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Global warming effect of leakage from CO₂ storage



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Global warming effect of leakage from CO₂ storage

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1 Introduction

Most assessments of carbon capture and storage (CCS) characterise technologies in terms of annual or cumulative avoided emissions. However, the ultimate purpose of CCS is not the abatement of emissions, but the avoidance of damages to be expected from climate change. Between emissions and damages, there is a causal chain of quantities such as radiative forcing, global warming, and sea level rise, amongst others. Further down this causal chain¹, quantities become successively better proxies for damages from climate change (Udo de Haes *et al.* 1999), however they also become more uncertain (Lenzen 2006). This is exemplified in the European Commission's ExternE study of monetary externalities from electricity generation (Krewitt 2002).

This recognition has led the Brazilian Government to propose a methodology that calculates the contribution of entities (countries, technologies) to global warming from their historical greenhouse gas emissions (Federative Republic of Brazil 1997). In essence, in order to model the time lag between emissions and warming, this methodology formulates an approximation of carbon cycle and climate models where the temperature increase $\Delta T(t)$ at a time t is an additive function of distinct emissions "parcels" $\varepsilon(t')$ (Meira and Miguez 2000):

$$\Delta T[\varepsilon_1(t') + \varepsilon_2(t'), t] = \Delta T[\varepsilon_1(t'), t] + \Delta T[\varepsilon_2(t'), t]. \quad (1)$$

Whilst the Brazilian Government had primarily the distinction between countries in mind, the idea of this work is to use the above mathematical formulation of the Revised Brazilian Proposal (RBP) to enumerate the effect on global warming of leakage from geological CO₂ storage.

Two CO₂ storage and monitoring projects have injected so far more than 5 Mt CO₂ into a depleted oil field (Canada) and more than 10 Mt CO₂ into a deep saline formation (off-shore Norway) without detectable leakage (IEA GHG 2008). However, technology assessments mention the possibility of rapid leakage paths, for example in form of failed wells. Lower rates of seepage can be expected through known and unknown permeable faults, with the CO₂ driven either passively through diffusion, or through active porous flow facilitated by tidal pumping (Lenzen and Neugebauer 1999).

Expected leakage rates are very likely to be less than 1% in 100 years, and likely to be less than 1% per thousand years (IPCC 2005a), and thus low enough to not lead to long-term increases of atmospheric CO₂ concentrations (IEA GHG 2008). However, understanding of leakage and long-term movement of CO₂ is so far mainly obtained from complex numerical simulations, and not from field measurements, and the IPCC 2005b acknowledges that experience with geological storage is still limited, and leakage risks still need to be confirmed in large-scale applications. Several authors emphasise the lack of knowledge and experience with underground storage (Pehnt and Henkel 2009; Riahi *et al.* 2005; Diesendorf 2006; Viebahn *et al.* 2007), and conclude that there is no

¹ Life-cycle assessment (LCA) uses the terms "mid-points" and "end-points" in order to characterise the causal distance of measured and reported quantities to the question asked (Bare *et al.* 2000; Heijungs *et al.* 2003; Hertwich and Hammitt 2001).

guarantee for the low leakage rates expected by the IPCC 2005a. However, these concerns are not necessarily shared by the public (Shackley *et al.* 2007).

Previous studies on CCS have only dealt with impermanent storage in terms of allowed CO₂ emissions in stabilisation scenarios (IPCC 2005a), but not in terms of resulting atmospheric CO₂ concentrations or global warming effects. Other studies (Marland *et al.* 2001; Dutschke 2002) have investigated the policy implications of temporary storage of CO₂ but with a focus on biospheric carbon pools (for example timber products). In addition, studies dealing with life-cycle emissions impacts of CCS do either not deal with leaks from storage at all (Pehnt and Henkel 2009; Hertwich *et al.* 2008; Odeh and Cockerill 2008), or suggest using weighting factors similar to discount rates (Viebahn *et al.* 2007). Given these shortcomings, a thorough analysis of the relationship between CO₂ leakage and global temperature change for a whole range of possible leakage rates is required. These findings can then be used as a basis for decision-making in defining acceptable upper limits for leakage rates.

This paper unfolds as following: Section 2 will explain the methodology of the Revised Brazilian Proposal (RBP) in translating emissions histories into temperature changes. Section 3 presents the results of simulations for a range of leakage rates, and sensitivity analyses. Section 4 discusses the results and concludes.

