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Natural mitigation of CO_2 leakage accumulations

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Abstract

This study aims at investigating the role played by the overlying aquifer formation as a safety barrier in case of CO_2 leakage accumulation (i.e. CO_2 accumulated in an overlying aquifer after its leakage from the CO_2 storage reservoir) by focusing on its natural capacity to prevent any further upward migration. Based on numerical simulations performed using TOUGH2/ECO2N incl. hysteretic module, we assess the processes influencing the quantity of mobile CO_2 within the leaking plume and perform a sensitivity analysis to point out the key-parameters and conditions for an efficient natural trapping by dissolution and residual trapping. Additional simulations of a leak - active remediation scenario on a complete system (storing reservoir connected to an overlying aquifer) show the importance of the natural trapping capacity of the overlying aquifer. This capacity could be integrated in the mitigation strategy, associating natural and engineered safety barriers. Gaining more knowledge on these formations can support first estimations of this natural capacity, and hence can help building the corrective measure plan and designing potential interventions during operations.

Keywords: CO_2 *geological storage; leakage;mitigation; overlying aquifer; trapping*

1. Introduction

Carbon dioxide geological storage in deep saline aquifer is considered as a promising solution to ensure the necessary decrease of CO_2 anthropogenic emissions (IPCC [1]). Its industrial development is above all conditioned by its safety demonstration. The flow behavior is notably driven by the interplay between viscous, capillary and gravity forces; these phenomena have been widely studied in the storage aquifer (e.g. Ide et al. [2]; Kopp et al. [3]), which is characterized by high advective forces during the injection period. Several modes of trapping ensure the confinement of the injected CO_2 within saline aquifers: structural and residual trapping (as a gas phase), solubility trapping (dissolved in the aqueous phase) and mineral trapping (Bachu et al., [4]; Juanes et al. [5]; Ennis-King and Paterson [6]; Gunter et al. [7]).

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Nevertheless, the risk of CO_2 leakage outside of the storage reservoir should be considered, as well as its possible migration to sensitive targets (e.g. potable groundwater, surface) should be studied. Such migration can occur through poorly sealed wells or undetected defects within the reservoir caprock leading to potential CO_2 accumulation in overlying formations before impacting shallower targets (Benson and Hepple [8]). Models describing and quantifying CO_2 leakage through one or multiple wells in a sequence a geological layers have been proposed (e.g. Chang et al. [9]; Celia et al. [10]). In addition to those global models, there is a need of understanding the processes occurring in intermediate formations where CO_2 would accumulate, such accumulation being potential sources for further upward migration. The trapping modes and interplay between forces also occur in overlying aquifers with different magnitudes than for the reservoir: advective forces are indeed likely to be less predominant. Therefore, depending on the processes, the leaking plume might either be immobilized or stay mobile, thus increasing the risk of additional leakage.

In this work, we consider a leakage plume accumulating in an overlying aquifer, and we study the conditions under which the natural flow processes acting within this aquifer lead to the immobilization of the buoyant mobile gas, i.e. the conditions that would allow an "efficient" natural mitigation of a potential accumulation. We focus here on the natural processes of dissolution and residual trapping and the concept of efficiency is understood in terms of time duration to trap a given amount of mobile accumulated CO_2. The term "natural" is used here in opposition to engineered barriers implying notably fluid management techniques as proposed in the literature (Réveillère et al. [11]; Esposito & Benson [12]) for an active remediation of a potential CO_2 leakage.

In a first section, we focus on the processes influencing the natural mitigation capacity and perform sensitivity analysis to point out the key-parameters for an efficient natural trapping of the leaking plume. In a second section, we model a leak scenario on a complete system (storing reservoir, leakage pathway and overlying aquifer) and assess how this capacity can be used according to the intervention strategy to be implemented.

2. Analysis of the physical processes during and after a leakage in the overlying aquifer

2.1. Objective and model set-up

In this section, we assess how the interplay between viscous, gravity and capillary forces influences the trapping in the overlying aquifer. Our discussion is based on a sensitivity analysis using a simple model which represents a leakage in a horizontal aquifer formation (see Figure 1). The formation properties (Table 1) are based on typical values that can be found for instance in the Paris Basin limestone aquifers. The virtual alternative cases considered for the sensitivity analysis are synthetized in Table 2. They have been built to study the widest range of situations, explaining that parameters or combination of parameters might be extreme. The following simulations have been done using TOUGH2 code (Pruess et al. [13]) including the EOS module ECO2N (Pruess [14]) and a hysteresis module (see Doughty [15]). In this last module, Land's trapping model (Land [16]) is used with hysteretic characteristic functions derived from van Genuchten's capillary pressure function (van Genuchten [17]) and based on Lenhard and Parker's relative permeabilities (Lenhard and Parker [18]). 2-D leakage simulations were performed with a 10 layers discretisation in z-axis and a discretisation in x-axis evolving according to the case being modelled in order to refine the mesh in the surroundings of the gaseous plume.

