes, filter porous volume and casing volume. The equivalent porosity for each filter is the fraction of the dead volume in the cylinder (diameter of 55.6 mm and a length of 6 cm) used in the modelling to represent the filter.

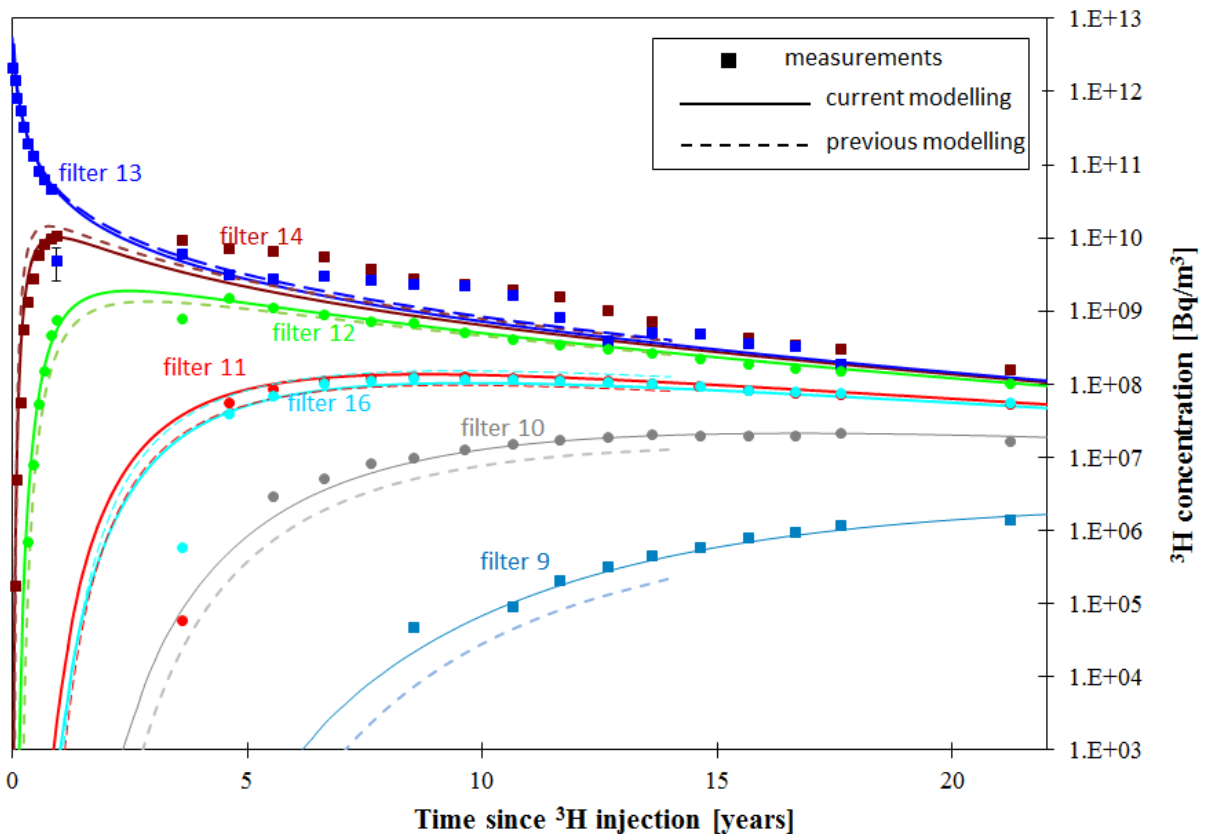


17.5 years after the HTO injection, concentration of HTO around filter 13 is presented in Figure 9:



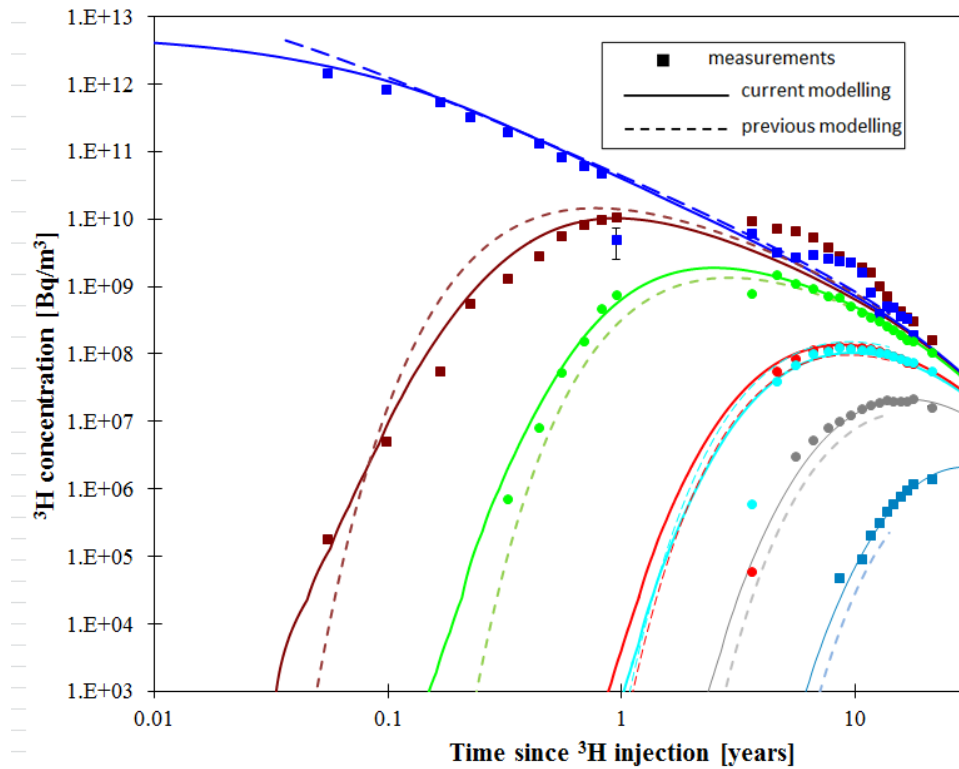
**Figure 9: concentration contour along one XZ section at piezometer C after 17.5 years since the HTO injection**

Comparisons between modelling results and field measurements at filters along piezometer C, together with previous modelling results are presented in Figure 10 and Figure 11. Results show that the current modelling results improve the simulation significantly. Note that the “jump” which is observed in e.g. the brown curve in Figure 11 is caused by an increasing rate of concentration, plotted on a log-Y scale.



