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# Global warming effect of leakage from CO<sub>2</sub> storage



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# Global warming effect of leakage from CO<sub>2</sub> 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 chain<sup>1</sup>, 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 CO<sub>2</sub> storage.

Two CO<sub>2</sub> storage and monitoring projects have injected so far more than 5 Mt CO<sub>2</sub> into a depleted oil field (Canada) and more than 10 Mt CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> concentrations (IEA GHG 2008). However, understanding of leakage and long-term movement of CO<sub>2</sub> 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

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<sup>1</sup> 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 CO<sub>2</sub> emissions in stabilisation scenarios (IPCC 2005a), but not in terms of resulting atmospheric CO<sub>2</sub> concentrations or global warming effects. Other studies (Marland *et al.* 2001; Dutschke 2002) have investigated the policy implications of temporary storage of CO<sub>2</sub> 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 CO<sub>2</sub> 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  $r^{\text{th}}$  of  $R$  fractions of gas  $g$  decaying in the atmosphere with characteristic time  $\tau_{gr}$ , normalised through  $\sum_{r=1}^R f_{gr} = 1$ ,
- $\beta_g$  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  $s^{\text{th}}$  of  $S$  fractions of radiative forcing that adjusts with characteristic time  $\tau_{cs}$ , 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 CO<sub>2</sub> leakage from geological storage sites, and also of residual CO<sub>2</sub> from the CCS process, the latter for example from recycling of solvents, compression, transport, etc. Assume that CO<sub>2</sub> emissions resulting from CCS ( $i = 1$ ) and CO<sub>2</sub> 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 CO<sub>2</sub> 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,  $\varepsilon$  emissions at year  $T_0$ , and  $\tau$  the time constant of emissions change.  $T_0 = 2050$  and  $\varepsilon = 25$  Gt CO<sub>2</sub> 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.<sup>2</sup> Note that any other scenario would be equally suitable to demonstrate the effect of leakage from CO<sub>2</sub> 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  $\kappa$ :

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

The capture rate  $\kappa$  in Equation 4 includes all life-cycle emissions arising out of the CCS process.<sup>3</sup> 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 CO<sub>2</sub> transport and injection.

Odeh and Cockerill 2008 review studies of life-cycle energy requirements and CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> emissions of 255 and 200 g/kWh, respectively. Pehnt and Henkel

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<sup>2</sup> According to the IEA 2008, fossil-fuel power plants emitted 11.4 Gt of CO<sub>2</sub> in 2006, 41% of the world total.

<sup>3</sup> 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%.<sup>4</sup>

Capturing and storing 80% of CO<sub>2</sub> emitted according to Equation 3 would result in about 1600 Gt CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub>, 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  $\lambda$  to cumulatively stored CO<sub>2</sub>:

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

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

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<sup>4</sup> 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> (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 CO<sub>2</sub> (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 CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> completely escapes in less than 1,000 years.