The simulations were divided in two steps: a leakage phase and a relaxation phase (i.e. we stop the leakage and see how the leaking plume evolves in the system). The influence of the parameters during these two phases is discussed.

Table 1: Properties for the limestone aquifer considered for the base case (adapted from André et al. [19])

General properties		*Van Genuchten relative permeability functions*	
Depth	1000 m	M	0.600
Height	50 m	S_{lr}	0.2
Temperature	45 °C	$S_{gr,\ max}$	0.2
Porosity	0.12	*Capillary pressure*	
Transversal Permeability	10^{-13} m²	M	0.600
Longitudinal Permeability	10^{-14} m²	S_{lr}	0.2
Salinity	0.5 %wt	$S_{gr,\ max}$	0.2
Groundwater flow (gw)	1 m/y	P_0	54000 Pa

Table 2: Definition of the Base case (case 1) and alternative cases presented in this study and corresponding trapping efficiency expressed as the time duration for trapping 95% of the leaked CO_2 ('-' refers the base case value)

Case number	1	2	3	4	5	6	7	8	9	10	11	12	13
Vertical k (m²)	10^{-14}	10^{-13}	-	10^{-15}	10^{-13}	10^{-15}	10^{-13}	-	10^{-15}	-	-	-	-
Groundwater flow (m/y)	1.0	-	-	-	-	-	-	-	-	-	-	0.1	0.1
P_0 (kPa)	54	540	540	540	-	-	5.4	5.4	5.4	-	-	-	5.4
m for k_r	0.6	-	-	-	-	-	-	-	-	0.9	-	-	-
$S_{gr,\ max}$	0,2	-	-	-	--	-	-	-	-	-	0.3	-	-
Time 95% trapping (months)	20	3	3	3	30	14	69	57	28	63	10	39	Uncomplete

2.2. Leakage phase

To date, the upward migration of CO_2 from a reservoir has been mostly studied through analogues assessment (natural or industrial) and/or modeling. According to de Lary et al. [20] who did the literature synthesis of such studies, the flow rate which reaches the surface following a leakage from the reservoir could be subject to substantial variations depending on the situation and the modality of CO_2 uprising. Some modeling works giving leaking rates in-depth exist (Humez et al. [21]; Réveillère et al. [11]; Oldenburg et al. [22]) but the rates and geometry of the leakage considered are highly variable as well. We decided here to model the leakage for the simple case study as follows: the leakage is pressure-driven (1 bar overpressure in the base case), with a constant CO2 saturation (0.5 for the base case) on all the aquifer height (Figure 1). This would represent for instance a leakage from a fault.

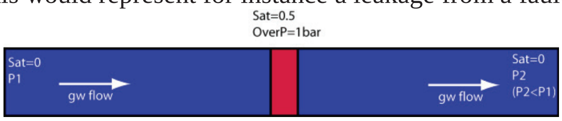


Figure 1: Overview and limit conditions of the simple case study

To compare the evolution of the leaking plume for different cases, the leakage is stopped after a given leaking mass per linear meter in the y-direction (100 tons/lm). Considering a length similar to the distances observed in Latera (Beaubien et al. [23]), this would correspond to a total leaking plume of 5,000-10,000 tons. This quantity may be compared to the monitoring possibilities given by Benson [24] for a 1000 m deep aquifer, who stated that a quantity of 1000-10000 t may be detectable.

The nine first cases were designed to test the effects of modifying the ratios of viscous, gravity and capillary forces. To quantify these changes, we use the following dimensionless numbers:

$$Macroscopic\ gravity\ number = N_{gv} = \frac{L}{v} \cdot \frac{\Delta\rho \cdot g \cdot k_v}{h \cdot \mu_l},$$

v being the total flow velocity in the horizontal direction, $\Delta\rho$ the density difference between the liquid and gas phases, k_v the vertical permeability, L and h are characteristic length and height of the flow and μ_l the fluid velocity (here brine). Many forms of gravity number could have been used, here we use the one proposed by Zhou et al. [25], and considered by Ide et al. [2] and Kopp et al. [3]. This number gives the relative significance of gravity forces compared to viscous ones (formally, it is the ratio of the time for fluid to flow in the horizontal direction due to viscous forces compared to that in the vertical one due to gravity forces). Depending on the time of observation and on the condition of leakage, v can be due to the pressure gradient induced by the leakage (case of a strong leakage) or by regional pressure gradient-groundwater flow (case of a smaller leakage, considered here).