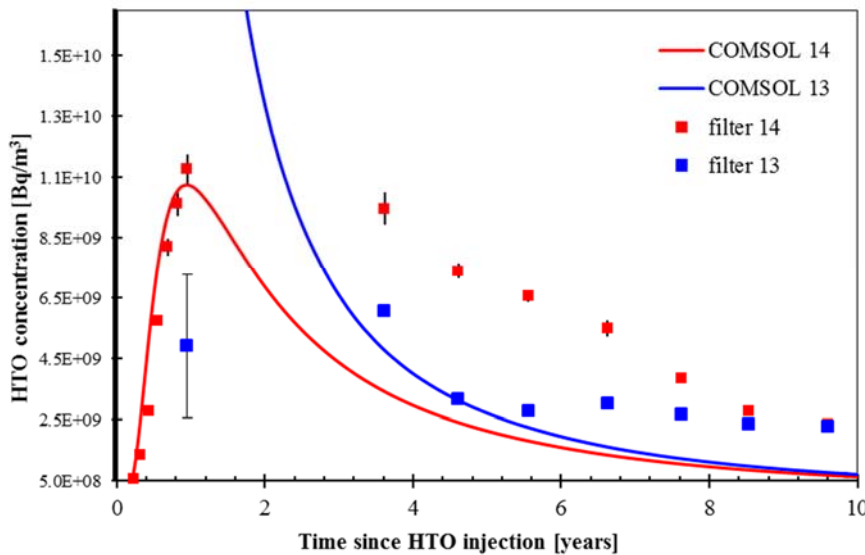
**Figure 10: comparisons between modelling results and field measurements at filters along piezometer C ( $D_H=4.1 \times 10^{-10} \text{ m}^2/\text{s}$  and  $D_V=2.05 \times 10^{-10} \text{ m}^2/\text{s}$ ) using semi-log**





**Figure 11: comparisons between modelling results and field measurements at filters along piezometer C ( $D_H=4.1 \times 10^{-10} \text{ m}^2/\text{s}$  and  $D_V=2.05 \times 10^{-10} \text{ m}^2/\text{s}$ ) using bi-log scale**

Although the comparison is agreeable for most of the filters, it is apparent that the measurements at injection filters 13 and 14 are higher than the modelling results during 3-14 years. This peculiar seems to be related to the pathway between filters 13 and 14 which was formed during the foregoing gas breakthrough test. Figure 12 presents a close-up view for filters 13 and 14.



**Figure 12: zoom of the HTO concentration at filters 13 and 14**

After the first year, monthly sampling was stopped and when the sampling was resumed after 2.5 years, a dramatic increase in filter 14 with a HTO level even higher than filter 13 was observed until the 9<sup>th</sup> year. The higher concentration at filter 14 than filter 13 can only be explained by some advective transport mechanism. During the sampling, it was observed that there are quite high initial flow rates when opening the filter 14, which could point to excess gas pressure induced by occluded gas bubbles. This may be of interest for further analysis/interpretation of the old gas breakthrough experiment.

It is regrettable that the long-term HTO concentrations were measured only along the piezometer C, which makes the anisotropic diffusion coefficients not being fully validated, nor the spatial coordinates of filters. Concentrations at all filters were measured in 2019 (data is listed in Table 2 in Annex I). Figure 13 till Figure 16 presents the comparison between modelling results and the measurements made in 2019 for all filters. Concentration at filters on piezometer C is successfully captured, however, discrepancies clearly exist at filters along the rest three piezometers.

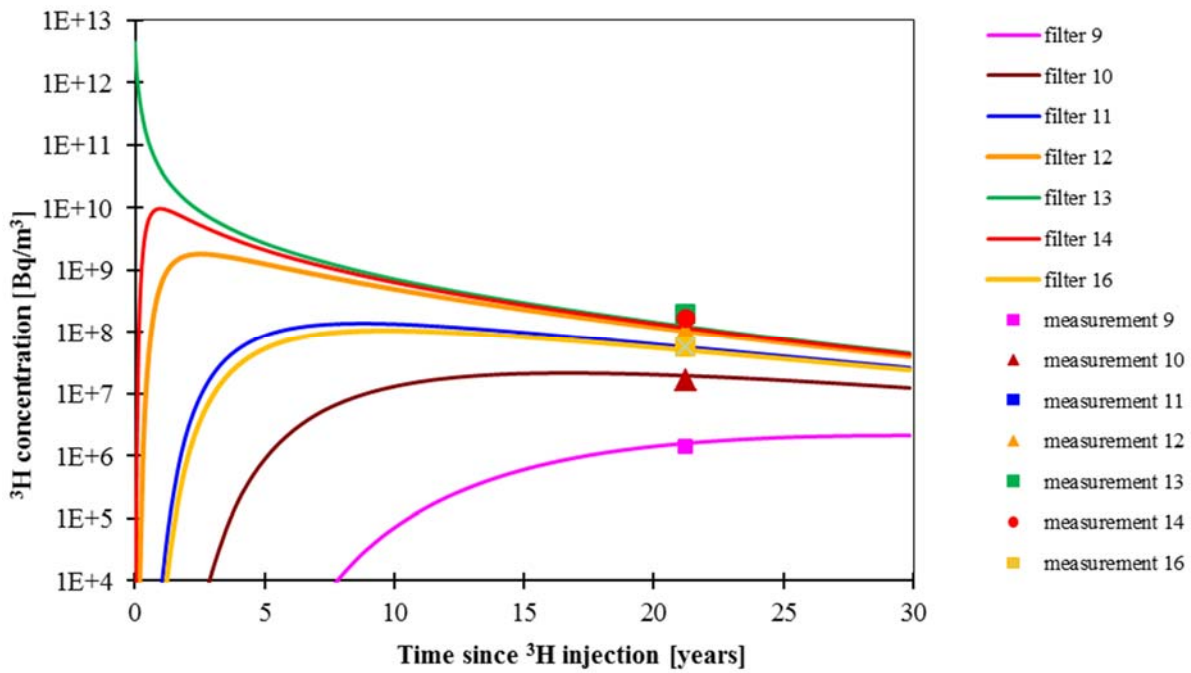


Figure 13: comparison between modelled HTO concentration at filters along piezometer C and the new measurements in 2019

