

1 **Multiregional environmental comparison of fossil fuel power**  
2 **generation – assessment of the contribution of fugitive emissions**  
3 **from conventional and unconventional fossil resources**

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21 **ABSTRACT**

22 In this paper we investigate the influence of fugitive methane emissions from coal, natural  
23 gas, and shale gas extraction on the greenhouse gas (GHG) impacts of fossil fuel power  
24 generation through its life cycle. A multiregional hybridized life cycle assessment (LCA)  
25 model is used to evaluate several electricity generation technologies with and without carbon  
26 dioxide capture and storage. Based on data from the UNFCCC and other literature sources, it  
27 is shown that methane emissions from fossil fuel production vary more widely than  
28 commonly acknowledged in the LCA literature. This high variability, together with regional  
29 disparity in methane emissions, points to the existence of both significant uncertainty and  
30 natural variability. The results indicate that the impact of fugitive methane emissions can be  
31 significant, ranging from 3 % to 56 % of total impacts depending on type of technology and  
32 region. Total GHG emissions, in CO<sub>2</sub>-eq./kWh, vary considerably according to the region of  
33 the power plant, plant type, and the choice of associated fugitive methane emissions, with  
34 values as low as 0.08 kg CO<sub>2</sub>-eq./kWh and as high as 1.52 kg CO<sub>2</sub>-eq./kWh. The variability  
35 indicates significant opportunities for controlling methane emissions from fuel chains.

36 **Keywords:** Carbon dioxide capture and storage; Life Cycle Assessment; fugitive emissions;  
37 coal; natural gas; electricity generation

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39        **1. INTRODUCTION**

40        With the increasing interest in power generation from unconventional fossil fuel resources,  
41        such as shale gas, and the large push for gas fired power plants as a clean form of electricity  
42        production (Stephenson et al., 2012), a more complete quantification of the (potential)  
43        environmental impacts of fossil fuel power generation life cycle is needed. Though the  
44        environmental impacts of the operation of most power generation technologies are relatively  
45        well described and quantified in Life Cycle Assessment (LCA) literature (Corsten et al., 2013;  
46        Heath et al., 2014; O'Donoghue et al., 2014; Whitaker et al., 2012), we argue here that  
47        attention should also be directed towards upstream processes, such as the extraction and  
48        transport of fossil fuel resources (Alvarez et al., 2012; Burnham et al., 2012; Weber and  
49        Clavin, 2012). The fuel supply is especially important when carbon dioxide capture and  
50        storage (CCS) technology is applied to reduce the greenhouse gas emissions of the power  
51        plant itself, a step which increases fuel consumption due to the inherent energy efficiency  
52        penalty related to the carbon dioxide capture and compression processes.

53        One of the major greenhouse gases (GHGs) emitted in natural gas and coal production is  
54        methane. As a major constituent of natural gas, methane emissions occur at all points during  
55        the natural gas extraction process: well drilling and completion, well operation, e.g. in the  
56        form of purges and vents, and through leakages of the entire natural gas infrastructure, e.g., at  
57        intermediate compressor and redistribution stations of the pipeline (Burnham et al., 2012).  
58        Coal bed methane is formed from bacterial degradation of coal and biomass residuals, and  
59        thermally through devolatilisation within the coalification process of organic matter (Moore,  
60        2012). It is released during coal extraction and removal of overburden. Methane emissions  
61        from fossil fuel origin are estimated to represent about 30% of the world anthropogenic  
62        methane emissions, although both fossil emissions and total anthropogenic emissions are  
63        quite uncertain (Kirschke et al., 2013).

64 A range of life cycle assessments (LCAs) of fossil fuel power generation with and without  
65 CCS has been published previously (Jaramillo et al., 2007; Koornneef et al., 2008; NETL,  
66 2010b, c, d, e; Odeh and Cockerill, 2008; Singh et al., 2011a; Zapp et al., 2012). Most studies  
67 were thoroughly reviewed in the papers by Whitaker et al. (2012), O'Donoughue et al. (2014),  
68 Heath et al. (2014), and Corsten et al. (2013). Whitaker et al. (2012) present a review and  
69 harmonization of LCA greenhouse gas emission results for coal based electricity generation.  
70 Coal methane emissions are discussed, and an interquartile range of the reviewed studies of  
71 54-73 g CO<sub>2</sub>-eq/kWh is presented (median 63 g CO<sub>2</sub>-eq/kWh). O'Donoughue et al. (2014)  
72 review and harmonize LCA greenhouse gas emission results for conventional gas based  
73 electricity generation. Heath et al. (2014) harmonize shale gas life cycle emissions. Methane  
74 leakage is discussed and ranges from 0.2 % to 6 % of natural gas production in the reviewed  
75 studies. Corsten et al. (2013) review the LCAs of both coal and natural gas based electricity  
76 generation in combination with CCS. They conclude that the upstream emissions of natural  
77 gas lead to large impacts on the overall GHG emissions, to the extent that electricity  
78 generated by a natural gas combined cycle power plant with CCS appears to have associated  
79 GHG emissions of the same order of magnitude as pulverized coal generated electricity with  
80 CCS.

81 Several recent studies focus on fugitive methane emissions from conventional and  
82 unconventional fossil fuel production. Weber and Clavin (2012) perform a Monte Carlo  
83 analysis based on six previous studies for natural gas from conventional and unconventional  
84 sources. Burnham et al. (2012) compare results for emissions related to coal and natural gas,  
85 shale gas and petroleum. Both studies conclude that upstream methane leakage and venting  
86 can reduce significantly the life cycle benefit from gas compared to coal, and that gas related  
87 emissions from conventional or shale production are statistically indistinguishable in a life  
88 cycle perspective. Laurenzi and Jersey (2013) study GHG emissions and water consumption

89 of Marcellus shale gas production, but indicate that for certain GHG emissions EPA emission  
90 factors are used. They find that the estimated ultimate recovery of shale wells is one of the  
91 major determinants in the life cycle GHG emissions of shale gas electricity generation.

