

Emission of non-CO₂ greenhouse gases

Jan Willem Storm van Leeuwen
Independent consultant

member of the Nuclear Consulting Group

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storm@ceedata.nl

Note

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Table 1 Greenhouse gases

Global warming potential

Carbon dioxide is not the only greenhouse gas, although it is the most important one due to the vast amounts being emitted. This is not to say that for any industrial process CO₂ is the most important greenhouse gas produced. Many other greenhouse gases have a global warming potential (GWP) thousands of times larger than CO₂, so even tiny emissions of such gases may have a significant effect. A zero-carbon process may have a significant contribution to anthropogenic global warming if it emits high-GWP greenhouse gases.

Table 1 shows that gaseous halocarbons and other gaseous halo-compounds, in particular, may be potent greenhouse gases, as well as ozone depletion substances.

Table 1

Greenhouse gases. The unit of the Global Warming Potential GWP is kg gas per kg carbon dioxide. ODS = ozone depleting substance. Time horizon 100 years. Some gases are classified as ODS, but are also potent greenhouse gases. Sources: [EPA 2002], [EIA-G 2001], [Blasing & Smith 2006] and [Blasing & Jones 2003].

gas	formula	ODS ?	GWP
carbon dioxide	CO ₂		1
methane	CH ₄		23
nitrous oxide	N ₂ O		296
chlorofluorocarbons, CFCs	e.g. ClCF ₂ CClF ₂	+	4600 - 10600
hydrochlorofluorocarbons, HCFCs	e.g. CHClF ₂	+	120 - 2400
hydrofluorocarbons, HFCs	e.g. CHF ₃		12 - 12000
perfluorocarbons, PFCs	e.g. C ₂ F ₆		5700 - 11900
halons	e.g. CF ₂ ClBr	+	1300 - 6900
carbon tetrachloride	CCl ₄	+	1800
sulfur hexafluoride	SF ₆		22200
trifluoromethyl sulfur pentafluoride	SF ₅ CF ₃		> 17500
nitrogen trifluoride	NF ₃		10800
ethers and halogenated ethers	e.g. F ₃ C-O-CHF ₂		1 - 14900

In all processes from uranium ore to nuclear fuel substantial amounts of fluorine, chlorine and compounds of these elements are used, often in combination with organic solvents. Fluoro-compounds are essential in these processes, because enrichment of uranium requires uranium hexafluoride (UF₆), the only gaseous compound of uranium.

Unknown are the amounts of fluoro and chloro compounds used in other processes of the nuclear process chain. In a nuclear power plant, for example, considerable quantities of numerous different high-grade materials are incorporated; what emissions are coupled to the production of those materials?

As with all chemical plants, significant amounts of gaseous and liquid compounds from the processes will be lost into the environment, due to unavoidable process losses, leaks and accidents. No chemical plant is leakproof. From a chemical point of view, it is likely that in several processes potent GHG's arise or are used, or that GHGs are formed when they react with materials in the environment after release. Notably halocarbons have GWPs many thousands of times stronger than carbon dioxide.

Fluorine consumption in the nuclear process chain

In the processes of uranium ore milling through fuel element fabrication fluorine and its compounds are involved, often in combination with organic solvents.

Yellow cake from the uranium mill, containing $\text{Na}_2\text{U}_2\text{O}_7$ and/or $(\text{NH}_4)_2\text{U}_2\text{O}_7$, contaminated with chemical species from the ore and the extraction process, is converted into uranium hexafluoride UF_6 , using fluorine and/or its compounds, for instance hydrogen fluoride HF and elemental fluorine (F_2).

The stoichiometric mass ratio implies that for conversion of each gram uranium, a minimum of 0.48 gram fluorine is needed. In practice the ratio will be significantly higher than the stoichiometric minimum, due to unavoidable losses and secondary reactions. Because the uranium hexafluoride UF_6 has to be extremely pure, the fluorine and its compounds used in the process have to be extremely pure too. The required purification processes of the product are unavoidably coupled to significant losses. Likely the conversion process of yellow cake into UF_6 generates substantial waste streams containing compounds of fluorine, some of which may be potent greenhouse gases.

