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^{14}C and ^{3}H Discharges from Pressurized Water Reactors and Boiling Water Reactors

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Lund University

Contents

Acronyms

ALARA	As Low As Reasonably Achievable.
BWR	Boiling Water Reactor.
IAEA	International Atomic Energy Agency.
LNPP	Large Nuclear Power Plant.
LWR	Light Water Reactor.
NPP	Nuclear Power Plant.
PHWR	Pressurized Heavy Water Reactor.
PRIS	Power Reactor Information System.
PWR	Pressurized Water Reactor.
RADD	European Commission RAdioactive Discharge Database.
TRS	Technical Report Series.

1 Introduction

1.1 Background

The ongoing electrification of our society requires a large expansion of the energy grid. Projections of different scenarios have estimated that Sweden's energy need will rise between 70-160 % by the year 2050 [1]. This puts forth a need for sustainable and reliable energy sources. Nuclear power has for the last few decades been an integral part of the energy grid in many countries including Sweden. Nuclear power plants continuously release small amounts of radioactive nuclides during normal operation, referred to as operational releases. These operational releases are monitored and minimized in accord with the ALARA principle in order to comply with dose constraints for the members of the public. ^{14}C and ^{3}H are two of these nuclides which contribute significantly to the effective as well as collective dose to members of the public after their release [2]. This is mainly due to two factors:

1) Their relatively long half-lives (^{3}H = 12.3 a and ^{14}C = 5730 a) give them sufficient time to spread far from the release point through natural distribution processes [2].

2) Their ease of assimilation into organic matter [2], owing to the fact that this is mostly made up of carbon and water.

Although ^{14}C and ^{3}H are weak beta emitters and don't pose an immediate radiation hazard unless highly concentrated [2], the above stated factors necessitate the surveillance of these nuclides as well as the predictions of their release rate.

The purpose of this paper is thus to investigate the release rate of ^{14}C and ^{3}H due to nuclear energy production. The constraint will be traditional Large Nuclear Power Plants (LNPPs), specifically Pressurised Water Reactors (PWRs) and Boiling Water Reactors (BWRs) as these are by far the most common type of operational reactors, constituting approximately 84 % of the reactors world-wide (306 PWRs, 41 BWRs, 415 total [3]).

1.2 Previous Work and Motivation

The topic has been studied previously in various publications. The following selection of publications have significantly informed this report, and they will be given special attention here as well as in chapter 3.

The IAEA report "Management of Waste Containing Tritium and Carbon-14" [2] was published in 2004. This report thoroughly covers various aspects of ^{3}H and ^{14}C releases for different types of reactors.

A more recent study, "Global and Regional Emissions of Radiocarbon from Nuclear Power Plants from 1972 to 2016" by Zazzeri et al. [4] (2016) extends the release rate study of ^{14}C in European nuclear power plants by about a decade, covering data until the year 2015.

In a very recent study (2024), Kim et al. [5] studied the ^{3}H and ^{14}C release rates from Korean PWRs and Pressurised Heavy Water Reactors (PHWRs) up until the year 2021.

While these reports are important in their own right, for the purpose of evaluating future nuclear power installments with regard to both ^{3}H and ^{14}C releases, they are not sufficent. The IAEA report [2] is about 2 decades old at this point (2025), and while the production rate of ^{3}H and ^{14}C in nuclear reactors is not expected to have decreased significantly, various changes in the systems surrounding the reactors could have had an effect on the release rates of the nuclides. The report by Zazzeri et al. [4] only covers ^{14}C releases, and only up until the year 2015. The report by Kim et al. [5] only includes 23 PWRs, and so the matter of statistical significance becomes a concern. The report does not contain any data on BWRs, and regional differences (Korea vs Europe) could have also had an effect on the reported releases.

These factors have therefore motivated a new study reported here.

2 Production and Operational Releases of ^{3}H and ^{14}C from LWRs

As already mentioned, this report restricts its attention to the most common type of LNPPs, namely to PWRs and BWRs, both of which are Light Water Reactors (LWRs). This chapter describes how LWRs generate operational releases of the radionuclides of interest. Since any operational releases necessarily constitute only a subset of all produced such radionuclides, the major production mechanisms of ^{3}H and ^{14}C in both types of LWRs are described as well. A brief description for PWRs is found in chapter 2.1, whereas a brief description for BWRs is found in chapter 2.2. For the remainder of this chapter, general information pertaining to the production and the operational releases of ^{3}H and ^{14}C in LWRs is provided.

In LWRs, any operational releases of radionuclides must esentially take a path through the coolant water, from which the nuclides are released atmospherically during venting or aqueously due to leakages of water. Therefore for any production pathway in a nuclear reactor, ^{3}H and ^{14}C must either be produced in or transferred to the coolant to be available for operational release.

For both types of LWRs, the majority of ^{14}C is released atmospherically, whereas ^{3}H is released both atmospherically and through liquid discharges [2]. Quantification of the release rates of ^{3}H and ^{14}C - in oxidized as well as in reduced form - to the atmosphere can be achieved by commercial stack air samplers followed by Liquid Scintillation Counting (LSC). Source monitoring of waterborne ^{3}H in the form of H_2O can also relatively easily be done by distillation followed by LSC analysis. Analysis of ^{14}C in water samples is significantly more challenging, due to the relatively low concentration of ^{14}C in discharged water and due to the more laborious sample pretreatment required, in particular if different chemical forms of the discharges should be considered [6]. LSC may thus require impractically large amounts of water to be taken through the sample preparation to give a sample with activity above the LSC detection limit. Therefore, most NPPs are not performing routine analysis of ^{14}C in liquid effluents. However, despite the lower amounts of ^{14}C discharged to water than to air, the local environmental effects due to waterborne ^{14}C discharges can be considerably larger than for airborne releases [7].

Some common design elements of LWRs imply corresponding commonalitites in their production and in their operational releases of ^{3}H and ^{14}C. As the name implies, LWRs use regular water as both moderator and coolant. This means that any production pathway and possible subsequent operational releases associated with the activation of regular water due to neutron irradiation will be present in any LWR. Both ^{3}H and ^{14}C are produced by such neutron activation of H_2O. ^{14}C is produced through the reaction $^{17}O(n,\alpha)^{14}C$ with a thermal neutron cross section of 0.24 barn [2] whereas ^{3}H is produced directly from neutron irradiation of ^{2}H with a thermal neutron cross section of 0.506 mbarn (PWRs have additional pathways which will be described later). Note that ^{2}H, besides appearing in the isotopic composition of natural hydrogen, is also created from neutron activation of ^{1}H with a thermal neutron cross section of 0.333 barn [2].