92 Though there are differences between the LCA studies of power plants with and without CCS  
93 in the literature, relatively little attention has been paid to fugitive emissions. These are  
94 mainly included by application of an emission factor and sometimes discussed as a subject of  
95 sensitivity analysis. In addition, most studies have a limited regional scope, evaluating power  
96 plants in Europe or the United States, with the shale gas literature focusing almost solely on  
97 the United States. This leads to the questions to what extent data are available with respect to  
98 fugitive methane emissions for both coal and natural gas, how they vary regionally, and  
99 consequentially what that implies for the environmental performance of fossil fuel power  
100 generation with and without CCS.

101 The aim of this paper is to make an inventory of the ranges of fugitive methane emissions  
102 available in the literature and assess the consequences these emissions have on the life cycle  
103 GHG impacts of fossil fuel power generation. We focus on fugitive methane emissions of coal  
104 mining, conventional natural gas production and shale gas production. The hybridized  
105 multiregional life cycle assessment model THEMIS (Technology Hybridized Environmental-  
106 economic Model with Integrated Scenarios) is used (Hertwich et al., 2014), in combination  
107 with a set of life cycle inventories for state-of-the-art fossil fuel power plants, both with and  
108 without CCS facilities. We allow for regional variation of fugitive emissions in order to  
109 increase understanding of the environmental consequences of implementation of fossil fuel  
110 power generation in different regions.

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## 113        **2. METHODS**

114    In this section we discuss the approach followed to assemble the fugitive emission datasets  
115    with special focus on the data reported in UNFCCC. We continue with a description of the  
116    HLCA model employed. The system description for the HLCA and life cycle inventories used  
117    are described separately in section 3 of this paper.

### 118    ***2.1 Dataset assembly fugitive emissions***

119    Three datasets were compiled containing a total of 227 entries for coal fugitive emissions, 34  
120    entries for conventional gas fugitive emissions and 19 entries for shale gas emissions, based  
121    on peer reviewed published literature as well as data reported as part of the United Nations  
122    Framework Convention on Climate Change (UNFCCC). The UNFCCC was established in  
123    1992 at the United Nations Conference on Environment and Development in Rio de Janeiro.  
124    The treaty has the objective to achieve ‘...*stabilization of greenhouse gas concentrations in*  
125    *the atmosphere at a level that would prevent dangerous anthropogenic interference with the*  
126    *climate system...*’ (United Nations, 1992). Annex I countries that have ratified the convention,  
127    report national greenhouse gas inventories yearly in the form of a national inventory report  
128    (NIR) and the common reporting format (CRF). The NIRs contain detailed information for  
129    each country and the CRF is an electronically submitted series of standardized data tables for  
130    all greenhouse gas emissions per sector. According to the guidelines governing the reporting  
131    on annual inventories, the estimates of emissions should be comparable among parties. In  
132    order to do so, countries have to follow the IPCC guidelines (IPCC, 2006) to estimate and  
133    report on anthropogenic emissions, but are free to use the different methods included in those  
134    guidelines (UNFCCC, 2004). Though data should be comparable between countries, there are  
135    different levels of uncertainty related to the UNFCCC data, which are related to the different  
136    calculation approaches accepted in the IPCC guidelines. Countries can report data using a tier  
137    1 approach. In this approach, associated with the highest level of uncertainty, total emissions

138 are calculated using a global average range of emissions factors and country-specific activity  
139 data. In the tier 2 approach, emissions are calculated using country or basin specific emissions  
140 factors. In the tier 3 approach, associated with the lowest level of uncertainty, direct  
141 measurements on a mine-specific basis are used (IPCC, 2006). Though not reported in the  
142 tables of the CRF, the NIRs contain information about the approaches used by Annex I  
143 countries (commonly mixes between tiers 1, 2, and 3) in reporting emissions data.

144 In this paper, we used the data provided by the Annex I countries in Tables 1.B.1 and 1.B.2 of  
145 the CRF, that describe the fugitive emissions from solid fuels (1.B.1) and oil, natural gas and  
146 other sources (1.B.2) (UNFCCC, 2012). We selected for each country the average, minimum  
147 and maximum emissions of the time series from the starting year of reporting (usually 1990,  
148 though there are variations between countries) until 2010. These country level data were  
149 subsequently aggregated to generate a list of regional estimates of methane emissions related  
150 to coal production and conventional natural gas production. The regions correspond to the  
151 regional division of our HLCA model, which is described in section 2.2.

152 In this study, values larger than 1.5 times the global interquartile range above the (global) 3<sup>rd</sup>  
153 quartile were considered outliers and were removed from the database. This was the case for  
154 natural gas data reported by Ukraine and Greece (respectively 1025 and 837 g CH<sub>4</sub>/m<sup>3</sup> natural  
155 gas) and some of the coal data for Russia and France. Such high numbers may be due to the  
156 application of too uncertain emissions factors in the tier 1 method and possibly aggregation of  
157 fugitive emissions related to the natural gas transportation infrastructure in the UNFCCC  
158 common reporting format.

159 Because the United States is the only country with significant past shale gas production and  
160 because there is no distinction in the UNFCCC natural gas data regarding the source

161 (conventional or shale) of methane emissions, we assumed that UNFCCC natural gas  
 162 emissions data are only relevant for the conventional natural gas system.

163 In addition to official emissions reports scientific literature sources were consulted. Coal  
 164 mining, conventional natural gas extraction, and shale gas extraction are described by  
 165 (Burnham et al., 2012). Shale gas is included in (Howarth et al., 2011; NETL, 2014 ; Weber  
 166 and Clavin, 2012). A set of emissions factors for coal mines was found for the regions China,  
 167 OECD Pacific and Economies in Transition (mainly Russia) (Bibler et al., 1998; EPA, 2006;  
 168 NETL, 2010f; Saghafi, 2012; Su et al., 2011; Sørstrøm, 2001), thus generating at least one  
 169 dataset for five different regions in the HLCA model. Table 1 shows the regional coverage of  
 170 the three datasets compiled based on the references consulted. The total number of data  
 171 points per region and source is presented in Table ST1 of the supporting information.