$$Macroscopic\ capillary\ number = N_{cv} = \frac{\mathrm{k} \cdot p_{cr}}{l_{cr} \cdot \mu_l \cdot v_{cr}},$$

p_{cr}, l_{cr} and v_{cr} being respectively the characteristic value of capillary pressure, length and velocities. This number represents the ratio between capillary forces and viscous ones (ratio between the capillary pressure difference and the viscous pressure difference on a characteristic length). Kopp et al. [3], who used this number in this form, insisted on the importance of the choice of these characteristic values.

Figure 2 presents the extent of the sensitivity study by describing the difference in terms of shapes of the leaking plume according to the capillary and gravity numbers for each case. The plume shape has been recognized as an important parameter for capillary trapping (see for instance MacMinn & Juanes [26]). A high N_{gv} leads to gravity segregation and plume tonguing. With a high N_{cv}, the segregation is decreased, the vertical extent of the plume is increased and the average saturation lowered (see for instance Golding et al. [27]). The influence of the groundwater flow (regional viscous forces) is observed when the ratio of viscous forces is more important. Please note that in our cases of relatively small leakage, the capillary and gravity numbers are relatively high, regarding for instance the values obtained for the storage plume in the storing reservoir which is characterized by strong viscous forces created by the injection.
The cases presented in Figure 2 only concern cases with similar mobility ratio, defined as such:

$$Mobility\ ratio = M = \frac{\lambda_g}{\lambda_l},$$

$\lambda_i = \frac{k_{r,i}}{\mu_i}$, $i = g$ or aq, being the mobility of each phase. It represents the ratio between the gas viscous forces and the liquid viscous forces. This ratio is a function of the saturation. The effect of lower mobility of CO_2 is a decrease of the gas flow. Then, the typical consequences are a decrease of the tonguing or of the extension and an increase of the saturations in the plume.

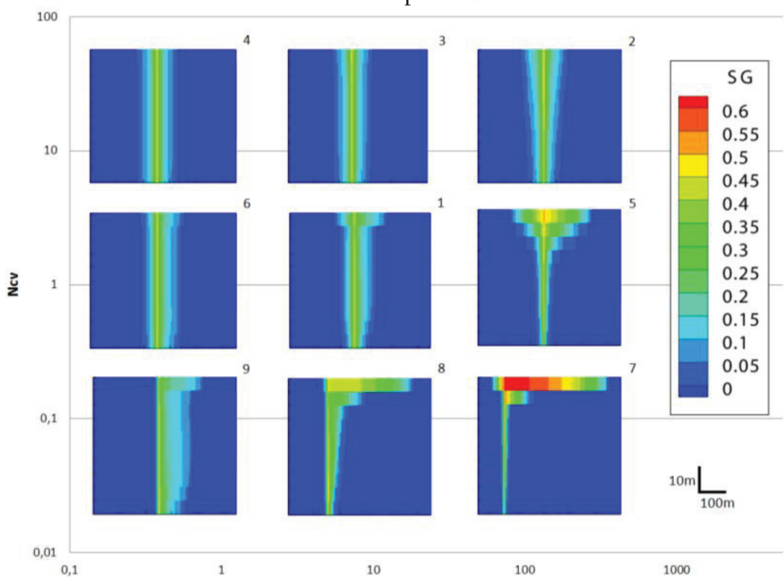


Figure 2: Leaking plume shape rough regions as a function of capillary and gravity numbers (cases 1 to 9).

2.3. Relaxation phase

After 100 kt/lm has leaked, the leakage is stopped in order to assess the immobilisation of the plume in different configurations. We evaluate the quantity of dissolved and capillary trapped gas to deduce mobile gaseous CO_2 left, which presents a risk of further migration. The comparison between the different cases is made through the relaxation duration needed to reach 5% of mobile gaseous mass fraction (denoted $t_{95\%}$). $t_{95\%}$ is given for each case in Table 2. Several parameters are involved in the plume evolution during the relaxation. We investigate the main parameters in the following of the study. Please note that we focus on the trends observed rather than on the time durations themselves, which might not have a real signification on such a theoretical case.