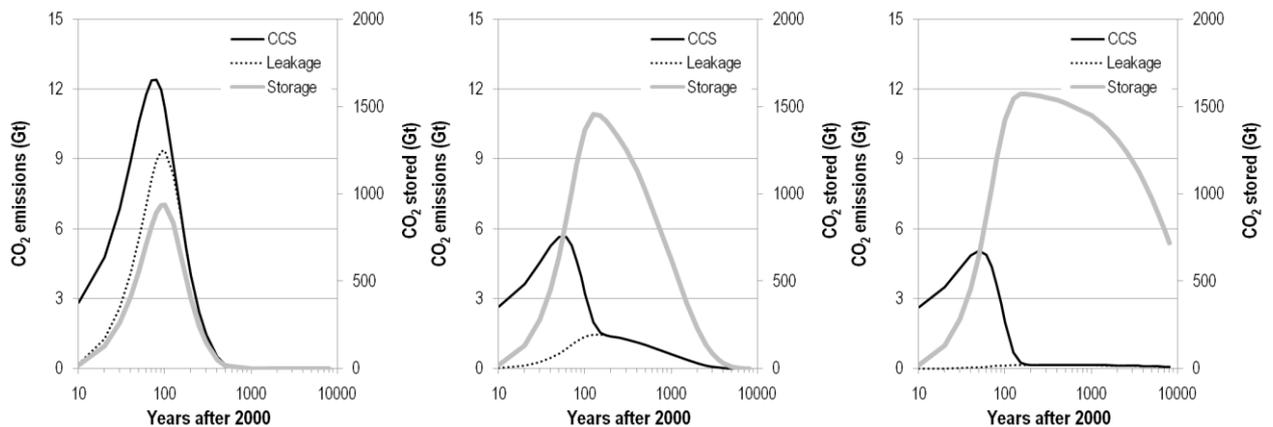


Fig. 1: Emissions over time from CO<sub>2</sub> leakage (dotted), the entire CCS system (solid black), as well as CO<sub>2</sub> stored subsurface (solid grey), for a leakage rate of 1%/y (left), 0.1%/y (middle) and 0.01%/y (right). The area between the dotted and solid black curves represents CO<sub>2</sub> 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 CO<sub>2</sub> 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).

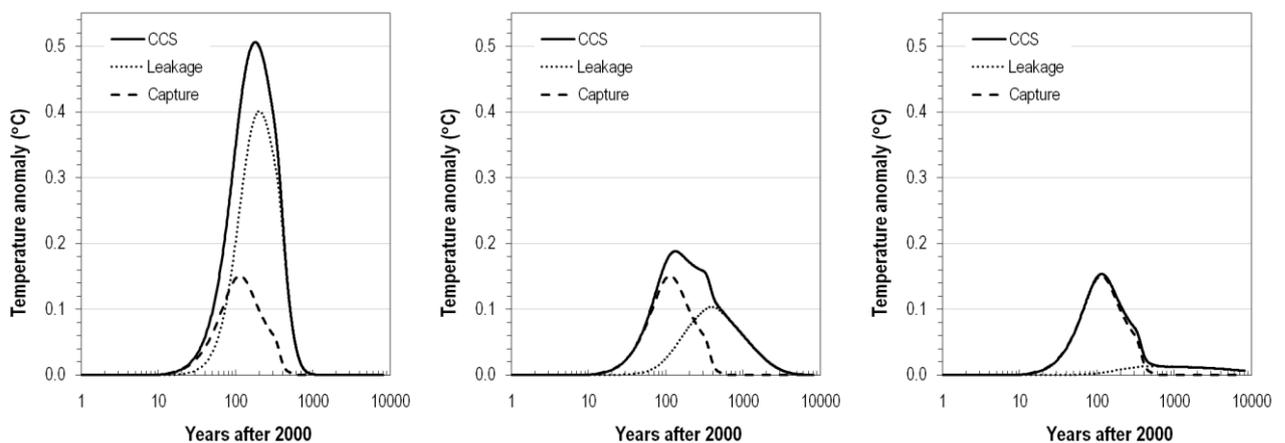


Fig. 2: Global temperature change over time due to CO<sub>2</sub> leakage (dotted), remaining CCS processes (capture, transport, injection, and CCS life-cycle; dashed), and the entire CCS system (solid black, sum of dotted and dashed curves), for a leakage rate of 1%/y (left), 0.1%/y (middle) and 0.01%/y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

The interplay of the magnitude and time span of CO<sub>2</sub> 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 CO<sub>2</sub> 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).