**Table 1**  
 Regional coverage of datasets investigated

| Reference              | Coal | Conventional gas | Shale gas | Regions <sup>a</sup> |
|------------------------|------|------------------|-----------|----------------------|
| UNFCCC, 2012           | X    | X                |           | RER;US;PAC;EIT       |
| Burnham et al., 2012   | X    | X                | X         | US                   |
| Weber and Clavin, 2012 |      | X                | X         | US                   |
| Howarth et al., 2011   |      | X                | X         | US                   |
| Sørstrøm, 2001         | X    |                  |           | RER;US;EIT           |
| Su et al., 2011        | X    |                  |           | CN                   |
| Bibler et al., 1998    | X    |                  |           | CN                   |
| EPA, 2006              | X    |                  |           | RER;US; EIT          |
| Saghafi, 2012          | X    |                  |           | PAC                  |
| NETL, 2010f            | X    |                  |           | US                   |
| NETL, 2014             |      | X                | X         | US                   |

172 <sup>a</sup> Region abbreviations are: CN = China, RER = OECD Europe, US = OECD North America, PAC = OECD Pacific, EIT = Economies in Transition

## 173 **2.2 HLCA model**

174 A multi-regional integrated hybrid life cycle assessment (HLCA) model was employed to  
 175 model the potential environmental impacts (Hertwich et al., 2014). We modeled a traditional  
 176 process based Life Cycle Assessment and complemented this with economic data where these  
 177 were available. The model was set-up as a tiered hybrid model, in which it is possible to select  
 178 for each unit process background data from both a physical inventory, ecoinvent 2.2 (Dones et



179 al., 2007), and an environmentally extended Input-Output database EXIOBASE (Tukker et  
180 al., 2013). In the THEMIS model, EXIOBASE is aggregated to nine regions from its original  
181 regional classification, but incorporates a disaggregated electricity sector (Hertwich et al.,  
182 2014). Potential environmental impacts were calculated on a per-kWh electricity produced  
183 functional unit basis. For the LCA we took a cradle-to-gate approach.

184 As methane is an important greenhouse gas, we evaluated GHG emissions using Global  
185 Warming Potentials (GWPs) over a 100-year time horizon. For each of the emission factors  
186 found in the literature the appropriate stressors were adapted and the LCA model was run,  
187 which generated a range of model outcomes for the climate change impact associated with the  
188 fossil electricity generation. The ecoinvent database contains nine unique processes that cover  
189 natural gas extraction and ten processes for the extraction of hard coal. A shale gas extraction  
190 process did not exist in the database, and therefore an inventory was built based on data from  
191 the Argonne National Laboratory (Clark et al., 2011). All modeling was performed in Matlab  
192 in combination with an Excel interface for data input.

193 The life cycle inventory data are based on state-of-the-art power plants described by several  
194 reports of the National Energy Technology Laboratory in the US. These studies present  
195 detailed cost economic modeling of power plants and life cycle inventories (NETL, 2010a, b,  
196 c, d, e), thus providing a suitable starting point for hybrid life cycle assessment. Where data  
197 were not sufficient, or too specific for a generic power plant, peer reviewed literature was  
198 consulted to provide input data (Koornneef et al., 2008; Singh et al., 2011a; Veltman et al.,  
199 2010).

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201

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### 203 3. LIFE CYCLE INVENTORY

204 Four different types of electricity production technologies were modeled. The investigated  
205 technologies are:

- 206 i) subcritical pulverized coal fired power (Sub-PC)
- 207 ii) supercritical pulverized coal fired power (SCPC)
- 208 iii) integrated gasification combined cycle (IGCC)
- 209 iv) natural gas combined cycle (NGCC)

210 Out of these technologies, three are connected to a post-combustion CO<sub>2</sub> capture process  
211 (using amine as solvent) and one is connected to a pre-combustion CO<sub>2</sub> capture process (using  
212 a solvent consisting of dimethyl ethers and polyethylene glycol). Key characteristics of these  
213 technologies are described in Table 2. We evaluate the power plants on a cradle-to-gate  
214 perspective. Electricity transport and distribution to the end users is outside the scope of the  
215 study. Each life cycle inventory is set up according to the following structure: fossil fuel  
216 extraction, fossil fuel transport, power plant operation and a separate foreground process for  
217 power plant infrastructure. For the inventories in which carbon capture and storage is  
218 included, the following foreground processes are added: CO<sub>2</sub> capture and compression, CO<sub>2</sub>  
219 transport by pipeline, and the CO<sub>2</sub> injection well. The key foreground processes are shortly  
220 discussed in the following sections. Information regarding specific emissions and the  
221 efficiencies of emissions reduction measures implemented with each power plant is given in  
222 the table ST2 in the Supporting Information.

**Table 2**  
Key power plant characteristics (NETL, 2010a)

|   | Unit                     | Sub-PC           | SCPC             | IGCC                  | NGCC                        |
|---|--------------------------|------------------|------------------|-----------------------|-----------------------------|
| <b>Net power output without CCS</b>                       | MW                       | 550              | 550              | 629                   | 555                         |
| <b>Net power output with CCS</b>                          | MW                       | 550 <sup>a</sup> | 550 <sup>a</sup> | 497                   | 474                         |
| <b>Capacity factor</b>                                    | %                        | 85               | 85               | 80                    | 85                          |
| <b>Net plant efficiency without CCS</b>                   | %                        | 38.2             | 40.7             | 43.6                  | 55.6                        |
| <b>Net plant efficiency with CCS</b>                      | %                        | 27.2             | 29.4             | 32.3                  | 47.4                        |
| <b>Fuel requirements</b>                                  | kg/kWh                   | 0.361            | 0.338            | 0.315                 | 0.187 (m <sup>3</sup> /kWh) |
| <b>Fuel requirements with CCS</b>                         | kg/kWh                   | 0.507            | 0.467            | 0.425                 | 0.219 (m <sup>3</sup> /kWh) |
| <b>CO<sub>2</sub> emissions from power plant</b>          | g/kWh                    | 856              | 802              | 723                   | 365                         |
| <b>CO<sub>2</sub> emissions from power plant with CCS</b> | g/kWh                    | 120              | 111              | 109                   | 42.6                        |
| <b>MEA consumption</b>                                    | kg/tonne CO <sub>2</sub> | 2.15             | 2.15             | 0.09 (dimethyl ether) | 2.15                        |
| <b>CO<sub>2</sub> capture efficiency</b>                  | %                        | 90               | 90               | 90                    | 90                          |
| <b>Lifetime</b>   | years                    | 30               | 30               | 30                    | 30                          |

<sup>a</sup> The nominal net output for the Sub-PC and SCPC cases was maintained at 550 MW for the cases with CCS. This is done by increasing the boiler and turbine/generator sizes to account for a larger auxiliary load due to the carbon capture process. For the IGCC and NGCC cases, the plant size was kept constant, leading to a lower net power output.

