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### Review

# The environmental impact and risk assessment of $CO_2$ capture, transport and storage – An evaluation of the knowledge base

Joris Koornneef<sup>a,\*</sup>, Andrea Ramírez<sup>b</sup>, Wim Turkenburg<sup>b</sup>, André Faaij<sup>b</sup>

<sup>a</sup> Ecofys Netherlands B.V., Kanaalweg 16-A, 3526 KL Utrecht, The Netherlands <sup>b</sup> Copernicus Institute, Faculty of Science, Utrecht University, Budapestlaan 6, 3584CD Utrecht, The Netherlands

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#### ABSTRACT

In this study, we identify and characterize known and new environmental consequences associated with CO<sub>2</sub> capture from power plants, transport by pipeline and storage in geological formations. We have reviewed (analogous) environmental impact assessment procedures and scientific literature on carbon capture and storage (CCS) options. Analogues include the construction of new power plants, transport of natural gas by pipelines, underground natural gas storage (UGS), natural gas production and enhanced oil recovery (EOR) projects. It is investigated whether crucial knowledge on environmental impacts is lacking that may postpone the implementation of CCS projects. This review shows that the capture of  $CO_2$  from power plants results in a change in the environmental profile of the power plant. This change encompasses both increase and reduction of key atmospheric emissions, being: NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub>, particulate matter, Hg, HF and HCl. The largest trade-offs are found for the emission of NO<sub>x</sub> and NH<sub>3</sub> when equipping power plants with post-combustion capture. Synergy is expected for  $SO_2$  emissions, which are low for all power plants with CO<sub>2</sub> capture. An increase in water consumption ranging between 32% and 93% and an increase in waste and by-product creation with tens of kilotonnes annually is expected for a large-scale power plant (1 GWe), but exact flows and composition are uncertain. The cross-media effects of CO<sub>2</sub> capture are found to be uncertain and to a large extent not quantified. For the assessment of the safety of CO<sub>2</sub> transport by pipeline at high pressure an important knowledge gap is the absence of validated release and dispersion models for CO<sub>2</sub> releases. We also highlight factors that result in some (not major) uncertainties when estimating the failure rates for CO<sub>2</sub> pipelines. Furthermore, uniform CO<sub>2</sub> exposure thresholds, detailed dose-response models and specific CO<sub>2</sub> pipeline regulation are absent. Most gaps in environmental information regarding the CCS chain are identified and characterized for the risk assessment of the underground, non-engineered, part of the storage activity. This uncertainty is considered to be larger for aquifers than for hydrocarbon reservoirs. Failure rates are found to be heavily based on expert opinions and the dose-response models for ecosystems or target species are not yet developed. Integration and validation of various sub-models describing fate and transport of  $CO_2$  in various compartments of the geosphere is at an infant stage. In conclusion, it is not possible to execute a quantitative risk assessment for the non-engineered part of the storage activity with high confidence.

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\* Corresponding author. Tel.: +31 30 66 23 396; fax: +31 30 66 23 301. *E-mail address:* j.koornneef@ecofys.com (J. Koornneef).

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#### 1. Introduction

The capture, transport and storage of  $CO_2$  (CCS) is currently being researched as a promising approach that may help to reduce anthropogenic  $CO_2$  emissions. The basic idea is that  $CO_2$  is captured, mainly from point sources in the industry and power sector, compressed, transported and injected in deep underground formations.

Several permits are required to realize CCS projects. Following the EU CCS Directive [1], commercial CO<sub>2</sub> capture, transport and storage activities *are highly likely to be subjected to an obligatory* Environmental Impact Assessment (EIA) to acquire these permits. The EIA is a procedural tool with the main goal to assess the environmental impacts of a proposed project. It is used to include environmental criteria into the decision making process for that project.

A complementary tool is the Strategic Environmental Assessment (SEA). This tool is used to facilitate policy decisions on a strategic level. Strategic policy plans are obligated to include a SEA when they contain the consideration or appointment of possible locations or routes of EIA obligated activities. Such considerations or appointments are typically the subject of spatial plans formulated by national, regional or local governments [2-4].

According to Finnveden et al. [3], both environmental assessments can be characterized by three elements: institutional arrangements, the procedure and applied methods. A fourth element would be the environmental impacts assessed in the procedure, i.e. the content of the environmental report or Environmental Impact Statement (EIS). In recent literature, increasingly attention has been given to the role of EIA and SEA procedures in the implementation of CCS activities. Zakkour and Haines [5] identify Environmental Impact Assessment (EIA) as a key process in regulatory and permitting procedures and attribute a role for SEA procedures to contribute to the formulation of CCS deployment policy. Mace et al. [6] identify EIA and SEA requirements in the implementation of CCS activities as a regulatory challenge to policymakers. In Koornneef et al. [2] parts of this challenge for administrative bodies and project initiators regarding the institutional arrangements and procedural elements of both assessments have been addressed [2]. There, the focus was aimed towards the identification of the scope of both procedures, yielding insight in the operational, technical, location and strategic alternatives that should be investigated in the assessments. No detailed attention was paid to the environmental impacts to be investigated in the assessments.

The challenge remains to take the existing assessment frameworks that are used in analogous EIAs and apply them on CCS activities. This includes the possibility to use existing tools to investigate the environmental consequences of CCS activities. Recently, this issue has also been addressed in a IEA GHG<sup>1</sup>

 $<sup>^{-1}</sup>$  IEA GHG = Greenhouse Gas R&D programme of the International Energy Agency.

Nomeno	clature	kWh MDEA	kilowatt-hour Methyldiethanolamine
ASU	Air Separation Unit	MEA	Monoethanolamine
BAT	Best Available Technology	MHI	Mitsubishi Heavy Industries
			5
BREF	Best Available Technology Reference Documents	MJ	Megajoule
CCS	Carbon Dioxide Capture (Transport) and Storage	Mt	Megatonne
DeNO <sub>x</sub>	Installation to remove $NO_x$ from flue gases	NGC	Natural Gas Cycle
EIA	Environmental Impact Assessment	NGCC	Natural Gas Combined Cycle
EIS	Environmental Impact Statement	PC	Pulverized Coal
EOR	Enhanced Oil Recovery	PM	Particulate Matter
ESP	Electrostatic Precipitator	ppm	parts per million
FEP	Feature, Event, Process	(Q)RA	(Quantitative) Risk Assessment
FGD	Flue Gas Desulphurization	S(N)CR	Selective (Non) Catalytic reduction
FGR	Flue Gas Recycling	SCR	Selective Catalytic Reduction
GC	Gas cycle	SEA	Strategic Environmental Assessment
Gt	Gigatonne	SOFC (+	GT) Solid Oxide Fuel Cell (+Gas Turbine)
HSE	Health, Safety and Environment	t	Tonne
HSS	Heat Stable Salt	UGS	Underground Gas Storage
IEA	International Energy Agency	VOC	Volatile Organic Compounds
IGCC	Integrated Gasification Combined Cycle	WGS	Water Gas Shift
IPPC	Integrated Pollution Prevention and Control (Bureau/	WWT	Waste Water Treatment
	Directive)	vr	Year
kt	Kilotonne	-	

programme study which was, next to reviewing international procedural EIA frameworks, oriented towards the identification of information requirements and possible knowledge gaps on environmental consequences when these frameworks are applied to CCS activities [7]. The results of that study indicate the presence of gaps in environmental guidelines, standards and knowledge required for the execution of environmental assessments. The study concludes that additional knowledge is required on:

- The environmental performance of large-scale CO<sub>2</sub> capture systems;
- The modeling of the dispersion of supercritical CO<sub>2</sub> releases;
- The probability, size and environmental consequence of CO<sub>2</sub> leakages resulting from CO<sub>2</sub> storage.

Especially the latter turns out to be a primary concern in the public debate about an onshore  $CO_2$  storage project, the Barendrecht project, in a small depleted gas field in the Netherlands [8]. This project has been cancelled and the results of the EIA turned out to be of very high importance for the (local) governmental bodies involved in the decision making process for that project. The environmental consequences and the way they are assessed and presented in an EIA procedure may be a pivot in the further deployment of CCS, especially when storage takes place onshore.

Information in environmental assessments is often captured in the form of environmental indicators. Such indicators can be used to report on complex phenomena in a simple form that in turn can be used in decision making [9]. In this study, specific attention is paid to quantified indicators that are or may be used to report on the environmental consequences of CCS activities.

The goal of this study is to identify and characterize known and new environmental interventions associated with CCS activities that are typically addressed in EIA procedures. We screen state-ofthe-art literature on available and missing quantitative information (and indicators) on environmental impacts and risks of CCS activities. In addition, it will be investigated whether crucial environmental information is lacking that may postpone the implementation of CCS projects. Specific emphasis is put on knowledge that should be available if CCS is to be implemented on a large-scale in the short-term. This focuses this study towards technologies that are available at present or in the near future.

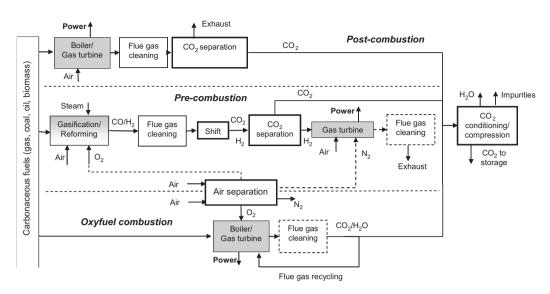
#### 2. Approach and research method

In order to fulfill the goal of this study we carried out a review of documents related to analogous EIA procedures and EIA procedures for CCS activities as well as scientific literature on CO<sub>2</sub> capture, transport and storage. Analogous EIA procedures were reviewed for three distinctive process steps of a CCS project: the power plant with capture, the transport and finally the underground storage of CO<sub>2</sub>. The selected analogues include the construction of new power plants, transport of natural gas by pipelines, underground natural gas storage (UGS), natural gas production and enhanced oil recovery (EOR) projects. For a comprehensive list of the reviewed EIA procedures see [2,10] and the supplementary material provided online. In addition, EIA procedures for CO<sub>2</sub> storage projects were reviewed.

With this information, the following research steps were carried out:

- Identify and characterize quantitative environmental indicators reported in EIA documents for CCS and analogous activities;
- Discuss new environmental information, possible indicators and assessment tools for CCS activities;

The results of these research steps are presented in the following sections. The structure of the article is as follows. In Section 3, 4 and 5 we assess the environmental information on power plants equipped with (and without)  $CO_2$  capture, its transport by pipelines and its storage in geological formations, respectively. In Section 6 we compare the significant risks of CCS activities. In Section 7 we summarize our main findings and we conclude with several recommendations for further research and regulatory efforts.



**Fig. 1.** Simplified overview of the three CO<sub>2</sub> capture systems for power plants: post-, pre- and oxyfuel combustion. Grey components indicate power generation processes. Components with highlighted borders indicate processes causing a drop in generating efficiency. Components with dashed borders indicate optional processes. Note that natural gas reforming using steam is an endothermic process and therefore not a power generation process, hence the altered shading.

#### 3. Power plants with CO<sub>2</sub> capture

When assessing the environmental consequences of a power plant it is important to consider the construction, operational and decommissioning phase. The reviewed EIAs focus on the operational phase of the power plant, see [11–16]. Earlier studies have indicated that environmental impacts that can be attributed to the infrastructure of CCS projects are limited compared to the impacts attributable to the operational phase [17–19]. Here we focus on the operational phase and the environmental themes that are expected to be affected the most by equipping power plants with CO<sub>2</sub> capture, being: energy, atmosphere, water, waste and by-products. resource consumption and external safety. This information is needed on the short term to allow the permitting of CCS projects. We thus focus on possible environmental impacts of CO<sub>2</sub> capture technologies that may be implemented at power plants in the nearterm and compare the performance of power plants equipped with CO<sub>2</sub> capture with reference power plants without CO<sub>2</sub> capture. We take into account the three main capture systems for the removal of CO<sub>2</sub> depicted in Fig. 1: post-combustion, pre-combustion and oxyfuel combustion.

Post-combustion  $CO_2$  capture encompasses the removal of  $CO_2$  from the flue gas of a combustion process. This can be (pressurized) combustion in a boiler or gas turbine.  $CO_2$  is removed by a solvent that chemically or physically traps the  $CO_2$ . A combination of both mechanisms is also possible. The  $CO_2$  can then be removed from the solvent by heating or a pressure reduction. The current focus is on using chemical absorption as separation technique. The chemical absorption technologies that we reviewed include technologies using alkanolamines, such as monoethanolamine (MEA), Fluor's Econamine FG+ and MHI's<sup>2</sup> KS-1 solvent. Other technologies reviewed are based on absorption using chilled ammonia (NH<sub>3</sub>), alkali salts (i.e. potassium carbonate - K<sub>2</sub>CO<sub>3</sub>) and amino salts. The post-combustion system can be applied to various energy conversion technologies. In this study, we review its application to Pulverized Coal (PC) and Natural Gas Combined Cycle (NGCC).

Pre-combustion CO<sub>2</sub> capture is aimed to remove the CO<sub>2</sub> before the fuel is combusted. This requires the fuel to be gasified or reformed into a syngas, which comprises mainly CO, H<sub>2</sub>O, H<sub>2</sub> and CO<sub>2</sub>. The water gas shift reaction catalytically shifts CO and H<sub>2</sub>O to H<sub>2</sub> and CO<sub>2</sub>. The CO<sub>2</sub> can then be removed, with chemical and physical solvents, adsorbents and membranes. The H<sub>2</sub> can be used for power production in a gas turbine. The current focus is on using chemical or physical (or a combination) solvents to separate the CO<sub>2</sub>. The energy conversion technology that is envisaged using precombustion that is mainly investigated in this study is the Integrated Gasification Combined Cycle (IGCC) power plant. We take into account IGCC systems based on Shell, GE and E-Gas gasifiers with pre-combustion capture based on Selexol and MDEA (methyldiethanolamine).

Oxyfuel combustion is based on denitrification of the combustion medium. The nitrogen is removed from the air through a cryogenic air separation unit (ASU) or membranes. Combustion thus takes place with nearly pure oxygen. The effect of which is a flue gas containing mainly  $CO_2$  and water. The  $CO_2$  is purified by removing water and impurities. The current focus is on using cryogenic air separation as the oxygen production technique. The energy conversion technologies that have been reviewed in this

Table 1

Simplified overview of energy conversion and CO<sub>2</sub> capture efficiencies of power plants equipped with various CO<sub>2</sub> capture technologies, after [20].

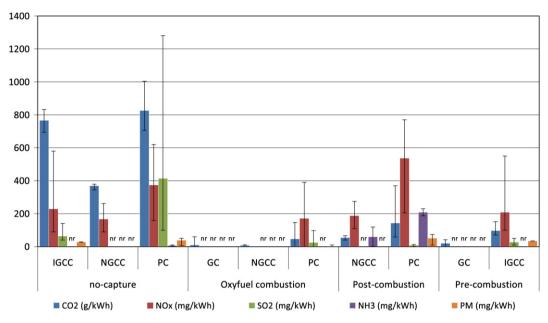
		-		
Capture process	Conversion technology <sup>a</sup>	Generating efficiency <sup>b</sup> (%)	Energy penalty of CO <sub>2</sub> capture (% pts.)	
Post-combustion (chemical absorption)	PC NGCC	30–40 43–55	8–13 5–12	85—90 85—90
Oxyfuel	PC GC and NGCC	33–36 39–62	9–12 2–19	90-100 50-100
Pre-combustion	IGCC GC	32–44 43–53	5–9 5–13	85–90 85~100

<sup>a</sup> PC = Pulverized Coal, NGCC = Natural Gas Combined Cycle, GC = Gas Cycle, IGCC = Integrated Gasification Combined Cycle.