The reference reactor in our study consumes 20.3 Mg fresh enriched uranium during each reload period (in practice about each year). To prepare 20.3 Mg enriched uranium 162.35 Mg natural uranium has to be mined. For conversion of 162.35 Mg natural uranium into UF_6 , a stoichiometric minimum of 77.9 Mg fluorine is required; in practice substantially higher amounts than the stoichiometric minimum are needed.

In the enrichment facility the total amount of UF_6 is separated into two fractions: one fraction is depleted in U-235 and one is enriched in U-235. In practice the enriched fraction, containing 20.3 Mg uranium, is converted into UO_2 for use in nuclear fuel. In this case 9.74 Mg fluorine is set free. It is unknown in which form it is disposed of, likely as calciumfluoride CaF_2 , also in this conversion process losses are inevitable.

Depleted uranium is stored generally as UF_6 in special vessels, often in storage facilities in the open air. UF_6 is very reactive, so when it leaks into the environment, various fluorine compounds arise. Of course this method of storage cannot be a permanent one, in view of deteriorating and leaking vessels and increasing chances for accidents or terroristic actions. For that reason this study assumes that all depleted UF_6 is reconverted into a stable compound, such as U_3O_8 , for permanent sequestering in a geologic repository. Reuse of depleted uranium in MOX fuel is not a viable option, as explained in report m15 *Plutonium recycling in LWRs*.

World wide some 66000 Mg natural uranium is fluorinated each year, consuming a stoichiometric minimum of about 32000 Mg fluorine. The actual amount certainly will be much larger, may be some 100 000 Mg. Purification of fluorine and its compounds to high specifications generates unavoidably large waste streams containing fluorine and its compounds. Separation processes never go to completion, as follows from the Second Law of thermodynamics.

Chlorine use for fuel fabrication

Nuclear fuel, uranium oxide UO_2 enriched in uranium-235, is clad in tubes of Zircalloy, an alloy of extremely pure zirconium and a small percentage of another metal, e.g. tin or nickel. Technical-grade zirconium always contains hafnium, which has adverse effects in the core of a nuclear reactor and therefore has to be removed.

Zirconium can be purified by chlorination of the metal and distillation of the resulting chlorides, to remove all traces of hafnium. The stoichiometric mass ratio chlorine/zirconium in the compound zirconium tetrachloride ZrCl_4 is 1.56. So a minimum of 1.56 grams of chlorine is consumed per gram of Zr to produce ZrCl_4 .

To produce the 20-40 Mg Zircalloy needed for each reload of 20.3 Mg enriched UO_2 a stoichiometric minimum

of about 31-62 Mg of exceedingly pure chlorine (in any chemical form) is needed. In practice the amount of chlorine may be much larger to obtain an extremely pure product, and large waste streams are unavoidable. World wide some 7600 Mg enriched uranium is converted into nuclear fuel each year, requiring some 7600-15200 Mg Zircalloy annually. Production of that amount of Zircalloy requires a stoichiometric minimum of 11700-23400 Mg annually chlorine.

About 80% of the world zirconium production is consumed by the nuclear industry. This is a one-way production flow, because Zircalloy cannot be recycled, due to the high radioactivity of the material after use in a nuclear reactor.

Nuclear emission of non-CO₂ greenhouse gases: not reported

In 2001 the US enrichment plants alone had a specific GHG (greenhouse gas) emission of 5 grams CO₂-equivalents per kilowatt-hour of freon 114 (CFC-114, ClCF₂CClF₂), as follows from data from [EIA-DOE 2005]. Apart from these we found no data in the open literature on the emissions of fluorine- and chlorine-related chemical compounds by the nuclear industry. [Vattenfall 2005] noticed the absence of data on emission of greenhouse gases by processes needed to convert uranium ore into nuclear fuel.

Unknown are the GHG emissions of the front end processes, the conversion of uranium ore into ready-to-use nuclear fuel.

Unknown are the GHG emissions of the construction of a nuclear power plant, with its large mass of high-quality and often exotic materials.

Unknown are the GHG emissions of the operation, maintenance and refurbishment of nuclear power plants and the production of the materials used in these activities.