223

224 The following sections describe our base inventory for the four investigated power plant  
 225 technologies. As the purpose of this paper is to show how varying emissions upstream can  
 226 influence the LCA results related to power generation we do not change the efficiency of the  
 227 power plants between regions. However, as our base inventory (presented in Tables ST5-  
 228 ST16) is based on fuels with very specific energy and carbon density, we assume a regional  
 229 specific lower heating value (LHV) for the fuel used in order to adapt the fuel requirement  
 230 and direct emissions of power plant operation for each region. The scaling factors we  
 231 developed to adapt these flows in the base inventory are presented in Tables ST3 and ST4 of  
 232 the Supporting Information. The regional specific LHV is used to calculate the fossil fuel  
 233 input for the power plant in each region. Direct emissions of power plant operation are scaled  
 234 with both regional specific LHVs and carbon content. To that extent, we assembled a set of  
 235 coal carbon content (CC) and LHV pairs (in the range of 18-31 MJ/kg coal), that were used in  
 236 previous LCAs and express CC as function of LHV (Whitaker et al., 2012). In the specified  
 237 LHV range we assumed that this function behaves linearly for all practical purposes. The  
 238 scaling factor for direct power plant emissions was calculated based on the relative change of  
 239 the ratio between calculated CC and regionally specified LHV. Since the variation in the LHV

240 of natural gas used in the model is relatively low, we have chosen to use the same scaling  
241 factor for both natural gas inputs and emissions.

242

### 243 ***3.1 Fossil fuel extraction***

244 Three types of extraction processes are modeled in this paper: coal mining, conventional  
245 natural gas extraction, and shale gas extraction. For coal and conventional natural gas the  
246ecoinvent processes *hard coal, at mine* and *natural gas, at production* are used, with the  
247 exception of making the fugitive methane emissions in these processes a model variable.  
248 Please note that, for coal, we do not explicitly distinguish between underground and surface  
249 coal mining processes, but use the underground/surface mine ratio in the ecoinvent database.

250 A shale gas extraction process was modeled based on data published by the Argonne National  
251 Laboratory (Clark et al., 2011). A well production over a lifetime of 30 years of 98 million  
252 cubic meters was assumed, though it should be noted that much shorter lifetimes have been  
253 reported (O'Sullivan and Paltsev, 2012). Material requirements for the drilling and  
254 construction of the well pads are taken as the non-weighted average of four shale gas plays in  
255 the United States (namely, Barnett, Marcellus, Fayetteville, and Haynesville). The fracking  
256 fluid is a mixture of water and sand with a range of organic and inorganic chemicals such as  
257 methanol, hydrogen chloride, formaldehyde, sodium hydroxide and ethylene glycol. The  
258 inventory for the fracking fluid is given in table ST5 of the Supporting Information.

259 Electricity and diesel fuel consumption for well operation are taken as an average of four  
260 wells described by Clark et al. (2011). Within our model, the emissions associated with well  
261 completion and well workovers are not explicitly stated, but are part of the well operation  
262 process, as many sources report well completion in percentage of natural gas during  
263 production. The methane emissions for well completion and workovers are reported to be 417

264 tonnes of methane per well over its life cycle (Clark et al., 2011). Table ST6 in the supporting  
265 information shows the required material inputs and methane emissions associated with the  
266 construction of a shale gas well as modeled in this study and Table ST7 shows the inventory  
267 for shale well operation.

### 268 **3.2 Fossil fuel transport**

269 In this study, the coal fired power plants are assumed to use the same coal transport unit  
270 process. Coal is transported by rail over a distance of 330 km from the excavation site to the  
271 power plant (NETL, 2010e). The material requirements for the trains are included in the  
272 inventory, as well as diesel required for transport. The rails themselves are assumed to be  
273 constructed and available and are not included in the inventory. During coal extraction and  
274 transport it is assumed that no coal is lost. The coal transport inventory is presented in Table  
275 ST8 of the supporting information.

276 Gas is assumed to be transported 1000 km by pipeline, connecting an offshore natural gas  
277 extraction site and the power plant location (ecoinvent process *transport, natural gas,*  
278 *pipeline, long-distance*). Although the shale gas wells are land based and it would be expected  
279 that the transport distance is shorter, it was chosen to keep the pipeline length constant, in  
280 order to make inventories more comparable. Methane leakage during transport is assumed at  
281 0.026 % of transported natural gas per 1000 km based on ecoinvent (Faist Emmenegger et al.,  
282 2007).

283 For Russia, the ecoinvent leakage rate is considerably higher at 0.23 % per 1000 km (1.4 %  
284 for a transport distance of 6000 km) (Faist Emmenegger et al., 2007). Leakage rates for  
285 transmission and distribution of 0.67 % (0.29 % - 1.05 %) to 1.5 % (0.8 % - 2.2 %) are  
286 reported for the US, but a specific transport distance is not reported (Burnham et al., 2012;  
287 Weber and Clavin, 2012). To study the increase in contribution of methane to the life cycle

288 impacts, the natural gas transport process was updated with the values for the EIT (0.23 %)  
289 and the US (0.67 %). We report the influence of different natural gas pipeline fugitive  
290 emissions rates in section 4.3.

### 291 ***3.3 Pulverized coal fired power plants***

292 The baseline inventory includes both sub- and supercritical coal fired power technology (see  
293 tables ST9 and ST10). Both coal fired power plants are based on designs from the National  
294 Energy Technology Laboratory (NETL, 2010a). Key plant characteristics are given in Table  
295 2. Main inputs are taken from the ecoinvent background. The largest flows are hard coal fuel,  
296 limestone for the flue gas desulphurization unit, ammonia for the selective catalytic reduction  
297 of NO<sub>x</sub> emissions and water for cooling duties. In addition, for the CCS processes,  
298 monoethanolamine (MEA), caustic soda, and activated carbon are also used. The treatment of  
299 waste generated by the power plants, is modeled following ecoinvent. Main emissions for the  
300 power plants without CCS are carbon dioxide, water vapor, particulate matter, sulfur dioxide  
301 and nitrogen oxides (NETL, 2010b, e). The flue gas desulphurization process in the coal fired  
302 power plants yields gypsum as an economic byproduct. In this paper, we take a conservative  
303 approach and no impacts are allocated to gypsum production. In power plants with CCS,  
304 ammonia and MEA emissions are also included. The CO<sub>2</sub> captured is transported in dense  
305 phase and is compressed on-site to 153 bar before transport. The electricity for compression is  
306 generated by the power plant and it is included in the energy penalty due to CO<sub>2</sub> capture. It is  
307 further assumed that no extra cleaning equipment is required and that dehydration during  
308 compression reduces the water content to at least 500 ppmv, making it suitable for transport.  
309 Power plant infrastructure, as well as chemicals that constitute minor inputs, are modeled  
310 using flows from the economic EXIOBASE background (see tables ST12 and ST13).