<sup>b</sup> Efficiencies are reported based on the Lower Heating Value (LHV) and assuming a CO<sub>2</sub> product pressure of 11 MPa.

<sup>&</sup>lt;sup>2</sup> Mitsubishi Heavy Industries.

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**Fig. 2.** Atmospheric emissions of substances CO<sub>2</sub>, NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub> and particulate matter for various conversion technologies with and without CO<sub>2</sub> capture, adapted from [24]. Ranges indicate maximum and minimum values reported. Note that emissions are based on various fuel specifications and on the configuration and performance of the power plant and CO<sub>2</sub> capture process. 'nr' = 'not reported'.

study more extensively are rather conventional PC and NGCC power plants.

#### 3.1. Energy

 $CO_2$  capture and compression requires energy which results in an energy penalty for the power plant, reducing the net conversion efficiency of the power plant. In Fig. 1, the processes are shown per capture system that are added to the power generation concepts and through their demand for thermal, chemical or electrical energy result in an efficiency penalty. The energy penalty varies with capture system and technologies, see Table 1. A detailed review of the thermodynamic performance of power plants equipped with  $CO_2$  capture technologies is presented by Damen et al. [20].

#### 3.2. Atmosphere

Key atmospheric emissions assessed in EIAs for biomass and coal fired concepts are  $CO_2$ ,  $NO_x$ ,  $SO_2$ , HCl, HF, VOC, PM, Hg, Cd, and other heavy metals. Additionally, the emission of NH<sub>3</sub> slip from flue gas cleaning and dust during the handling of the fuel are assessed. For gas fired concepts  $CO_2$  and  $NO_x$  are the most dominant atmospheric emissions. Equipping power plants with  $CO_2$  capture technologies affects both the formation and fate of many of these emissions. Tzimas et al. [21] reviewed  $NO_x$  and  $SO_2$  emissions in fossil fuel fired power plants equipped with  $CO_2$  capture and found a trade-off in atmospheric emissions. A detailed review of the effect of post-, pre- and oxyfuel combustion  $CO_2$  capture on the substances  $NO_x$ ,  $SO_2$ , VOC, PM and  $NH_3$  is provided in [21–24]. The reported emission factors per kWh for these substances are presented in Fig. 2. The main effects of  $CO_2$  capture on atmospheric emissions are summarized below per capture system.

#### 3.2.1. Post-combustion

In Fig. 1, it is shown that the CO<sub>2</sub> capture process is situated after the flue gas cleaning section. Depending on the type of solvent that is used, impurities need to be removed from the flue gas in order to limit operational problems. Examples are solvent degradation, foaming and fouling. Impurities that need to be removed are typically acid gases (NO<sub>x</sub>, SO<sub>x</sub>, HCl and HF) and particulate matter (PM). Power plants equipped with CO<sub>2</sub> capture should thus be equipped with highly efficient flue gas desulphurization (FGD), DeNO<sub>x</sub> installations and electrostatic precipitators (ESP) and/or fabric filters to remove PM. Also, the flue gas typically requires cooling before it is processed in the CO<sub>2</sub> capture installation. In the CO<sub>2</sub> capture process also some of these substances are partially removed, see Table 2.

Depending on the increase in primary energy use due to the capture process, the net result may be that non-CO<sub>2</sub> emissions to air increase per kWh, like NO<sub>x</sub>. For some post-combustion variants additional atmospheric emissions are expected. This encompasses the emission of solvent or degradation products of the solvent. This may be NH<sub>3</sub> for the chilled ammonia process [29–31]. The

#### Table 2

Overview of removal efficiencies of flue gas conditioning and post-combustion capture technologies removing atmospheric substances.

Sorbent/power plant	Removal efficiency <sup>a</sup> (%)	Remarks	
	Flue gas conditioning	Reduction in capture process	
Amine based MHI KS-1/PC power plant	PM: 40% and 50% SO <sub>2</sub> : > 98% HCl and	PM: 40–60% SO <sub>2</sub> : "almost all" NO <sub>x</sub> : 1–3%.	Cooling and desulphurization with NaOH scrubber [25,26].
Alkanolamines/PC power plant	HF: ~complete –	$SO_2:$ 40–85% uptake of total sulphur $NO_x:$ 0.8%	No additional flue gas conditioning installed [27,28]

<sup>a</sup> This indicates the extra removal of impurities compared to existing flue gas cleaning equipment and does not take into account the efficiency penalty.

Box 1. Potential environmental impacts of amines and their degradation products.

Amine based solvents used for post-combustion capture are usually produced from basic chemicals like ammonia, methanol and ethylene oxide. MEA is distilled from a mixture of MEA, DEA and TEA (mono-, di- and tri-ethanolamine) and produced in a batch mode from ethylene oxide and ammonia. Amines and degradation products are found to be emitted by the stack, causing potential environmental impacts. MEA (2-aminoethanol) is emitted in small quantities (1–4 ppmv) due to entrainment in the scrubbed flue gas. This corresponds to 40–160 t/yr for a plant capturing 1 Mt per annum, but is possibly lower for capture facilities with mitigation measures implemented [44].

The toxicity of MEA is well documented and exposure guidelines are set [45]. However, research towards understanding chronic exposure effects and other toxicity end-points seems to be lacking. According to the National Research Council [45], no relevant studies were identified for the carcinogenicity of MEA.

Another potential concern that was already raised by Rao et al. [33] is the formation of (carcinogenic) nitrosamines, nitramines and amides that are products of the reaction of ethanolamines and atmospheric oxidants (e.g.  $NO_x$ ) under the influence of sunlight. Unlike diethanolamine, MEA has not been found to form a stable nitrosamine [44,45].

There is growing awareness on the possible environmental impacts of  $CO_2$  capture and both desktop studies as measurement campaigns are deployed to address potential concerns [46]. Several are listed below:

• In 2007, the Norwegian Institute for Air Research (NILU) initiated a project to study the effects of amine emissions to the environment. The amines studied are MEA, AMP, MDEA and piperazine (MEA (2-aminoethanol):  $H_2NCH_2CH_2OH$ ; AMP (2-amino-2-methyl-1-propanol): (CH<sub>3</sub>)<sub>2</sub>C(NH<sub>2</sub>)CH<sub>2</sub>OH; MDEA (2,2'-(methylimino)bis-ethanol): CH<sub>3</sub>N(CH<sub>2</sub>CH<sub>2</sub>OH)<sub>2</sub>; Piperazine: HN(CH<sub>2</sub>CH<sub>2</sub>)<sub>2</sub>NH).

• In 2009, Shao and Stangeland [47] advised to focus research on the determination of atmospheric degradation paths, precise degradation yields, and degradation products' life time in the atmosphere. Another advice was to focus research on developing both acute and chronic human toxicity exposure limits for amines and associated substances.

• In 2010, a workshop on this topic was organized by IEA GHG to identify measurement campaigns and knowledge gaps to structure R&D activities.

• CESAR, Emission measurements at Dong's pilot plant for CO<sub>2</sub> capture in Esbjerg: Oxidative degradation products of MEA are found in gas and liquid phase but a water wash reduces the amount of emitted MEA and formaldehyde

• Mitsubishi Heavy industries, MHI Amine emission control technology: Pilot plants test results indicate that degraded amine was less than 0.2 ppm as vapor. R&D topics actively pursued are the evaluation of the environmental effects, photogenic reaction in the air of released amine and the effect of nitrosamines into aquatic environment.

• Fluor, Econamine FG+ Process, recent advances in emissions control: A new scrubbing system has been developed by Fluor with reduced solvent emissions of 0.1 to 0.2 ppm in the vent. This process will be tested in a demonstration plant in Germany in 2011.

• Aker Clean Carbon, emissions measurements and analysis from Mobile Carbon Capture Test facility: Results from various measurement campaigns indicate that sampling and analytical methods will give different results and are challenging due to the low concentrations of the compounds. Given the uncertainties, more campaigns and results are needed in order to provide rigorous conclusions on emission levels.

alkanolamine-based solvents may result in the emission of VOC and NH<sub>3</sub> due to the degradation of the solvent [28,32,33]. Korre et al. [34] report that NH<sub>3</sub> emission from using the MHI KS-1 solvent is expected to be higher than from using MEA or potassium carbonate.<sup>3</sup> Contrarily, IEA GHG [35] reports lower values for NH<sub>3</sub> emissions for the MHI KS-1 process compared to Fluor's process based on MEA.

Also, the direct emission of MEA has been reported. The exact quantity of this 'MEA slip' (estimates range between 1 and 4 ppmv) and possible effects on the environment, including human safety, are not fully known, see Box 1. By contrast, the exact composition of solvents with additives is classified, as this is part of competition sensitive information. Data on exact emissions of reaction products from these additives or emissions of the additives themselves are also not known to be publicly available. In addition, solvent additives (e.g. corrosion inhibitors) may result in trace emissions of heavy metals [36].

For the  $K_2CO_3$  sorbent the slip to the atmosphere is considered negligible. Furthermore, this substance is considered less toxic to the environment [37,38].  $K_2CO_3$  may however require the addition

of promoters to increase the reaction rate. Some promoters, like arsenic trioxide and piperazine, are known to be toxic [38].

Allaie and Jaspers [39] claim that the use of amino salts does not result in ammonia formation, losses due to evaporation and virtually nil emissions of the solvent. Furthermore, amino acids are according to Hetland and Christensen [40] biodegradable.

The emissions of gas fired power plants equipped with postcombustion  $CO_2$  capture are also affected.  $NO_x$  emissions<sup>4</sup> are expected to be reduced per primary energy input but are expected to increase per kWh. NH<sub>3</sub> emissions increase for both, due to the emission of solvent or its degradation products, see Fig. 2. The higher oxygen concentration in the flue gas from natural gas combustion possibly results in higher oxidative degradation of solvents. MEA is for instance susceptible to this type of degradation [43]. However, as other impurities such as SO<sub>2</sub> and PM are virtually not present in the flue gas, overall degradation is considerably lower compared to coal fired power plants.

<sup>&</sup>lt;sup>3</sup> In this case piperazine, an amine, is added to the potassium carbonate sorbent as an activator to increase reaction rate.

<sup>&</sup>lt;sup>4</sup> The main fraction of NO<sub>x</sub> is formed by NO which is expected to be unaffected by the CO<sub>2</sub> capture process. NO<sub>2</sub> fraction of NO<sub>x</sub>, which is typically about 5–10%, may react with the solvent resulting in a reduction of NO<sub>x</sub> emission per MJ<sub>primary</sub>. However, also not all of the NO<sub>2</sub> is expected to react, i.e. only 25% [41,42].

Raw water usage<sup>a</sup> in conversion technologies equipped with various CO<sub>2</sub> capture technologies.

Conversion technology/CO <sub>2</sub> capture technology	Source	Water usage w/o capture (L kWh <sup>-1</sup> )	Water usage with capture (L kWh <sup>-1</sup> )	Annual increase <sup>b</sup> million (m <sup>3</sup> yr <sup>-1</sup> )	Relative increase in water use (%)	Relative increase in primary energy use (%)
IGCC/pre-combustion	[60] <sup>c</sup>	2.57-3.12				
	[61] <sup>d</sup>	0.6	0.9	1.97	50%	16%
	[59] <sup>e</sup>	1.35-1.42	1.81-2.00	3.02-3.81	32-48%	18-28%
NGCC/post-combustion	[60] <sup>c</sup>	1.88				
	[59] <sup>e</sup>	1.02	1.84	5.39	81%	16%
PC subcritical/post-combustion	[60] <sup>c</sup>	4.43				
	[61] <sup>d</sup>	3.1				
	[59] <sup>e</sup>	2.56	5.04	16.30	96%	48%
PC supercritical/post-combustion	[60] <sup>c</sup>	3.94				
	[61] <sup>d</sup>	3.1	4.1	6.57	32%	31%
	[59] <sup>e</sup>	2.25	4.34	13.74	93%	44%
Oxyfuel combustion with CO2 removal	[62] <sup>f</sup>	-	2.97-3.01	4.84-5.13 <sup>g</sup>	33–35% <sup>g</sup>	39–41% <sup>g</sup>

<sup>a</sup> Raw water usage is defined as the total internal water consumption minus internal recycling.

<sup>b</sup> This is calculated as the difference between a 1 GWe power plant with capture and a 1 GWe power plant without capture, both with a capacity factor of 75% (6575 full load hours yr<sup>-1</sup>).

<sup>c</sup> Based on power plants equipped with evaporative cooling towers. Ranges for IGCC represent various gasifier technologies (GE, Shell and E-Gas).

<sup>d</sup> Reflect life cycle emissions. Not specified whether figures are based on power plants with evaporative cooling tower(s) or once through cooling configuration.

<sup>e</sup> Based on power plants equipped with evaporative cooling towers. Ranges represent various gasifier technologies: GE, Shell and E-Gas. For cases from this source it is reported that 71–99% of water use is due to cooling tower water make-up.

<sup>f</sup> Based on power plants equipped with evaporative cooling towers. Ranges represent variations in the purity of the oxygen supply for combustion.

<sup>g</sup> Compared to supercritical PC power plant without CO<sub>2</sub> capture as presented in [62].

#### 3.2.2. Pre-combustion

In coal and biomass fired pre-combustion concepts using solvents, no solvent emission to the air is expected during normal operation as any slip of the solvent would be combusted in the gas turbine or end up in the CO<sub>2</sub> stream. NO<sub>x</sub> emissions are still an area of research for the turbine manufacturers of the IGCC with pre-combustion CO<sub>2</sub> capture. The hydrogen rich fuel may increase NO<sub>x</sub> emissions from the gas turbine section due to the different combustion characteristics of hydrogen compared to natural or syngas. Therefore, in Fig. 1, an additional flue gas cleaning step, a DeNO<sub>x</sub> installation, is included after the gas turbine section. This can be installed if NO<sub>x</sub> emissions are required to be lower than can be achieved by turbine development alone. A possible trade-off is that this results in NH<sub>3</sub> emissions from the DeNO<sub>x</sub> installation.

Co-sequestration of  $H_2S$  is technically possible. Acid gas coinjection is common in, for instance, Canada [48]. In essence, the CO<sub>2</sub> capture unit is an acid gas removal unit. Such units are already applied in IGCC configurations for the removal of  $H_2S$ . The removal of  $H_2S$  from the syngas may be enhanced by adding CO<sub>2</sub> removal. Some  $H_2S$  may also end up in the CO<sub>2</sub> stream.

#### 3.2.3. Oxyfuel combustion

The main effect of oxyfuel combustion is the change in the composition of the flue gas. For we refer to [22,49-51]. In Fig. 1, it is shown that flue gas recycling (FGR<sup>5</sup>) in the oxyfuel concept is needed to reduce the temperature in the combustion step. The cleaning of flue gas in coal fired oxyfuel concept has the additional purpose of limiting fouling, erosion and corrosion further down the chain. Removal of particulate matter, NO<sub>x</sub> and SO<sub>x</sub> may therefore be necessary.

FGR also leads to an additional reduction of NO<sub>x</sub> formation during the combustion process. Typically less (24–40%) NO<sub>x</sub> is formed in the boiler as NO<sub>x</sub> is now virtually limited to fuel bound NO<sub>x</sub> formation and some possible formation due to air in-leakage. For gas fired concepts NO<sub>x</sub> is virtually eliminated as fuel bound NO<sub>x</sub> is virtually nil [52].