A second commonality is the similarity of the fuel rod designs among LWRs. The fuel rods in both types of LWRs consist of Uranium Dioxide fuel pellets surrounded by a very thin gaseous gap (helium), which is enclosed by a Zirconium alloy cladding. The Zirconium alloy is usually Zircaloy-2 or Zircaloy-4. The radial and axial dimensions of the fuel pellets, gap and cladding are comparable in different LWRs. A common production pathway of ^{3}H in LWRs is from ternary fission in the fuel. Associated to this source of ^{3}H are operational releases of the same nuclide due to its diffusion out of the fuel rod materials and into the surrounding coolant. This fraction of the ^{3}H produced in the fuel that diffuses into the coolant varies significantly, ranging from 0.013% all the way to 1% [8].

2.1 Pressurized Water Reactors

The main source of released ^{3}H in a PWR is from neutron activation of ^{6}Li and ^{10}B in the primary coolant. In PWRs boron is used for reactivity control due to the large neutron absorption cross section of ^{10}B, with the reaction ^{10}B(n,α)^{7}Li having a cross section of 3837 barn for thermal neutrons. For this nuclide however there is a possibility of producing ^{3}H in the reaction ^{10}B(n,2α)^{3}H with a cross section of 8.029 mbarn for thermal neutrons [9]. The boron is added to the coolant as boric acid which reduces the pH of the coolant. To balance the pH in the water, lithium hydroxide, which contains ^{6}Li in its naturally occurring amount, is added. ^{6}Li has a large chance to produce ^{3}H with the reaction ^{6}Li(n,α)^{3}H having a cross section of 940 barn for thermal neutrons [10]. The production rate of ^{3}H in the primary coolant is therefore much higher in a PWR than in a BWR [2]. Due to the reducing chemical conditions of the coolant water, having an excess of hydrogen, gaseous releases of ^{14}C in PWRs is mainly in the form of hydrocarbons such as methane. ^{3}H is released mainly as HTO (tritiated water, i.e. a water molecule with 1 hydrogen atom replaced by a tritium atom). The gaseous discharges in a PWR come mainly from the primary off-gas system, which lets out gases in pulses and not continuously as in a BWR [2].

2.2 Boiling Water Reactors

Besides the neutron activation of ^{2}H that was previously mentioned, the main production pathways of ^{3}H in BWRs comes from ternary fission in the fuel, as well as neutron activation of the boron in control rods used for reactivity control. A portion of the ^{3}H produced in the fuel may escape from the fuel, either by diffusing through the cladding [8], or during fuel leakage. Depending on the magnitude of the leakage, ternary fission ^3H may constitute the majority of ^3H available for operation releases from BWRs [11]. The diffusion of ^3H from control rods is generally considered insignificant [11]. Due to the oxidizing chemical conditions in the coolant, ^{14}C is mainly released as CO_2 [6], and ^3H is released mainly as HTO [2] also in BWRs.

3 Previous Studies of ^{14}C and ^3H Release Rates

The release rates of ^{14}C and ^3H have been studied and presented previously in various reports. The purpose of the current chapter is to present a selection of the data that has been published in these papers. The upcoming chapter will present values for the release rates of ^{14}C and ^3H in a similar fashion to those found in the tables in this chapter. The total release rate is often normalized to energy production and presented as [$GBq \cdot GW(e)^{-1} \cdot a^{-1}$], hereafter referred to as the normalized release rate. Note that the unit ($GBq \cdot GW(e)^{-1} \cdot a^{-1}$) is also used for the production rate. This will instead be referred to as the "normalized production rate".

3.1 Technical Report Series 421: Management of Waste Containing Tritium and Carbon-14 (2004)

The technical report "Management of Waste Containing Tritium and Carbon-14" produced by IAEA and released in 2004 in the Technical Report Series 421, hereafter referred to as TRS-421, attempted to, amongst other things, identify the magnitude of production, release paths and rate of release of both ^{14}C and ^3H. This was done by compiling and analyzing at the time available information.

The results showed a normalized ^{14}C production rate of approximately 1480 $GBq \cdot GW(e)^{-1} \cdot a^{-1}$ in PWRs and 1290 $GBq \cdot GW(e)^{-1} \cdot a^{-1}$ in BWRs. The small discrepancy is mainly due to a slightly higher ^{14}C production in the fuel cladding and coolant/moderator of the PWR. The average normalized release rate of ^{14}C (airborne) was summarized for both PWRs and BWRs for the period 1975-1989. The results are shown in Table 2. As for ^3H, the production was estimated to be $5.55 \cdot 10^5$ and $5.18 \cdot 10^5$ $GBq \cdot GW(e)^{-1} \cdot a^{-1}$for PWRs and BWRs respectively. The liquid normalized release rates were calculated from discharge data before 1980 to be $3.70 \cdot 10^3$ and $2.59 \cdot 10^4$ $GBq \cdot GW(e)^{-1} \cdot a^{-1}$ for BWRs and PWRs respectively. The atmospheric normalized release rates were calculated also before 1980 to be $1.85 \cdot 10^3$ and $3.70 \cdot 10^3$ $GBq \cdot GW(e)^{-1} \cdot a^{-1}$ for BWRs and PWRs respectively, but were also calculated for the period 1975-1989 just as for ^{14}C. The data is summarized in Table 2 [2].

Table 2: Calculated production rate [GBq·GW(e)$^{-1}$·a^{-1}] and average normalized release rate [GBq·GW(e)$^{-1}$·a^{-1}] of ^{14}C and ^{3}H during the period 1975-1989 [2].

^{14}C	PWR	BWR
Production rate [GBq·GW(e)$^{-1}$·a^{-1}]	1480	1290
Normalized release rate (gaseous) 1975-1979 [GBq·GW(e)$^{-1}$·a^{-1}]	222	518
Normalized release rate (gaseous) 1980-1984 [GBq·GW(e)$^{-1}$·a^{-1}]	345	330
Normalized release rate (gaseous) 1985-1989 [GBq·GW(e)$^{-1}$·a^{-1}]	120	450
^{3}H		
Production rate [GBq·GW(e)$^{-1}$·a^{-1}]	$5.55 \cdot 10^5$	$5.18 \cdot 10^5$
Normalized release rate (gaseous) 1975-1979 [GBq·GW(e)$^{-1}$·a^{-1}]	$7.8 \cdot 10^3$	$3.4 \cdot 10^3$
Normalized release rate (gaseous) 1980-1984 [GBq·GW(e)$^{-1}$·a^{-1}]	$5.9 \cdot 10^3$	$3.4 \cdot 10^3$
Normalized release rate (gaseous) 1985-1989 [GBq·GW(e)$^{-1}$·a^{-1}]	$2.8 \cdot 10^3$	$2.5 \cdot 10^3$
Normalized release rate (liquid) -1980 [GBq·GW(e)$^{-1}$·a^{-1}]	$2.59 \cdot 10^4$	$3.70 \cdot 10^3$

As can be seen in Table 2, the gaseous discharges of ^{3}H from PWRs decreased during the period 1975-1989. This was due to better maintenance and management of the reactor systems [2]. The liquid ^{3}H and gaseous ^{14}C discharges showed no apparent decrease during this time period, however for the liquid effluents only data prior to the year 1980 was accounted for. For this data set, about 10-25 % of the produced ^{14}C was released from the PWRs during normal operation, while the same figure for the BWRs was 25-40 %. For ^{3}H, about 0.5 % of that which was produced was released as gas in both PWRs and BWRs, while about 5 % was released as liquid from the PWRs and about 0.7 % was released from the BWRs. As can be seen, most of the produced ^{3}H and ^{14}C is not released during normal operation. As previously stated, most of the production takes place inside components and the fuel, and is therefore mostly contained therein (although some of the activation products diffuse into the coolant).