2 Methodology

We follow the RBP in decomposing global temperature increase $\Delta T(t)$ at a time t into contributions by historical emissions “parcels” (t'). The calculus proceeds in three steps: a) from historical emissions $\varepsilon_g(t'')$ of gases g to their atmospheric concentrations $\Delta q_g(t')$ above pre-industrial levels, b) from concentrations to mean radiative forcing $\Delta Q_g(t')$, and c) from forcing to contributions $\Delta T_g(t')$ to temperature increase (see Meira and Miguez 2000)

$$\Delta T_g(t') = \frac{1}{C} \int_{-\infty}^t \left[\bar{\sigma}_g \left(\beta_g \int_{-\infty}^{t'} \varepsilon_g(t'') \sum_{r=1}^R f_{gr} e^{-\frac{t'-t''}{\tau_{gr}}} dt'' \right) \right] \sum_{s=1}^S \frac{l_s}{\tau_{cs}} e^{-\frac{t-t'}{\tau_{cs}}} dt', \quad (2)$$

where

- $\varepsilon_g(t'')$ are emissions of gas g avoided by a certain technology, in the past, or under a certain future scenario,
- f_{gr} is the r^{th} of R fractions of gas g decaying in the atmosphere with characteristic time τ_{gr} , normalised through $\sum_{r=1}^R f_{gr} = 1$,
- β_g is the above-pre-industrial atmospheric concentration of gas g per unit annual emission of that gas,
- the term in the round brackets is atmospheric concentration $\Delta q_g(t')$,
- $\bar{\sigma}_g$ is the change in mean radiative forcing by gas g per unit atmospheric concentration of that gas,
- the term in the square brackets is mean radiative forcing $\Delta Q_g(t')$,
- l_s is the s^{th} of S fractions of radiative forcing that adjusts with characteristic time τ_{cs} , normalised through $\sum_{s=1}^S l_{cs} = 1$, and
- C is the heat capacity of the climate system.

Meira and Miguez 2000 point out that Equation 2 ignores non-linearities in the warming response to emissions, due to saturation of carbon fertilisation and ocean surface uptake (meaning f_{gr} is a function of t''), and due to saturation of radiative forcing (meaning $\bar{\sigma}_g$ is a function of $\Delta q_g(t')$). In their review of the RBP, Den Elzen *et al.* 1999 note that the calculus only considers oceanic but not terrestrial carbon dynamics, and that the atmospheric lifetime of methane is concentration-dependent. Based on these criticisms, the integral calculus in Equation 2 was replaced with an iterative, differential calculus (Rosa *et al.* 2004). The parametrisation and calibration of Equation 2 with historical data is described in the Appendix.

This work focuses on the effect on global temperature increase of CO₂ leakage from geological storage sites, and also of residual CO₂ from the CCS process, the latter for example from recycling of solvents, compression, transport, etc. Assume that CO₂ emissions resulting from CCS ($i = 1$) and CO₂ leakage ($i = 2$) are $\varepsilon_{i,g}(t'')$. Then, the temperature increase at time t' attributable to the use of this technology over the period $[t_0, t']$ is calculated using Equation 2, but with specific emissions $\varepsilon_{i,g}(t'')$, and with the two lower integral bounds of $-\infty$ set to t_0 .

The $\varepsilon_{i,g}(t'')$ were specified as follows: First, without loss of generality, only CO₂ is considered, and it is assumed that CCS is only applied to power plants. A hypothetical emissions scenario for electricity generation that is similar to the IPCC SRES B1 and B2 scenarios (Nakicenovic and Swart 2000) can be approximated by

$$E_p(t'') = \varepsilon \exp\left(\frac{t''-T_0}{\tau}\right)^2, \quad (3)$$

where T_0 denotes the year of peak emissions, ε emissions at year T_0 , and τ the time constant of emissions change. $T_0 = 2050$ and $\varepsilon = 25$ Gt CO₂ are chosen in order to approximate both the SRES B1 and B2 scenarios, and $\tau = 50$ years was set so that Equation 3 correctly reproduces 2008 emissions from electricity generation.² Note that any other scenario would be equally suitable to demonstrate the effect of leakage from CO₂ storage on global warming.

CCS is applied to all emissions over a time interval [2000, 12000]. Emissions from the CCS process including capture, transport and injection, but excluding leakage, are modelled using a capture rate κ :

$$\varepsilon_{1,\text{CO}_2}(t'') = (1 - \kappa)E_p(t''). \quad (4)$$

The capture rate κ in Equation 4 includes all life-cycle emissions arising out of the CCS process.³ This includes the so-called energy penalty resulting from a) the additional energy requirements for capture, and b) conversion efficiency decreases. Energy penalties (see Tab.TS.10 in IPCC 2005a, Rubin *et al.* 2007, Odeh and Cockerill 2008, and Davison 2007) are typically 25% in post-combustion systems (due to an 8-10% efficiency decrease, and scrubbing agent regeneration), and 15% in pre-combustion (due to a 6-8% efficiency decrease, and to the water-gas shift reaction). The life-cycle component represents CO₂ transport and injection.

Odeh and Cockerill 2008 review studies of life-cycle energy requirements and CO₂ emissions of carbon capture and storage technologies. They cite a study by Spath and Mann 2004 on a pulverised coal and natural gas combined cycle plant, where post-combustion capture and storage decreased the life-cycle CO₂ emissions from 847 to 247 g/kWh, and from 499 to 245 g/kWh, respectively. Viebahn *et al.* 2007 report comparable values of 274 and 200 g/kWh, respectively. For post-combustion capture applied to pulverised coal and natural gas combined cycle plants Odeh and Cockerill 2008 report specific CO₂ emissions of 255 and 200 g/kWh, respectively. Pehnt and Henkel

² According to the IEA 2008, fossil-fuel power plants emitted 11.4 Gt of CO₂ in 2006, 41% of the world total.