 $SO_x$  composition in the flue gas changes for the coal fired power plants, i.e. higher concentrations of  $SO_3$  and higher retention of

sulphur in ashes are reported. This enables the use of other or adapted desulphurization technologies; none of which has been demonstrated at commercial scale, however. According to [53,54] a high removal of SO<sub>2</sub> (64 and ~100%) and NO<sub>x</sub> (48–90%) is possible in the CO<sub>2</sub> conditioning and compression section. A detailed review of the impacts of sulphur impurities on the coal fired oxyfuel cycle by Stanger and Wall [51] yielded the insights that the choice for proposed desulphurization will strongly depend on the regulations that are to be set for transport and storage of CO<sub>2</sub>, and perhaps co-storage of sulphur compounds.

The estimates for  $NO_x$  emissions from oxyfuel combustion of solid fuels vary considerably, mainly due to the various  $CO_2$  purification configurations proposed.

More insight into the effect on emissions comes from the results of a coal fired demonstration project in Germany [55]. There, possible configurations for flue gas cleaning are predominantly based on (adapted) conventional flue gas cleaning technologies. The additions compared to a conventional configuration consisting of an SCR, ESP and FGD, are a flue gas cooler (FGC) and CO<sub>2</sub> compression & purification process. The FGC is aimed to reduce the temperature, acidic substances (SO<sub>2</sub> between 93 and 97%, SO<sub>3</sub> between 58 and 78%), water content (>85%) and particulates (>90%) in the flue gas prior to compression. In the following compression & purification step, additionally NO<sub>x</sub>, SO<sub>x</sub>, HCl, water and heavy metals are removed as condensate from the compressors and with the use of an activated carbon filter and an adsorber [56–58]. Overall, a deep reduction of  $SO_2$  and  $NO_x$  emissions is expected to be possible with oxyfuel combustion, although R&D is required to better understand the behavior of these substances in the CO<sub>2</sub> compression & purification process. This includes attaining better understanding of the thermodynamic properties of mixtures of SO<sub>2</sub>, H<sub>2</sub>O and supercritical CO<sub>2</sub>; and insights into the effect of SO<sub>3</sub> formation on heat exchanger operation and material selection [51].

#### 3.3. Water

#### 3.3.1. Water consumption

Water consumption increases due to the energy penalty and the additional water demand by the CO<sub>2</sub> capture system.

Table 3 shows an overview of several studies reporting the raw water use per kWh. The relative increase in water use is in most studies higher than the relative increase in primary energy. This is

<sup>&</sup>lt;sup>5</sup> FGR is applied to control the combustion temperature, as this is limited by materials currently applied.

Table	4
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Waste streams and by products of coal fired power plants with and without CO<sub>2</sub> capture.

Waste/by-product	Technology	Source	W/o capture (g kWh <sup>-1</sup> )	With capture (g kWh <sup>-1</sup> )	Annual increase <sup>a</sup> (kt yr <sup>-1</sup> )	Relative increase (%)
Solvent waste	PC post-combustion	[67]	_	2.63 (Fluor)	17.29	_
		[67]		0.26 (MHI KS-1)	1.71	
		[19]		2.1 (MEA)	13.81	
	IGCC pre-combustion	[67]	0.01	0.02	0.07	100%
Gypsum	PC post-combustion	[19]	9.08	11.91	18.61	31%
		[35]	15.23	21.15	38.92	39%
		[67]	13.8	18.8-19.1	32.87-125.57	36%/38%
		[59]	53.6 <sup>b</sup>	77 <sup>b</sup>	153.84	44%
		[59]	47.8 <sup>c</sup>	70.3 <sup>c</sup>	147.93	47%
Sulphur'	IGCC pre-combustion	[67]	2.78 <sup>d</sup>	3.48 <sup>d</sup>	4.60	25%
		[67]	3.16 <sup>e</sup>	3.81 <sup>e</sup>	4.27	21%
		[59]	8.7 <sup>e</sup>	10.4 <sup>e</sup>	11.18	20%
		[59]	8.5 <sup>f</sup>	10 <sup>f</sup>	9.86	18%
		[59]	8 <sup>d</sup>	10.3 <sup>d</sup>	15.12	29%
Bottom-/fly-ash	PC post-combustion	[67]	39.3	48.9 (Fluor)	63.12	24%
	-	[67]	_	48.3 (MHI KS-1)	59.17	23%
		[59]	26.5/6.6 <sup>b</sup>	37.2/9.3 <sup>b</sup>	70.35/17.75	40%/41%
		[59]	24.8/6.2 <sup>c</sup>	35.4/8.9 <sup>c</sup>	69.69/17.75	43%/44%
Slag	Oxyfuel combustion	[67]	39.3	48	57.20	22%
-	IGCC pre-combustion	[67]	44.7 <sup>d</sup>	55.8 <sup>d</sup>	72.98	25%
	-	[67]	54.1 <sup>e</sup>	65.3 <sup>e</sup>	73.63	21%
		[59]	38 <sup>e</sup>	45 <sup>e</sup>	46.02	18%
		[59]	34.4 <sup>f</sup>	42.5 <sup>f</sup>	53.25	24%
		[59]	32.2 <sup>d</sup>	41.4 <sup>d</sup>	60.49	29%

<sup>a</sup> This is calculated as the difference between a 1 GW<sub>e</sub> power plant with capture and a 1 GW<sub>e</sub> power plant without capture, both with a capacity factor of 75% (6575 full load hours  $yr^{-1}$ ).

<sup>b</sup> Subcritical steam parameters.

<sup>c</sup> Supercritical steam parameters.

<sup>d</sup> Based on Shell gasifier.

<sup>e</sup> Based on GE (General Electric) gasifier.

<sup>f</sup> Based on ConocoPhillips E-Gas gasifier.

most distinctive for the post-combustion capture cases for which the water consumption at present almost doubles as a result of the large additional cooling requirement of the  $CO_2$  capture process. For the IGCC with pre-combustion the additional water use is due to the water requirement in the water gas shift reaction [59]. For oxyfuel combustion the limited available data suggest an increase in water usage, although the increase is less then proportional to the increase in primary energy use.

#### 3.3.2. Emissions to water

The effect of equipping power plants with  $CO_2$  capture on the emissions to water bodies is currently an insufficiently researched subject. Cross-media effects<sup>6</sup> are likely as gaseous emissions are transformed into the liquid phase [55]. Trade-offs thus will occur with the decrease in gaseous emissions as mass flows must balance. Quantification of this trade-off is not possible due to lack of publicly available data. Qualitatively some issues can however be addressed.

For example, a liquid waste stream for amine based postcombustion capture processes may come from the reclaimer section [63]. Quantities and exact compositions of this waste stream are however not known to be reported in public available literature. Increased removal efficiency in emission control technologies (e.g. FGD and pre-scrubbing) and the additional reduction in the  $CO_2$  capture process are possible processes that likely results in a shift from air emission to water or solid stream emissions. For the post-combustion process with potassium carbonate it is possible that potassium based minerals, usually fertilizers, may be discharged with the waste water if not recovered [64].

For an IGCC without  $CO_2$  capture (1.2 GW<sub>e</sub>) an emission to surface water of the solvents MDEA and Sulfolane of approximately 26 t yr<sup>-1</sup> is estimated [16]. This may increase due to the implementation of pre-combustion CO<sub>2</sub> capture.

Yan et al. [55] suggest that due to a change in the configuration of the flue gas cleaning system in coal fired oxyfuel plants contaminants may be transferred to liquid waste streams. These liquid waste streams may in turn affect overall emissions to water bodies.

#### 3.4. Waste and by-products

The formation of waste streams and by-products in power plants firing coal and biomass is affected by the application of  $CO_2$  capture. Waste and by-product formation is typically not an issue for natural gas fired power plants without  $CO_2$  capture [65]. This may change when equipped with post-combustion  $CO_2$  capture.

#### 3.4.1. Post-combustion

Table 4 shows that in PC plants with post-combustion  $CO_2$  capture more ash (bottom-ash and fly-ash) is formed per kWh. In the  $CO_2$  capture unit impurities in the flue gas such as  $SO_x$  and halogen compounds react with amine-based solvents to form heat stable salts.<sup>7</sup> These salts reduce the  $CO_2$  binding capacity of the solvent and are corrosive compounds that are harmful for equipment. Degradation products and other impurities are therefore separated from the solvent in a reclaimer where also solvent is recovered. Results from a study analyzing the composition of reclaimer waste implies that  $CO_2$  capture influences the distribution of trace element emissions (Se, As, Cr, Cu, Ni, Zn and Hg) over the various waste streams from a coal fired power plant [36]. The residues from the reclaimer are to be considered as hazardous

<sup>&</sup>lt;sup>6</sup> Possible shift of environmental pressure from one environmental media (water, atmosphere, soil) to the other.

 $<sup>^{\,7}\,</sup>$  Heat stable salt: a salt that is not capable of being regenerated by the addition of heat.

waste [32,33,36] and can be in the order of several kilotonnes per year for a commercial scale power plant [66], see Table 4.

The most appropriate treatment of the reclaimer sludge has yet to be determined. It can possibly be treated in the wastewater treatment installation (WWT), which means that a fraction of the sludge is emitted to the surface water and the other fraction (WWT sludge) is to be disposed of. Co-firing is an option similar to that of the optional treatment of WWT sludge [68]. Re-introducing the sludge into the boiler will redistribute the elements of the sludge over other waste streams such as fly-ash, bottom-ash and gypsum and WWT effluent and sludge. As there are limits of acceptance regarding the concentration of impurities, such as mercury and other heavy metals, valorization of by-products can become a problem [69].

For coal fired PC power plants also a larger stream of solid byproduct from the FGD unit, primarily gypsum, is expected due to the required improved  $SO_x$  removal efficiency and the energy penalty, see Table 4.

In the chilled ammonia concept ammonium sulphate can be a by-product that is theoretically recoverable and usable as fertilizer. This is the reaction product of SO<sub>2</sub> in the flue gas with the ammonia solution [31].

In the concept using potassium carbonate possible new waste or by-product streams include: nitrates, nitrites, sulphates and sulphites formed by the reaction of the sorbent with SO<sub>2</sub> and NO<sub>2</sub> [38]. If recovered, these substances can be used as fertilizers. When using sodium carbonate, it is likely that SO<sub>2</sub> that still remains in the flue gas reacts to sodium sulphite, -bisulphite and-sulphate, comparable with the reaction in a sodium alkali FGD scrubbing system [cf. 70]. These salts in solutions are liquid waste streams that should be treated properly. Amino acids are reported by Allaie and Jaspers [39] to be stable and show low degradation rates which would imply that waste and by-product formation is low. It should however be noted that the results of the pilot plant test are confidential and that these results cannot be verified.

#### 3.4.2. Pre-combustion

Typical waste streams and by-products from IGCC power plants are: fly-ash, bottom-ash, slag and sulphur or sulphuric acid. The amount and composition of these often marketable streams depend on the gasifier and desulphurization technologies applied and the fuel utilized [71]. Table 4 shows that sorbent waste increases with a factor 2 for the pre-combustion concept. Furthermore, the production of the marketable elemental sulphur increases per kWh. For the production of slag an increase between 18% and 29% is expected in literature, depending on the type of gasifier implemented.

#### 3.4.3. Oxyfuel combustion

Davidson et al. [72] suggest that oxyfuel combustion characteristics affect the speciation and further removal of mercury from the flue gas. Oxidized mercury is more easily captured in existing flue gas control systems. Additionally captured Hg would then end up in the waste streams of flue gas control technologies such as FGD and dust control (ESP and filters). However, some flue gas control technologies may be omitted when applying oxyfuel combustion. White et al. [53,54] suggest a technology that removes SO<sub>2</sub> and NO<sub>x</sub> in the form of sulphuric (H<sub>2</sub>SO<sub>4</sub>) and nitric acid (HNO<sub>3</sub>), respectively. The latter substance may react with oxidized mercury (Hg<sup>2+</sup>) in the flue gas producing mercuric nitrate. This is a toxic substance and should be considered a hazardous waste.

Table 5

Resource consumption by energy conversion technologies equipped with and without CO<sub>2</sub> capture.

Resource (process)	Technology	Source	No capture (g kWh <sup>-1</sup> )	Capture (g kWh <sup>-1</sup> )	Annual increase <sup>a</sup> (kt yr <sup>-1</sup> )	Relative Increase (%)
Sorbent make-up (CO <sub>2</sub> capture)	PC/post	[61]	_	3.6 MEA	23.67	_
		[19]		2.04 MEA	13.41	
		[67]		1.31 Fluor	8.61	
		[67]		0.13 MHI KS-1	0.85	
		[35]		1.31 MEA	8.61	
		[59]		0.37 Fluor (sub crit.)	2.43	-
		[59]		0.33 Fluor (super crit.)	2.17	
		[74,75]		0.18 AA <sup>b</sup>	1.18	
		[27]		2.16 MEA <sup>c</sup>	14.2	
		[28]		1.26 MEA <sup>d</sup>	8.28	
		[37]		0.45 K <sub>2</sub> CO <sub>3</sub> /PZ <sup>e</sup>	2.96	
	NGCC/post	[61]	_	1.33 MEA	8.74	-
		[35]		0.61 MEA	4.01	
		[59]		0.12 Fluor	0.79	
	IGCC/pre	[61]	0.02 Selexol	0.03 Selexol	0.07	50%
		[67]	0.01 Selexol	0.02 Selexol	0.07	100%
		[35]		0.005 MDEA	0.03	-
Limestone <sup>f</sup> (FGD)	PC/post	[55]	16.9	27.2	67.72	61%
		[19]	5.6	7.5	12.49	34%
		[67]	8.4	11.4–11.6	19.72-21.04	36%-38%
		[35]	8.4	01.6	21.04	38%
		[59]	33.6-35.9	48.2-52.7	95.99-110.45	43%-47%
Ammonia (SCR)	PC/Post	[61]	0.61	0.80	1.25	31%
		[19]	0.31	0.41	0.66	32%
	NGCC/post	[61]	0.20	0.23	0.20	15%

Note: sub crit. = subcritical steam parameters; super crit. = supercritical steam parameters indicating higher generating efficiency, i.e. a lower capture penalty.

<sup>a</sup> This is calculated as the difference between a 1 GW<sub>e</sub> power plant with capture and a 1 GW<sub>e</sub> power plant without capture, both with a capacity factor of 75% (6575 full load hours yr<sup>-1</sup>).

<sup>b</sup> AA = Aqueous Ammonia. Based on the assumption of 0.9 kg CO<sub>2</sub> captured kWh. Original value 0.2 g/kg captured [74,75].

<sup>c</sup> Based on the assumption of 0.9 kg CO<sub>2</sub> captured/kWh. Reported value 2.4 g/kg captured [27].

<sup>d</sup> It is reported that similar ranges were found for alternative solvents 'CASTOR 1' and 'CASTOR 2'. Based on the assumption of 0.9 kg CO<sub>2</sub> captured/kWh. Reported value 1.4 g/kg captured [28].

<sup>e</sup> Piperazine promoted potassium carbonate. Based on the assumption of 0.9 kg CO<sub>2</sub> captured/kWh. Reported value 0.5 g/kg captured [37].

<sup>f</sup> Limestone use depends mainly on FGD efficiency and sulphur content of the fuel.

Summary of risk assessments for CO<sub>2</sub> transport by pipeline showing the failure scenarios assessed, pressure, the pipeline diameter, section length, assumed critical CO<sub>2</sub> exposure threshold, the calculated maximum distance to this threshold and the distance to the individual risk contour.