3.2 "Global and Regional Emissions of Radiocarbon from Nuclear Power Plants from 1972 to 2016" - Zazzeri et al. (2017)

In a paper published in 2017, Zazzeri et al. analyze emission data from various nuclear power plants worldwide, and calculate normalized release rates [4]. The emission data was collected from the European Comissions RAdioactive Discharges Database (RADD) [12], while operational data from the Power Reactor Information System (PRIS) database, operated by IAEA, was used [3]. Different normalized release rates were produced based on reactor type and country (also based on model and age but this is not included here). The results are summarized in Tables 3 and 4. Tabulated is the median value along with the inter-quartile range (in brackets).

Table 3: ^{14}C median normalized release rate [GBq·GW(e)$^{-1}$·a^{-1}] by reactor type from 1995-2015 [4].

Reactor type	1995-2015 ^{14}C median normalized release rate [GBq·GW(e)$^{-1}$·a^{-1}]
PWR	248 [151-360]
BWR	471 [371-630]

Table 4: ^{14}C median normalized release rates 1995-2005 and 2005-2015 for a number of European countries [4].

Location	Reactor type	1995-2005 ^{14}C median normalized release rate [GBq·GW(e)$^{-1}$·a^{-1}]	2005-2015 ^{14}C median normalized release rate [GBq·GW(e)$^{-1}$·a^{-1}]
Germany	PWR	193 [108-284]	256 [185-321]
France	PWR	209 [208-210]	209 [208-210]
Spain	PWR	49 [42-50]	161 [76-232]
UK	PWR	183 [101-196]	190 [176-276]
Other Europe	PWR	330 [208-477]	379 [257-485]
Germany	BWR	401 [293-593]	390 [355-473]
Spain	BWR	-	485 [361-546]
Europe	BWR	539 [469-732]	600 [475-738]

The values shown in Table 3 are similar to those of Table 2, although one of the ^{14}C normalized release rates from PWRs in Table 2 (1985-1979) falls outside the inter-quartile range for PWRs in Table 3. What can be seen as well, and as Zazzeri et al. also note, is that there is a large variation in the median values of the normalized release rate between countries, but also within the same country (large inter-quartile range). Notably, some countries have significantly lower normalized release rate than others, such as for example Spain. In general, the countries specifically listed in Table 4 have a significantly lower normalized release rate than all of Europe (or the rest of Europe). Zazzeri et al. also remark that the small variation in normalized release rate of the French reactors is indicative that these were calculated based on energy production and not measured. Some outliers were also found such as for example Swedish Ringhals 2 reactor, which for the entire period of 1995-2016 showed consistently higher normalized release rates than other reactors. It was also noted that the year-to-year variation in emissions of ^{14}C in all reactors did not show a strong correlation to the year-to-year variation in energy production. Previous studies (e.g. [13]) have shown that emissions can be elevated during periods of outages, which Zazzeri et al. note is the opposite relation expected in the normalized release rate approach [4] where it is assumed that normalized release rate increases linearly with produced energy.

3.3 "Analysis of carbon-14 discharges from Korean nuclear power plants" - Hwapyoung et al. (2024)

In the recently published (2024) paper "Analysis of carbon-14 discharges from Korean nuclear power plants", Kim et al. study the discharges of radionuclides, including both ^{14}C and ^{3}H, from Korean NPPs during the period 2002-2021. Korea currently has 26 operational nuclear reactors, 23 of them being PWRs and 3 being PHWRs (Pressurised Heavy Water Reactors). The time period was in the report divided into 2 intervals, 2002-2011 and 2012-2021. The reasoning behind this was that in 2012, regulations regarding the monitoring of ^{14}C were implemented. Prior to 2012, only the gaseous ^{14}C effluents in the 4 PHWRs that were operational at the time were measured. From 2012 and onwards, measurement of the gaseous ^{14}C effluents in the operational PWRs as well as liquid effluents in the PHWRs were also implemented. The annual average discharges of all PWRs during the period 2012-2021 along with their power generation have been compiled in Table 5. The data for PHWRs have been excluded as they are not relevant to this report.

Table 5: Annual average discharges (GBq and normalized per GWa) of ^{14}C and ^{3}H and power generation (MW(e)a) during 2012-2021 by Korean NPPs summarized from [5]. The original data of ^{3}H discharges by Kim et al. have been normalized to energy production, and the normalized discharges have been converted from MWh to GWa.

Data	2012-2021 annual average
Produced electrical energy (GWa)	3.1035
^{14}C gaseous discharges (GBq)	400
^{14}C normalized release rate (gaseous) [$GBq \cdot GW(e)^{-1} \cdot a^{-1}$]	129 [64-142]
^{3}H gaseous discharges (GBq)	$1.22 \cdot 10^4$
^{3}H normalized release rate (gaseous) [$GBq \cdot GW(e)^{-1} \cdot a^{-1}$]	3930 [898-4047]
^{3}H liquid discharges (GBq)	$4.13 \cdot 10^4$
^{3}H normalized release rate (liquid) [$GBq \cdot GW(e)^{-1} \cdot a^{-1}$]	$1.33 \cdot 10^4$ [7785-$1.2759 \cdot 10^4$]

The ^{14}C normalized release rates of this study corresponded fairly well with both the TRS-421 study as well as the study by Zazzeri et al, and those of the ^{3}H are not noticeably different either. To see the spread in data, the annual normalized release rates were calculated for each site that had PWRs (4 different sites) by using the emission data in [5] and the energy production found in the PRIS database [3]. The lowest and highest of these calculated normalized release rates are given in table 5 inside brackets. As can be seen, the spread in data is quite large. The lower bounds were all found for the site "Shin-Wolsong" which only has two PWRs, and which had considerably lower normalized release rates than other sites. This could either be due to an error in the measured discharges, or it could be that the two PWRs present at this site (which were built recently) utilize some newer technology to reduce discharges. In either case, the lower measured emissions from this site contributes to bringing down the average normalized release rates from the Korean PWRs.