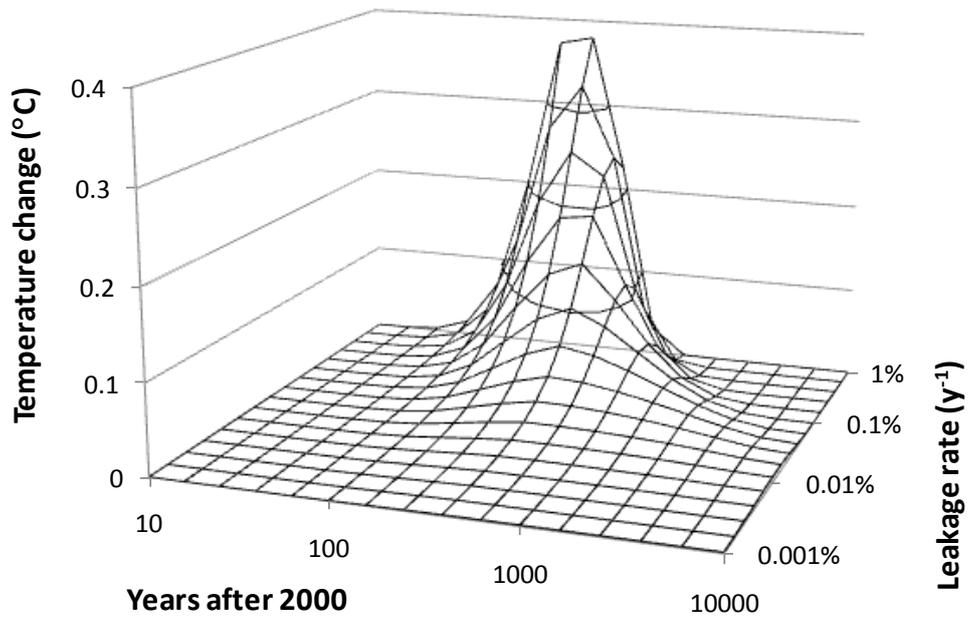


Fig. 3: Temporal profile of global warming due to CO<sub>2</sub> leakage alone.

### 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 CO<sub>2</sub> 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 CO<sub>2</sub> is stored, so that more CO<sub>2</sub> can escape.

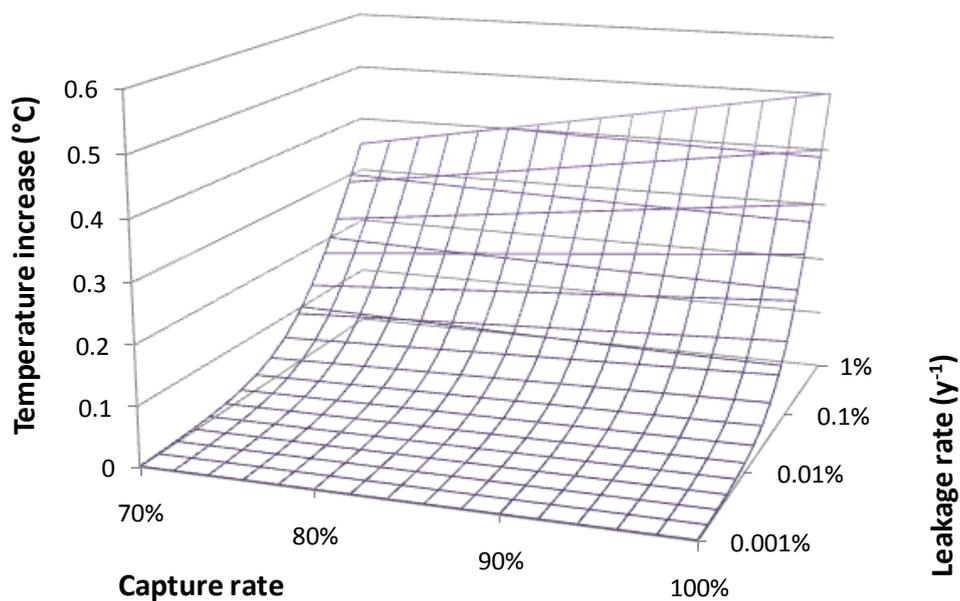


Fig. 4: Dependence of the peak temperature increase due to leakage, on capture and leakage rates.

Fig. 5 shows the dependence of the global warming effect due to all CCS processes and well as CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> is stored thus increasing absolute leakage, if percentage leakage rates are assumed constant.