Gravity and capillary effects influence: in a first step, we evaluate the influence of capillary forces and gravity, keeping the viscous forces due to groundwater flow constant. We simulate the relaxation of the nine first cases corresponding to the capillary and gravity numbers of the cases presented in Figure 2. The results of $t_{95\%}$ are presented on Figure 3. We observe the following trends:

1/ Increasing the gravity effects intensifies the gravity tongue that forms both during the leakage and after it and leads to both a reduction of the dissolution and of the trapping quantity. To understand more deeply this statement, we display on Figure 4 the trapping evolution between cases n°7 and n°9, which have similar conditions excepted different gravity forces. The differences observed in $t_{95\%}$ come from both trapping modes. Dissolution is increased when gravity is less important (both during the leakage, see at t=0, and after due to a larger volume of brine contacted), as well as residual trapping, which is more efficient mainly due to the lower CO_2 average saturation in the leaking plume.

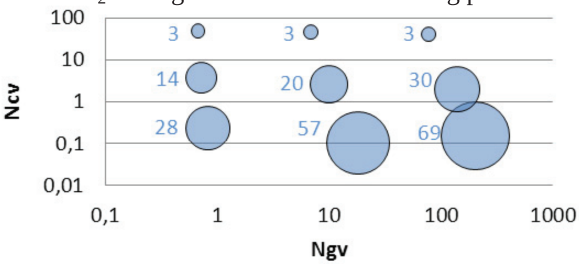


Figure 3: Time of total trapping (blue number, in months) as a function of Ncv and Ngv (with constant viscous forces) (case 1 to 9)

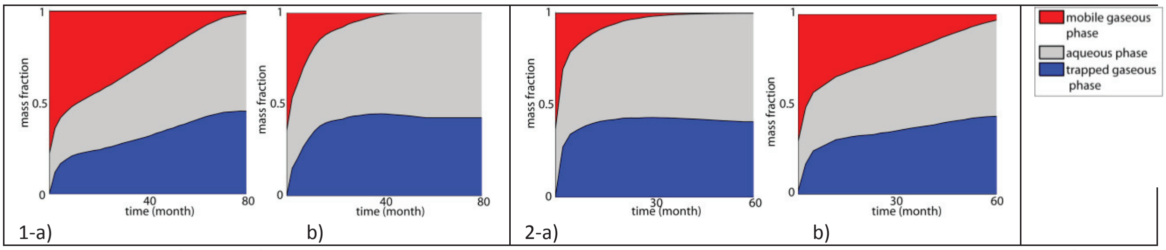


Figure 4: 1-Influence of gravity - comparison between case 7 (1-a, higher gravity) and 9 (1-b, lower gravity) / 2- Influence of capillary forces – comparison between case 1 (2-a, higher capillary forces) and 8 (2-b, lower capillary forces)

2/ High capillary forces induce a significant vertical and horizontal extension of the plume and lower average saturation. The initial shape and saturation distribution of the plume associated to the higher rate of capillary-driven imbibition accelerates both the dissolution and the residual trapping as shown in Figure 4, which depicts the evolution of mobile gaseous, trapped gaseous and aqueous mass fractions of CO_2 over time for case n°1 and case n°8 (lower capillary forces).

Viscous effects influence: In this study the leakage is rather small and we assume that the viscous forces are dominated by the groundwater flow. The contact of the plume with fresh brine is therefore improved when the viscous forces are increased, leading logically to an increase of the dissolution and of the capillary trapping. We can then note that the capillary number variations (if both capillary and viscous forces change) cannot be directly used for the comparison of the trapping in two cases where both viscous and capillary forces change since both forces lead to a larger trapping. As an illustration of this point, we compare a case similar to a rapid trapping case (case n°2) with regards to the capillary number (the groundwater flow and capillary forces were decreased): this is case n°12. $t_{95\%}$ equals 39 months for case n°12 compared to 3 months for case n°2. The same operation was done with case n°5 ($t_{95\%}$=30 months) with a lower groundwater flow and capillary pressure. In that last case (n°13), there is still 20% of mobile gaseous CO_2 after 150 months, meaning that the trapping is very slow.

Mobility ratio influence: at given capillary, gravity and viscous forces, an increase in the mobility ratio leads to a more stretched-out plume, increasing the quantity of CO_2 dissolved both during the leakage and during the relaxation. The increased mobility of CO_2 also leads to a more rapid residual trapping, the residual saturation being reached more rapidly since gas flow increases with the mobility ratio. This is enhanced by the lower initial average saturation in the plume before imbibition.