4 Present Study

To see the development in normalized release rate of ^{3}H and ^{14}C from NPPs, discharge data received directly from Swedish NPPs was first studied (Barsebäck [14], Oskarshamn [15], Forsmark [16] and Ringhals [17]). The NPPs were initially studied individually, after which the data was aggregated by reactor type. In the second part of the chapter the entire European nuclear fleet is considered, and the total releases as well as the calculated normalized release rates are presented. The release data was gathered from the RADD[1] database. RADD contains data from European NPPs from 1995 to 2023. For operational data of the NPPs, the PRIS database has been used.

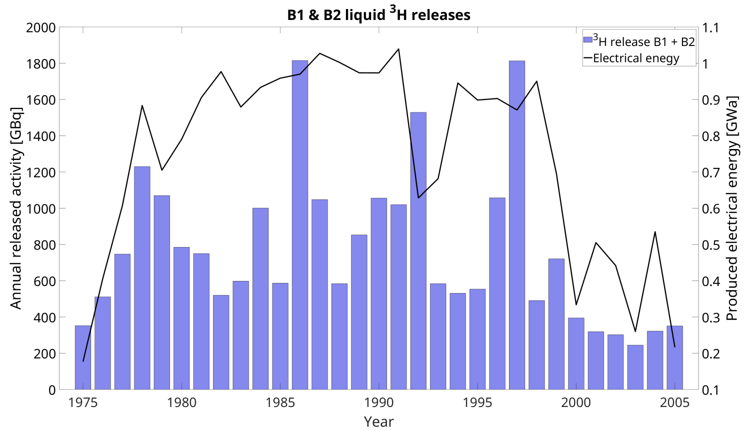
4.1 Swedish NPPs

Sweden has a total of 12 nuclear reactors, 6 of which are operational and 6 of which have been permanently shut down. The reactors are distributed over 4 NPPs; Barsebäck, Forsmark, Oskarshamn and Ringhals.

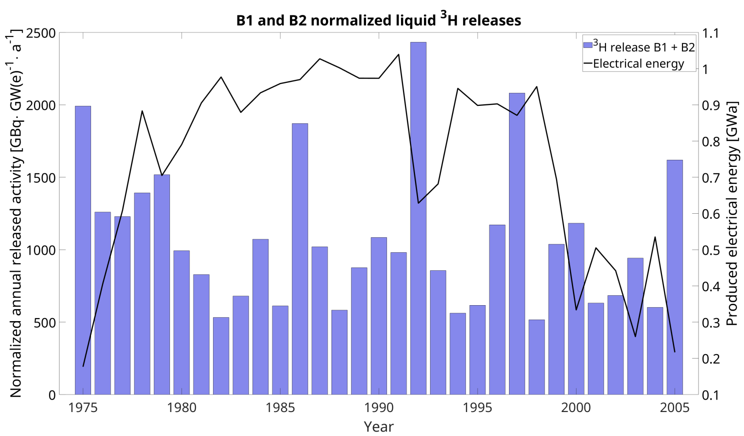
4.1.1 Barsebäck

Barsebäck NPP consists of 2 BWR reactors, B1 and B2 which were taken into operation in 1975 and 1977, and shut down in 1999 and 2005 respectively. Both reactors were of the same model, AA-II developed by ASEA-ATOM, and each with a net-power output of 600 MW. Discharge data for the NPP was recieved directly from Barsebäck Kraft AB [18]. The ^{3}H discharges to water have been measured for the entire operational period of the NPP, and even a few year after B2 shut down (until 2014) [14]. After 2005 the discharges were however only measured during some parts of the year (between 3-9 months), which is why this data has been excluded as to not be misrepresentative. The total ^{3}H discharges along with the discharges normalized to energy production are shown in Figures 1a and 1b. As can be seen, there is a large variation in the normalized emissions. In particular, the normalized release rate seems to increase significantly when the energy production drops. This seems to indicate a delay in the emissions, resulting in a disproportionate normalized release rate during periods of lower energy production.

[1]In fact, also the DIRATA dataset was considered together with RADD. However, some problems with the former (multiple entries for the same discharge, wrong units, missing data, etc...) lead to us using RADD only as our source of release data



(a)



(b)

Figure 1: Aggregated ^{3}H a) total discharges to water (GBq), b) Normalized release rate (GBq·GW(e)$^{-1}$·a^{-1}), from B1 and B2 during 1975-2005. Produced energy shown as black line.

Measurement of atmospheric ^{3}H and ^{14}C releases started in 2002, after B1 had already shut down. This means there was only annual data for 4-5 years for the atmospheric release of these nuclides (^{3}H was measured also in 2006). The measurements, normalized to produced energy, are shown in Figure 2, where it can be seen that the chemical form of the nuclides was also measured. As there are only 4 measuring points it is very difficult to ascertain any particular trend in this data set.

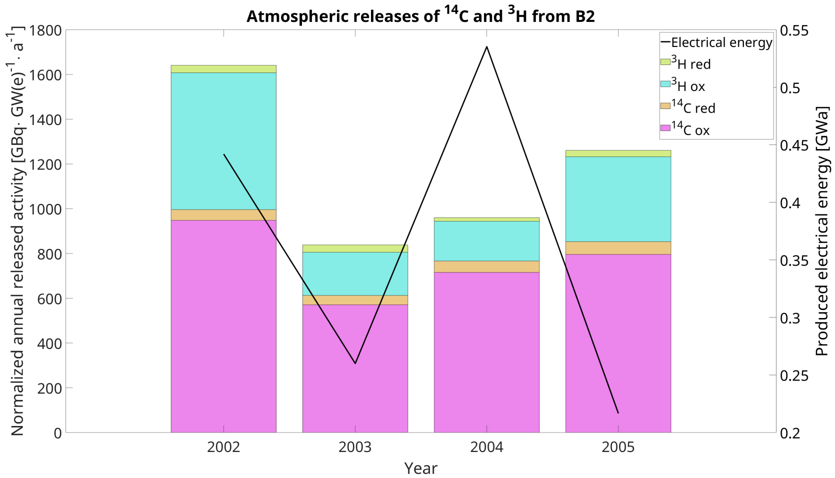
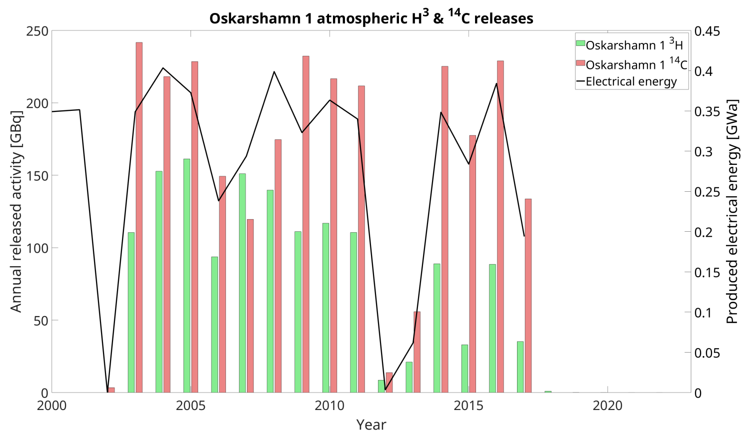