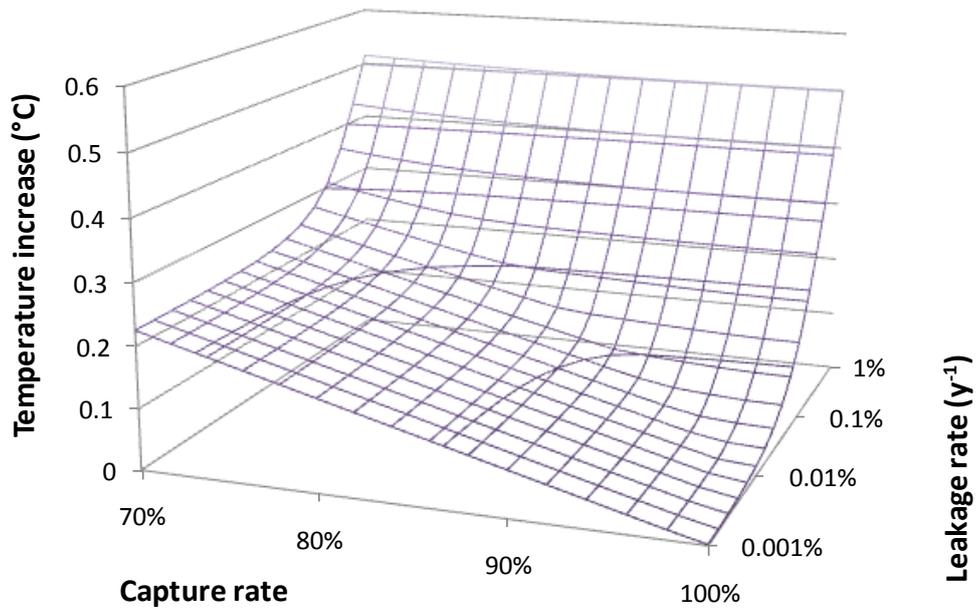


Fig. 5: Dependence of the peak temperature increase due to CCS, on capture and leakage rates.

### 3.3 Sensitivity to model parameters

As discussed previously, the higher relevance of the temperature increase variable for understanding CO<sub>2</sub> 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  $\tau$  (Rosa *et al.* 2004). It is therefore necessary to understand the sensitivity of our results with regard to all parameters.