$S_{gr,max}$ *influence:* Regarding the residual trapping, a classical model was used in this study (Land, [16]). This model gives a final residual saturation from the initial gas saturation reached at the end of the drainage stage. This model depends notably on the maximum residual gas saturation ($S_{gr,max}$), which is obtained when the irreducible water saturation is reached during drainage. The other parameters being constant, a lower $S_{gr,max}$ decreases the immobilized gas saturation in the pores and decreases the residual trapped quantity at a given time. The CO_2 mass fraction dissolved in brine is not highly impacted in the flow by this change.

The simulations in this simple model allow understanding the phenomenology involved in the trapping of the CO_2 plume leaking from a storing formation to an overlying aquifer and assessing the natural mitigation capacity of such aquifer. In some situations, total trapping occur after a short relaxation time and this capacity may be used to prevent further migration towards a sensitive target in the subsurface or at the surface.

3. Complete migration and relaxation model

This section investigates how the overlying aquifer mitigation capacity can be integrated in the mitigation strategy in order to prevent impacting migration of mobile CO_2 in case of gaseous leakage. For this purpose, an injection in a storage aquifer, a leak through a given pathway and the resulting accumulation in an overlying aquifer, and intervention strategies likely to be implemented in such a case are simulated.

3.1. Description

The case study was designed by Réveillère et al. [11], who simulated the leakage from a storage aquifer within an overlying one (the reader is referred to this article for more precisions, especially for the parameter values, which are the same than for the base case). TOUGH2/ECO2n code and associated hysteresis module was used for the simulations. To compare different situations, we simulated the leakage with two capillary pressure cases (capillary strength P_0 = 5,000 and 20,000 Pa) and for two groundwater (gw) flow velocities (0 m/y and 1 m/y) in the overlying aquifer. CO_2 is injected in the storing aquifer (50 kg/s), reaches the leak, migrates upwards and forms a leakage plume in the upper aquifer. Figure 5 presents the leaking plume after 6.3 years of injection.

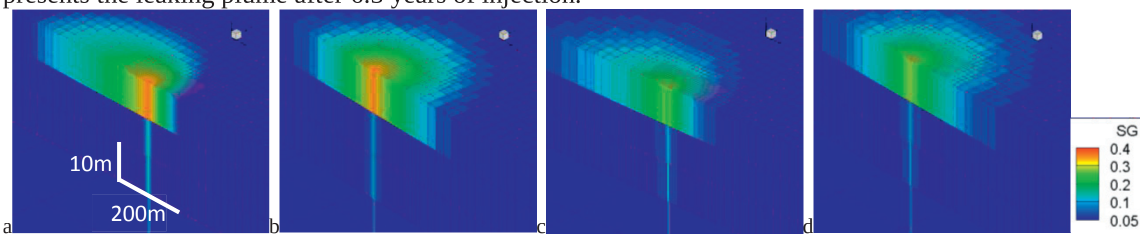


Figure 5: overlying aquifer after 6.3 years of injection in the main aquifer – a) 1m/y gw flow + lower P_{cap}, b) no gw flow + lower P_{cap}, c) 1m/y gw flow + higher P_{cap}, d) no gw flow + higher P_{cap}

We logically observe a significant segregation (no anisotropy was considered), meaning that the gravity forces are important relatively to the capillary and viscous ones (due to both natural pressure gradient and leakage induced pressure gradient). The groundwater flow influences the plume shape. The influence of the capillary pressure is observed by a smoothing of the saturation gradient along the plume. The leaking CO_2 mass after 6.3 years of CO_2 injection in the main aquifer is sensibly similar for all cases: around 9,500 kg of CO_2. We consider, for this study, that this quantity can be detected (reasonable choice according to the detection thresholds discussed by Benson [24]). After the detection, we assume that two main decisions could be taken for the management of the risk associated with the secondary accumulation:

1- Intervention on the transfer pathway if its location has been characterized and repair. The leakage immediately stops.
2- No intervention on the transfer pathway but the injection in the reservoir is turned off. The leakage continues at a decreasing rate, as the pressure in the storage aquifer decreases.

In the following, we assess the behavior of the mobile gaseous CO_2 in the overlying aquifer for both cases in order to evaluate the necessity of an additional intervention in the overlying aquifer if the risks of subsequent migration are important.