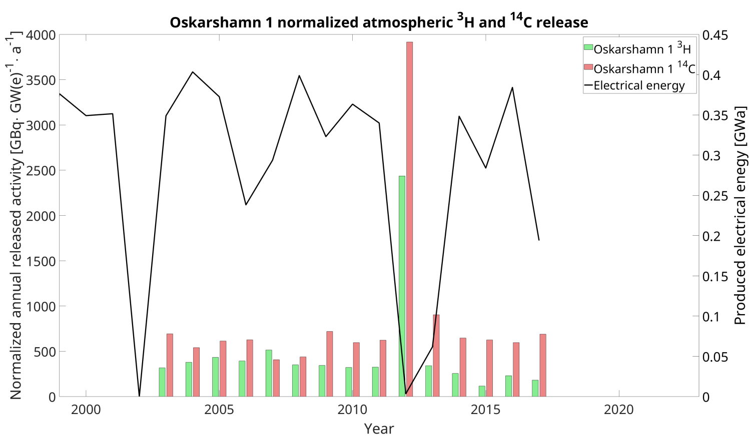
Figure 2: Atmospheric releases of ^{3}H and ^{14}C from B2 during 2002-2005 normalized to produced electrical energy (GBq/GWa). The produced energy is shown as a black line.

4.1.2 Oskarshamn

Oskarshamn NPP consists of 3 BWR reactors O1-O3, 2 of which have already shut down (O1 and O2). O1, O2 and O3 were also produced by ASEA-ATOM (models AA-I, AA-II and AA-IV respectively), and first started producing power in 1971, 1974 and 1985 with respective power outputs of 440 MW_e, 565 MW_e and 1055 MW_e. By the end of its life in 2017, O1 had gone through 2 power upgrades, with a final power output of 473 MW_e. O2 shut down some years prior in 2013, also having recieved several power upgrades during its lifetime, the last of which was still underway when it was shut down. Its final power output was 638 MW_e. O3 is still operating with operation planned until at least 2045. Just like O1 and O2, O3 has had its power upgraded several times since it was first built, with the latest one increasing the power output to 1400 MW_e making it one of the worlds largest BWRs in this regard [3]. Measurements on release data from O1, O2 and O3 was provided by OKG AB [15]. The atmospheric releases of O1 are shown in Figure 3. As can be seen, the total releases follow the produced energy quite well, but when examining the normalized releases, these spike when the energy production is especially low just as was seen for the Barsebäck liquid releases. The same effect can also be seen in Figure 4 which shows the liquid releases of ^{3}H from both O1 and O2. These are shown together since the measurement is done on a channel through which both reactors release ^{3}H. It is thus impossible to distinguish between the releases from each reactor.

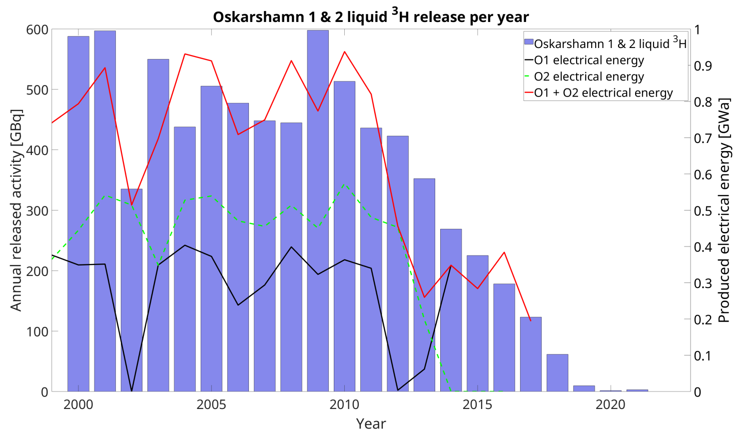


(a)

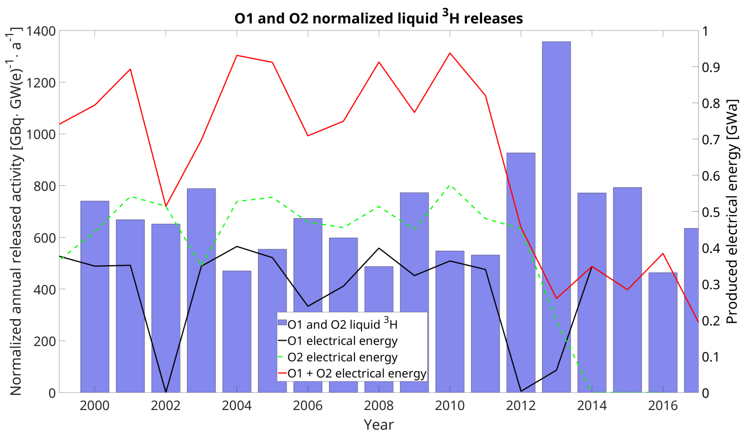


(b)

Figure 3: a) O1 atmospheric releases (GBq) after the year 2000 (bar graphs). b) O1 atmospheric normalized release rates (GBq·GW(e)$^{-1}$·a^{-1}) after the year 2000 (bar graphs). Produced energy shown as a black line in both graphs.

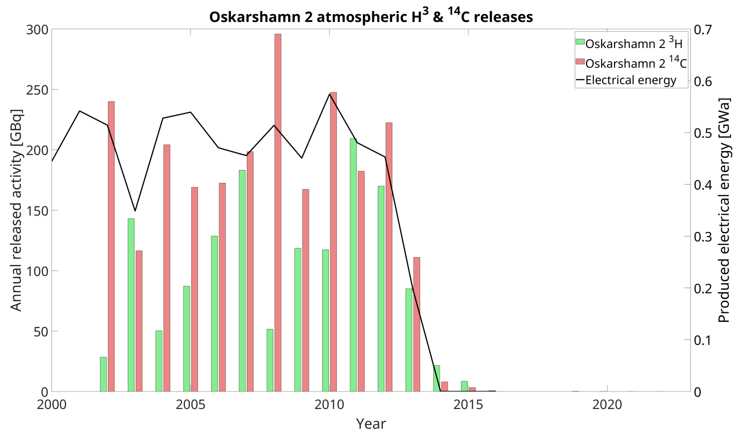


(a)