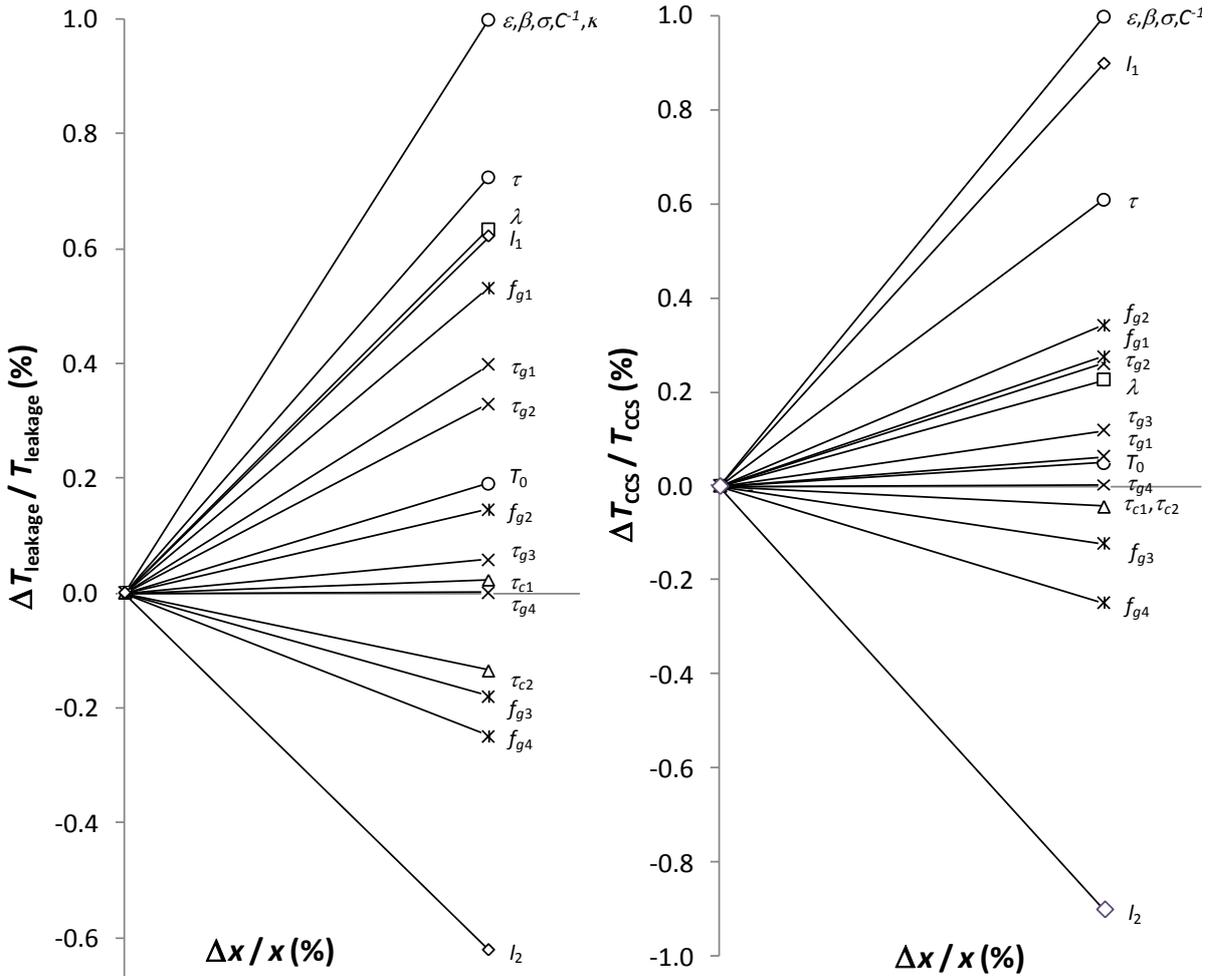


Fig. 6: Sensitivity of peak temperature increases due to CO<sub>2</sub> leakage (left) and CCS including all processes (right) with regard to parameters  $x = \{\varepsilon, T_0, \tau, \lambda, \kappa, \beta, \sigma, C^{-1}, f_{gr}, \tau_{gr}, l_s, \tau_{cs}\}$ , varied around the base case of  $\lambda = 0.01\%$  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  $\varepsilon$ ,  $\beta$ ,  $\sigma$ , and  $C^{-1}$ , so that for every 1%-increase in those parameters, the peak temperature increase also increases by 1%. The parameters  $\tau$

and  $T_0$  determine the shape of the emissions curve. With increasing characteristic time  $\tau$  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 CO<sub>2</sub> slightly increases, and with it the temperature (○ in Fig. 6).

The influence of the capture rate  $\kappa$  has been discussed in Section 3.2. Fig. 6 left corresponds to Fig. 4 in that the leakage is directly proportional to stored CO<sub>2</sub>, which is directly proportional to the capture rate. The sensitivity of  $T_{CCS}$  with regard to  $\kappa$  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  $\lambda$  on peak temperature increases (□ in Fig. 6) is positive, but is obviously restricted to the effect due to CO<sub>2</sub> 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 CO<sub>2</sub> 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.<sup>5</sup>

The sensitivity of peak temperature increases on characteristic times  $\tau$  depends on whether only CO<sub>2</sub> leakage is considered or all emissions due to CCS. In the case of CO<sub>2</sub> leakage, the peak temperature increase is always positively affected by CO<sub>2</sub> lifetimes (× in Fig. 6), because longer lifetimes mean more radiative forcing. CO<sub>2</sub> leakage is a long-term process, and so the influence of the long-lived CO<sub>2</sub> 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  $\tau_{gr}$  with the higher CO<sub>2</sub> fractions  $f_{gr}$  ( $\tau_{g2}$  and  $\tau_{g3}$ ) have a stronger influence on peak temperature increases.

There are two competing influences of the characteristics times  $\tau_{cs}$  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).

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<sup>5</sup> The longest characteristic time in the temperature response functions is 990 years, compared to 330 years for the CO<sub>2</sub> 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  $\beta_g$  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.