311

### 312 *3.4 Integrated gasification combined cycle*

313 The integrated gasification combined cycle power plant is modeled based on the designs of  
314 NETL (NETL, 2010a). The key plant characteristics are given in Table 2. Main inputs are  
315 taken from theecoinvent background (see table ST8). Before combustion, coal is gasified  
316 producing a mixture of hydrogen and carbon monoxide. As noted before, the coal transport  
317 process is assumed to be the same as the transport process for the sub- and supercritical power  
318 plants. Due to its higher efficiency, the fuel requirements for the IGCC are somewhat lower  
319 than those for the pulverized coal power plants. Besides coal, the main inputs to plant  
320 operation are process water for cooling duties, catalyst for the COS hydrolysis unit (in the  
321 case of the IGCC without CCS) and activated carbon for the removal of mercury. In the case  
322 of IGCC with CCS, the water gas shift reactor also hydrolyses carbonyl sulphide (COS) into  
323 H<sub>2</sub>S, hence no separate COS hydrolysis reactor is needed. A mixture of dimethyl ethers and  
324 polyethylene glycol is used as a physical solvent for both the IGCC plant with and without  
325 CCS and is used for mainly sulfur removal (single stage) or for both sulfur and CO<sub>2</sub> removal  
326 (dual stage). Though sulfur is a byproduct of the IGCC power plant, the same approach as  
327 with the gypsum production in the supercritical power plant is followed, thus impacts are not  
328 allocated with respect to sulfur. The solvent has a low vapor pressure, and spent solvent is  
329 therefore assumed to end up in the solid waste output of the power plant (Singh et al., 2011b).  
330 Main emissions for the IGCC power plant are carbon dioxide, water vapor, particulate matter,  
331 sulfur dioxide and nitrogen oxides. The CO<sub>2</sub> captured is compressed to 153 bar before  
332 transport. Power plant and CCS infrastructure, as well as chemicals that constitute minor  
333 inputs, are modeled using flows from the economic EXIOBASE background (see tables ST15  
334 and ST16).

335

### 336 ***3.5 Natural gas combined cycle***

337 The natural gas plant is modeled as a combined cycle plant (NETL, 2010a). Besides natural  
338 gas, the main plant inputs are ammonia for the selective catalytic reduction (SCR) of NO<sub>x</sub>,  
339 process water for cooling duties and chemicals such as the catalyst of the SCR unit. Inputs to  
340 the CO<sub>2</sub> capture process are activated carbon and MEA. Main emissions for the NGCC power  
341 plants are carbon dioxide, water vapor, ammonia, and nitrogen oxides (see table ST9). The  
342 CO<sub>2</sub> captured is compressed to 153 bar before transport. Similar to the other inventories,  
343 infrastructure is modeled using the EXIOBASE economic background (see tables ST15 and  
344 ST16).

### 345 ***3.6 CO<sub>2</sub> transport and storage***

346 Captured carbon dioxide is assumed to be transported to an underground aquifer by pipeline.  
347 CO<sub>2</sub> is transported in dense phase over a transport distance of 150 km. As the inlet pressure  
348 was 153 bars, the pressure drop over the 150 km pipeline is small enough to prevent two-  
349 phase formation and therefore intermediate booster stations are not required. Following the  
350 approach by Singh et al. (Singh et al., 2011a), pipeline inventory data are modeled after a high  
351 capacity offshore natural gas pipeline fromecoinvent (see table ST13). Carbon dioxide  
352 leakage from the pipeline is assumed to be 0.01% of transported CO<sub>2</sub> (see table ST14,  
353 (Koornneef et al., 2008)).

354 Captured CO<sub>2</sub> is injected in an aquifer at a depth of 1200 m. It is assumed that no booster  
355 compression is required at the wellhead, though this will be determined by site specific  
356 pressure conditions in the bottom well. The CO<sub>2</sub> injection rate per well is 9.4 kt CO<sub>2</sub> per day  
357 and is modeled as an offshore drilling well fromecoinvent (Singh et al., 2011a). In this study  
358 it is assumed that the reservoir is large enough to store the CO<sub>2</sub> over the lifetime of the power  
359 plant and that CO<sub>2</sub> is stored permanently (that is, there is no leakage from the reservoir).



## 360 4. RESULTS

### 361 4.1 Dataset analysis

362 Figure 1 **Error! Reference source not found.** shows the fugitive methane emissions within  
363 the data assembled. As can be seen for both coal and natural gas, fugitive emissions vary by  
364 orders of magnitude. The figure shows the outlier-adjusted minimum and maximum values  
365 for the different regions in the dataset (indicated by the lines), and the first and third quartile  
366 of the data (indicated by the box). In addition to the different regions, the global range is also  
367 presented. The regions China and Economies in Transition show clearly higher emissions  
368 associated with coal than the United States and Europe. There is a large spread in the  
369 European data as the result of some very low emissions (0.01 g CH<sub>4</sub>/kg coal) reported in the  
370 UNFCCC data. Methane emissions from gas production in North America are larger than  
371 those in Europe and the Economies in Transition. This divergence raises the question to what  
372 extent higher reported emissions in the US are due to difference in practice and specific site  
373 conditions and to what degree it could be the result of more attention to the issue, as indicated  
374 by the relatively high attention for (US) fugitive emissions in scientific literature. The results  
375 also indicate that fugitive emissions associated with shale gas are on average higher than for  
376 conventional natural gas production. This can be due to the large uncertainty involved in the  
377 emissions associated with well completion and workover emissions. For example, these  
378 emissions are reported to be in the range of 0.006-2.75% of natural gas production (Burnham  
379 et al., 2012). Dataset analysis did not reveal an obvious distribution of the emissions factors in  
380 the UNFCCC data, even though a lognormal (Dones et al., 2007) and triangular distribution  
381 (Weber and Clavin, 2012) have been assumed previously for the purpose of Monte Carlo  
382 simulations.