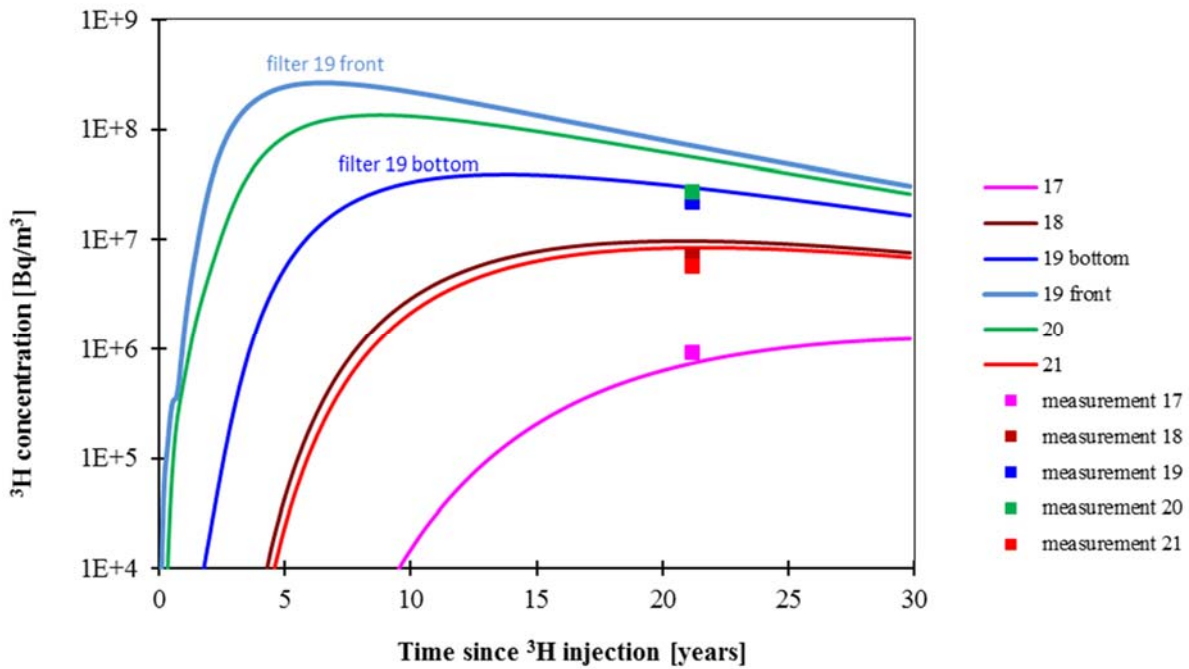


Figure 14: comparison between modelled HTO concentration at filters along piezometer A and the new measurements in 2019

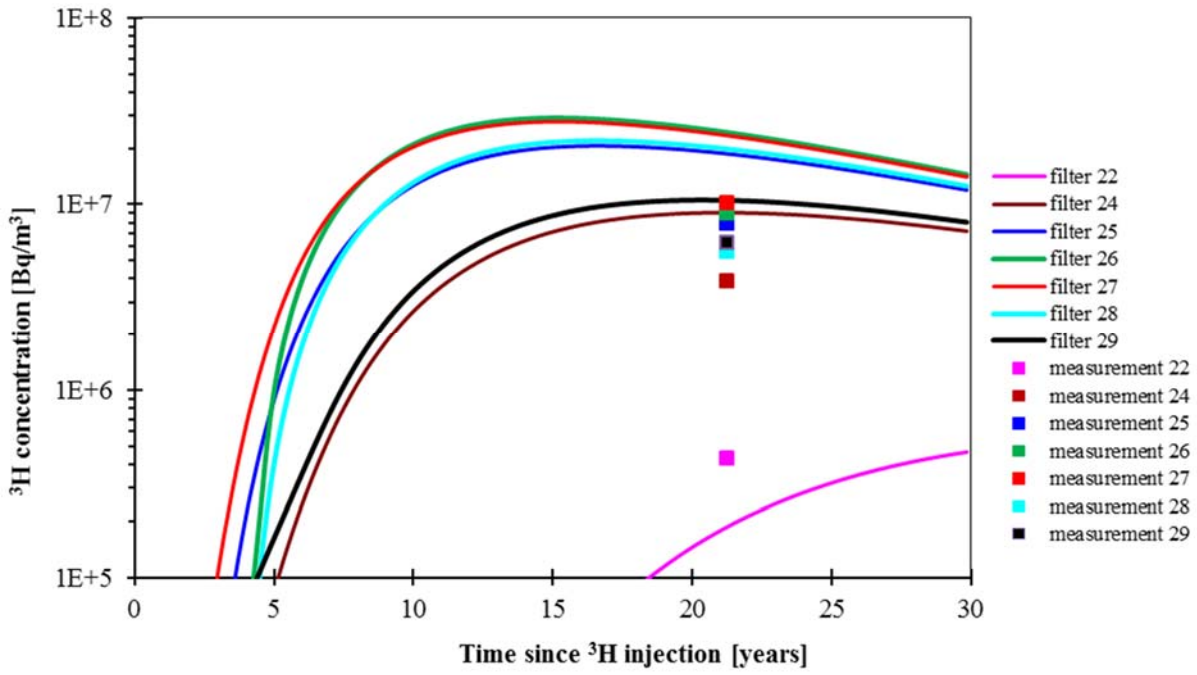


Figure 15: comparison between modelled HTO concentration at filters along piezometer B and the new measurements in 2019

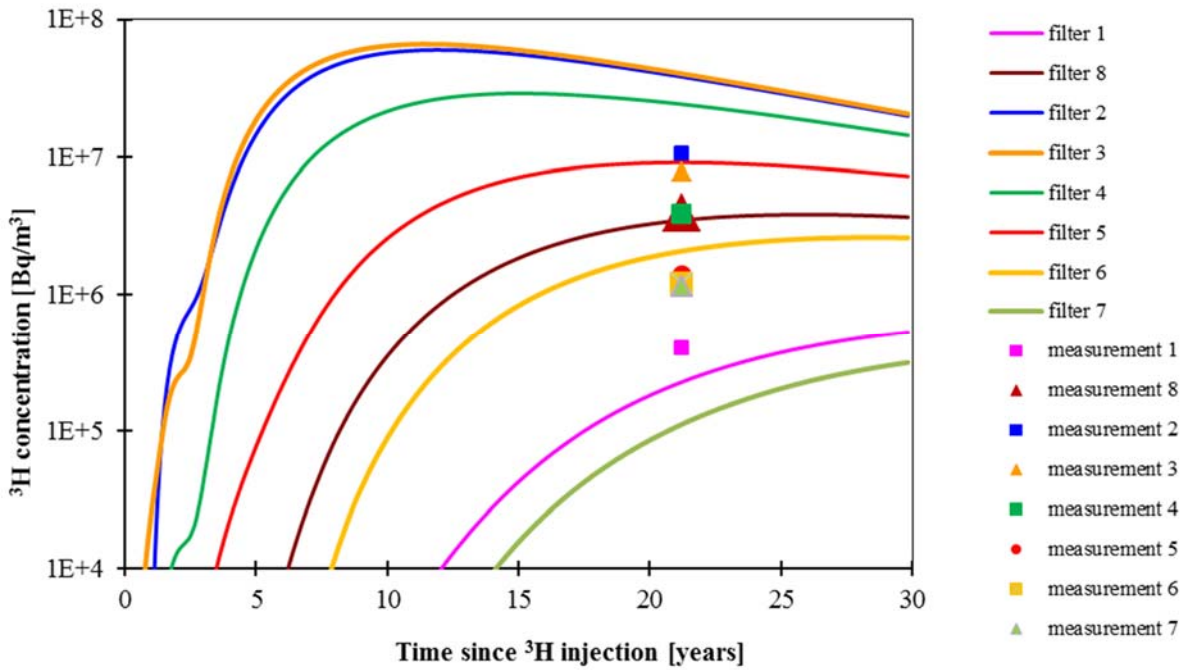
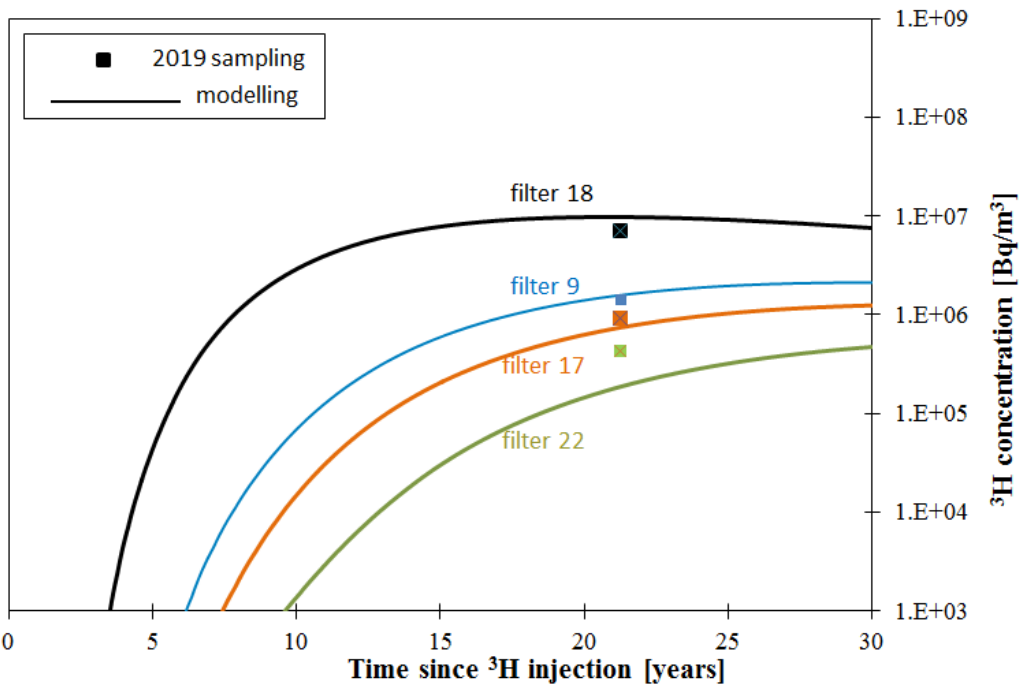