## References

- Bare, J. C., P. Hofstetter, D. W. Pennington, and H. A. Udo de Haes. 2000. Life cycle impact assessment workshop summary. Midpoints versus endpoints: the sacrifice and benefits. *International Journal of Life Cycle Assessment* 5(6): 319-326.
- Davison, J. 2007. Performance and costs of power plants with capture and storage of CO<sub>2</sub>. *Energy* 32(7): 1163-1176.
- Den Elzen, M., M. Berk, M. Shaeffer, J. Olivier, C. Hendriks, and B. Metz. 1999. *The Brazilian Proposal and other options for international burden sharing: an evaluation of methodological and policy aspects using the FAIR model*. 728001011. Bilthoven, Netherlands: National Institute of Public Health and the Environment.
- Diesendorf, M. 2006. Can geosequestration save the coal industry? In *Transforming Power: Energy as a Social Project*, edited by J. Byrne, et al.: Energy and Environmental Policy Series.
- Dutschke, M. 2002. Fractions of permanence - squaring the cycle of sink carbon accounting. *Mitigation and Adaptation Strategies for Global Change* 7: 381-402.
- Etheridge, D. M., L. P. Steele, R. J. Francey, and R. L. Langenfelds. 2002. Historical CH<sub>4</sub> records since about 1000 A.D. from ice core data. In *Trends: A Compendium of Data on Global Change*. Oak Ridge, Tenn., U.S.A.: Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy.
- Federative Republic of Brazil. 1997. *Proposed elements of a protocol to the United Nations Framework Conventions on Climate Change, presented by Brazil in response to the Berlin Mandate*. FCCC/AGBM/1997/MISC.1/Add.3, Internet site <http://www.mct.gov.br/clima/ingles/quioto/propbra.htm>. Brasília, Brazil:
- Heijungs, R., M. J. Goedkoop, J. Struijs, S. Effting, M. Sevenster, and G. Huppes. 2003. *Towards a life cycle impact assessment method which comprises category indicators at the midpoint and the endpoint level*. <http://www.pre.nl/download/Recipe%20phase1%20final.pdf>. Amersfoort, Netherlands: PRé Consultants.
- Hertwich, E. G. and J. K. Hammitt. 2001. Decision-analytic framework for impact assessment, Part 2: Midpoints, endpoints and criteria for method development. *International Journal of Life Cycle Assessment* 6: 265-272.
- Hertwich, E. G., M. Aaberg, B. Singh, and A. H. Strømman. 2008. Life-cycle Assessment of carbon dioxide capture for enhanced oil recovery. *Chinese Journal of Chemical Engineering* 16(3): 343-353.
- Houghton, R. A. 2008. Carbon flux to the atmosphere from land-use changes: 1850-2005. In *Trends: A Compendium of Data on Global Change*. Oak Ridge, Tenn., U.S.A.: Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy.
- IEA. 2008. *World Energy Outlook 2008*. Paris, France: International Energy Agency.
- IEA GHG. 2008. *Geologic Storage of Carbon Dioxide - Staying Safely Underground*. [www.ieagreen.org.uk/glossies/geostoragesfty.pdf](http://www.ieagreen.org.uk/glossies/geostoragesfty.pdf). Stoke Orchard, Cheltenham, UK: International Energy Agency Greenhouse Gas R&D Programme.
- IPCC. 2005a. *Technical Summary*. Carbon Dioxide Capture and Storage. Geneva, Switzerland: Intergovernmental Panel on Climate Change.
- IPCC. 2005b. *Summary for Policymakers*. Carbon Dioxide Capture and Storage. Geneva, Switzerland: Intergovernmental Panel on Climate Change.
- Jones, P. D., D. E. Parker, T. J. Osborn, and K. R. Briffa. 2009. Global and hemispheric temperature anomalies--land and marine instrumental records. In *Trends: A Compendium of Data on Global Change*. Oak Ridge, Tenn., U.S.A.: Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy.
- Keeling, R. F., S. C. Piper, A. F. Bollenbacher, and J. S. Walker. 2008. Atmospheric CO<sub>2</sub> records from sites in the SIO air sampling network. . In *Trends: A Compendium of Data on Global Change*. Oak Ridge, Tenn., U.S.A.: Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy.