383 It is important to note here that the large ranges of fugitive emissions shown are caused by  
384 both natural variation and uncertainty in the data. For example, differences in coal grade and  
385 rank between mines have an influence on the methane emissions included in the coal bed  
386 (Moore, 2012). Furthermore, surface mines are more likely to have been vented by natural  
387 processes and can therefore be expected to have lower associated fugitive emissions than  
388 underground mines. In addition, natural gas is captured from coal seams (coal bed methane)  
389 thereby reducing the potential fugitive emissions of to-be extracted coal (NETL, 2014 ). The  
390 range of emissions related to gas infrastructure is most likely a result of the inherent  
391 uncertainty involved in the quantification of emissions using the tier 1 and 2 methods.

392 <FIGURE 01>

393

#### 394 ***4.2 Life Cycle Impact Assessment***

395 In this section, the results of the life cycle impact assessment are presented. Figure 2 presents  
396 a boxplot of impact assessment results for the climate change impact category in g CO<sub>2</sub>-eq per  
397 kWh for all technologies investigated and based on a global warming potential evaluated at a  
398 100 year time horizon (Solomon and Miller, 2007). It is shown that the results vary  
399 considerably, with China and the Economies in Transition showing the highest impacts, as  
400 can be expected from the fugitive emissions range presented in Figure 1. The full range of  
401 results for coal fired technology without CCS lies between 747 and 1303 g CO<sub>2</sub>-eq./kWh of  
402 electricity produced. Not surprisingly, for the cases without CCS, natural gas power plant  
403 emissions are lower than coal fired power emissions, and lie between 367 and 533 kg CO<sub>2</sub>-  
404 eq./kWh. For the coal fired power plants, the average contribution of methane emissions  
405 varies considerably between 4% in the OECD Pacific region and 15% in China. For the

406 natural gas fired power plants this range is wider with the average contribution of methane  
407 ranging from 3 % in Europe up to 16 % for shale gas in the US.

408 Though there are large differences in the contribution of methane to GWP between regions,  
409 we see no significant difference in relative methane contribution for the three different coal  
410 technologies without CCS. It is important to note here that the difference between regions has  
411 a three-fold origin. The first one is the variation in the fugitive emissions rates between  
412 regions according to the data ranges shown in Figure 1. The second is due to the introduction  
413 of the regional specific LHVs for coal and natural gas. In regions with relatively low LHV  
414 (e.g. China) the higher fuel requirements translate into a higher contribution of methane to the  
415 GWP. Thirdly, the regionalized background contained in THEMIS introduces some variation  
416 in regional GWPs. For example, the electricity mix used in the production of the diesel fuel  
417 used in the transport of coal varies between regions. In the case of fossil fuel power plants the  
418 contribution of the regionalized background is small, as most of the emissions are associated  
419 with the foreground processes.

420 The inclusion of CCS decreases the environmental impacts of power plants considerably, with  
421 GHG results ranging from 128 to 747 g CO<sub>2</sub>-eq./kWh for coal fired power plants. Results for  
422 natural gas plants lie between 75 and 250 g CO<sub>2</sub>-eq./kWh. The average contribution of  
423 fugitive methane emissions after installing CCS technology ranges from 23 % to 50 % for  
424 coal and 19 % to 56 % for natural gas. Contrary to the cases without CCS, we can observe  
425 differences in the average contribution of methane emissions between technologies (for equal  
426 regions) since the direct emissions of the power plant become less dominant.

427 In the interest of comparability we have not included intra-regional changes in both LHV and  
428 efficiency of the power plant. An increase in power plant efficiency will shift the entire range  
429 of GWPs proportional to the decrease in fuel requirements. An increase in LHV would also

430 result in lower fuel requirements, but most likely would affect direct power plant emissions  
431 much less due to the associated increase in carbon content. The opposite is valid for decreases  
432 in both efficiency and LHV. The above presented numbers show the importance of mitigation  
433 of methane emissions in the fossil fuel extraction process, as these emissions contribute  
434 largely to the emissions associated with fossil fuel power generation, especially for fuels with  
435 a relatively low LHV. It should be noted here that results for gas fired power plants and coal  
436 fired power plants partially overlap when carbon capture technology is installed, a conclusion  
437 also reached by for instance Corsten et al. (2013).

438 <FIGURE 02>

#### 439 ***4.3 The influence of natural gas transport emissions***

440 So far, we have explored only the fugitive emissions associated with the extraction of fossil  
441 fuels. However, emissions also occur in the transport of natural gas. As described before, the  
442 natural gas transport process was updated with new values for EIT (0.23%) and the US  
443 (0.67%). The results are presented in Table 3. We see a small increase for the EIT, even  
444 though emissions associated with transport are increased by an order of magnitude. Not  
445 surprisingly, the change is more apparent for North America, due to the high rate of emissions  
446 assumed to be associated with transport. However, it is not clear whether the 0.67% natural  
447 gas loss would be consistent with the pipeline length of 1000 km that is used in our model.  
448 Rather than estimating the contribution of emissions associated with natural gas transport, the  
449 purpose here is to show that emissions associated with transport have to become relatively  
450 high (as indicated by the US emissions rate) in order to become significant compared to  
451 fugitive emissions during extraction.

**Table 3**

The contribution of methane to the life cycle GHG emissions of power production when including region-specific transport emissions<sup>a</sup>

|                         | <b>EIT</b> | <b>US</b> |
|-------------------------|------------|-----------|
| <b>NGCC</b>             | 9% (8%)    | 16% (12%) |
| <b>NGCC + CCS</b>       | 34% (29%)  | 54% (45%) |
| <b>NGCC shale</b>       | n.a.       | 20% (16%) |
| <b>NGCC + CCS shale</b> | n.a.       | 61% (56%) |

<sup>a</sup> Values in brackets indicate the methane contribution with generic transport emissions previously used.