³ Compare with a definition in Rubin *et al.* 2007, p. 4451 and footnote 3.

2009 state values of around 200 g/kWh. In this work it is assumed that – including all life-cycle emissions – the capture rate for all CCS applications is 80%.⁴

Capturing and storing 80% of CO₂ emitted according to Equation 3 would result in about 1600 Gt CO₂ under geological storage, which is within the range of estimated global storage capacity. Using a spatial inventory of large point-emitters (above 0.1 Mt CO₂ per year) and comparing this to the global distribution of sedimentary basins, the IPCC 2005a (Tab. TS.6) has estimated global storage capacity at about 200-2000 Gt CO₂, or equivalent to about 5-50 years of global emissions.

Emissions from leakage in geological storage sites is modelled iteratively by applying a constant leakage rate λ to cumulatively stored CO₂:

$$\varepsilon_{2,\text{CO}_2}(t'') = \lambda \varepsilon_{\text{cumulative,CO}_2}(t'') = \lambda \sum_{t=2008}^{t''-1} \varepsilon_{1,\text{CO}_2}(t) - \varepsilon_{2,\text{CO}_2}(t). \quad (5)$$

Several sensitivity analyses are undertaken by varying leakage rates λ between 1% per year and 1% per 100 years, as well as the fractions f_{gr} and l_s , and their corresponding characteristic times τ_{gr} and τ_{cs} , according to various specifications in Rosa *et al.* 2004 and UNFCCC 2009a, 2009b.

⁴ For example, the emissions from 1 kWh generated in a pulverised-coal power plant with CCS are typically composed of 880 g (combustion) + 79 g (9% efficiency penalty) + 141 g (16% remaining energy penalty) – 935 g (85% capture of 880+79+141 g) + 20 g (remaining CCS life cycle) = 185 g.

3 Results

3.1 Effect of CO₂ leakage on global warming

As expected, the lower the leakage rate, the more carbon ends up being stored in subsurface reservoirs, and the more drawn out and lower-peaking the global temperature increase resulting from CO₂ being released back into the atmosphere.

Fig. 1 shows the total emissions from applying CCS at $\kappa = 80\%$ capture rate (solid black curves, $\varepsilon_{1,\text{CO}_2}(t'') + \varepsilon_{2,\text{CO}_2}(t'')$ in Eqs. 4 and 5), which are initially mainly comprised of the uncaptured and life-cycle CO₂ (area between the solid black and dotted curves, $\varepsilon_{1,\text{CO}_2}(t'')$ in Eq. 4, not dependent on the leakage rate), but increasingly complemented by leaking CO₂ (dotted curves, $\varepsilon_{2,\text{CO}_2}(t'')$ in Eq. 5). The solid grey curves ($\varepsilon_{\text{cumulative,CO}_2}(t'')$ in Eq. 5) show the amount of CO₂ stored subsurface over time.

At 0.01%/y leakage (right graph), about 1600 Gt are stored subsurface following the phase-out of fossil power around 2100 resulting in a small leakage of 160 Mt/y, which is considerably below the emissions from the CCS process itself. Subsurface CO₂ is released slowly over more than 10,000 years. At 0.1%/y leakage (middle graph), only about 1500 Gt are stored due to increased leakage (1.5 Gt/y around 2100). Subsurface CO₂ completely escapes over a few thousands of years. At 1%/y leakage (left graph), only about 900 Gt can be stored due to significant leakage (900 Mt/y around 2100). Subsurface CO₂ completely escapes in less than 1,000 years.