- Krewitt, W. 2002. External cost of energy - do the answers match the questions? Looking back at 10 years of ExternE. *Energy Policy* 30: 839-848.
- Lenzen, M. 2006. Uncertainty of end-point impact measures: implications for decision-making. *International Journal of Life-Cycle Assessment* 11(3): 189-199.
- Lenzen, M. and H. J. Neugebauer. 1999. Measurements of radon concentration and the role of earth tides in a gypsum mine in Walferdange, Luxembourg. *Health Physics* 77(2): 154-162.
- Marland, G., K. Fruit, and R. Sedjo. 2001. Accounting for sequestered carbon: the question of permanence. *Environmental Science & Policy* 4(6): 259-268.
- Marland, G., T. A. Boden, and R. J. Andres. 2008. Global, Regional, and National CO<sub>2</sub> Emissions. In *Trends: A Compendium of Data on Global Change*. Oak Ridge, Tenn., U.S.A.: Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy.
- Meira, L. G. 2009. Personal Communication with Meira, L. G., Consulta sobre o valor de alguns parâmetros. 30 March 2009.
- Meira, L. G. and J. D. G. Miguez. 2000. *Note on the time-dependent relationship between emissions of greenhouse gases and climate change*. <http://www.mct.gov.br/clima/ingles/quioto/propbra.htm>. Brasília, Brazil: Ministry of Science and Technology.
- Nakicenovic, N. and R. Swart, eds. 2000. *Special Report on Emissions Scenarios*. Geneva, Switzerland: Intergovernmental Panel on Climate Change.
- Neftel, A., H. Friedli, E. Moor, H. Lötscher, H. Oeschger, U. Siegenthaler, and S. B. 1994. Historical CO<sub>2</sub> record from the Siple Station ice core. In *Trends: A Compendium of Data on Global Change*. Oak Ridge, Tenn., U.S.A.: Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy.
- Odeh, N. A. and T. T. Cockerill. 2008. Life cycle GHG assessment of fossil fuel power plants with carbon capture and storage. *Energy Policy* 36(1): 367-380.
- Pehnt, M. and J. Henkel. 2009. Life cycle assessment of carbon dioxide capture and storage from lignite power plants. *International Journal of Greenhouse Gas Control* 3(1): 49-66.
- Riahi, K., L. Barreto, S. Rao, and E. S. Rubin. 2005. Towards fossil-based electricity systems with integrated CO<sub>2</sub> capture: Implications of an illustrative long-term technology policy. In *Greenhouse Gas Control Technologies*. Oxford: Elsevier Science Ltd.
- Rosa, L. P., S. K. Ribeiro, M. S. Muylaert, and C. P. de Campos. 2004. Comments on the Brazilian Proposal and contributions to global temperature increase with different climate responses - CO<sub>2</sub> emissions due to fossil fuels, CO<sub>2</sub> emissions due to land use changes. *Energy Policy* 32(13): 1499-1510.
- Rubin, E. S., C. Chen, and A. B. Rao. 2007. Cost and performance of fossil fuel power plants with CO<sub>2</sub> capture and storage. *Energy Policy* 35(9): 4444-4454.
- Shackley, S., H. Waterman, P. Godfroij, D. Reiner, J. Anderson, K. Draxlbauer, and T. Flach. 2007. Stakeholder perceptions of CO<sub>2</sub> capture and storage in Europe: Results from a survey. *Energy Policy* 35(10): 5091-5108.
- Spath, P. L. and M. Mann. 2004. *Biomass power and conventional fossil systems with and without CO<sub>2</sub> sequestration - comparing the energy balance, greenhouse gas emissions and economics*. TP-510-32575. Golden, USA: US National Renewable Energy Laboratory.
- Steele, L. P., P. B. Krummel, and R. L. Langenfelds. 2003. Atmospheric CH<sub>4</sub> concentrations from sites in the CSIRO Atmospheric Research GASLAB air sampling network (October 2002 version). In *Trends: A Compendium of Data on Global Change*. Oak Ridge, Tenn., U.S.A.: Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy.
- Stern, D. I. and R. K. Kaufmann. 1998. Annual estimates of global anthropogenic methane emissions: 1860-1994. In *Trends: A Compendium of Data on Global Change*. Oak Ridge, Tenn., U.S.A.: Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy.