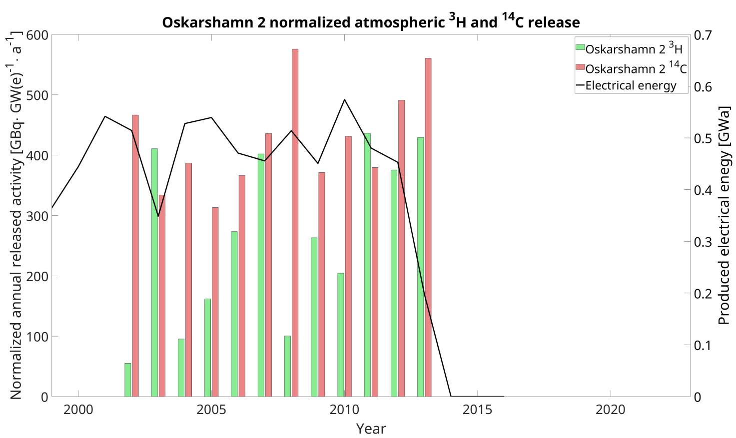


(b)

Figure 4: O1 and O2 liquid releases (GBq) after the year 2000 (bar graphs) a) total, and b) normalized to produced energy (GWa). Produced energy for O1, O2 and O1+O2 shown as different lines.



(a)

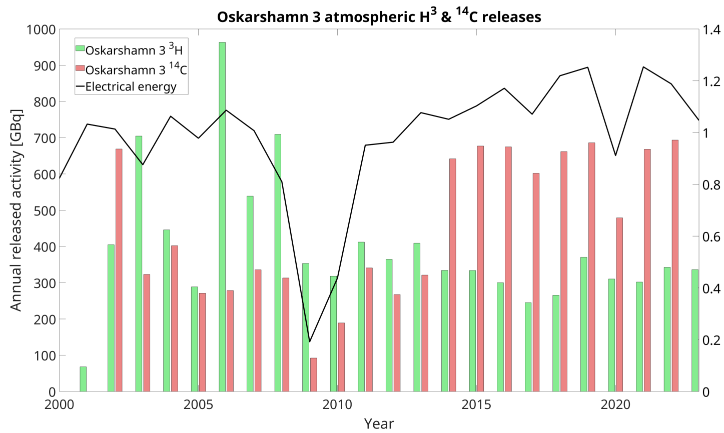


(b)

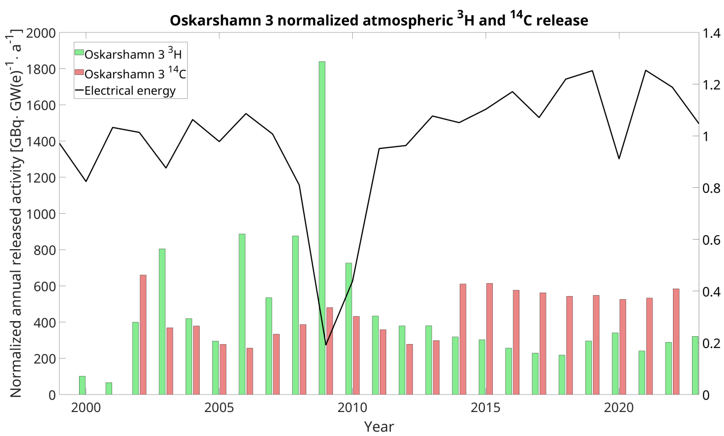
Figure 5: O2 atmospheric releases (GBq) after the year 2000 (bar graphs) a) total, and b) normalized to produced energy (GWa). Produced energy shown as black line.

Figure 5 shows the atmospheric releases from O2. Just like it did for O1, the total discharges seem to follow the energy production quite well. Examining O3 instead, the releases of which are shown in Figures 6 and 7, the same effects as before can be seen. The total discharges seem to follow the energy production, but the normalized releases spike when the energy production drops by a large amount, as can be seen especially for the year 2009. This was the year the last power-upgrade of O3 was initiated, meaning the energy production was halted. The primary loop was also most likely emptied somewhat to allow for the work being carried out. This in turn caused the spike in liquid releases seen, which coupled with the halted energy production lead to an abnormally large spike in normalized liquid release rate.

The liquid (total) releases of ^{3}H seem to be about a factor 2-5 higher than for the atmospheric ^{3}H releases, which is consistent with the values in Table 2 from TRS-421 [2]. The atmospheric ^{14}C discharges however seem to be higher than the atmospheric ^{3}H discharges which is not consistent with the same table. Comparing the normalized discharges from Oskarshamn to those of Table 2 and 3, it is evident that the atmospheric ^{14}C discharges are within the same range, or perhaps slightly higher in the Oskarshamn reactors than for a general European BWR. The ^{3}H discharges are, however, significantly lower (factor 5 approximately) in the Oskarshamn reactors than those tabulated in Table 2 which indicates that these discharges have decreased since the values from TRS-421 were calculated.

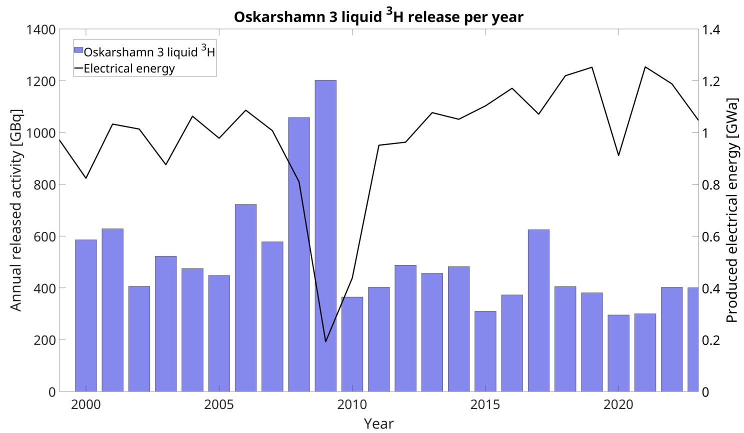


(a)

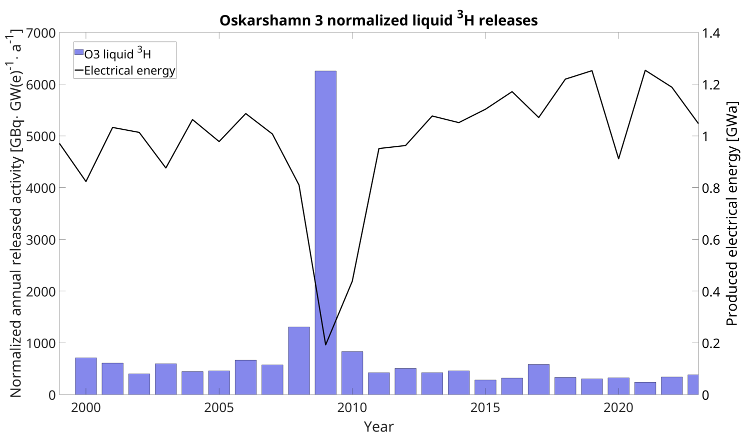


(b)

Figure 6: O3 atmospheric releases (GBq) after the year 2000 (bar graphs) a)total, and b) normalized to produced energy (GWa). Produced energy shown as black line.