Source	Failure scenario <sup>a</sup>	Pressure	Pipeline	Isolable Section	Exposure threshold		Distance to exposure	Distance to individual
		(MPa)	diameter (cm)	length (km)	Concentration (ppm)	Duration (min)	threshold (m)	risk contour <sup>b</sup> (m)
[96]	Rupture	3.5	66	5-30	50 000	1	250-750	
	Rupture	6	41	5-30	50 000	1	150-600	
[95,98]	Cumulative	13-20	61-107	30	40 000	30	1350 1900	1900-2450
[97]	Cumulative	1.7	66	17	54 656	60		<3.5
[91] <sup>c</sup>	Rupture	6.9	8-41		5000	10	310-1246	
	Rupture	6.9	8-41		30 000		59-89	
[94]	Rupture	15.2	36-51	8 <sup>d</sup>	30 000	15	<1-202	
	Rupture	15.2	36-51	8	40 000	15	<1-136	
	Rupture	15.2	36-51	8	70 000	15	<1-66	
	Puncture	15.2	36-51	8	15 000	>180	265-272	
	Puncture	15.2	36-51	8	20 000	>180	168-197	
	Puncture	15.2	36-51	8	60 000	>180	44-46	
	Puncture	15.2	36-51	8	70 000	>180	35-38	
	Rupture $H_2S$ (0.01%)	15.2	36-51	8	0.51	15	1271-6885	
	Rupture $H_2S$ (0.01%)	15.2	36-51	8	27	15	40-593	
	Rupture $H_2S$ (0.01%)	15.2	36-51	8	50	15	4-373	
	Rupture $H_2S$ (0.01%)	15.2	36-51	8	0.20	>180	2136-2356	
	Rupture $H_2S$ (0.01%)	15.2	36-51	8	0.33	>180	1628-1741	
	Rupture $H_2S$ (0.01%)	15.2	36-51	8	17	>180	167-169	
	Rupture $H_2S$ (0.01%)	15.2	36-51	8	31	>180	115-116	
[90]	Puncture	20.4 <sup>e</sup>	33	30	40 000-100 000	30	70-110	
11	Rupture	20.4 <sup>e</sup>	33	30	40 000-100 000	30	170-210	
	Puncture H <sub>2</sub> S (2%)	20.4 <sup>e</sup>	33	30	100	30	290	
	Puncture $H_2S(2\%)$	20.4 <sup>e</sup>	33	30	100	30	1180	
	Puncture H <sub>2</sub> S (2%)	20.4 <sup>e</sup>	33	30	800	5	100	
	Puncture H <sub>2</sub> S (2%)	20.4 <sup>e</sup>	33	30	800	5	390	
[88]	Rupture	14	102	0.5-6.5	100 000		321-750	
[89]	Cumulative	20	102	160	2000-15 000	15	2500-7200	1500-3300
1	Cumulative	10	76	50	2000-15 000	15	2000-3800	1250-2650
[112]	Puncture	1.5-3.2	76	18	SLOT DTL <sup>f</sup>		3-149	
	Puncture	1.5-3.2	76	18	SLOD DTL <sup>g</sup>		3-107	
	Rupture	1.5-3.2	76	18	SLOT DTL <sup>f</sup>		100-160	
	Rupture	1.5-3.2	76	18	SLOD DTL <sup>g</sup>		71–107	
	Cumulative	1.5-3.2	76	18	SLOT DTL <sup>f</sup>		3–160	0-20 <sup>h</sup>
	Cumulative	1.5-3.2	76	18	SLOD DTL <sup>g</sup>		3–107	0 <sup>h</sup>
[99] <sup>i</sup>	Cumulative	4-20	41	20	27 000	10	194-800	0-204
	Cumulative	4-20	41	20	55 000	10	0-524	

<sup>a</sup> 'Cumulative' encompasses multiple scenarios, i.e. both rupture and puncture scenario.

<sup>b</sup> The individual risk contour here indicates the probability of adverse impact (in  $1.0 \times 10^{-6}$  yr<sup>-1</sup> km<sup>-1</sup>) on an ever-present and unprotected person. Note that the probability of occurrence is taken into account when determining the individual risk contour contrary to when determining the distance to the exposure threshold. The adverse impact is considered to be 'fatality' by [95,98] (70 000 ppm for several minutes) and [97] (assumed 1% fatality at 100 mg/m<sup>3</sup> for 60 min); 'non-fatal' is the impact assumed in [89] (at 15 000 ppm).

<sup>c</sup> In [91] also significant shorter distances are calculated for receiving the shown concentration levels at 1.5 m above ground level additional distances received at ground level shown here.

<sup>d</sup> One of the pipelines in this study has a length of 0.8 km which equals in that case the isolable section length.

<sup>e</sup> Maximum operating pressure of the pipeline.

<sup>f</sup> Is determined as Specified Level of Toxicity Dangerous Toxic Load which equals 1% mortality and is set at  $1.5 \times 10^{40} = (ppm^8 \times min)$ .

 $^{g}$  Is determined as Significant Likelihood of Death Dangerous Toxic Load which equals 50% mortality and is set at 1.5  $\times$  10<sup>41</sup> = (ppm<sup>8</sup>  $\times$  min).

<sup>h</sup> Indicating the distance to the pipeline at which the chance of receiving the Dangerous Toxic Load equals  $1 \times 10^{-6}$  yr<sup>-1</sup>.

<sup>i</sup> In [99], a sensitivity analysis was performed varying the type of release (instantaneous, horizontal and vertical jet, dry-ice bank sublimation), the failure rate  $(0.7 \times 10^{-4}-6.1 \times 10^{-4} \text{ km}^{-1} \text{ yr}^{-1})$  and dose-response (probit) function.

Also, the ash formation per kWh increases (see Table 4) and the composition of fly and bottom-ash may change as a consequence of oxyfuel firing [73]. A significant change in composition could pose problems for its qualification as usable by-product. Yan et al. [55] also state that due to oxyfuel combustion more gaseous contaminants will be transferred to liquid, solid waste or by-product streams. Quantitative data are however not available.

#### 3.5. Resource consumption

For NGCC the main resources used, besides fuel during operation, are ammonia and catalyst make-up for the removal of  $NO_x$  in an SCR [12,13]. Furthermore, chemicals are used for the conditioning of the cooling water and production of demineralised water for the steam cycle. Substances typically used in the normal operation of a PC power plant are: limestone, ammonia, sodium hypochlorite, lubricants, caustic soda, hydrochloric acid and sulphuric acid.

For the post-combustion capture concepts, the consumption per kWh of most of the above mentioned substances will increase with the energy penalty, see Table 5. The exception may hold for ammonia and limestone if the efficiency of the SCR and FGD section is required to improve, e.g. in the case of a retrofit. Amine based capture technologies require deep removal of both  $NO_x$  and  $SO_x$  to minimize solvent loss, the latter being the dominant target substance. Supap et al. [43] report that higher MEA concentrations in the solvent, next to  $O_2$  and  $SO_2$ , also increase the degradation rate. High  $CO_2$  concentrations were found to decrease the degradation rate. For gas fired concepts the degradation rate and solvent consumption are expected to be lower.

Caustic soda may be used to remove acid components in a scrubber prior to  $CO_2$  removal. In addition, NaOH may be used too in the  $CO_2$  capture process to reclaim part of the solvent that reacted with impurities [19,41]. Both lead to an increase in its consumption compared to a power plant without  $CO_2$  capture.

The consumption of solvent in the capture process is an important driver for solvent development as solvent loss deteriorates operational economics and has environmental consequences.

Table 5 clearly shows that the consumption of the sorbent varies per type of sorbent. Typically, the consumption of MEA is higher compared to its alternatives. Moreover, the consumption of sorbents used in IGCC with or without pre-combustion concepts can considered to be very low, although an increase is expected when  $CO_2$  capture is applied.

In the coal fired oxyfuel combustion concepts ammonia and limestone are used. Quantitative details on their consumption in adapted flue gas cleaning configurations are not known to be publicly available.

#### 3.6. Findings CO<sub>2</sub> capture at power plants

We found that depending on the applied  $CO_2$  capture technology, trade-offs and synergies can be expected for key atmospheric emissions. An increase in water consumption ranging between 32% and 93% and an increase in waste and by-product creation with tens of kilotonnes is expected for a 1 GW<sub>e</sub> power plant, but exact flows and composition are uncertain. Further, we found that there is considerable uncertainty on how the environmental fate of emissions may shift when equipping power plants with CO<sub>2</sub> capture. Information on cross-media effects when capturing CO<sub>2</sub> is underexposed at present and not quantified. We recommend that environmental monitoring programmes for demonstration plants should help to fill this knowledge gap on cross-media effects.

An important consideration in the EIA for power plants is that its design should be benchmarked against the Best Available Technology (BAT) described in the BAT Reference documents<sup>8</sup> (BREF) issued under the IPPC Directive for energy efficiency, pollution control and cooling water discharge, see [65,76,77]. Benchmarking is not yet possible as neither a BAT for CO<sub>2</sub> capture options is established nor is CO<sub>2</sub> capture considered BAT for large combustion plants. In fact, no elaboration on the environmental impacts of CO<sub>2</sub> capture is included.

This also includes the absence of emission performance standards for key (solid, atmospheric and liquid) emissions that take into account the efficiency penalty due to capture. Human safety norms do exist for some of the additional emitted substances, like amines and their degradation products. However, in general, the development of exposure limits for these type of substances has been identified as an important knowledge gap by Shao and Stangeland [47].

The knowledge base, from which a BREF is distilled, still has to be created for  $CO_2$  capture. The compilation of test results from the various (pilot and demo)  $CO_2$  capture facilities worldwide can be a valuable source of information to gradually expand and improve the BREF for Large Combustion Plants regarding capture options and its relation with other emission reduction techniques.

In the BREF for economic and cross-media effects a truncated version of the Life Cycle Analysis approach is proposed to determine the BAT for an individual activity taking into account multiple environmental themes.<sup>9</sup> The approach is truncated in the sense

that it, in principle, limits the system boundaries of the study to the proposed activity and its possible alternatives and thus does not include up- and downstream effects of the process [78]. That BREF and our review are not aimed at identifying life cycle effects of implementing  $CO_2$  capture options, but this should not be neglected when reviewing the environmental performance of complete CCS chains, from cradle to grave. Recent studies namely indicate that some direct emissions, like  $SO_x$ , may decrease due to  $CO_2$  capture; but that additional life cycle emissions by up- and downstream process may result in a deterioration of the overall environmental performance of the CCS chain compared to a power plant without CCS, see for instance [17–19,35,61,79–87].

#### 4. Transport of CO<sub>2</sub> by pipelines

In the international arena, primarily in the United States, there is significant experience with transporting large quantities (i.e. several Mt per pipeline) of  $CO_2$  by pipelines at high pressure, primarily for EOR projects. Several thousands kilometers of pipeline are being operated for this purpose. High-pressure transport is required as economics are not favorable for transporting large amounts of  $CO_2$  over considerable distances in the gas phase. The  $CO_2$  is therefore transported in the dense liquid or supercritical phase (i.e. above 31 °C and 7.38 MPa). The modeling of the dispersion of high-pressure  $CO_2$  releases in risk assessments was in the introduction of this article already identified as a knowledge gap. In the following section, we will focus the assessment on the external safety of high-pressure  $CO_2$  pipelines as it is indicated that this is one of the most important issues in the environmental assessment of  $CO_2$  transport pipelines.

Various quantitative risk assessments (QRA) have been performed for  $CO_2$  pipelines, see e.g [88–98]. A summary of the results of these studies is presented in Table 6. A review of these studies was performed by Koornneef et al. [99] yielding insight in the knowledge gaps and their impacts on the assessment of external safety of  $CO_2$  transport by pipeline. Also, Eldevik et al. [100] and UK's Health and Safety Executive [101] provide insight into the current knowledge base on the safety of high-pressure  $CO_2$  pipelines. The main conclusions of these studies are summarized below.

#### 4.1. Failure rates for CO<sub>2</sub> pipelines

Failure rates used in QRAs range between 0.7 and  $6.1 \times 10^{-4} \text{ yr}^{-1} \text{ km}^{-1}$  and are often based on experience with natural gas pipelines. A failure is predominantly caused by third party interference, corrosion, construction or material defects (e.g. welds), ground movement or operator errors [98,102]. Terrorism is presumably an underexplored factor in risk assessments for CO<sub>2</sub> pipelines. This factor should not be ignored although we presume that CO<sub>2</sub> pipelines are less likely targeted than hydrocarbon pipelines.

Currently, empirical data on the operation of CO<sub>2</sub> pipelines is not sufficient to determine the probability of failure of a pipeline section with the same accuracy as for natural gas pipelines. Furthermore, the presence of impurities and water influences the corrosion rate of CO<sub>2</sub> pipelines. Depending on the CO<sub>2</sub> capture process, the process flow may constitute toxic and corrosive impurities. Expected impurities are H<sub>2</sub>O, SO<sub>x</sub>, NO<sub>x</sub>, N<sub>2</sub>, O<sub>2</sub>, H<sub>2</sub>S, CO and H<sub>2</sub>. Current models seem not to be appropriate to accurately estimate corrosion rates when taking these impurities into account and with it fall short in providing quantitative information to determine the possibility of failure due to internal corrosion, see also [100]. The presence of free water is the dominating factor here and should be minimized to restrain corrosion to a high extent [103]. The presence of impurities may also influence

<sup>&</sup>lt;sup>8</sup> The BREFs for Large Combustion Plants (LCP) [65] for Industrial Cooling Systems and for Monitoring are applicable.

<sup>&</sup>lt;sup>9</sup> It includes 7 environmental themes: human toxicity, global warming, aquatic toxicity, acidification, eutrophication, ozone depletion and photochemical ozone creation potential.

General description of EIA procedures for activities in the geosphere reviewed this study.

Project short name	Description	Source
Analogous projects		
UGS Norg (Netherlands)	Underground gas storage project in the Netherlands in an empty gas field at a depth of $\sim$ 2700 m.	[113,114]
EOR Schoonebeek (Netherlands)	Enhanced oil recovery project using steam injection including the injection of produced water in nearby empty hydrocarbon reservoirs and aquifer at a depth of 1500 and 3000 m.	[115]
Gasselterenijveen (Netherlands)	Gas production project including the injection of produced water in nearby empty hydrocarbon reservoirs at a depth of 800 m and >3000 m.	[116]
CO <sub>2</sub> storage projects		
Frio, Texas (United States)	$CO_2$ storage pilot in saline aquifer at a depth of about 1500 m.	[117]
Gorgon Gas development (Australia)	Gas production project including the removal of CO <sub>2</sub> from the natural gas and injection into an aquifer at a depth of 2000 m.	[118,119]
AMESCO (Netherlands)	Generic environmental impact assessment for $CO_2$ storage in Dutch onshore gas fields.	[120]
CO <sub>2</sub> storage Barendrecht (Netherlands)	CO <sub>2</sub> storage in depleted onshore natural gas reservoirs at a depth of 1700 m (phase 1) and 2500 m (phase 2).	[8,108,121]
FutureGen (United States)	Integrated $CO_2$ capture, transport and storage project to be located in the United States (four storage sites pre-selected with reservoirs at a depth of 0.6–2.6 km).	[122]

thermodynamics (i.e. the phase) of the  $CO_2$  flow resulting in energy losses [100,102–105]. This influence on thermodynamic properties is also important in case of a sudden release or leakage from the pipeline.

#### 4.2. Release and dispersion of $CO_2$

The maximum CO<sub>2</sub> release rate from a failing pipeline is estimated in [99] to range between 0.001 and 22 t s<sup>-1</sup> depending mainly on the diameter of the pipeline and the size of the puncture. Other studies report somewhat lower rates of 8.5 t s<sup>-1</sup> [96] and 15 t s<sup>-1</sup> [95].