Figure 16: comparison between modelled HTO concentration at filters along piezometer D and the new measurements in 2019

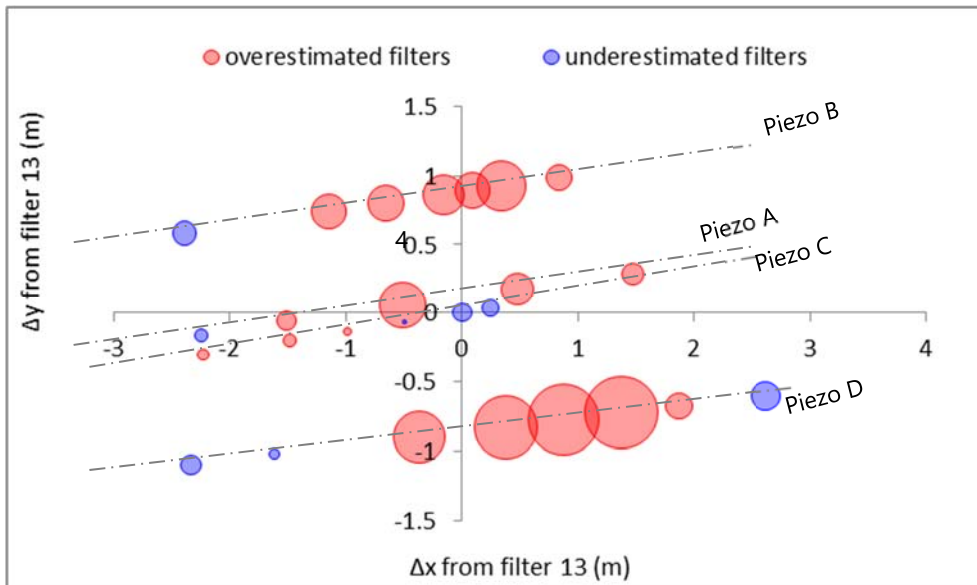
Figure 17 compares at those filters to be used in the new *in situ* gas test.



**Figure 17: concentration of HTO at filters to be used in the new *in situ* gas diffusion test**

Figure 18 illustrates the relative errors at all filters, defined as  $\frac{C_{modeled} - C_{measured}}{C_{measured}}$ , based on field measurements of 2019. The size of the bubble represents error amount. The figure indicates that most filters are overestimated, except that several remote filters and filters 13 & 14 are underestimated. This could be induced by either measurement error, or the error related to filter coordinates, or the biased diffusion coefficients used in the modelling.

Taking into account the lower concentrations than actual at filters 13 and 14 in the modelling, and overestimations still occurring at most surrounding filters, one reason could be diffusion coefficients used in the modelling are higher than the actual values.



**Figure 18: relative errors between modelling results and measurements in 2019**

Next, a local gradient method is used to find the optimized diffusion coefficients. Excluding the abnormal points during 3-14 years at filters 13 & 14, all the rest measurements, including measurements of 2019 on all filters, are used for the purpose (the first measurement of filter 11 is also excluded).  $D_H$  starts from  $4.1 \times 10^{-10}$  ( $m^2/s$ ), with lower and upper bound defined as  $1 \times 10^{-10}$  and  $8 \times 10^{-10}$  ( $m^2/s$ ). Anisotropic ratio starts from 2.05, with lower and upper bound defined as 1.2-6.0. The horizontal diffusion coefficient and anisotropic ratio start from the best estimate values and loop with various combinations within the lower and upper bound.