452

453 **5. DISCUSSION**

454 The direct comparison of LCA results between different studies is always hampered by  
 455 differences in system boundaries, plant type investigated, and background database used. For  
 456 example, Burnham et al. (2012) use an NGCC power plant efficiency of 47% and a  
 457 supercritical coal power plant efficiency of 41.5% (compared to respectively 55.6 % and  
 458 40.7% used in this paper). Modelling is performed with the GREET model, and not with  
 459 ecoinvent. In this section we therefore compare qualitative results rather than quantitative  
 460 results.

461 Burnham et al. (2012) have concluded that total upstream emissions can reduce the life-cycle  
 462 benefit for natural gas compared to coal, which the current study also indicates. There is no  
 463 agreement in the literature on the comparison on the magnitude of shale gas emissions  
 464 compared to conventional natural gas emissions and appears to be strongly tied to the shale  
 465 well lifetime and associated ultimate recovery (Laurenzi and Jersey, 2013; O'Sullivan and  
 466 Paltsev, 2012). In our modeling we see on average a larger impact for US shale than for US  
 467 conventional gas, but we would like to point out that the ranges overlap to a considerable  
 468 extent. Both Weber and Clavin (2012) and Laurenzi and Jersey (2013) conclude that the  
 469 relative difference in GWP between conventional and shale gas production is smaller than the

470 uncertainty in either estimate. As gas is increasingly extracted from unconventional sources  
471 special attention to methane emissions could provide a significant mitigation opportunity.

472 While fossil fuel power plants with high GHG emissions are reported in the literature, these  
473 emissions are generally caused by a low efficiency of the power plants (Whitaker et al.,  
474 2012). Our results show that even modern power plants can have high life cycle GHG  
475 emissions due to fuel chain methane releases. They also show that fuel energy density and  
476 associated carbon content are an important parameter in determining fuel requirement, and  
477 hence the contribution of fugitive emissions, and direct emissions of power plant operation. It  
478 should be noted that the non-methane upstream contribution is in the order of 1-4 %, mainly  
479 diesel combustion during operation of machinery and transport of coal, or carbon dioxide  
480 emissions associated with combustive processes during natural gas extraction and transport.

481 All impact results in this paper are reported using global warming potentials with a 100-year  
482 time horizon and a characterization factor for methane of 25 kg CO<sub>2</sub>-eq/kg CH<sub>4</sub>. In the latest  
483 round of IPCC reports, the characterization factor was updated to 34 kg CO<sub>2</sub>-eq/kg CH<sub>4</sub>. For  
484 GWPs evaluated over a 20-year time horizon the methane characterization factor is  
485 considerably larger at 86 CO<sub>2</sub>-eq/kg CH<sub>4</sub> (Myhre et al., 2013). The methane characterization  
486 factors show that the contribution of methane to radiative forcing is significant, especially in  
487 the short term. Several authors have tried to capture this by developing alternative models  
488 such as Technology Warming Potential (Alvarez et al., 2012) and Time Adjusted Warming  
489 Potential (Kendall, 2012).

490

## 491 **6. CONCLUSION**

492 The aim of this paper was to provide a better understanding of methane emissions associated  
493 with the extraction of fossil fuels and assess their effect on the life cycle impacts of fossil fuel

494 power generation. A set of life cycle inventories was assembled and combined with a dataset  
495 of fugitive methane emissions in a multiregional hybrid LCA model. The results of the dataset  
496 analysis reveals that fugitive emissions can vary by orders of magnitude, both inter- and  
497 intraregional. Our impact assessment results indicate that fuel chain methane emissions can  
498 constitute a substantial portion of the total emissions from fossil fuel power, and both their  
499 absolute magnitude and relative importance will increase with the deployment of CCS. In the  
500 most extreme cases, emissions from the fuel chain could be of equal importance to emissions  
501 from a power plant with CCS.

502 We see that methane emissions from fossil fuel production vary more widely than commonly  
503 acknowledged in the LCA literature, and that there are distinct regional disparities. By  
504 including the regionalization in our model we provide a more detailed picture of the  
505 contribution of fugitive methane emissions to the total life cycle impact. Coal methane  
506 emissions are more relevant for power plants in the regions China and Economies in  
507 Transition, with contributions over 40% for plants with CCS technology included, than for  
508 similar plants in Europe and North America. This is a result from higher fugitive emissions  
509 during extraction and the increased fuel requirements related to the use of fuel with a lower  
510 energy density. However, in the case of natural gas extraction, the contribution of fugitive  
511 emissions is significant for the North American region, with an average contribution that can  
512 exceed 50 % for the plants with CCS technology. European conventional natural gas  
513 production appears to have the lowest amount of fugitive emissions associated. The inclusion  
514 of higher emissions associated with natural gas pipeline transport becomes only significant  
515 when gas leakage rates increase by at least an order of magnitude compared to leakage from  
516 the European grid, which was used as the defaultecoinvent process.

517 The regional disparities may not reflect differences in geological factors, technologies, and  
518 practices employed. Most emissions estimates in both the UNFCCC data and literature are

519 based on engineering calculations and not measurements, with only one paper utilizing actual  
520 measured shale gas production data. More measurements and an in-depth review of the  
521 engineering calculations are required to illuminate whether reported differences reflect actual  
522 variation in emissions or our uncertainty about them. A more clear approach on how many of  
523 the data points are generated using tier 1, 2, 3 or mixed methods. In addition, most literature  
524 seems to focus on processes in the United States, but as this study shows, there is a need for  
525 detailed empirically determined emissions data in both North America and other regions, as  
526 the uncertainties related to the data reported under the UNFCCC common reporting format  
527 are not sufficiently quantified.

528 Given the large impact of methane emissions on LCA results we recommend practitioners to  
529 be aware of the sensitivity and to always perform a sensitivity analysis addressing uncertainty  
530 related to the upstream processes. Depending on timeframe and scope, there are examples of  
531 detailed inventories (NETL, 2014 ) in which fugitive emissions are addressed on a component  
532 specific level that could be adapted to specific conditions.