Impurities may affect the phase, temperature and pressure during the accidental release and dispersion the CO<sub>2</sub>. Another important aspect is that expanding CO<sub>2</sub> may involve phase changes that result in (dry) ice formation in the surrounding of the pipeline, see also [106]. This in turn affects the release and dispersion of the  $CO_2$ . Eventually, this has effect on the concentration of  $CO_2$  and impurities in the surrounding of a failing pipeline. These effects are currently not rigorously addressed in existing models. Field-testing and (further) validation of release and dispersion models is thus necessary for a more accurate assessment of the external safety of CO<sub>2</sub> pipelines. Field scale CO<sub>2</sub> release and dispersion experiments have been undertaken by BP and Shell in the recent years, respectively in 2006 and 2010. In the joint industry project CO2PIPETRANS these data are used to validate release and dispersion models. Experiments are also planned within the Dutch CATO2 programme.

The models that are being used to estimate the dispersion of  $CO_2$  can typically be divided into Gaussian/dense-gas models and CFD (computational fluid dynamics) models. The first group of models is more widespread and has typically shorter computation times. It also requires a smaller data set to perform the calculations. Recent studies do however suggest that CFD models can more accurately assess the dispersion of  $CO_2$  and indicate that Gaussian/dense-gas models tend to over-estimate (up to one order of magnitude) concentrations of dispersing  $CO_2$  [107].

Another aspect is that release characteristics, such as the direction (vertical or horizontal) and momentum (impinged or unimpinged jet or instantaneous release), have a significant impact on the outcomes of a QRA. Currently, no uniform assessment methodology prescribes how to cope with assumptions on the direction and momentum of the release.

#### 4.3. Possible impact of accidental release

The estimation of the impact of an accidental release on human safety is highly determined by the methodology used. Some studies assume a concentration threshold for  $CO_2$  and impurities, while other methodologies include a dose-response function.

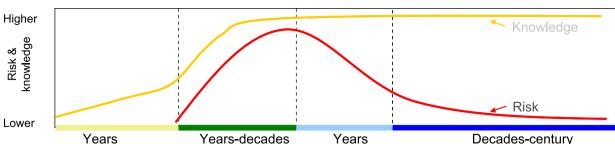
Table 6 shows that the assumed type of threshold has large influence on the outcome of the RA in literature. Consequently, effect distances to these thresholds vary orders of magnitude. Thresholds are often incommensurable as they vary in three main characteristics of the threshold: the concentration, the duration and the effect. The level of the effect belonging to the various thresholds in the reviewed literature varies between 'adverse effect on the environment' and 'fatality'. In the dose-response function, concentration and duration are used to estimate the fraction of fatally injured people. Currently, a variety of concentration thresholds is used worldwide and no formal dose-response function is adopted yet [95,99,108]. Work has been done by ter Burg and Bos [109] to establish such a dose-response (probit) function.<sup>10</sup> They however conclude that more scientific research is needed aimed at understanding the complexity of the relationship between CO<sub>2</sub> concentration, duration of the exposure and the resulting fatality in humans. A probit function could not be proposed in absence of this understanding. Instead, ter Burg and Bos [109] propose to use conservative concentration thresholds<sup>11</sup> as long as a reliable probit function is absent. Reviewing Table 6 also yields the observation that impurities like H<sub>2</sub>S may dominate the risk of CO<sub>2</sub> pipelines.

#### 4.4. Findings CO<sub>2</sub> transport by pipeline

Overall, these limitations of current risk assessment methodologies and models limit the possibility to compare outcomes of QRAs case by case and with existing industrial activities. Difficulties

<sup>&</sup>lt;sup>10</sup> The probit function has the form:  $Pr = a + b \times ln (C^n \times t)$ . Pr is a representation of the response fraction, e.g. percentage of people fatally injured. In this equation a, b and n are substance specific constants describing the lethality related to a dose of a toxic substance, explosion or heat, C is the concentration (in kg/m<sup>3</sup>) and t is the exposure time (in s) [110].

 $<sup>^{11}</sup>$  ter Burg and Bos [109] propose to use the following thresholds: no deaths are expected at CO<sub>2</sub> concentrations of up to 50,000–100,000 ppm, serious effects and possible mortality may start to occur at about 100,000–150,000 and a high level of mortality may occur at about 200,000–250,000 ppm.



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Time

Decades-century

	Time									
Phase	Preparation and construction	Operation (injection)	Dismantling and abandonment	Post closure						
Detection/ Monitoring	-Site screening and ranking; -Geological characterization: capacity, injectivity, containment; -Seismicity baseline; -Baseline monitoring biosphere.	<ul> <li>High frequency and high resolution monitoring of subsurface (incl. well): injection characteristics (pressure, temperature composition, rate) and fate of CO<sub>2</sub>;</li> <li>Seismicity;</li> <li>Monitoring biosphere.</li> </ul>	-Targeted monitoring of geosphere and biosphere.	Decreasing occasional monitoring of geosphere and biosphere. Less monitoring tools available due to closure of well.						
Modeling	-Long/short term simulation: hydrodynamics, geomechanics and geochemistry; -Risk assessment.	Validate/update models and re-evaluate: -Injection plan; -Monitoring plan; -Risk assessment and mitigation/remediation plan.	Validate/update models and re- evaluate: -Monitoring plan; -Risk assessment and mitigation /remediation plan.	Validate/update models and re-evaluate long term : -Monitoring plan; -Risk assessment and mitigation/remediation plan.						
Dominating trapping mechanism		Primary mechanisms (structural, stratigraphic and hydrodynamic trapping).	Increasingly secondary r	nechanisms (residual, solubility and mineral trapping).						

Fig. 3. Indicative graphical representation of the typical four phases of a CO<sub>2</sub> injection project with its relation to monitoring and modeling efforts, dominating trapping mechanisms and the development of knowledge (inverse of uncertainty) and risk over time. Based on [123-125]. Note that the 'knowledge curve' for aquifers in general starts below that of oil and gas reservoirs.

also persist in drawing risk contours<sup>12</sup> for CO<sub>2</sub> pipelines, see also Table 6.

It is recommended that efforts should be undertaken to improve the accuracy of a QRA for CO<sub>2</sub> pipelines. These efforts should be focused on 1) the validation of release and dispersion models for high-pressure CO<sub>2</sub> including impurities and 2) the development of a universal dose-response model for CO<sub>2</sub>.

Best practice guidelines for the design and operation of CO<sub>2</sub> pipelines have been developed recently, providing first guidance steps [111]. We recommend further development and implementation of detailed guidelines for assessing the risk of (highpressure) CO<sub>2</sub> pipelines. These should include a definition of the type of failures that should be assessed, the methodological choices to be made, uniform exposure thresholds and dose-response model, and safety distances for CO<sub>2</sub> pipelines.

#### 5. Activities in the geosphere – storage of CO<sub>2</sub>

Analogous activities to CO<sub>2</sub> storage in the underground are underground gas storage (UGS), acid gas injection and hydrocarbon production projects like EOR. There is extensive experience with these activities worldwide and also with concluding EIA procedures for such activities.

These activities can be used as a point of reference for the EIA procedure for CO<sub>2</sub> storage projects. In addition to concluded and

ongoing EIA procedures for CO<sub>2</sub> storage projects, we have studied analogous activities like storage of natural gas in gas fields and in salt caverns, and EOR projects. The EIA procedures assessed in more detail in our study are presented in Table 7. Worldwide there are more CO<sub>2</sub> storage projects<sup>13</sup> being operated or planned than presented in this table. For these projects either no EIA procedures have been concluded or the accessibility of the related documents was limited, hence they were not reviewed in detail.

The storage of CO<sub>2</sub> in the deep underground encompasses various options: aquifers, (nearly) depleted hydrocarbon reservoirs, underground (unminable) coal layers and salt caverns. The focus in our study is on the first two options as these have the highest estimated storage potential [42].

CO<sub>2</sub> storage in these geological formations encompasses the injection of CO<sub>2</sub> into porous rocks that may hold or have held fluids like gas, oil and brine. Important considerations for choosing a suitable single formation, or geological formations at all for that matter, are the injectivity, capacity and containment [123,124]. The first requires that the permeability of the rock is sufficiently high to enable that the CO<sub>2</sub> can be injected. The second, capacity, is determined by the available space in the rock which is mainly dependent on the dimensions of the formation, the porosity of the rock and the density<sup>14</sup> of the CO<sub>2</sub>. Finally, the containment of CO<sub>2</sub>

<sup>&</sup>lt;sup>12</sup> An individual risk contour depict the probability per year on a topographical map that an unprotected ever-present person dies at a certain distance from the pipeline due to the accidental release of the CO<sub>2</sub>.

<sup>&</sup>lt;sup>13</sup> Currently (2010) operating CO<sub>2</sub> storage projects are: Sleipner and Snohvit, Norway; Weyburn, Canada; In-Salah, Algeria; K-12B, Netherlands; Ketzin, Germany; Otway, Australia.

<sup>&</sup>lt;sup>14</sup> The density of CO<sub>2</sub> increases with increasing depth, i.e. increasing pressure. Therefore, geological storage is considered in formations from a depth of 800 m as CO<sub>2</sub> is, in general, in the supercritical phase from that depth.

should be safeguarded to inhibit the  $CO_2$  from moving outside the target formation. Several short and long-term trapping mechanisms prevent the  $CO_2$  from leaking, see Fig. 3.

One of the most important leakage barriers is the presence of an impermeable rock layer, or caprock, which seals the formation. This feature makes formations that held fluids such as natural gas and oil for geological time attractive for  $CO_2$  storage. This sealing capacity is however not proven for all of the aquifers. These formations are also less studied compared to hydrocarbon formations rendering more uncertainty regarding sealing capacity (containment), injectivity and storage capacity [42].

In this section, we will not address all environmental consequences of  $CO_2$  storage, but only focus on 'new' possible environmental consequences that are most likely to be assessed in an EIA. Furthermore, the focus is on the environmental indicators and tools that are used to determine and communicate these consequences. We will distinguish between several environmental compartments: the underground (including target storage reservoir and wells), the overburden and the biosphere (including atmosphere, groundwater, vadose zone,<sup>15</sup> and surface water).

If we divide the storage activity simply into above ground activity and underground activity, the new environmental concerns are related to the latter. The above ground activity including construction, operation and dismantling of infrastructure can be considered current practice. A clear difference between the EIA procedures for  $CO_2$  storage projects and those for analogous activities is the extensive additional attention to the performance of the geological reservoir. More specific, the safe and long-term storage of  $CO_2$  is an important new issue in these assessments compared to current activities in the geosphere. In the following section we will therefore focus on the risks of  $CO_2$  storage, as this is one of the most important remaining issues in the environmental assessment of  $CO_2$  storage.

We will assess the tools and indicators that are used to assess the Health, Safety and Environmental (HSE) consequences of  $CO_2$ storage. Also, an overview will be presented of tools that are available to monitor the performance and possible effects of  $CO_2$ storage. Finally, we will assess the measures that can be applied to mitigate and remediate HSE consequences in the case of a failure in the containment of  $CO_2$ .

#### 5.1. Leakage scenarios

Several scenarios conceivable may result in the leakage of  $CO_2$  from the target reservoir. Often investigated scenarios are: leakage through existing or induced faults and fractures, leakage along a spill point, caprock failure or permeability increase and leakage along a well and wellhead failure.

Injecting  $CO_2$  in the targeted reservoirs will result in pressure changes in the reservoir which may re-activate faults and fractures. These may result in seismic events. Another consequence may be the creation of preference pathways for  $CO_2$  migration from the reservoir into the overburden which eventually could result in leakage of  $CO_2$  into the biosphere [119,126]. In the Gorgon project therefore a mitigating action has been proposed to limit pressure build-up. This includes the production of water from the reservoir to lower the pressure. This water is planned to be re-injected into another pressure depleted reservoir (as proposed), discharged directly or to be treated and then the effluent is discharged in surface water bodies. This risk mitigating activity at least requires the drilling, operation and abandonment of additional wells, with attached environmental consequences. More detailed information on the environmental interventions and impacts associated with produced water from the analogous oil and gas production projects can be found elsewhere, e.g [127–130].

Injection of  $CO_2$  in the reservoir may also result in a pressure build-up beyond the boundaries of the  $CO_2$  plume [126,131]. In the case of a depleted oil or gas field the pressure will increase towards the original pressure of the reservoir. In case of an aquifer, the pressure will increase above the original pressure in the reservoir. This may result in displacement of brine out of the target reservoir. This brine, including its contaminants, theoretically may come in contact with potable water layers. This indicates that with respect to this matter pressure depleted reservoirs (such as depleted oil and gas reservoirs) are in general favorable over aquifers. For aquifers, the hydrodynamic effect of injecting large volumes of  $CO_2$ needs further scrutiny.

Leakage along a spill point, which is the lowest structural trap of a reservoir, is possible when more  $CO_2$  is injected into the reservoir than can be hold in that reservoir [132].

Another scenario often assessed is leakage through the caprock due to a failure or due to increased permeability of this caprock. The sealing capacity of a hydrocarbon reservoir for  $CO_2$  is in general considered to be high as the caprock has proven to hold the hydrocarbon for geological times. Such a proof is often not available for aquifers. The sealing capacity with respect to aquifers is considered less certain. There are several geochemical or geomechanical processes that may trigger a scenario that results in the failure of  $CO_2$  containment. These are not detailed further here but more information can be found elsewhere [42,132].

Finally, a well can be a pathway for  $CO_2$  to leak into non-targeted environmental media, including the biosphere. This may be  $CO_2$ injection wells, but also old abandoned wells. The corrosion of materials (i.e. cement degradation) used to construct or plug the wells after abandonment is an important process that should be considered for the long time horizon of  $CO_2$  storage. It is therefore necessary to characterize all existing wells before  $CO_2$  injection, including: the location, type and age, in addition to the completion technique and type of materials used. The difference between hydrocarbon reservoirs and aquifers is in this respect that the number of wells drilled through aquifers is in general lower, which renders fewer pathways for leakage [132].

#### 5.2. Effects of fluxes from the underground into the biosphere

Although several trapping mechanisms (see Fig. 3) significantly hinder  $CO_2$  transport through geological strata, it cannot be ruled out on forehand that  $CO_2$  does not end up in the biosphere. When this occurs,  $CO_2$  fluxes will change the concentration of  $CO_2$ in the soil, water bodies and/or atmosphere depending on the size of the flux. As a result, the pH of the (ground)water may decrease and with that possibly mobilizing heavy metals<sup>16</sup> [133–135]. The  $CO_2$  may also act as a carrier gas, transporting other gases such as radon and H<sub>2</sub>S into the biosphere. An extensive review of effects of elevated  $CO_2$  concentrations in abovementioned compartments is provided in [132,136–144]. In some studies also tolerances for selected organisms to  $CO_2$ exposure are presented [136,137].

 $<sup>^{15}</sup>$  The vadose zone is the unsaturated zone between land surface and the groundwater table (saturated zone).

<sup>&</sup>lt;sup>16</sup> Apps et al. [132] conclude that dissolution of pyrite and solubilization of arsenic are the most important concerns for shallow groundwater. Other elements (Ba, Pb and Zn) may in the case of high CO<sub>2</sub> partial pressures also approach or exceed US regulatory concentration limits. This is considered to be unlikely for Cd, and Sb. For Hg, Se and U concentrations are found to be unaffected by CO<sub>2</sub> intrusion.

Maul et al. [139] present a model based on observations from a natural leaking site. This model simulates the response (both toxic and fertilizing) of organisms to elevated  $CO_2$  concentrations. However, these effects are site specific. Beaubien [145] specifically notes that the impact of  $CO_2$  leakage (altered vegetation type and presence, microbial activity) depends on the pathways and spatial distribution of the flux.

Furthermore, environmental impacts depend on the response of local organisms to elevated  $CO_2$  concentrations or changes in groundwater composition. For this response it is important to distinguish between long-term chronic and short-term acute exposure.

For healthy humans the effect of short-term acute exposure is in general well known. The effect of long-term chronic exposure to healthy subjects and the effect of both types of exposure for more sensitive subjects (children, elderly and the infirm) do require further investigation [144].