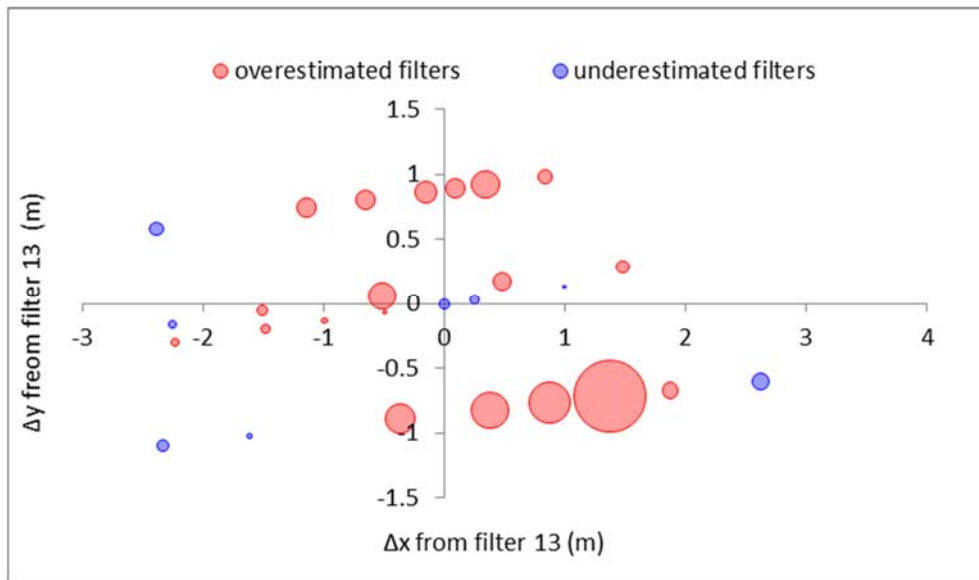
In optimization, gradient method is an algorithm to solve problems of the form  $\min_{x \in \mathbb{R}^n} f(x)$  with the search directions defined by the gradient of the function at the current point. The coefficient of determination ( $R^2$ ) is computed from the sums-of-squares terms as  $R^2 = 1 - \frac{\sum(y-y')^2}{\sum(y-\bar{y})^2}$ , where  $y$  is the log(concentration) values.

Results show that the optimized values for  $D_H$  and  $D_V$  are  $4.12 \times 10^{-10} m^2/s$  and  $1.85 \times 10^{-10} m^2/s$ , with anisotropic ratio of 2.23. Table 3 lists the  $R^2$  variations with  $D_H$  and  $D_V$  values. Figure 19 present results using these optimized diffusion coefficients. Comparing with previous figures, a little decreasing of the vertical diffusion coefficient alleviates the overestimation at most surrounding filters, as shown in Figure 19. But very less noticeable improvements are for the other two figures.

The current HTO simulation not only provides information that assists preparing the new in-situ gas diffusion test, but also validates the numerical tool to be used for the gas test.

**Table 3: optimization of diffusion parameters**

$D_H$ ( $\times 10^{-10}$ )	$D_V$ ( $\times 10^{-10}$ )	Anisotropic ratio	$R^2$
4.1	2	2.05	0.97
4.101	2.0005	2.05	0.97
4.1	1.999	2.05	0.97
<b>4.1181</b>	<b>1.8504</b>	<b>2.23</b>	<b>0.97</b>
4.1191	1.8508	2.23	0.97
4.1181	1.8495	2.23	0.97



**Figure 19: relative errors of simulation results compared with measurements in 2019 based on optimized diffusion coefficients:  $D_H=4.12 \times 10^{-10} m^2/s$  and  $D_V = 1.85 \times 10^{-10} m^2/s$**

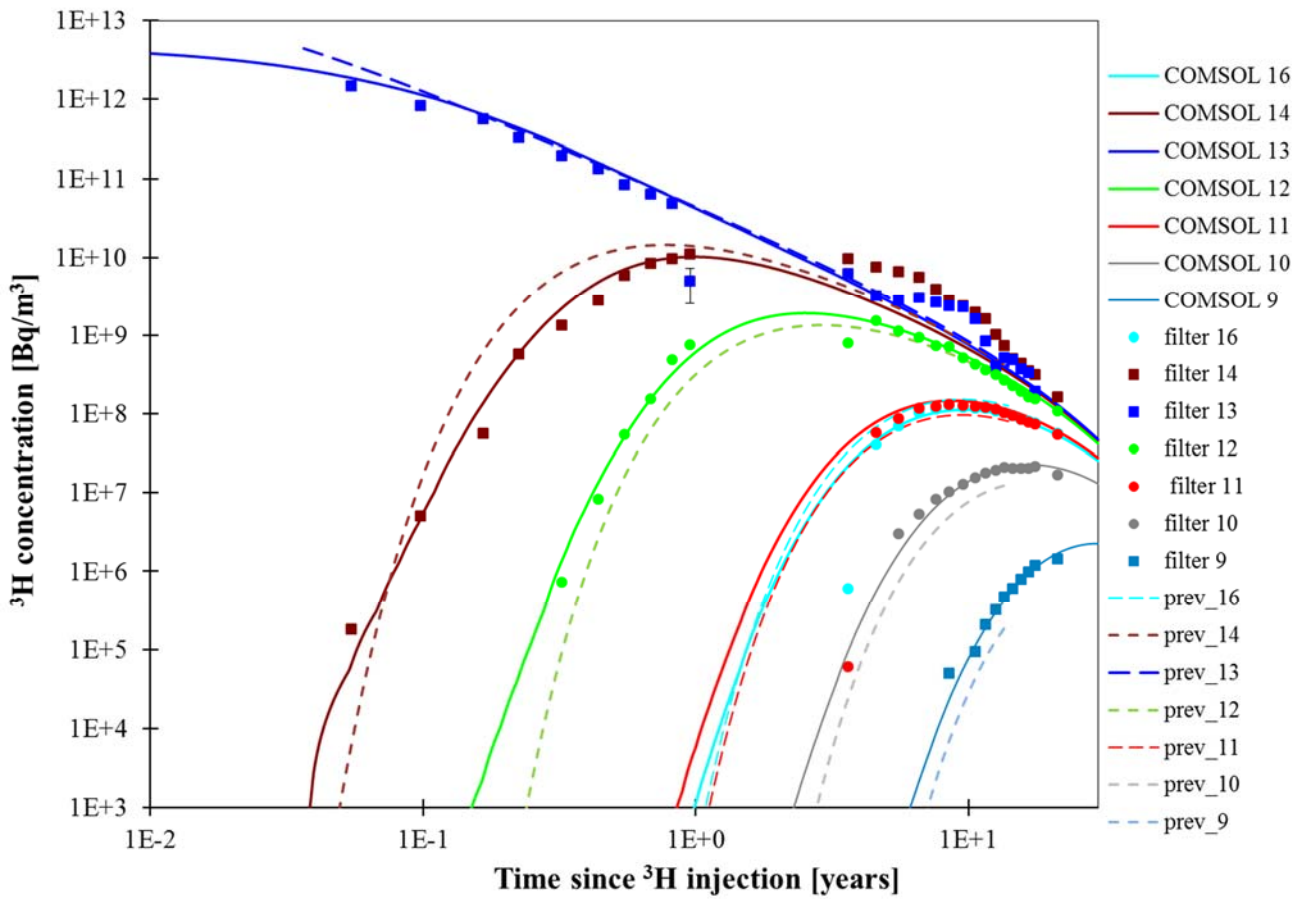


Figure 20: concentrations with optimized parameter values  $D_H=4.12 \times 10^{-10} \text{ m}^2/\text{s}$  and  $D_V=1.85 \times 10^{-10} \text{ m}^2/\text{s}$  (bi-log scale)