(a)



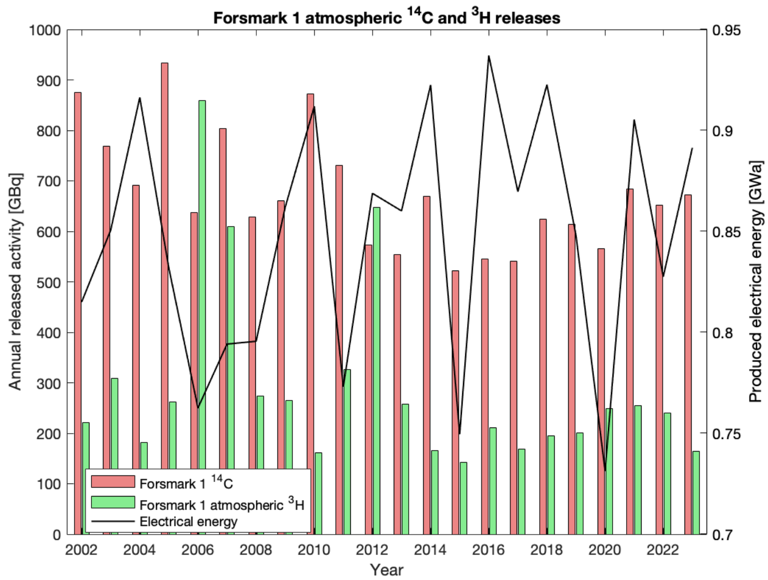
(b)

Figure 7: O3 liquid releases (GBq) after the year 2000 (bar graphs) a) total, and b) normalized to produced energy (GWa). Produced energy shown as black line.

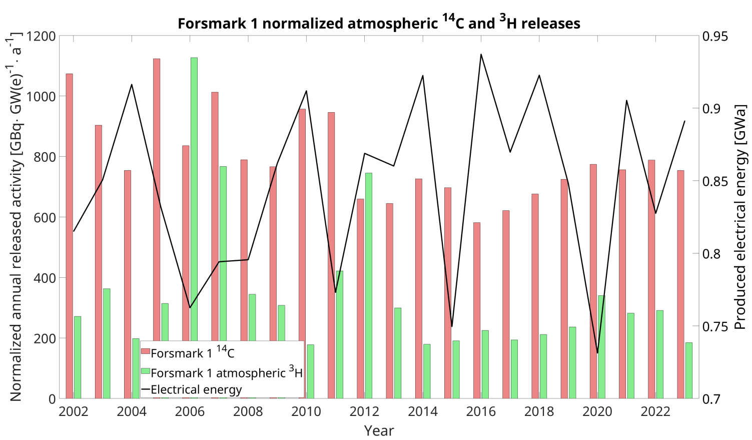
4.1.3 Forsmark

Forsmark NPP consists of 3 BWR reactors. Just as for Barsebäck and Oskarshamn, the Forsmark reactors were produced by ASEA-ATOM. F1 and F2 are of the same model (AA-III) with an initial power output of 900 MW_e, while F3 is the same model as O3 (AA-IV) with an initial power output of 1050 MW_e. The reactors first started producing power in 1980, 1981 and 1985 respectively and all are currently still operational. Due to power upgrades the power output of F1 is now 1014 MW_e, the power output of F2 is 1121 MW_e and the power output of F3 is 1172 MW_e [19]. The liquid releases of ^3H have been measured for most of the NPP's operational period. As for the atmospheric releases of ^{3}H and ^{14}C, these have been measured since 2002. These were collected in a database recieved directly from the NPP [16]. In regards to this data, Forsmark themselves have deemed that the equipment used to measure the atmospheric ^{14}C release data is sometimes prone to errors. The previous measuring equipment which has been used since 2002 has therefore recently been exchanged (2023) [20]. The liquid releases are measured in terms of total ^{3}H, while for the atmospheric releases only the oxidized forms of the nuclides are measured. This is reasonable for a BWR as the vast majority of the releases are in the oxidized form (see for example R1 in Figures 13 and 14 in the next sub-chapter). The atmospheric releases from F1 and F2 are shown in Figures 8 and 10 respectively. The liquid releases from F1 and F2 (aggregate) are shown in Figure 9. The liquid and atmospheric releases from F3 are shown in Figures 11 and 12.

As can be seen from the atmospheric releases of F1-F3, these are all quite similar, both in shape and in values. Compared to the atmospheric releases from O1-O3, the releases from F1-F3 are much more stable and spike less with changing energy production. Comparing the values between the Forsmark and Oskarshamn reactors, the normalized ^{3}H release rates are quite similar, but the normalized ^{14}C release rates from F1 and F2 are somewhat higher, in the range of 50 %. This also means that they are higher than those shown in Table 2. The normalized release rates from F3 are however more similar to those from the Oskarshamn reactors, as are the normalized liquid releases from F1-F3.

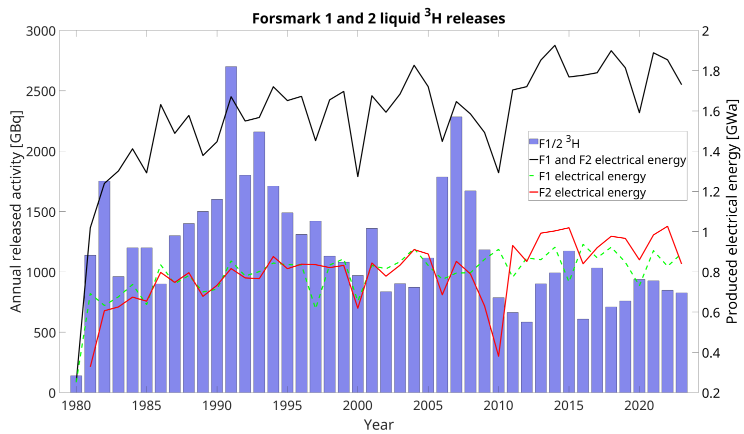


(a)

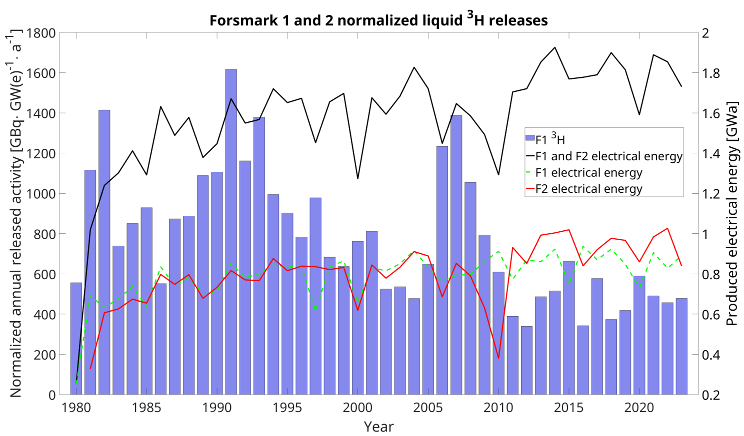


(b)

Figure 8: F1 atmospheric releases (GBq) after the year 2002 (bar graphs) a) total, and b) normalized to produced energy (GWa). Produced energy shown as black line.