The effect of long-term exposure on ecosystems can be indicated as a knowledge gap [146]. In addition, it is important to understand and quantify ecosystem recovery rates after remediation of a leakage [144]. Ideally, dose-effect relationships should be known for ecosystems or target species to model the impact of  $CO_2$ releases taking into account the level, duration and location of exposure to the  $CO_2$ .

#### 5.3. Safety of CO<sub>2</sub> storage—the assessment and results

The conceivable scenarios for the leakage of  $CO_2$  discussed above are typically assessed in a risk assessment. The used methodologies for  $CO_2$  storage are mainly based on existing methodologies and tools from the hydrocarbon industry and from underground storage of nuclear waste. Here we focus on the type of indicators they provide us and how these are determined and reported in the EIA procedures.<sup>17</sup> When available, quantitative results of these risk assessments are presented. A concise summary of the approach and results of the reviewed risk assessments is presented in Table 8.

## 5.3.1. Methodologies used in risk assessments of CO<sub>2</sub> storage projects

The information presented in Table 8 yields the insight that there is currently no uniform risk assessment methodology or approach in place for the assessment of possible HSE effects due to  $CO_2$  storage in geological formations, although there are similarities among the methodologies. One similarity is that the approach differs from RAs for 'normal' industrial activities as in the case of  $CO_2$  storage a non-engineered system is assessed. Both the FutureGen and Barendrecht RA therefore split the assessment into an engineered and non-engineered part. The bottleneck for this latter part is however that the performance of that system cannot be assessed with high certainty on forehand.

5.3.1.1. Site characterization. Another similarity is that a site characterization is included in all reviewed studies. Accuracy of the performance assessment of the system, i.e. the assessment of the containment of  $CO_2$ , increases with increasing knowledge of the characteristics of the reservoir and its surroundings, see Fig. 3. In that figure it is shown that the knowledge curve for aquifers would in general start below that of hydrocarbon reservoirs as the latter are already extensively investigated prior and during hydrocarbon removal. The most important characteristics to be assessed are the capacity, injectivity and the containment [124]. This is followed by the identification and characterization of possible leakage pathways in the overburden. The characterization of the reservoir makes it possible to construct a reservoir model to describe the current state of the reservoir and predict possible future states resulting from CO<sub>2</sub> injection.

5.3.1.2. Hazard identification and failure scenarios. Next, an identification of hazards is typically performed by a panel of experts based on the characterization. A tool that is often used is a database<sup>18</sup> that contains several hundreds of Features, Events and Processes - or FEPs - for geological formations. Features are defined as factors that describe the current state of the reservoirs and its surroundings. Events and Processes can be described as factors that change the state of the sequestration system [147].

Expert panels are used to identify relevant FEPs and prioritize these. In this way scenarios can be developed and selected that are based on the relevant FEPs and that may be critical for the safety of  $CO_2$  storage. In the RA for Barendrecht the FEP method has been applied in combination with the Bow-Tie method to systematically order FEPs in cause-consequence chains for the injection and postclosure phase [121].

Table 8 shows that, although there is no RA standard, studies assess comparable failure scenarios. They encompass the leakage scenarios already discussed: leakage along a well and wellhead failure, caprock failure or permeability, leakage along a spill point and leakage through existing or induced faults and fractures.

5.3.1.3. Scenario modeling. The selected scenarios can then be modeled in a (extended) reservoir model to assess the transport and fate of the  $CO_2$  in the reservoir and other environmental compartments. However, data uncertainty is omnipresent and results in uncertain estimates for current and future states. Furthermore, although the behavior of  $CO_2$  in reservoirs has been modeled in EOR projects and experience thus exists, these models were not developed for modeling the fate of  $CO_2$  taking into account detailed (geochemical, geophysical and hydrodynamic) interactions with the reservoir. As a result, these models are not calibrated yet for long-term  $CO_2$  storage [94,121,124,155]

Also, the level of detail of the applied models varies between studies. First, different reservoir models and various differentiations of existing reservoir models are being applied to cope with the special properties of  $CO_2$  and the long-term storage of it. Second, the amount of environmental compartments and the amount of sub-models taken into account, as well as the environmental compartment that is targeted in the RA, also varies.

5.3.1.4. Alternatives to modeling – expert panels and natural analogues. A second general approach is to assess the probability and consequences of failure scenarios qualitatively with the use of an expert panel. A risk matrix with these two dimensions can be used to score the risks. Such an approach was used in the Barendrecht and Gorgon projects. A similar but quantitative approach, called RISQUE (Risk Identification and Strategy using Quantitative Evaluation), is described in [156]. This approach has been applied partially in an assessment of a CO<sub>2</sub> storage project in Latrobe Valley, Australia [98]. In the methodology, first a set of risk events<sup>19</sup> was identified. With the use of an expert panel a qualitative description of the likelihood was attributed to each event which was then converted to a quantitative

<sup>&</sup>lt;sup>17</sup> Not all reviewed risk assessments are necessarily part of an EIA procedure.

<sup>&</sup>lt;sup>18</sup> A comprehensive database containing FEPs can be found at http://www. quintessa.org/consultancy/index.html?co2GeoStorage.html.

<sup>&</sup>lt;sup>19</sup> The risk events identified are: leakage from exploration, production, and injection wells, leakage from permeable zone in the caprock, leakage from faults through caprock, leakage due to regional over pressurization of the reservoir, leakage due to local over pressurization of the reservoir, spill points due to limited storage capacity, leakage due to earthquake induced fractures, leakage due to failure of surface installations (pipeline, compressor and platform).

 Table 8

 Overview of methodologies and results of risk assessments of CO<sub>2</sub> storage projects, including natural analogues and proposed thresholds.

Project description	Methodology	Failure scenarios	Receiving environmental compartment	Results and indicators (L, S or I) <sup>a</sup>
Weyburn CO <sub>2</sub> monitoring & storage project (hydrocarbon) [148]	<ul> <li>Site characterization</li> <li>FEP</li> <li>Deterministic/stochastic scenario</li> <li>Probabilistic scenario (CQUESTRA)</li> <li>Reservoir model (ECLIPSE E-300)</li> </ul>	- Migration from geosphere - Leakage through (abandoned) well bores	Biosphere (including 300 m subsurface)	(L) 0.001(mean)-0.2% CO <sub>2</sub> ip <sup>b</sup> (L) 0.001(mean)-0.14% CO <sub>2</sub> ip (well bore) (L) 16 g day <sup>-1</sup> (well bore) (L)~0.04-2 × 10 <sup>-4</sup> t yr <sup>-1</sup> m <sup>-2</sup> from reservoir (S) CO <sub>2</sub> concentration in layers in geosphere <sup>6</sup>
Safety assessment for Schweinrich structure (aquifer) [149]	- Site characterization - FEP - Simulation discrete scenarios with stochastically varied	- Leakage through caprock - Leakage through faults	Shallow subsurface including groundwater (–80 m to 0 m)	$ \begin{array}{l} \text{(L)} \sim 0 \text{ tyr}^{-1} \text{ m}^{-2} \\ \text{(L)} \ 2.5 \times 10^{-4} - 6.2 \times 10^{-1} \text{ tyr}^{-1} \text{ m}^{-2} \\ \text{(S)} \ < 4\% \text{ concentration in} \\ \text{groundwater at depth of 80 m} \end{array} $
EIA for Barendrecht	parameters in reservoir model (SIMED-II) - Site characterization	- Leakage through well - Leakage through caprock	Subsurface	(L) 60% CO <sub>2</sub> ip (L) 15–350 t yr <sup>-1</sup> m <sup>-2</sup> (L) 0.03 Mt (cumulative)
storage project (hydrocarbon)	- FEP and BowTie - Reservoir model	- Caprock breach - Caprock seepage	(non-target aquifer)	(L) ~1.5% $CO_2ip^d$ (L) <0.1% $CO_2ip^d$
[121,150]	(PETREL, MoRes) - Characterization of risk (qualitative)	<ul> <li>Leakage along spill point</li> <li>Leakage through the</li> <li>well bore along the</li> </ul>	Subsurface/surface	(L) 0-5% CO <sub>2</sub> ip <sup>d</sup> (L) Very long cement leak >800 m: >40% CO <sub>2</sub> ip <sup>d</sup>
	nsk (quintative)	well casing	Subsurface/well	(L) Long cement leak 200-800 m: 9-40% CO <sub>2</sub> ip <sup>d</sup>
			Subsurface/well	(L) Short cement leak $<200$ m: $0-9\%$ CO <sub>2</sub> ip <sup>d</sup>
			Not specified	(L) During operational phase: 1.9 kg-4.4 t/yr
		<ul> <li>Leaking well (potential leak rates through narrow cracks or conduits)</li> </ul>	Not specified	(L) 0.6–1.8 t CO <sub>2</sub> day <sup>-1</sup>
	- Release model - Atmospheric dispersion	- Well blow out and release	Atmosphere	(L) 9–150 kg/s (I) $1 \times 10^{-6}$ Risk contour at
Risk Assessment for the FutureGen Project (sandstone and saline aquifer) [94]	model (SafetiNL) Split in pre- and post-sequestration risk assessment for multiple sites <sup>e,f</sup> Post sequestration	- Leakage into non-target aquifers due to unknown structural or stratigraphic connections and lateral migration	Subsurface (non-target aquifers)	~30–60 m from well (L) 1.39 × 10 <sup>-3</sup> –2.36 × 10 <sup>-1</sup> t yr <sup>1</sup> m <sup>-1</sup>
	risk assessment: - Site characterization	- Leakage due to CO <sub>2</sub> , oil, gas and undocumented wells	Atmosphere	( <b>S</b> ) 60–1490 ppmv at 100 meter from well
	<ul> <li>Analogue database</li> <li>Extrapolation</li> <li>Reservoir model (STOMP)</li> <li>Atmospheric dispersion</li> <li>model (SCREEN3)</li> </ul>	Leakage through: - Caprock failure - Existing and pressure induced faults	Atmosphere	(L) $1.39\times 10^{-3}{-}4.17\times 10^{-2}$ t yr $^{-1}$ m (S) 0.076–4.1 ppmv at 1 m from source
	Pre-sequestration risk assessment: - Release model - Atmospheric dispersion model (SLAB)	- Wellhead equipment failure	Atmosphere	(L) $85-510 \text{ kg s}^{-1} \text{ CO}_2$ (L) $8-51 \text{ g s}^{-1} \text{ H}_2\text{S}$ (S) $2-8 \text{ meter to no effect level}$ (30 000 ppmv CO <sub>2</sub> ) (S) $290-788 \text{ m to no effect level}$ (0.5 ppmv H <sub>2</sub> S)
Environmental Assessment for the Frio Formation (aquifer) [117]	- Methodology not reported in detail	- 10% of CO2ip (max 3750 t) returned to the surface over a 1-year period		<ul> <li>(S) pH of 5.28 (drop of 1.5) in overlying aquifer</li> <li>(S) 100% vapor concentration in the shallow soil</li> <li>(S) nearly 100% vapor concentration in atmosphere near leakage site</li> </ul>
Environmental assessment for Gorgon storage project (aquifer) [118]	<ul> <li>Site characterization</li> <li>Hazard identification</li> <li>Receptor identification</li> <li>Characterization of risk (qualitative)</li> </ul>	<ul> <li>Failure of compressors, pipelines</li> <li>or wellheads</li> <li>Migration along well penetrations, faults or fractures</li> <li>Failure of structural seals</li> </ul>	Surface	(L) $1.4 \times 10^{-3} - 1.4 \times 10^{-1}$ t yr <sup>-1</sup> m <sup>-2</sup>
Risk assessment for Ohio River Valley CO <sub>2</sub> Storage Project (sandstone) [151,152]	<ul> <li>Site characterization</li> <li>FEP</li> <li>Scenario selection</li> <li>Integrated geosphere model (STOMP-CO<sub>2</sub>)</li> <li>Quantification of risk<sup>g</sup></li> </ul>	<ul> <li>Major wellhead failure</li> <li>Moderate wellhead failure, sustained leak</li> <li>Minor wellhead failure, leaks of joints</li> <li>Fractured caprock</li> <li>High permeable zones in caprock</li> <li>Seismic induced</li> </ul>	Atmosphere (A) Buildings (Bl) Groundwater (GW) Surface water (SW) Vadose zone (VZ)	(I) 0.2 (A, Bl, VZ) (I) 0.1 (GW, SW) (I) 5 (A, Bl, VZ) (I) 2 (GW, SW) (I) 1 (A, Bl, VZ) (I) 0.5 (GW, SW) (I) 3 (A, Bl, VZ, GW, SW) (I) 1 (A, Bl, VZ, SW) (I) 2 (GW)

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#### Table 8 (continued)

Project description	Methodology	Failure scenarios	Receiving environmental compartment	Results and indicators (L, S or I) <sup>a</sup>
	Natural analogues <sup>h</sup>			
[153]	Natural flux from		Atmosphere	(L) $2.78 \times 10^{-3}$ – $2.78 \times 10^{-2}$ t yr <sup>-1</sup> m <sup>-2</sup>
	soil to atmosphere			
	Natural flux at		Atmosphere	(L) $2.5 \times 10^{-1}$ – $5 \times 10^{-1}$ t yr <sup>-1</sup> m <sup>-2</sup>
	volcanic active area			
	(Mammoth Mountain)			
[154]	Natural flux at Solfatara, Italy		Atmosphere	( <b>L</b> ) 1.10 t yr <sup><math>-1</math></sup> m <sup><math>-2</math></sup>
	Natural flux at		Atmosphere/	(L) $4.43 \times 10^{-1} \text{ t yr}^{-1} \text{ m}^{-2}$
	Albani Hills, Italy		Groundwater	
	Natural flux at		Atmosphere	(L) $1.46 \times 10^{-1}$ -7.31 × $10^{-2}$ t yr <sup>-1</sup> m <sup>-2</sup>
	Mátraderecske, Hungary			
	Natural flux at		Atmosphere	(L) $3.65 \times 10^{-2} \text{ t yr}^{-1} \text{ m}^{-2}$
	Paradox Basin, UT, USA			
	Natural flux at Latera, Italy <sup>i</sup>		Atmosphere	( <b>L</b> ) 28 t yr <sup><math>-1</math></sup> m <sup><math>-2</math></sup>
	Thresholds			
[143]	Threshold based on		Groundwater	(L) $1.7 \times 10^{-4} \text{ kg d}^{-1}$
	Pb mobilization			$6.21 \times 10^{-5} \text{ t yr}^{-1}$
	Threshold based on 3500		Atmosphere	(L) 5.4 kg $d^{-1}$
	ppmv in air			$1.97 \text{ t yr}^{-1}$

<sup>a</sup> L = Leakage indicator (flux or total amount of  $CO_2$  leaked), S = Indicator for the state of the environment (e.g.  $CO_2$  concentration), I = Impact indicators measuring the possible impact on target species.

<sup>b</sup>  $CO_2ip = CO_2$  in place.

<sup>c</sup> Time dependent aqueous CO<sub>2</sub> concentration profiles beyond the boundaries of the reservoir are presented; a simple range cannot be presented here. See for details [148].

<sup>d</sup> Theoretical leak quantity if no barrier or time limits would apply.

<sup>e</sup> Pre-sequestration risk assessment encompasses the engineered system, including pipelines and wellhead failure. In this table only a selection of results of the RA for the wellhead failure is presented. In Table 6, a selection of results of the RA for pipelines is presented. Post-sequestration encompasses leakage due to storage failure.