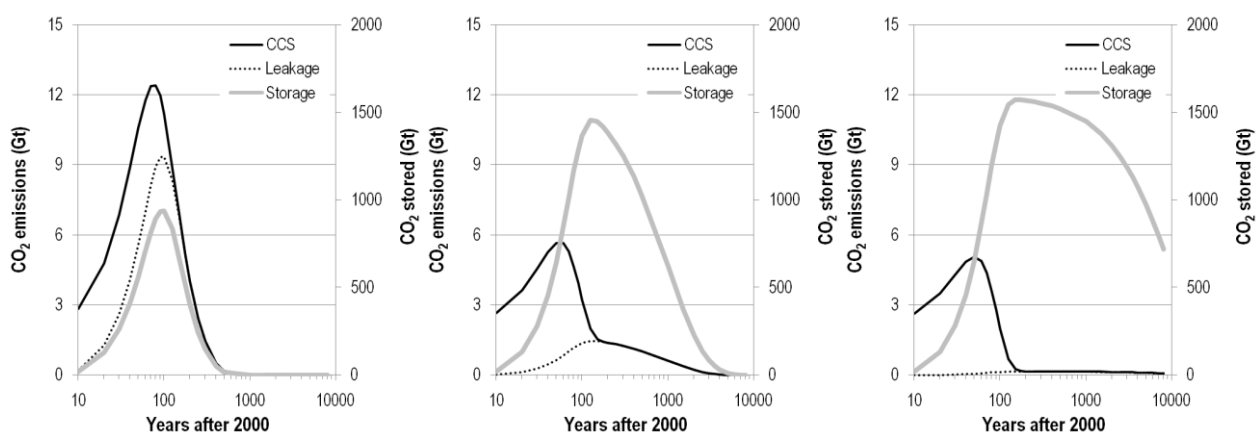


Fig. 1: Emissions over time from CO₂ leakage (dotted), the entire CCS system (solid black), as well as CO₂ stored subsurface (solid grey), for a leakage rate of 1%/y (left), 0.1%/y (middle) and 0.01%/y (right). The area between the dotted and solid black curves represents CO₂ emissions from capture, transport, injection, and CCS life-cycle.

Fig. 2 shows the global temperature increases to be expected from applying CCS at $\kappa = 80\%$ capture rate, corresponding to the scenarios in Fig. 1, calculated by applying the RBP in Eq. 2 to emissions from CCS processes $\varepsilon_{1,\text{CO}_2}(t'')$ and leakage $\varepsilon_{2,\text{CO}_2}(t'')$.

At 0.01%/y leakage (right graph), leakages contributes negligibly to the 0.16°C temperature increase caused by uncaptured and life-cycle CO₂ from CCS processes. The global warming effect of the CCS emissions pulse ceases according to a weighted combination of time constant in the climate system (up to 990 years). At 0.1%/y leakage (middle graph), leakages contributes a peak 0.1°C temperature increase occurring with a delay of a few hundred years after build-up of leakage (compare Fig.1 middle graph). The temperature increase continues as the leakage draws out over a few thousand years. At 1%/y leakage (left graph), leakages leads to a peak 0.4°C temperature increase, thus outweighing CCS processes. The warming effect ceases after about 1000 years because of the rapid leakage (compare Fig.1 left graph).

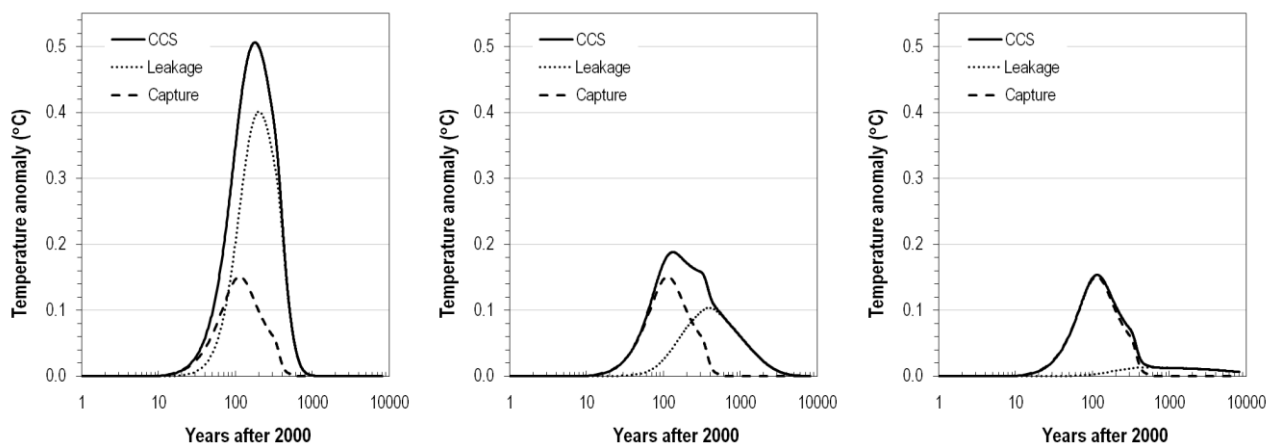


Fig. 2: Global temperature change over time due to CO₂ leakage (dotted), remaining CCS processes (capture, transport, injection, and CCS life-cycle; dashed), and the entire CCS system (solid black, sum of dotted and dashed curves), for a leakage rate of 1%/y (left), 0.1%/y (middle) and 0.01%/y (right).

The interplay of the magnitude and time span of CO₂ leakage from subsurface reservoirs hence leads to an interesting behaviour of the resulting warming effect. At both extremes, the warming effect is relatively short-lived (< 1000 years) because it is dominated either by the CCS and fossil power phase-out ($\lambda = 0.01\%/y$), or by the rapid CO₂ escape ($\lambda = 1\%/y$). At intermediate leakage rates (0.1%), the warming effect extends for a few thousand years. The peak temperature increase decreases with decreasing leakage rates (Fig. 3).