533

534



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542

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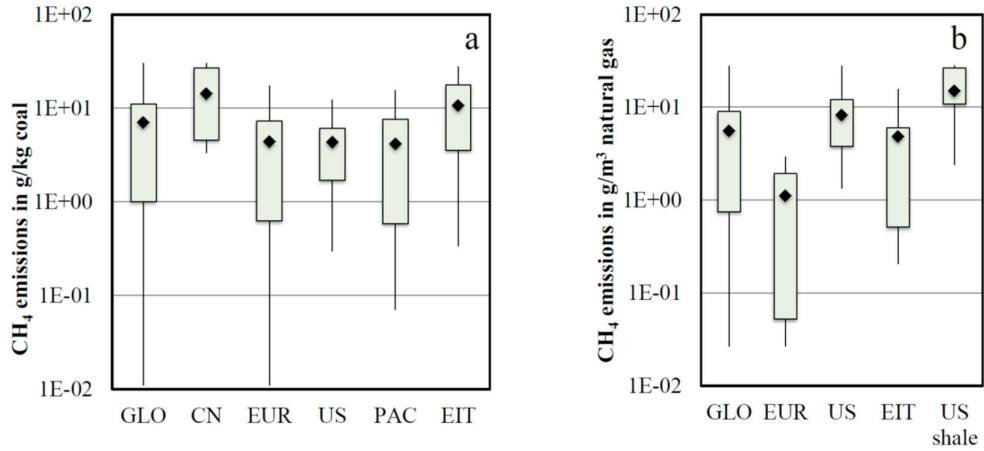
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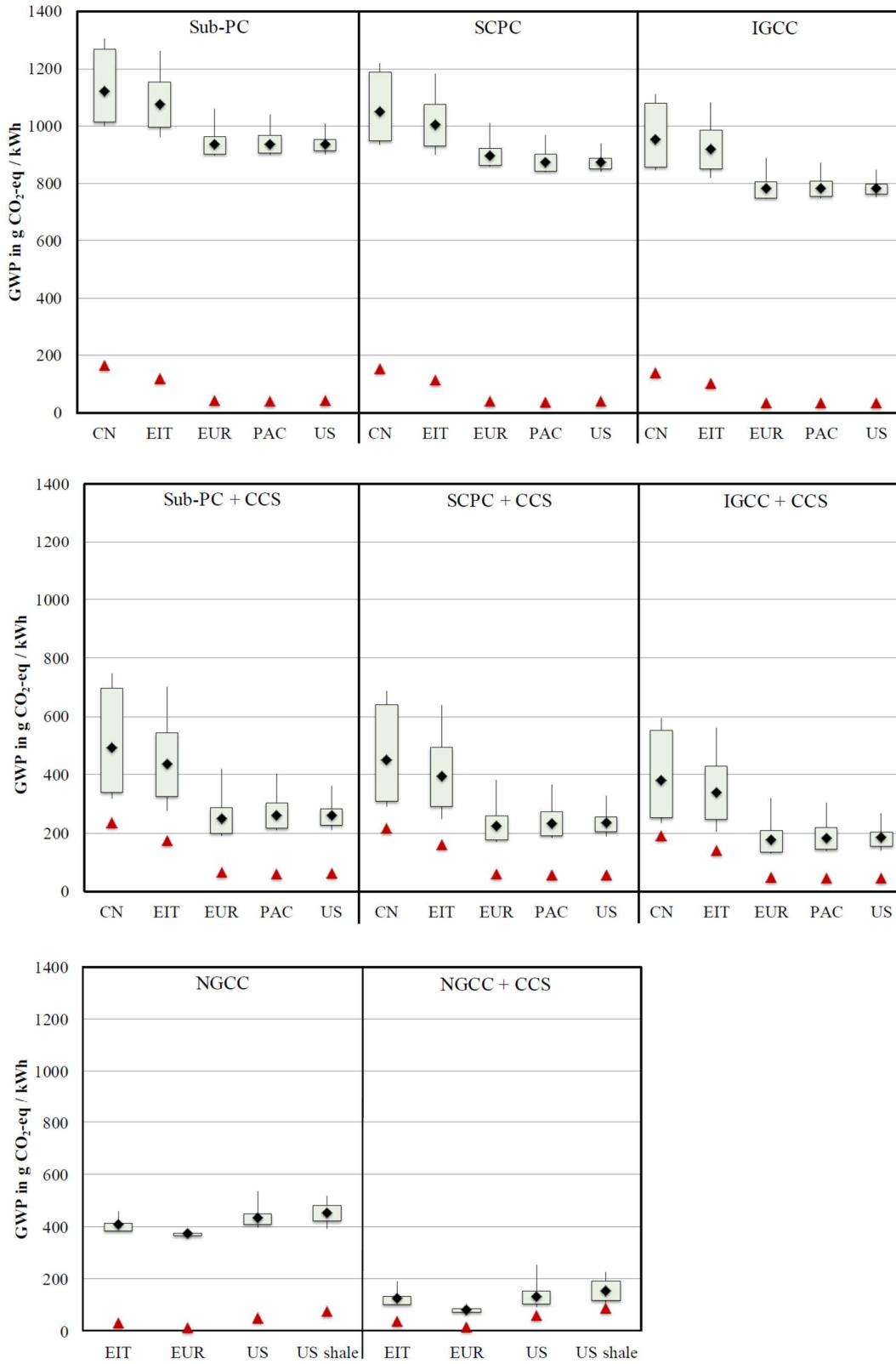
657

658

659 FIGURE CAPTIONS



660  
 661 **Figure 1(a-b): Reported fugitive methane emissions for the extraction of coal (a) and**  
 662 **extraction of natural gas (b). GLO = global, CN = China, EUR = OECD Europe, US =**  
 663 **OECD North America, PAC = OECD Pacific, EIT = Economies in Transition. N.B.**  
 664 **Emissions associated with natural gas production from conventional and shale source is**  
 665 **presented separately in columns US and US shale of panel b.**



666

667 **Figure 2: Calculated Global Warming Potential per kWh energy produced in sub-,**  
 668 **supercritical, integrated gasification coal fired power plants, and natural gas fired**

669 **power plants for the year 2010. Results are based on different fugitive emissions during**  
670 **fossil fuel extraction. Sub-PC = subcritical pulverized coal, SCPC = supercritical**  
671 **pulverized coal, IGCC = integrated gasification combined cycle, NGCC = natural gas**  
672 **combined cycle. The plotted triangles indicate the average contribution of methane**  
673 **emissions to the impact assessment. The plotted diamonds indicate the average GWP.**

674