<sup>f</sup> All values show the range reported for the four assessed sites/reservoirs. The additional indicator measuring the impact on target species reported in this study is not presented here, as this indicator is a risk ratio that is derived through dividing the calculated concentration by various toxicity thresholds for CO<sub>2</sub> (and H<sub>2</sub>S), i.e. no dose-effect relationship is used.

g Risk is defined as Risk = Frequency of occurrence × Consequence × 100 000. Frequency of occurrence is defined as "events/year for well failures, and percent area of a 50 km radial zone around the injection well occupied by fault zones/high permeability features in the case of cap rock failures". Consequence is characterized Low (0.1), Moderate (0.5) and Severe (1) based on concentrations calculated for the various environmental compartments [151,152].

<sup>h</sup> More natural analogues are reported in [94].

<sup>i</sup> Flux estimate comes from [94].

probability (probability/1000 yr). Leakage rates per event and per year, as well as the total duration of the leakage, were quantified by the expert panel. Then, a risk quotient was defined as the product of likelihood and consequence. The outcomes were compared with a pre-determined maximum acceptable risk quotient.

A third RA methodology is to match the target storage formation with natural analogue sites where  $CO_2$  is contained in the underground or where leaks into the biosphere occur. Characteristics of candidate site are first matched with those of natural sites. The release characteristics (pathways, magnitude, probability and duration) of those 'best fit' analogues are then extrapolated to the candidate site. Thus, based on similar geological characteristics possible leakage fluxes are estimated. This approach has been applied in the FutureGen study.

5.3.1.5. Quantification of risks. Theoretically, a quantitative risk assessment can be performed for a  $CO_2$  storage activity. Based on results from geosphere modeling and quantitative estimates for the probability of each scenario, a quantitative score for the risk (i.e. a product of probability and consequence) of a storage failure can be presented. The additional step performed here is that the concentrations of  $CO_2$  or pH values in environmental compartments are translated into indicators to measure possible impacts. A simple approach based on a simple dose-effect relationship has been suggested and applied by Saripalli et al. [151,152], see Table 8.

Regarding the environmental compartments, it can be seen that only the RA for the FutureGen and Ohio River site include an atmospheric dispersion model to assess the concentration of  $CO_2$  in the atmosphere. In the other RAs atmospheric dispersion is not included. Exposure of target species (e.g. humans) to  $CO_2$  is then difficult to determine and quantify. We deem the inclusion of atmospheric dispersion models in RAs for geological storage desirable when failure scenarios suggest possible leakages to the atmosphere.

5.3.1.6. Post closure phase uncertainties. Fig. 3 shows that for  $CO_2$  injection projects an additional phase is included compared to typical analogous projects, the post-closure phase. This brings forth additional uncertainties as current practice in the oil and gas production and injection sector is not aimed at assessing the long-term performance of the underground reservoir. Typical challenges mentioned by Cooper [124] related to this extended time horizon are: data limitations, dynamic modeling of  $CO_2$ , long-term subsurface interactions and caprock characterization.

Not shown in Table 8 is the timeframe that is taken into consideration when assessing the risks. For these studies the mentioned timeframe ranges between 100 and 10,000 years. This suggests that if results from these studies are to be compared, this difference may have an effect on the results. That is, the cumulative probability of failure will increase when longer time horizons are considered. However, annual failure probabilities will likely decrease with time as secondary trapping mechanisms like mineralization and dissolution will play a more important role, see Fig. 3.

5.3.1.7. Estimation of failure frequencies. The estimation of failure rates (quantitative of qualitative) for the non-engineered part of the storage system relies heavily on expert judgment. Certainly compared to the assessment of failure rates used in QRAs for industrial installations, which is more based on historic figures for failure rates. For the failure scenarios for  $CO_2$  storage in general no historic data are available [151]. The expert judgments used instead are mainly based on experience in the oil and gas industry, from natural analogue studies and through modeling [94,98,151,152]. This provides

CO2 containment issues and their mitigation/remedial measures suggested in EIA procedures for CO2 storage projects and general literature on CCS.

Environmental concerns regarding CO <sub>2</sub> storage	Mitigation/remediation	Source
Leakage and seepage through/alongside Caprock: -Catastrophic failure and quick release -Gradual failure and slow release	<ul> <li>-Injection/reservoirs pressure</li> <li>lower than initial pressure</li> <li>-Injection pressures up to 85</li> <li>percent of fracture gradient<sup>a</sup></li> <li>-Control composition CO<sub>2</sub></li> <li>-Avoid fracturing conditions</li> <li>-Stop injection, remove CO<sub>2</sub> from reservoir</li> <li>-Detecting leakage (mass balance, seismic monitoring, monitoring impact zone)</li> </ul>	[94,121]
-Leakage along wells including shallow accumulation -Upward leakage through existing deep oil and gas wells	Monitoring: -Pressure in annulus of the well -Analysis for gas in well annulus Mitigation: -Use state of the art drilling and completion techniques -(Re)completion of unused wells -Reworking deep wells -Appropriate plugging of wells -Early abandonment if well integrity is doubtful. -Common O&G industry mitigation techniques for leaking wells	[94,113,120,121]
Release through induced faults resulting from increased pressure (local over-pressure)	-Determine induced/activated fractures through seismic monitoring -Detect micro-seismicity -Alter injection strategy -Reduce injection pressure -Venting CO <sub>2</sub> from reservoir -Move to another injection well -Water production from reservoir	[94,120,121]
Leakage into non-target aquifers due to unknown structural or stratigraphic connections and due to lateral migration beyond spill point	-Stop injection -Remove $CO_2$ accumulation	[94,121]
Upward leakage through undocumented, abandoned, or poorly constructed wells	-Survey field for existing wells -Remote sensing (through satellites), atmospheric monitoring, surface and near surface monitoring and subsurface monitoring.	[94]
Induced fracturing (as consequence of UGS injection/production cycle)	-Monitor and control of temperature and pressure of $\rm CO_2$ -Minimal distance injection well and fault: -200 m	[113,126,162]
Effects of pressure development due to injection/production -Earth subsidence or uplift -Seismicity	Monitoring: -Seismographic -Water leveling Mitigation: -Production of reservoir fluids	[113,119]
Leakage to near-surface environmental compartments	-Sanitize groundwater -Sealing well zone -Building modification	[120,141]

<sup>a</sup> The pressure required to induce fractures in rock at a given depth.

a perspective on the order of magnitude of the probability of failure, but cannot be compared with the more certain failure rates used in QRAs for engineered systems. The failure rates for non-engineered system are in addition highly site specific as they depend on sitespecific geological characteristics. The geographical extent and natural heterogeneity of a failing system makes it also possible that there is a spatial distribution of the rate of failure and thus of risks.

#### 5.3.2. Results and indicators

The indicators that are used to report on the consequences are typically: the fraction of injected amount leaked (in %), the total amount leaked (in t  $CO_2$ ) and the flux (in t  $m^{-2}$   $yr^{-1}$  or t  $yr^{-1}$ ). Table 8 shows that the RAs deal with various environmental compartments. The subdivision of environmental compartments is not equal in all studies. The indicator (most often the flux) is sparsely presented for multiple environmental compartments nor

is the entire pathway of the CO<sub>2</sub> including multiple compartments presented with the use of quantitative indicators.

A performance indicator for the reservoir for which also a safety limit is suggested is the reservoir pressure. In the Barendrecht project it was stated that the reservoir pressure may not exceed 95-97.5% of the initial reservoir pressure before gas production due to injection of CO<sub>2</sub>. For aquifers such a limit should also be developed.<sup>20</sup> Indicators that have been presented in EIAs and RAs for compartments other than the reservoir comprise the concentration of CO<sub>2</sub> expected in the groundwater and atmosphere [94,149]. Based

 $<sup>^{20}</sup>$  In the Gorgon project where the CO<sub>2</sub> is injected into an aquifer, a pressure management program is developed to ensure that reservoir pressure is 'below acceptable levels'. Quantitative description of what such a level would be, or what the expected reservoir pressures are, is however lacking.

on literature some basic performance indicators per compartment can be suggested, although site specificity of geological storage may require an unique set of performance indicators:

- Reservoir: pressure, temperature and location of CO<sub>2</sub>;
- Well: annular pressure, gas composition well annulus;
- Groundwater: pH, CO<sub>2</sub> partial pressure, concentration of As, Ba, Pb and Zn;
- Overburden: amount of CO<sub>2</sub> (or other displaced substances) in non-target reservoirs;
- Soil: CO<sub>2</sub> concentration, earth subsidence/uplift;
- Atmosphere: CO<sub>2</sub> concentration.

As far as we can ascertain, no formal guidelines are set to limit the concentration of  $CO_2$  in compartments other than the atmosphere. Stenhouse et al. [143] use existing (US) limits for groundwater and implicitly suggest using the concentration of lead in groundwater as an indicator for the performance of the  $CO_2$  storage reservoir. Then it is possible to estimate the maximum amount of  $CO_2$  that is acceptable to leak into the groundwater from the geosphere, yielding a limit for the indicator. Wilson and Monea [148] propose limits (see Table 8) for the flux of  $CO_2$  from the geosphere into the atmosphere based on concentration limits for  $CO_2$  in the atmosphere. Saripalli et al. [151,152] have set concentration thresholds for several environmental compartments and biota. These studies thus provide preliminary tools to develop risk acceptance criteria.

There are very few indicators for concentrations or consequences reported in RAs and EIAs. These indicators are reported in the studies that include an atmospheric dispersion model. For instance, in the Ohio River Valley case indicators for consequences are presented. Although a simple dose-effect relationship was assumed, this is the only RA that reports quantitative indicators up to the level of 'impact' (not just flux or effect on concentration) for several relevant environmental compartments, with the note that consequences of elevated CO<sub>2</sub> concentrations are highly dependent on local conditions. For human safety also an 'impact' indicator is suggested in the FutureGen project. This is however not an indicator based on dose-response modeling.

Furthermore, it is important to stress that it is very difficult, if not impossible, to assess the location where the  $CO_2$  may enter the biosphere in case of a leakage other than a leaking well. Drawing iso-risk contours, an indicator for the possible impact used in some countries, to depict external safety on a topographical map is thus not possible for all leakage scenarios. When setting norms for such indicators they should be tailored to  $CO_2$  storage to cope with uncertainties regarding the spatial distribution of probabilities of failure and its impact.

It can be conservatively concluded from Table 8 that leakage along or through a well bore is the scenario with the highest indicator values, suggesting the highest risk. Furthermore, if co-sequestration of  $H_2S$  is to be applied it should be taken into account that the RA outcomes presented here show higher risks for  $H_2S$  than for CO<sub>2</sub>, even when present in low concentrations.

It should be noted that the results of the RAs presented in Table 8 represent the results of worst-case scenarios and not that of likely scenarios for the evolution of the storage reservoir. These results can be used to conservatively compare failure scenarios and to develop appropriate monitoring plans, as well as plans for mitigation and remediation of risks.

#### 5.4. Monitoring, mitigation and remediation

#### 5.4.1. Monitoring

Monitoring various environmental compartments before, during and after closure of CO<sub>2</sub> injection projects is crucial to understand the fate of the injected  $CO_2$ , the effects it has on the reservoir and surrounding, and possible impacts of leakages. It is furthermore essential to calibrate and possible improve the models that are used to assess the future (short- and long-term) state of the  $CO_2$  in the subsurface, see Fig. 3. Finally, monitoring is required to assess the effectiveness of remedial actions. It is however stressed in literature that monitoring of the deep subsurface inherently comes with uncertainty and is expected to remain so in the future, despite developments in monitoring tools [124,155].

The 'new' part of the monitoring plan for  $CO_2$  storage projects is aimed at monitoring the containment of  $CO_2$  in the underground. Several techniques<sup>21,22</sup> are proposed for different phases of the project (see also Fig. 3) to monitor various environmental subcompartments, being: the underground (including reservoir and wells), the overburden and the biosphere (including atmosphere, groundwater and surface water). From these three subcompartments the reservoir and wells are predominantly monitored, with the principal function to control the injection process. Monitoring of the biosphere is new compared to the standard in oil and gas industry [121]. Further additions compared to the oil and gas industry include: remote sensing (with satellites), more frequent seismic surveys and the employment of more monitoring wells in the soil layers above the reservoir [122].

An observation that we made from reviewing the monitoring plans is that the monitoring of  $CO_2$  storage projects focuses on assessing indicators for containment (i.e. possible leakages) and measuring the state of the reservoir and overburden. Considerably less attention is paid to indicators measuring possible impacts on target species. An example for this observation is presented in the EIA documents for the Gorgon project for which no definite monitoring plan<sup>23</sup> is yet provided. However, provisionally it is aimed at: monitoring the  $CO_2$  plume migration in the subsurface with time lapse (4D) seismic techniques, surveillance of surface  $CO_2$  fluxes and monitoring injection characteristics (e.g. pressure and rates) [118,119].

Monitoring tools are essential to measure the performance indicators and make benchmarking against the norms possible. It should be stressed however that the accuracy of measuring these indicators for the deep subsurface is challenging and comes with uncertainties. When formulating norms for the various environmental compartments this should be properly acknowledged. The development of norms and a site-specific optimal set of monitoring tools and plans should thus be in close harmony.

#### 5.4.2. Mitigation and remediation

In conjunction with monitoring also preventing, mitigating or remedial actions are crucial. These actions may counteract the occurrence, effects and impacts of failure of the injection and

<sup>&</sup>lt;sup>21</sup> This encompasses techniques to monitor: the injection and monitoring well (annular pressure, integrity of casing and cement,  $CO_2$  concentration), pressure around the well plugs, presence and distribution of  $CO_2$  near the well, small leakages around the wells with acoustic surveys, surface injection flux (debit, quality, temperature and pressure), subsurface pressure and temperature in well and reservoir, seismicity, ground movement and  $CO_2$  concentration in air and ground-water near wells.

<sup>&</sup>lt;sup>22</sup> An overview of monitoring techniques that are (to be) used at currently operating or planned injections sites (In Salah, Sleipner and Snøhvit) is presented in [157]. It encompasses surface and subsurface monitoring, the latter being subdivided into seismic and non-seismic monitoring. More extensive overviews and selection support tools for monitoring techniques are presented elsewhere, i.e. see [158,159].
<sup>23</sup> The following statement is made: "While not directly aimed at detecting

<sup>&</sup>lt;sup>23</sup> The following statement is made: "While not directly aimed at detecting impacts from migration of reservoir CO<sub>2</sub> on the environment, these monitoring programs will provide verification with respect to any impacts from reservoir CO<sub>2</sub> leakage."[119].

storage activity. Furthermore, improving mitigating techniques for CO<sub>2</sub> storage is according to Singleton et al. [160] crucial for facilitating public acceptance.

For CO<sub>2</sub> pipelines, an engineered system, risk mitigation options are readily available. This is different for the non-engineered subsurface part of the CO<sub>2</sub> storage activity for which the mitigation options are still under development. Minimizing risks starts with the screening and selection of suitable reservoirs. This is followed by detailed characterization of the reservoir and identification of faults, fractures and (abandoned) wells (i.e. possible leakage pathways) which comes with uncertainty [94]. The configuration, location, design and completion of wells form also an important part of a strategy to reduce the risk of leakage. The injection strategy then also should take into account the expected pressure development in the reservoir to prevent unwanted processes in the reservoir and its surroundings. Then mitigation and remediation can be aimed at the possible source of the CO<sub>2</sub>, which are the wells and reservoir, or aimed at the environmental compartment that is affected by the leakage. Benson and Hepple [153] as well as Cooper [124] have presented an overview of possible mitigating and remedial actions that can be summarized and simplified as actions to:

- Lower reservoir pressure to mitigate CO<sub>2</sub> leakage and other consequences by altering the injection strategy (pressure, rate, total volume) and producing water or eventually CO<sub>2</sub> from the reservoir;
- Carry out the recompletion, workover or plugging of (abandoned) wells to mitigate CO<sub>2</sub> leakage;
- Remove the accumulated CO<sub>2</sub> from the environmental compartment in the subsurface where it has leaked into. (In the case of leakage into groundwater, the groundwater can be produced, cleaned and reinjected);
- Dilute the CO<sub>2</sub> to remediate impacts of exposure to CO<sub>2</sub>, e.g. with fans or even helicopters<sup>24</sup> in the case where CO<sub>2</sub> has leaked into the atmosphere.