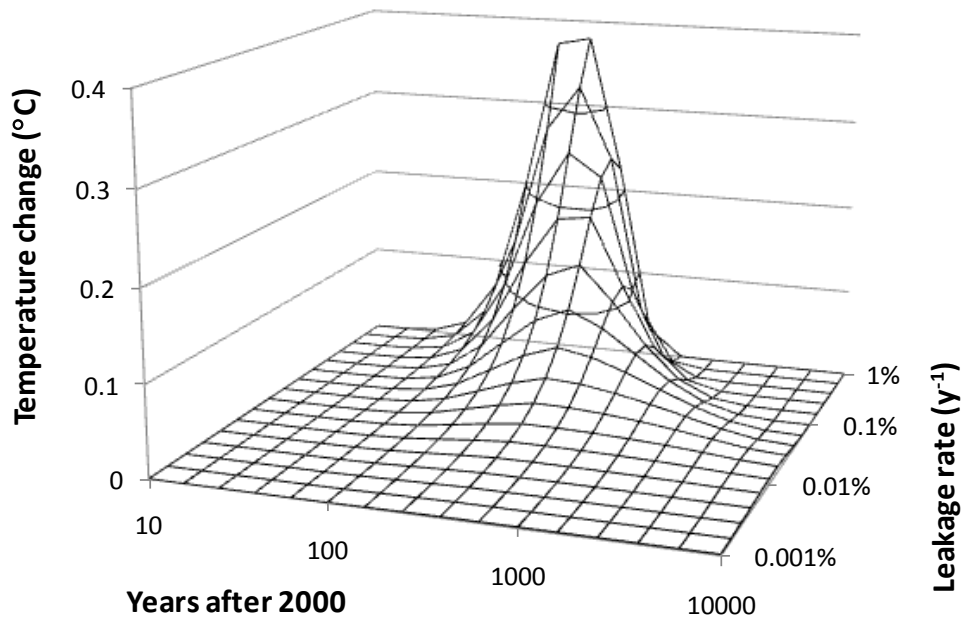


Fig. 3: Temporal profile of global warming due to CO₂ leakage alone.

3.2 Sensitivity to capture rate

With continuing research and development of CCS technology, capture rates may increase in the future. In theory, if all auxiliary and life-cycle processes were operated using zero-carbon technologies (ie, renewable power built using renewable power), capture rates could be close to 100%.

Fig. 4 shows the dependence of the global warming effect due to CO₂ leakage on both capture and leakage rates. The figure depicts on the vertical axis the peak temperature increase (compare Fig. 3). As seen before, low leakage rates lead to negligible temperature increases. For significant leakages rates, the temperature increase is larger at high capture rates, simply because more CO₂ is stored, so that more CO₂ can escape.

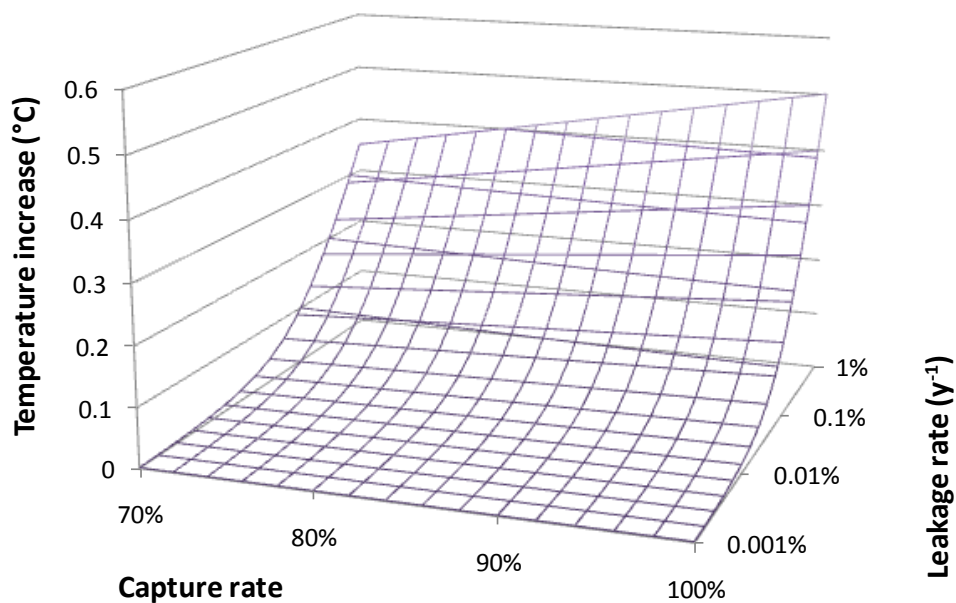


Fig. 4: Dependence of the peak temperature increase due to leakage, on capture and leakage rates.

Fig. 5 shows the dependence of the global warming effect due to all CCS processes and well as CO₂ leakage on both capture and leakage rates. At low leakage rates, the temperature increase is negatively proportional to the capture rate, because CCS processes are the dominant cause for CO₂ emissions. At high leakage rates, the temperature increase is rather independent of the capture rate, which is due to two counteracting effects: On one hand, high capture rates mean low emissions from CCS processes, however on the other hand they mean that more CO₂ is stored thus increasing absolute leakage, if percentage leakage rates are assumed constant.