- Udo de Haes, H. A., O. Jolliet, G. Finnveden, M. Hauschild, W. Krewitt, and R. Mueller-Wenk. 1999. Best available practice regarding impact categories and category indicators in life cycle impact assessment. *International Journal of Life Cycle Assessment* 4(2): 1-15.
- UNFCCC. 2009a. *Parameters for tuning a simple carbon cycle model*. <http://unfccc.int/resource/brazil/carbon.html>. Bonn, Germany: United Nations Framework Convention on Climate Change.
- UNFCCC. 2009b. *Parameters for tuning a simple climate model (plus aerosol forcing)*. <http://unfccc.int/resource/brazil/climate.html>. Bonn, Germany: United Nations Framework Convention on Climate Change.
- Van Aardenne, J. A., F. J. Dentener, J. G. J. Olivier, C. G. M. Klein Goldewijk, and J. Lelieveld. 2001. A high resolution dataset of historical anthropogenic trace gas emissions for the period 1890-1990. *Global Biogeochemical Cycles* 15(4): 909-928.
- Viebahn, P., J. Nitsch, M. Fishedick, A. Esken, D. Schüwer, N. Supersberger, U. Zuberbühler, and O. Edenhofer. 2007. Comparison of carbon capture and storage with renewable energy technologies regarding structural, economic, and ecological aspects in Germany. *International Journal of Greenhouse Gas Control* 1(1): 121-133.

## Appendix: Calibration of the RBP

Equation 2 was parametrised using the RBP values for the fractions  $f$  and  $l$  and their corresponding lifetimes  $\tau$ , as listed in Rosa *et al.* 2004 and UNFCCC 2009a, 2009b. The parameters  $\bar{\sigma}_g$  and  $\beta_g$  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 (CO<sub>2</sub> Keeling *et al.* 2008, CH<sub>4</sub> Steele *et al.* 2003) and ice core samples (CO<sub>2</sub> Neftel *et al.* 1994, CH<sub>4</sub> Etheridge *et al.* 2002), as well as against historical measurements of global temperature anomalies (Jones *et al.* 2009).

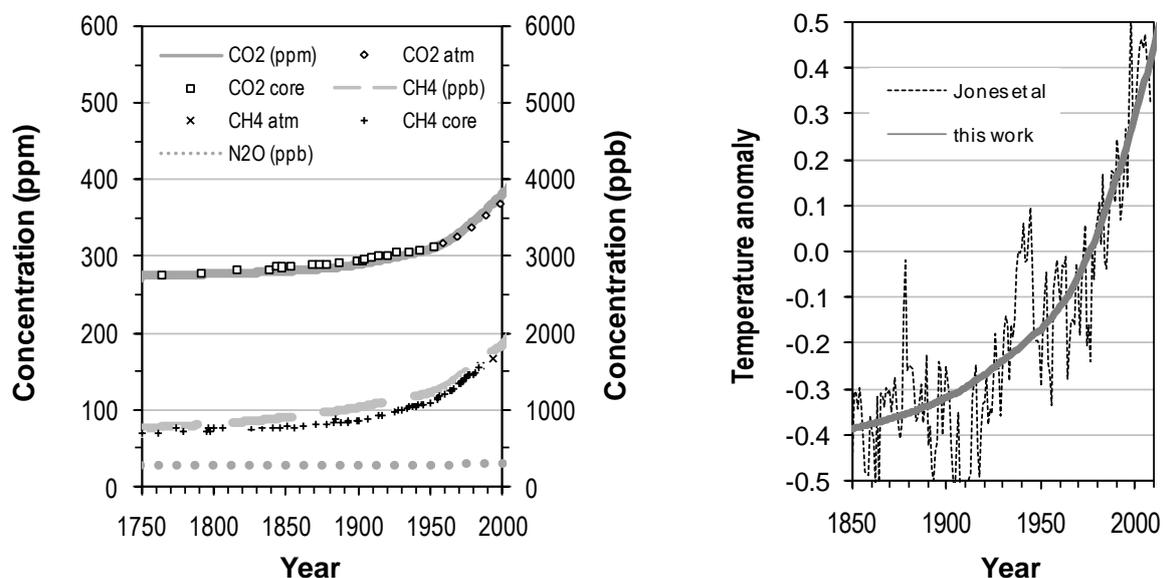


Fig. 2: Calibration of the RBP (grey curves) in terms of atmospheric concentrations of two greenhouse gases (left) and temperature anomaly (right) against measurements (left: markers, right: dashed curve).

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 CO<sub>2</sub> emissions from fossil fuel usage, cement production and gas flaring between 1750 and 2005 were taken from Marland *et al.* 2008, on CO<sub>2</sub> emissions resulting from land use change between 1850 and 2005 from Houghton 2008, on CH<sub>4</sub> emissions between 1860 and 1994 from Stern and Kaufmann 1998. N<sub>2</sub>O 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.