Unknown are the GHG emissions of the back end of the nuclear process chain: the handling and storage of spent fuel and other radioactive waste.

As pointed out above it is inconceivable that the nuclear process chain does not emit a gamut of fluoro and chloro compounds and it is also inconceivable that no greenhouse gases are among them.

Not reported' does not mean 'no emissions.

False comparison

Emissions of greenhouse gases other than CO₂ are not quantified in this study, due to the absence of data. For that reason this study explicitly uses the unit gCO₂/kWh and avoids the unit gCO₂eq/kWh (gram CO₂-equivalent per kilowatt-hour). The latter would imply that other greenhouse gases also are included in the emission figures.

Comparing, for instance, solar PV energy systems with nuclear power, using the unit gCO₂eq/kWh, the nuclear industry gives a false and misleading impression of things, comparing apples with oranges. The greenhouse gas emission of solar PV are partly due to the losses of fluorinated gases during the production of the silicon cells.

The nuclear industry mentions only the emission of CO₂ by nuclear power, albeit at an unrealistically low rate, but never mentions emissions of other GHGs. Nevertheless the nuclear industry incorrectly uses the unit gCO₂eq/kWh.

Krypton-85, another nuclear climate changer

Krypton-85 (symbols ^{85}Kr or Kr-85) is a radioactive isotope of the noble gas krypton. Although krypton is not a greenhouse gas in itself the presence of krypton-85 in the atmosphere gives rise to unforeseeable effects for weather and climate. Kr-85 is a beta emitter and is capable of ionizing the atmosphere, leading to the formation of ozone in the troposphere. Tropospheric ozone is a greenhouse gas, in addition it damages plants, it causes smog and health problems.

According to [WMO 2000]:

“The present background concentrations of ^{85}Kr in the atmosphere are about 1 Bq/m³ and are doubling every 20 years. At this level, ^{85}Kr is not dangerous for human beings, but the air ionization caused by ^{85}Kr decay will affect atmospheric electric properties. If ^{85}Kr continues to increase, changes in such atmospheric processes and properties as atmospheric electric conductivity, ion current, the Earth’s magnetic field, formation of cloud condensation nuclei and aerosols, and frequency of lightning may result and thus disturb the Earth’s heat balance and precipitation patterns.”

By nature krypton-85 is present in minute quantities in the atmosphere due to natural processes. In nuclear reactors massive amounts of krypton-85 are produced, as one of the major fission products. A small portion of it escapes into the atmosphere at the reactor site during operation, more will escape during storage of spent fuel in cooling pools and dry casks, for the number of leaking fuel elements increases with time due to unavoidable ageing processes. When spent fuel is reprocessed all Kr-85 is discharged from the spent fuel into the atmosphere. As a result of human nuclear activities the inventory of Kr-85 in the atmosphere has risen by a factor of 10 million and this quantity shows a rising trend [Ahlsvede *et al.* 2012], see also [Seneca 2015].

Materialization of the scenarios of the nuclear industry would lead to increased emissions of Kr-85, greatly increasing its atmospheric inventory. The Kr-85 discharges may be seen as another argument against reprocessing of spent fuel.

Health hazards of krypton-85

Being chemically inert, krypton and the other noble gases are not usually involved in biological processes. They are, however, absorbed into the tissues of the body via inhalation and dissolution in body fluids and tissues. Xenon has been shown to combine with specific sites in the body with certain protein molecules. Krypton is characterized by low blood solubility, high lipid solubility and rapid diffusion in tissue. Exceptions to the the biologically inert characterization of inert gases have been noted by numerous studies. A comparatively high uptake of krypton by the adrenal gland has been reported. These phenomena are not understood [NCRP-44 1975].

On a global scale the genetic and overall carcinogenic effects from Kr-85 are calculated to be small as compared with other possible sources of deleterious effects.

The possible interaction of radiation from krypton-85 and solar ultraviolet (UV) should be mentioned. In order to better understand the implications of long-term ^{85}Kr releases to the atmosphere, epidemiological and laboratory studies should be undertaken to define the nature and degree of interaction, if any, of UV radiation with ionizing radiation in the induction of skin cancer [NCRP-44 1975].

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