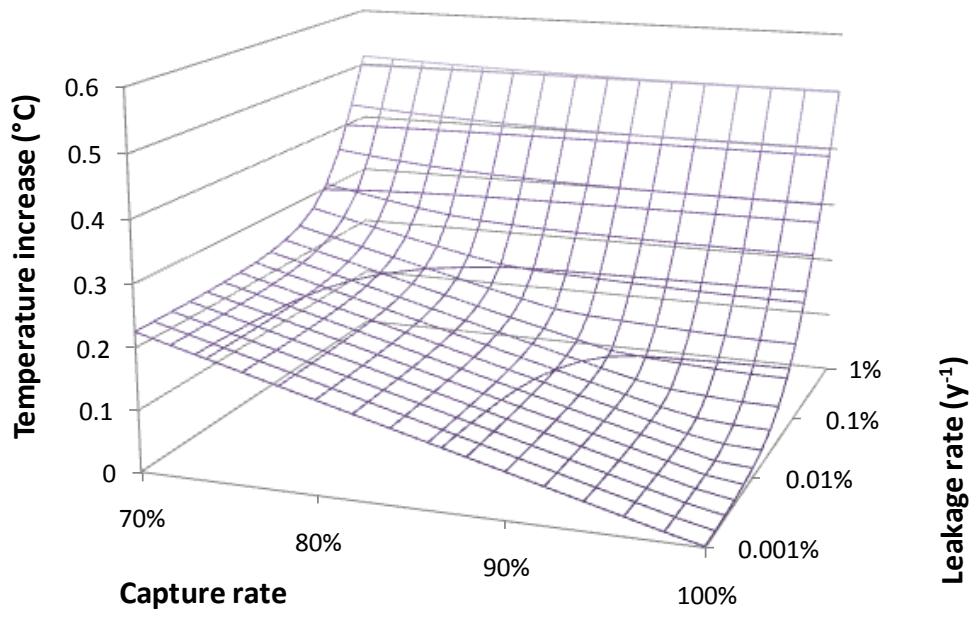


Fig. 5: Dependence of the peak temperature increase due to CCS, on capture and leakage rates.

3.3 Sensitivity to model parameters

As discussed previously, the higher relevance of the temperature increase variable for understanding CO₂ leakage is traded off against higher uncertainty of temperature estimates, compared to emissions. For example, the RBP formulation is only an approximation of the global carbon cycle, and even for this approximation there exist different sets of fractions f and l , and characteristic times τ (Rosa *et al.* 2004). It is therefore necessary to understand the sensitivity of our results with regard to all parameters.