3.2. Results and discussion

The results in terms of mass balance are presented in Table 3. For the case with a lower capillary pressure, one can notice the importance of the viscous effects due to groundwater flow; without any regional pressure gradient, a significant amount of gaseous CO_2 still remains mobile in both cases of intervention. A groundwater flow of 1 m/s leads to a quasi-total trapping of the mobile gas after 10 years if the injection is stopped and after 30 months if the leakage is stopped. Both residual and solubility trapping are necessary to the total trapping, the increase of residual trapping being more important. Regarding the case with higher capillary strength, when intervention on the leaking pathway occurs, the induced-spreading leads to the residual and solubility trapping of the CO_2 in the same time frame whether or not there is a groundwater flow. When the injection is stopped, the groundwater flow leads to a faster total trapping even at the beginning of the period studied; there is even a stagnation of the total trapped quantity for the no-groundwater flow case. The conclusions are identical to the ones stated for the simple case: when viscous regional forces are present, the CO_2 is trapped both by capillarity and dissolution; an increase in capillary pressure increases the trapping. The dependence of the trapping on the aquifer characteristics and properties is important and can play a major role in the mitigation of further CO_2 gaseous migration.

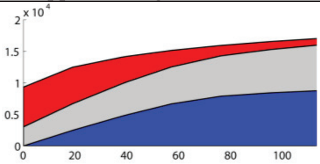
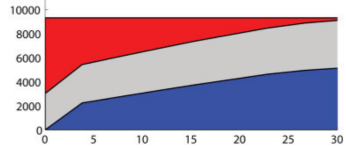
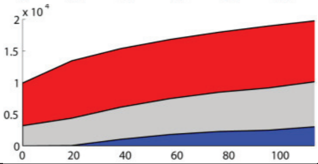
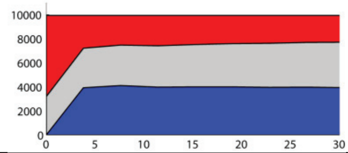
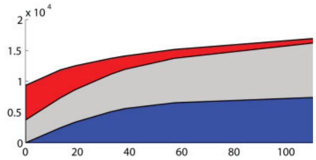
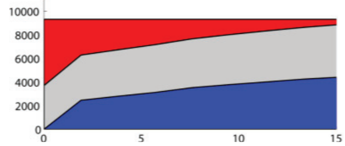
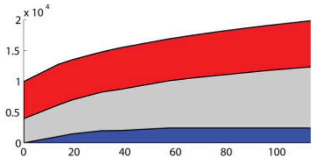
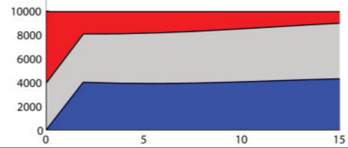
From an operational point of view, the intervention mode is important since the time frame needed to trap all the mobile CO_2 is lower when the transfer pathways is remediated; however, according to our case study, a complex intervention on the leaking pathways is not always necessary: stopping the injection can lead to the leaking plume trapping in acceptable time frame when enough viscous forces exist.

4. Conclusion and perspective

We have studied the evolution of a secondary accumulation in an overlying aquifer (non-sensitive) in case of CO_2 leakage from the main reservoir; this second accumulation being potentially the source of additional upward migration leading to environmental impacts, this work has been focused to the natural capacity of the intermediate aquifer to decrease the quantity of mobile CO_2.

The trapping of a leaking plume is dependent on the interplay between forces in an overlying aquifer (most of time different from those occurring in the reservoir). A theoretical study associated to both simple and more complex modeling work have shown that in some conditions, overlying aquifers could be an integral part of the mitigation strategy. Knowledge of overlying aquifers seems essential in this view: the characterization of the regional hydrology of the sedimentary basin could provide a better picture of the transfers between aquifer and first estimations for groundwater flow and permeability anisotropy. We acknowledge the difficulties to characterize multiphase flow properties, but insist that these are expected to play a significant role. The knowledge of the overlying aquifer might indeed not be as precise as it would be for the main reservoir but first estimations of the natural capacity to trap mobile gaseous CO_2 will help building the corrective measure plan and designing potential interventions during operations. The bases laid in this study could be used in the development of a screening tool for such estimations.

Table 3: CO_2 mass balance in the overlying aquifer (in x-axis of the charts, time in months; in y-axis, mass in tons)

	Injection is stopped; leakage continues		**Intervention on the leak; leakage stopped**	
Po= 5,000Pa	*Gw flow*		*Gw flow*	
	No gw flow		*No gw flow*	
Po= 20,000Pa	*Gw flow*		*Gw flow*	
	No gw flow		*No gw flow*	

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