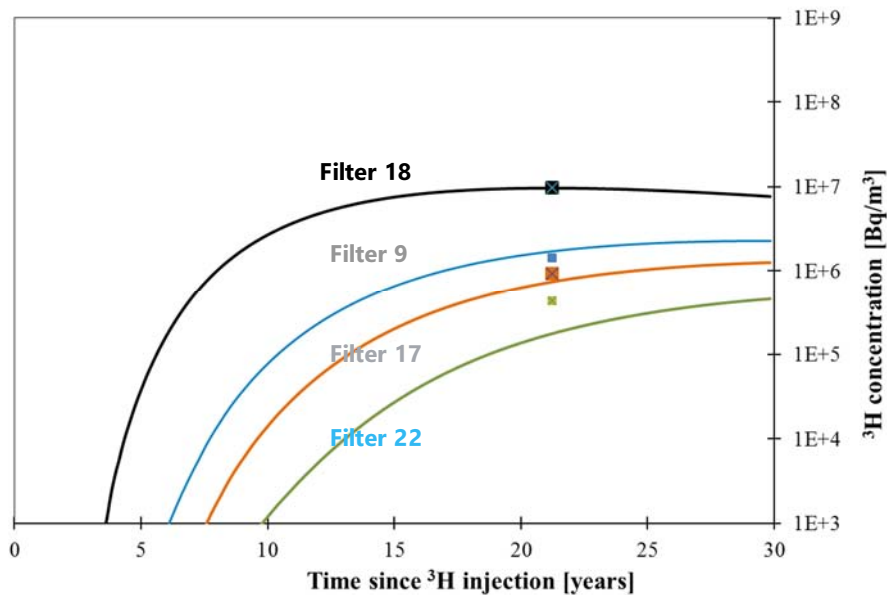


Figure 21: concentration of HTO at filters to be used in the new in situ gas diffusion test:  $D_H=4.12 \times 10^{-10} \text{ m}^2/\text{s}$  and  $D_V = 1.85 \times 10^{-10} \text{ m}^2/\text{s}$



## Annex III: Updated MEGAS *in situ* gas diffusion experiment

Annex III is an updating of the gas modelling in the report of Jacops et al. (2018). Considering the gas monitoring system to be used in the *in situ* gas diffusion test (see appendix IV for more information), the filter is changed from zero flux to zero concentration boundary. Similar to the modelling of HTO migration experiment, a sequential workflow is adopted to enhance the numerical calculation efficiency.

The excavation of the HADES URL galleries in 1982 is taken as the start of the drainage period. After 38 years of drainage by the time of the new *in situ* gas diffusion experiment starting, the water flow around filter 17 has reached steady state with a water velocity in the order of  $10^{-4}$  (m/yr). Figure 22 presents the evolution of the water velocity at filter 17 obtained from the hydraulic modelling. Actually, such a small velocity has negligible effects on the subsequent diffusion modelling results. The steady-state water velocity field is extracted from the hydraulic modelling and serves as input in the subsequent advection-diffusion modelling.

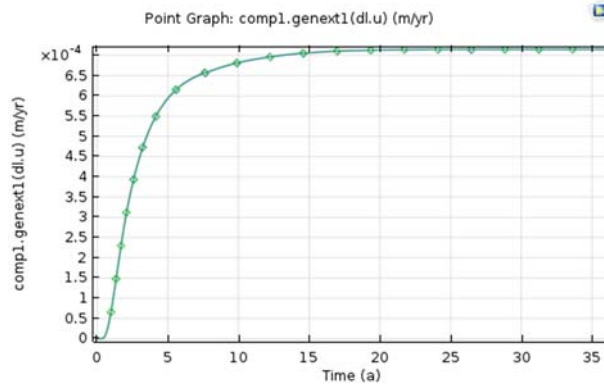


Figure 22: evolution of water velocity at filter 17

In order to get a suitable gas diffusion domain size, various domain sizes (2m, 5m and 10 m cube) around the filter 17 have been tested. Numerical results show that results from the 5-meter cube are similar to that from the 10-meter cube (Figure 23), therefore a clay cube of 5m X 5m X 5m around the injection filter 17 is used for gas diffusion modelling. Figure 24 illustrates the geometry of the modelling. Totally, 985825 quadratic tetrahedral elements are used.

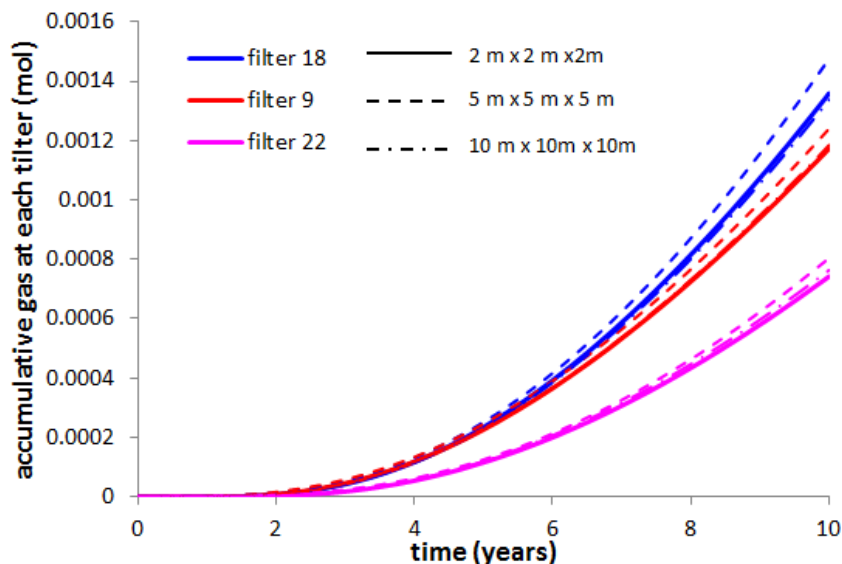
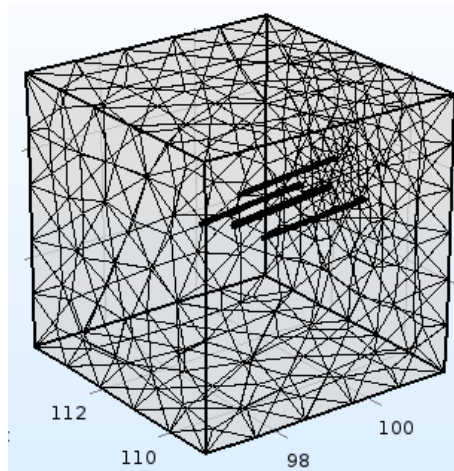


Figure 23: determination of domain size for the *in situ* gas diffusion modelling