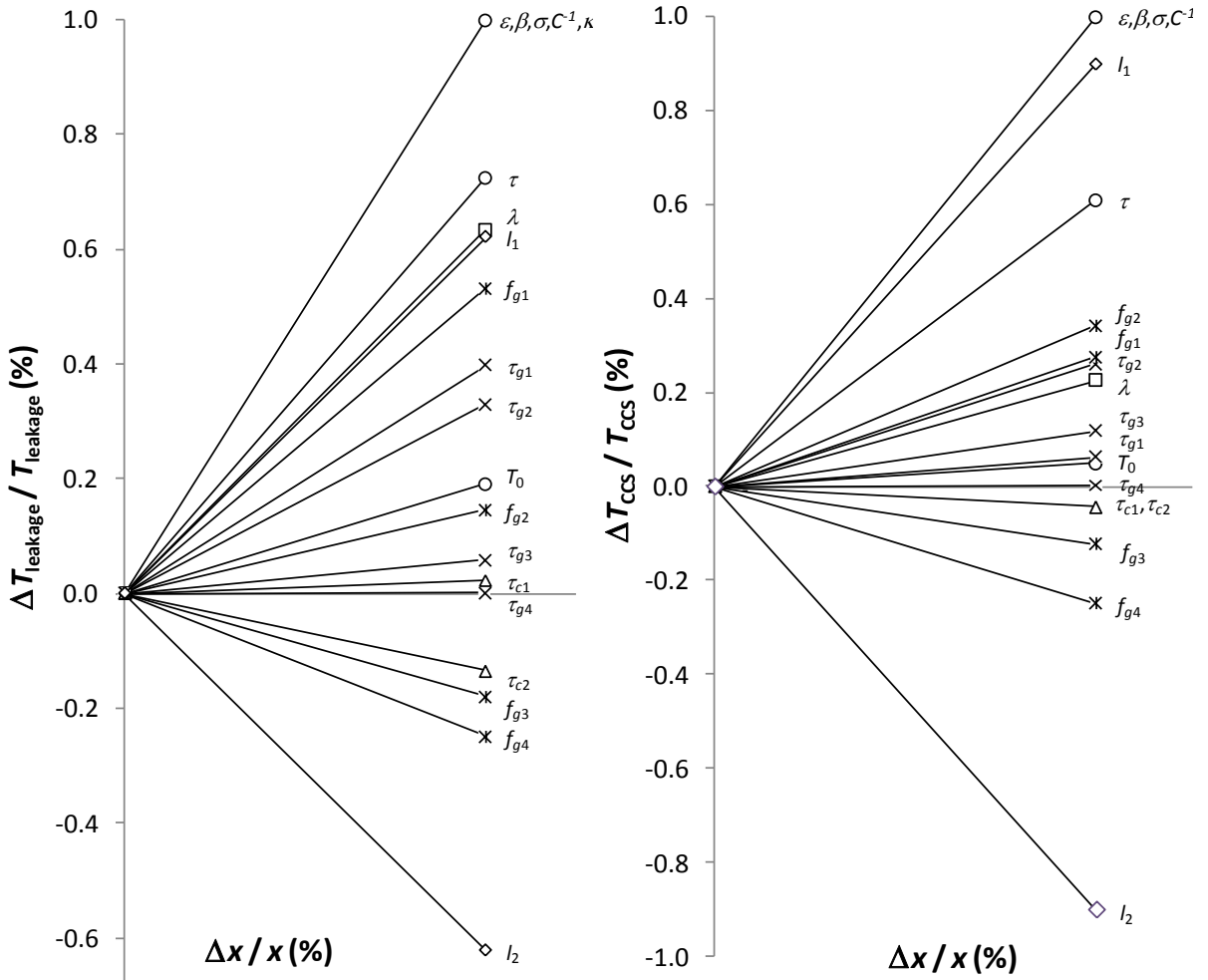


Fig. 6: Sensitivity of peak temperature increases due to CO₂ leakage (left) and CCS including all processes (right) with regard to parameters $x = \{\varepsilon, T_0, \tau, \lambda, \kappa, \beta, \sigma, C^{-1}, f_{gr}, \tau_{gr}, l_s, \tau_{cs}\}$, varied around the base case of $\lambda = 0.01\%$ and $\kappa = 80\%$. All points have horizontal coordinates $\Delta x/x = 1\%$.

First of all, inserting Eqs. 3-5 shows that Eq. 2 is linear in ε , β , σ , and C^{-1} , so that for every 1%-increase in those parameters, the peak temperature increase also increases by 1%. The parameters τ

and T_0 determine the shape of the emissions curve. With increasing characteristic time τ and peak year T_0 , the bell-shaped emissions curve becomes more drawn out, and overlaps more with the CCS application interval [2000,12000], so that in both cases the amount of captured and stored CO₂ slightly increases, and with it the temperature (○ in Fig. 6).

The influence of the capture rate κ has been discussed in Section 3.2. Fig. 6 left corresponds to Fig. 4 in that the leakage is directly proportional to stored CO₂, which is directly proportional to the capture rate. The sensitivity of T_{CCS} with regard to κ is -2.14 and hence beyond the negative y-axis in Fig. 6. This corresponds to Fig. 4 in that more capture means less emissions overall. The influence of the leakage rate λ on peak temperature increases (□ in Fig. 6) is positive, but is obviously restricted to the effect due to CO₂ leakage, and hence a lower sensitivity is recorded for the overall CCS system.

The variables discussed so far mainly relate to assumptions about future emissions profiles determined by future energy demand and CCS technology performance. In the following, the sensitivity if the calculations with respect to climate model parameters is discussed.

The sensitivity of peak temperature increases with regard to the four CO₂ fractions f_{gr} (* in Fig. 6) is determined by the condition that these sum up to 1. The influence of the two dominant fractions f_{g1} and f_{g2} is positive, but increasing the lesser fractions f_{g3} and f_{g4} means that the dominant fractions decrease, hence the negative influence. The same behaviour occurs for the radiative forcing fractions l_1 and l_2 (◇ in Fig. 6), however the effect is much stronger because of the comparatively long characteristic times involved.⁵

The sensitivity of peak temperature increases on characteristic times τ depends on whether only CO₂ leakage is considered or all emissions due to CCS. In the case of CO₂ leakage, the peak temperature increase is always positively affected by CO₂ lifetimes (× in Fig. 6), because longer lifetimes mean more radiative forcing. CO₂ leakage is a long-term process, and so the influence of the long-lived CO₂ fractions $\tau_{g1} = 30$ years and $\tau_{g2} = 80$ years is stronger than that of the short-lived fractions $\tau_{g3} = 20$ years and $\tau_{g4} = 1.6$ years. Once the relatively short-term CCS emissions “bell” is taken into account, those characteristic times τ_{gr} with the higher CO₂ fractions (τ_{g2} and τ_{g3}) have a stronger influence on peak temperature increases.

There are two competing influences of the characteristics times τ_{cs} on peak temperature increases, which are expressed in Eq. 2 by the factors $\frac{1}{\tau_{cs}}$ (negative) and $e^{-\frac{t-t'}{\tau_{cs}}}$ (positive), and hence the overall sensitivities related to these variables are low (Δ in Fig. 6).