In Table 9, a more detailed overview is presented of monitoring, mitigation and remedial actions that have been proposed in scientific literature and in EIA procedures for CO<sub>2</sub> storage projects.

Reviewing existing mitigating and remedial actions yields the insight that mitigation is aimed at controlling leakages from the well and reservoir as these are considered the most important failures. Mitigating actions for the overburden and near surface are addressed only very limited at present. Considering the importance of public acceptance in CO<sub>2</sub> storage projects and the role risk plays in that process it is deemed necessary to focus on developing more possibilities to mitigate and remediate risks, taking into account all environmental compartments.

#### 5.5. Findings geological CO<sub>2</sub> storage

In the risk assessments and EIA procedures typically the following failure scenarios are assessed: well failure, caprock failure and leakage through faults or fractures. It is, however, not formally put down in risk assessment guidelines whether or how these scenarios should be derived and assessed. Currently, different approaches for the RA exist with their own merits and limitations. Moreover, the execution of a risk assessment of CO<sub>2</sub> storage is highly site specific and for a considerable part based on expert judgment. This implies that significant emphasis should be put on

how - in addition to which - failure scenarios are developed, selected and assessed.

Exhibit AC-2, Page 20 of 25

Further, models of sub-compartments (vadose zone groundwater, surface water bodies, atmosphere, reservoir, overburden and well) are extremely limited and are being improved or being developed to deal with CO<sub>2</sub>. Maul et al. [146] however rightfully conclude that 'the development of models that satisfactorily represent the whole system remains at an early stage'. The next step is thus to integrate or correctly couple these models to make an assessment possible for all relevant performance indicators (e.g. pressure, CO<sub>2</sub> concentration, pH, amount of CO<sub>2</sub> leaked, temperature etc.). This also stresses the importance of formulating clear performance indicators and thresholds for them. With it, also a clear distinction between the environmental compartments possibly affected by CO<sub>2</sub> storage should be pursued.

To assess the possible consequences of leakage scenarios, fluxes of  $CO_2$  between environmental compartments can be modeled or estimated, though with significant uncertainty. However, using these fluxes to assess effects and impacts on the various organisms and ecosystems present in the various environmental compartments is currently a missing link. We recommend therefore that dose—response models for ecosystems or target species are developed and applied, taking into account site specificity.

The assessment of failure rates for most of the possible leakage scenarios lacks an empirical base and is heavily dependent on expert judgment. There is also no methodological standard on whether and how these scenarios should be modeled to estimate the risk using quantitative indicators.

To deal with the uncertainties mentioned above, we recommend a stepwise approach starting with an intensive (e.g. annual) evaluation cycle of  $CO_2$  storage activities, including: planning, modeling, monitoring, verification and calibration, evaluation, planning etc. This iterative cycle should focus on the operational phase and postclosure phase. With assuring monitoring results it then can be decided to gradually reduce the frequency of this cycle and reduce the intensity of monitoring depending on the outcomes of an evaluation using above recommended performance indicators.

A best practice guide could be a platform to implement these recommendations. For aquifers a best practice guide for the design and operation of  $CO_2$  storage projects is developed by Chadwick et al. [163]. The development and integration of best practice guides for other geological reservoirs would be valuable too, as it would reduce the uncertainty for both operators and regulators regarding the design and operation of the  $CO_2$  storage project. Furthermore, it would on a more strategic level ease the screening and selection of storage reservoirs. This is pursued to be filled with the  $CO_2$  QUALSTORE project [164–166]. The  $CO_2$ QUALSTORE

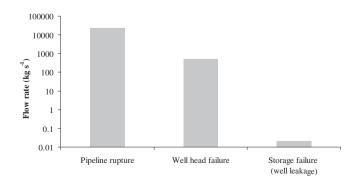


Fig. 4. Maximum flow rates reported for failure scenarios in risk assessments for  $CO_2$  transport and storage activities reviewed in this study.

 $<sup>^{24}</sup>$  Following an unwanted release from the CO<sub>2</sub> fire suppressing system in a factory in Mönchengladbach (Germany) helicopters were used to force dispersion of the CO<sub>2</sub> [161].

Key issues in the assessment of environmental interventions regarding CO <sub>2</sub> capture, transport and storage	ge.
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	Indicator	Models/tools	Regulations
Capture	<ul> <li>Atmospheric emissions quantified but uncertain.</li> <li>Co-benefits (PM, SO<sub>x</sub>, HCl, HF) and trade-offs (NO<sub>x</sub>, NH<sub>3</sub>) probable due to application of CO<sub>2</sub> capture. Depends on applied capture technology.</li> <li>Emissions of solvents and degradation products (focus: post- combustion).</li> <li>Limited quantitative data available on emissions to water and solid waste streams.</li> <li>Water consumption increase due to capture.</li> </ul>	<ul> <li>No reliable emission factors for emissions to water and air. No model seems available that models waste generation for capture technologies (focus recommended: coal fired post-combustion and oxyfuel).</li> <li>Possibly adaptation to atmospheric models needed to cope with 'new' emissions due to capture.</li> <li>See transport for issues of release and dispersion modeling of CO<sub>2</sub> from the engineered system.</li> </ul>	<ul> <li>No BREF and BAT</li> <li>Should emission standards take into account efficiency penalty?</li> <li>Emission and concentration norms for solvent emission and their degradation products should be formulated</li> </ul>
Transport	- Characteristics of released content are, within boundaries, uncertain. Maximum reported release rate is 22 t s <sup>-1</sup> . - Concentration of CO <sub>2</sub> and impurities in surrounding of a failed pipeline is assessed to be above concentration thresholds at up to 7.2 km. - Impact ( $1 \times 10^{-6}$ risk contour) of CO <sub>2</sub> pipelines is assessed to be possible up to 3.3 km based on a concentration threshold. With a preliminary probit function this contour extends up to 204 m.	<ul> <li>Probability of infrastructure failure requires scrutiny.</li> <li>Release models should include impurities and thermophysical properties.</li> <li>Release/dispersion model validation for high-pressure CO<sub>2</sub> release.</li> <li>Dose-response models (e.g. probit function) for target species (or ecosystems) should be developed depending on environmental compartment. Currently, these models are not (yet) available.</li> </ul>	<ul> <li>Pipeline standards are absent, although work is performed in this area.</li> <li>In QRA no standardized failure scenarios are formulated.</li> <li>No formal limits for release of CO<sub>2</sub> and impurities.</li> <li>Uniform atmospheric concentration limits for CO<sub>2</sub> to be used in RA.</li> <li>No formally adopted safety distances for CO<sub>2</sub> pipelines.</li> </ul>
Storage	<ul> <li>Characteristics (total amount and speed) of fluxes (e.g. CO<sub>2</sub> and brine) between environmental compartments can be quantified, although with high uncertainty. Maximum release rate from storage activity in reviewed RAs is 0.5 t s<sup>-1</sup>.</li> <li>The state (e.g. CO<sub>2</sub> concentration, pH) of a compartment is not frequently reported.</li> <li>Impact indicators per compartment are reported in RAs although sparsely for risks caused by failure of the geological storage system. No risk contours can be drawn as not all leakage pathways are known.</li> <li>Overall: No clear performance indicators per environmental compartment.</li> </ul>	<ul> <li>Failure scenarios are typically: leakage along well and wellhead failure, caprock failure and leakage through faults or fractures and leakage along spill point.</li> <li>CO<sub>2</sub> dispersion and transport models, reservoir models are not validated for long-term CO<sub>2</sub> storage.</li> <li>Integration of models for subsurface and biosphere is at an infant stage.</li> <li>See 'Transport' for issues of release and dispersion modeling of CO<sub>2</sub> from the engineered system.</li> <li>RA Tools rely highly on expert panel to (depending on approach):</li> <li>Identify and select failure scenarios;</li> <li>Characterize/quantify failure rates;</li> <li>Characterize consequences.</li> </ul>	<ul> <li>Best practice manuals for CO<sub>2</sub> injection are being developed</li> <li>Monitoring and reporting guidelines for, and prescription of, the exact characteristics of the injected CO<sub>2</sub> are not formulated</li> <li>Standardized methodology for the development of failure scenarios and reporting.</li> <li>Monitoring/reporting standards and limits for fluxes between compartments are absent.</li> <li>Monitoring/reporting standards and norms specific for the various environmental compartments are absent.</li> <li>Uniform atmospheric concentration limits CO<sub>2</sub> to be used in RA</li> <li>Standard Safety distances not formulated.</li> </ul>

guideline for selection, characterization and qualification of storage projects stipulates once more that development of project specific performance indicators, targets and norms is an iterative process that requires a dialogue between project developer(s) and regulators. This guideline also recommends that permit review or renewal includes the re-assessment of the risk profile and uncertainties of the storage project. This may include the addition and the up/downgrading of risks. It also may result in the revision of the set of site specific performance indicators and norms.

In conclusion, levels of acceptable risk and the methodologies to assess, measure, monitor and report on those risks should be defined on a case-by-case basis. An iterative and interactive dialogue between the key stakeholders is recommended to ensure that state of the art knowledge is included in the risk management of storage projects. This also contributes to a transparent process that demonstrates the general public how risks and uncertainties are managed.

#### 6. Comparing risks of CCS activities

Comparing the risk of  $CO_2$  transport and storage activities can be done to place these risks into perspective. It is however not judicious to use the results of such a comparison to provide any argument for the acceptance of these risks, see also [167]. A systematic comparison between the risks of  $CO_2$  pipelines and  $CO_2$  storage is rather difficult and could not be done within this study. The outcomes of RAs reviewed in this study for the CCS activities are incommensurable as not all RAs use and present a risk indicator in the form of the product of likelihood and consequence of a failure. It is however possible to compare maximum reported flow rates as is done in Fig. 4.

This flow rate is reported for various failure scenarios of CCS activities and can be considered a proxy for the consequence of the failure. Depending on the magnitude of the flow rate different HSE issues are of importance. High and local flow rates may have an acute effect on human safety. Contrarily, low and dispersed flow rates may have an effect in the case of long-term chronic exposure. This yields the insight that acute effects on human safety are, if at all, more likely<sup>25</sup> for CO<sub>2</sub> transport activities compared to CO<sub>2</sub> storage activities. Furthermore, Fig. 4 indicates that the maximum flow rates resulting from a pipeline or wellhead failure are orders of

<sup>&</sup>lt;sup>25</sup> This should not be confused with the probability of occurrence.

magnitude higher than that of a leaking well. This neglects however the probability of occurrence and local conditions which are crucial when determining risk with quantitative indicators. Clearly, an equal comparison can thus not be made at this moment.

#### 7. General conclusions

The goal of this study was twofold: (1) to identify and characterize existing and new environmental interventions associated with  $CO_2$  capture, transport and storage that are typically addressed in EIA and SEA procedures; (2) to assess whether crucial environmental information is lacking that may postpone the implementation of CCS projects and plans. To fulfill the goal of this study we carried out a literature study reviewing (analogous) EIA procedures and scientific literature on  $CO_2$  capture, transport and storage.

It should be stressed that it was not the goal of our study to assess whether the knowledge on environmental consequences of CCS is satisfactory to allow competent authorities to issue the permit(s) for CCS activities. This is up to the competent authority or, eventually, the judicial system to decide.

We have however identified several knowledge gaps that deserve proper acknowledgement in a formal decision making process for CCS activities. In Table 10, the key issues regarding the assessment of environmental interventions of the considered CCS activities are summarized. If unresolved, they may have the potential to postpone the implementation of CCS.

For the first step in the CCS chain, CO<sub>2</sub> capture from power plants, we found that changes in key atmospheric emissions ( $NO_x$ , SO<sub>2</sub>, NH<sub>3</sub>, particulate matter, Hg, HF and HCl) are expected. The largest increase is found for the emission of NO<sub>x</sub> and NH<sub>3</sub> when equipping power plants with post-combustion capture. A decrease is expected for SO<sub>2</sub> emissions, which are low for all power plants with CO<sub>2</sub> capture. Additional research (measurements and modeling) and regulatory efforts (norm setting) are required to cope with 'new' emissions from predominantly post-combustion CO<sub>2</sub> capture technologies. Furthermore, an increase in water use (32%–93%), resources, and waste and by-product formation is expected per net generated kWh. The composition, volume and mass of these waste streams is not fully known and thus environmental trade-offs by shifting for instance substances from atmospheric to aqueous emissions or to a solid waste stream are not fully acknowledged. We recommend that environmental monitoring programmes for pilot/demonstration plants should help to quantify these issues in further detail.

For the second step in the CCS chain, high-pressure  $CO_2$  transport by pipelines, we found several important knowledge gaps to be present in the assessment of risks of  $CO_2$  pipelines. The foremost gap is the absence of validated release and dispersion models for high-pressure  $CO_2$  pipeline failures. Another challenge is the assessment of the effects of impurities on operation, failure rates and HSE impacts. Considerable research efforts are being undertaken to close these gaps.

We recommend the further development and implementation of guidelines for assessing the risk of (high-pressure) CO<sub>2</sub> pipelines. These should include a definition of the type of failures that should be assessed, the methodological choices to be made, uniform exposure thresholds and dose-response model, and eventually safety distances for CO<sub>2</sub> pipelines.

For the final step in the CCS chain, we found that the safe and long-term storage of  $CO_2$  could be an important issue compared to environmental assessments for current proficient activities in the geosphere. This study has identified several challenges with respect to the assessment of risks. One of these challenges is a detailed characterization of storage formations and overburden. Subsequently, the validation of reservoir models is needed to make the assessment of performance indicators possible. Guidelines have been published to support project developers and regulators in developing site-specific norms and associated performance indicators. This set of performance indicators and norms should inherently linked with action plans for monitoring, mitigation and remediation.

The execution of a risk assessment of  $CO_2$  storage is highly site specific and for a considerable part based on expert judgment. We further conclude that it is currently not possible to execute a QRA for the non-engineered part of the storage activity with high confidence. Uncertainty is however expected to be reduced when learning-by-injecting increases. An iterative and interactive dialogue between the key stakeholders is therefore recommended to ensure that state of the art knowledge is included in the risk management of geological storage projects. This also contributes to a transparent process that demonstrates the general public how risks and uncertainties are managed.

We recommend the further development of guidelines for risk assessment. In absence of a methodological standard, the focus of the guidelines should be on the development of uniform reporting standards, especially, concerning parts of the assessment that heavily rely on expert judgment.

In conclusion, most gaps in environmental information regarding the CCS chain were identified and characterized for the underground part of the storage activity. This holds especially for aquifers in comparison with hydrocarbon reservoirs. This should however not be confused with an assertion on the magnitude of environmental consequences. That is, most environmental interventions and impacts are expected to be induced in the operational phase of the power plants with CO<sub>2</sub> capture.

Regarding the safety of CCS, it is found that the  $CO_2$  release in case of a failure is reported to be the highest for the transport activity. Although the failure of the underground  $CO_2$  storage system appears to have limited consequences, suggesting a low risk, the uncertainty regarding the assessment of the risk has the potential to become a bottleneck for wide scale implementation of CCS if not properly addressed.

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#### Appendix. Supplementary data

Supplementary data associated with this article can be found in online version at doi:10.1016/j.pecs.2011.05.002.

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