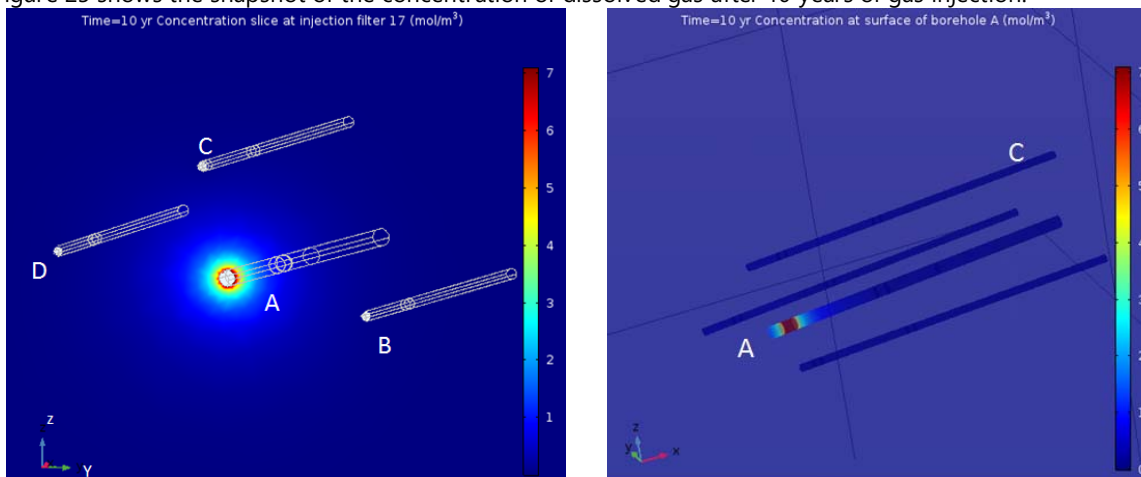
The external surface of the filter 17 is assigned with a constant concentration of dissolved Ne under a pressure of 1.58 MPa (recently measured in Oct. 2018):  $c=0.00045 \text{ mol}/(\text{kg}\cdot\text{bar})\times 15.8 \text{ bar}=7.11 \text{ mol}/\text{m}^3$ . Except for filters 9, 18, 22, which serve as zero concentration boundary, the rest part of the piezometers is considered impermeable. Other parameters used in the advection-diffusion modelling include:

- Porosity of the Boom Clay: 0.38
- Anisotropic apparent diffusion coefficient of the dissolved Ne in the Boom Clay:  $D_h=22.9 \times 10^{-11} \text{ (m}^2/\text{s)}$  &  $D_v=17.5 \times 10^{-11} \text{ (m}^2/\text{s)}$  (Jacops et al., 2017b);
- The steady-state water velocity field from hydraulic modelling is used in advection-diffusion simulation.



**Figure 24: geometry and mesh used for advection-diffusion modelling**

Figure 25 shows the snapshot of the concentration of dissolved gas after 10 years of gas injection.



**Figure 25: Concentration of dissolved gas after 10 years of gas injection: YZ Cross section at filter 17(left); Along the borehole surface of A (right)**

Cumulative dissolved gas that will be collected at some neighbouring filters is predicted in Figure 26. Among them, the three nearest filters to the injection filter 17 collect more gas and are suitable for monitoring filters: filter 18 with a distance of 0.75 m (along the piezometer), filter 9 with a distance of 0.55 m (vertical); filter 22 with a distance of 0.705 m (horizontal). The total amount of Ne injected into filter 17 reaches 0.3 mol after 10 years (Figure 27).



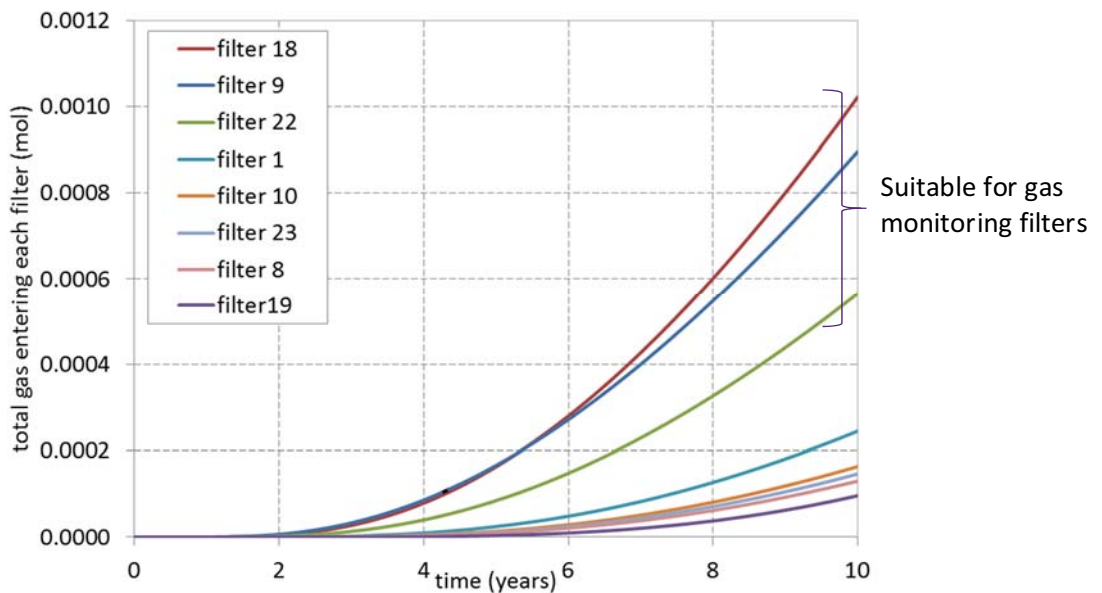


Figure 26: cumulative gas diffused into neighbouring filters around the gas injection filter