⁵ The longest characteristic time in the temperature response functions is 990 years, compared to 330 years for the CO₂ concentration response function (Rosa *et al.* 2004).

4 Discussion and conclusions

Evidently, the transformation of effects from emissions via concentrations and radiative forcing to temperature increase is associated with a higher level of uncertainty (Lenzen 2006). However, at the same time the temperature variable is more relevant to the question about the impacts of CCS for humans, which in a sense reduces uncertainty.

Clearly, the most sensitive climate model parameters in the calculations demonstrated here are the β_g and $\bar{\sigma}_g$ parameters, the heat capacity of the climate system C , and the two fractions of radiative forcing l_1 and l_2 .

Irrespective of the uncertainties involved in the calculations, this work has shown some qualitative relationships between capture rate and leakage rate in their effect on global warming and the temporal behaviour of the global temperature increase. These findings will still hold, no matter the magnitude of the actual effect.

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Appendix: Calibration of the RBP

Equation 2 was parametrised using the RBP values for the fractions f and l and their corresponding lifetimes τ , as listed in Rosa *et al.* 2004 and UNFCCC 2009a, 2009b. The parameters $\bar{\sigma}_g$ and β_g were obtained from Meira 2009. C was calculated according to Equation 28 in Meira and Miguez 2000, using a climate sensitivity of 3°C. The model was then calibrated and fine-tuned (Fig. 2) against historical measurements of atmospheric concentrations (CO₂ Keeling *et al.* 2008, CH₄ Steele *et al.* 2003) and ice core samples (CO₂ Neftel *et al.* 1994, CH₄ Etheridge *et al.* 2002), as well as against historical measurements of global temperature anomalies (Jones *et al.* 2009).

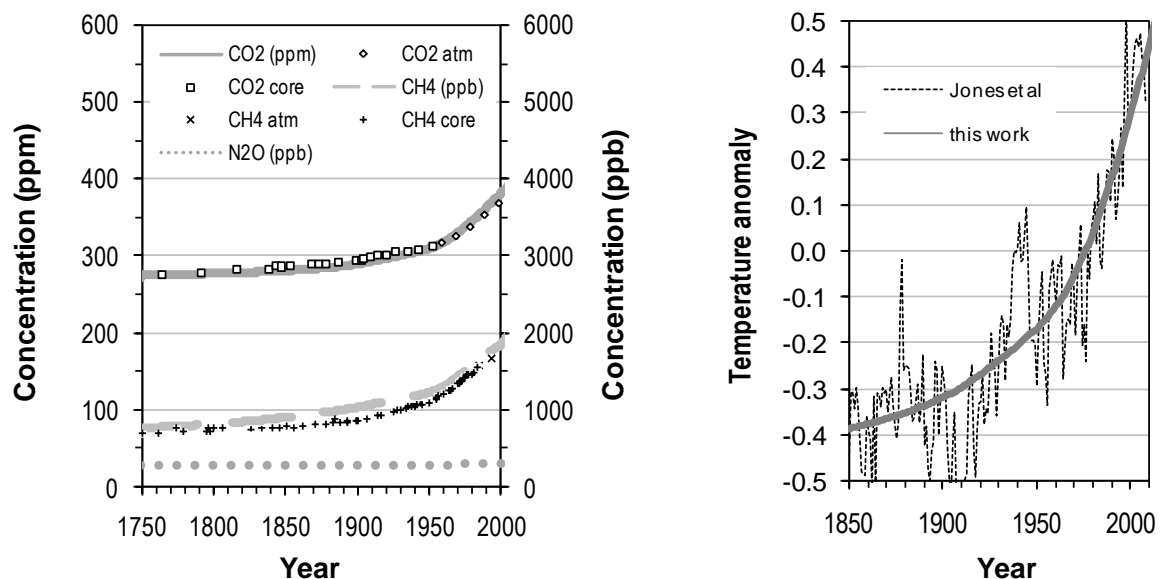


Fig. 2: Calibration of the RBP (grey curves) in terms of atmospheric concentrations of two greenhouse gases (left) and temperature anomaly (right) against measurements (left: markers, right: dashed curve).

Present mean radiative forcing and global warming are a function of greenhouse gas emissions reaching back into the past as far as 300 years. Therefore, the calibration and fine-tuning of the RBP model requires historical emissions data starting 1750. CDIAC data on global CO₂ emissions from fossil fuel usage, cement production and gas flaring between 1750 and 2005 were taken from Marland *et al.* 2008, on CO₂ emissions resulting from land use change between 1850 and 2005 from Houghton 2008, on CH₄ emissions between 1860 and 1994 from Stern and Kaufmann 1998. N₂O emissions between 1890 and 1995 were taken from the EDGAR-HYDE model, documented in Van Aardenne *et al.* 2001. Values prior to these periods were extrapolated using pre-1900 growth rates. These extrapolations are not expected to exert major influence on the results obtained here, since pre-1890 emissions are small compared to post-1890 emissions.