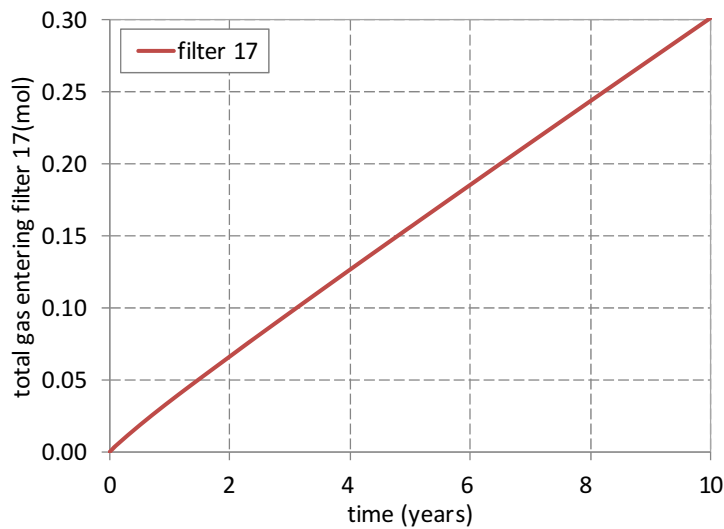
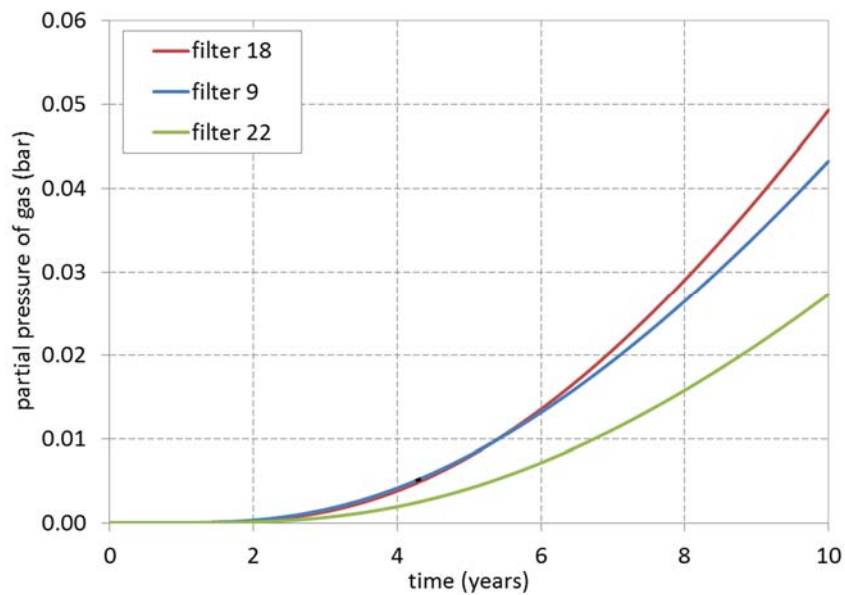
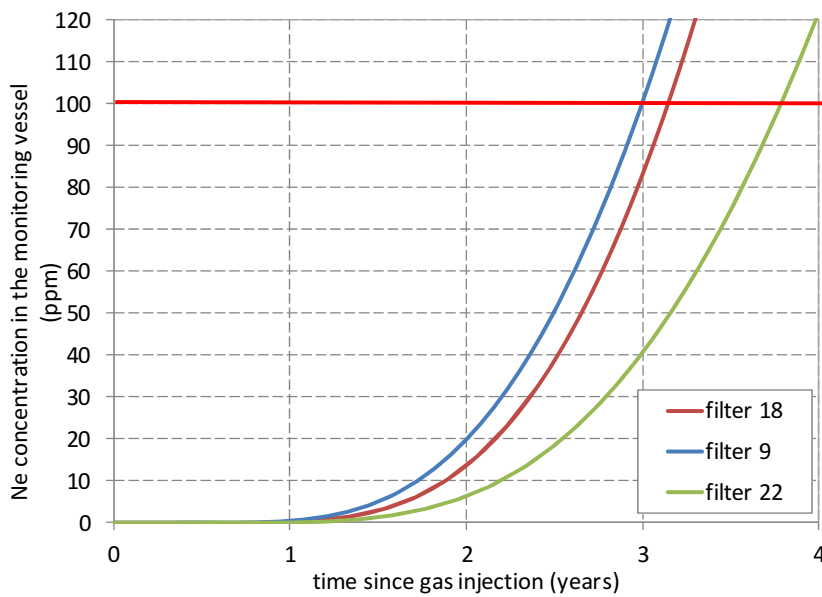


Figure 27: total gas flowing into the gas injection filter 17

Knowing the total amount of dissolved gas entering each monitoring filter, the partial gas pressure of Ne (bar) and concentration of Ne (ppm) in the gas phase can be calculated based on the equilibrium in the monitoring vessel (see appendix IV), as shown in Figure 28. When taking into account a detection limit of 100 ppm, Ne is estimated to be detected between 3-4 years (Figure 29).



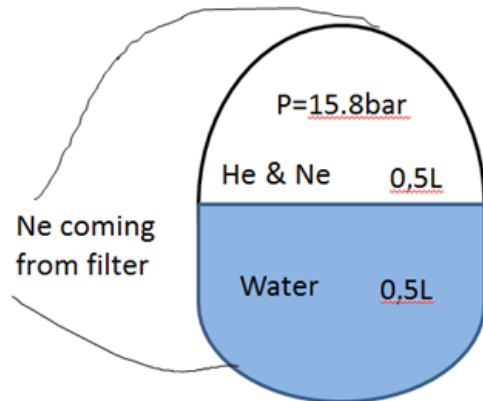
**Figure 28: partial pressure of Ne in three monitoring vessels**



**Figure 29: Zoom of Ne concentration in the gas phase (in ppm) to indicate when the detection limit of 100 ppm is passed considering a total gas pressure of 15.8 bar in the monitoring vessel.**

## Annex IV: gas monitoring vessel

Figure 30 illustrates the gas monitoring vessel to be used in the MEGAS *in situ* gas test. Initially, the vessel is filled with 0.5L gas (He at 15.8 bar) and 0.5L artificial pore water. The two tubs are connected to the monitoring filter and water keeps circulating during the test. With the test going on, dissolved Ne will flow into the filter and then to the vessel. Ne concentration in the gas phase and in the liquid phase will become equilibrated. Gas sampled will be regularly taken from the gas phase to measure the Ne concentration. The total gas pressure in the vessel will keep constant to be 15.8 bar.



**Figure 30: illustration of the gas monitoring vessel to be used in the in situ gas test**

From the numerical simulations, the total amount of Ne flowing into the vessel  $n_{total}$  is obtained, among which, part of them stays in the liquid phase and the rest goes into the gas phase. Based on the ideal gas law, the gaseous Ne (mol) in the gas phase is  $n_g = \frac{P_{Ne}V_g}{RT}$ . Henry's law gives the dissolved Ne in the liquid phase:  $n_l = P_{Ne}H_{Ne}V_l$ . Suppose a total of 0,1 mol dissolved Ne entering the vessel, the partial pressure of Ne in the gas phase should be

$$P_{Ne} = \frac{n_{total}}{H + \frac{V_g}{RT}} = \frac{0.1 \text{ mol}}{4,5 \times 10^{-4} \frac{\text{mol}}{\text{L} \cdot \text{bar}} \times 0,5 \text{ L} + \frac{0,5 \times 10^{-3} \text{ m}^3}{8,314 \times 293,15} \times 10^5 \text{ Pa} / \text{bar}} = 4,87 \text{ bar}$$

The Ne in the gas phase is:

$$n_g = \frac{P_{Ne}V_g}{RT} = \frac{4,87 \text{ bar} \times 0,5 \text{ L}}{8,314 \times 293,15} \times 100 \approx 0,1 \text{ mol}$$

Results indicate that nearly all the dissolved Ne goes into the gas phase and the concentration of Ne in the liquid phase should be around zero. Therefore, a zero concentration prescribed at the monitoring filters is used in the numerical modelling.

The relative partial pressure of Ne in the gas phase is consequently calculated to be:

$$\frac{P_{Ne}}{P_{total}} \times 1e6 = \frac{4,87}{15,8} \times 1e6 = 300000 \text{ ppm}$$

### Reference:

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## ER-0531 A new in situ gas diffusion experiment - objectives, design and experimental protocol

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