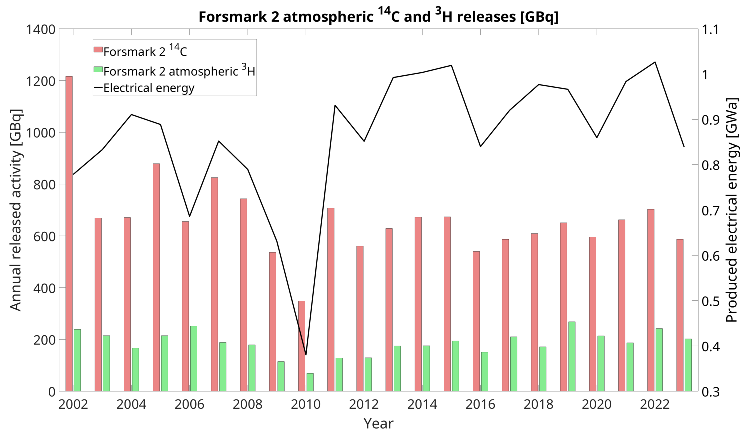


(a)

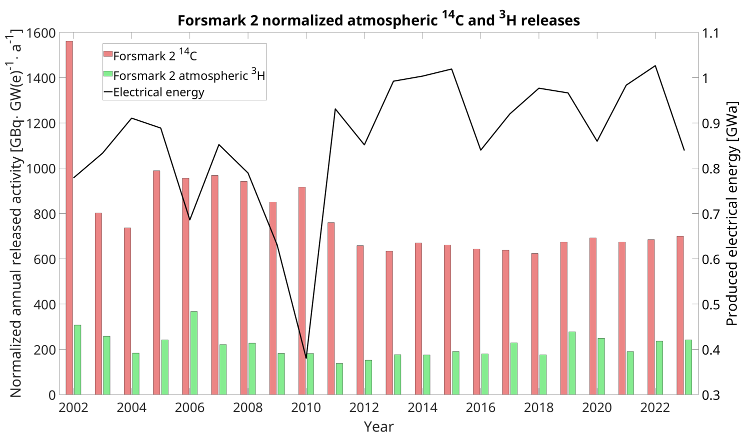


(b)

Figure 9: F1 & F2 liquid releases (GBq) a) total, and b) normalized to produced energy (GWa). Produced energy for F1, F2 and F1+F2 shown as different lines.

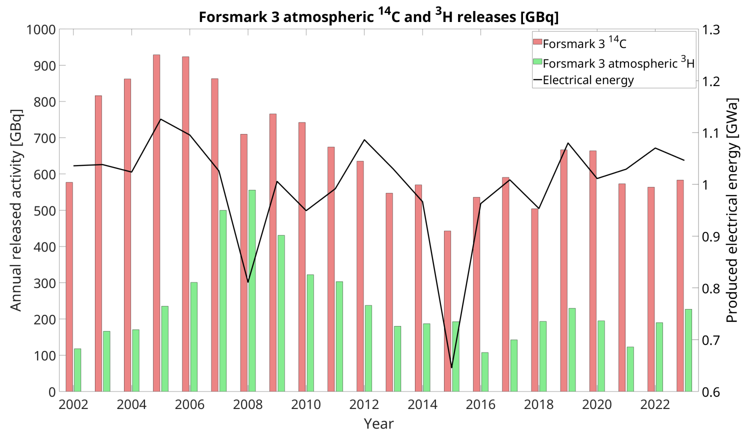


(a)

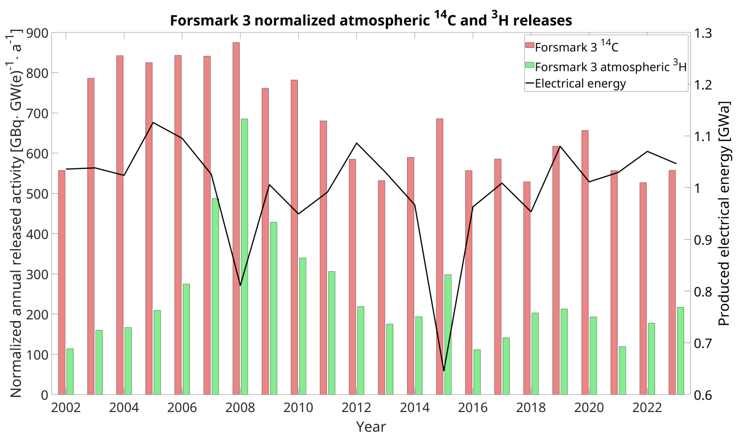


(b)

Figure 10: F2 atmospheric releases (GBq) after the year 2002 (bar graphs) a) total, and b) normalized to produced energy (GWa). Produced energy shown as black line.

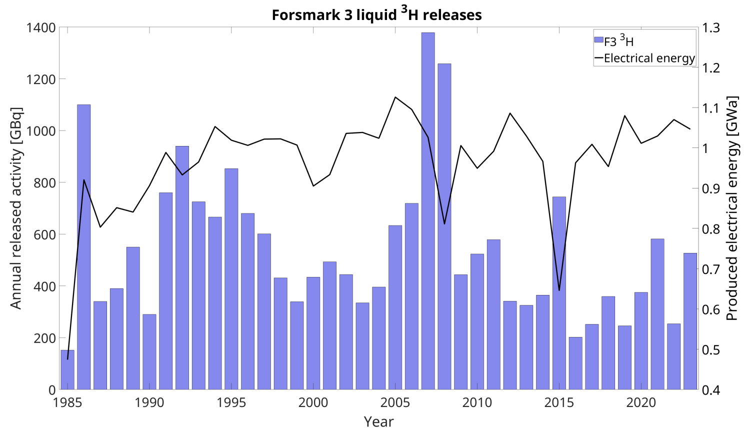


(a)

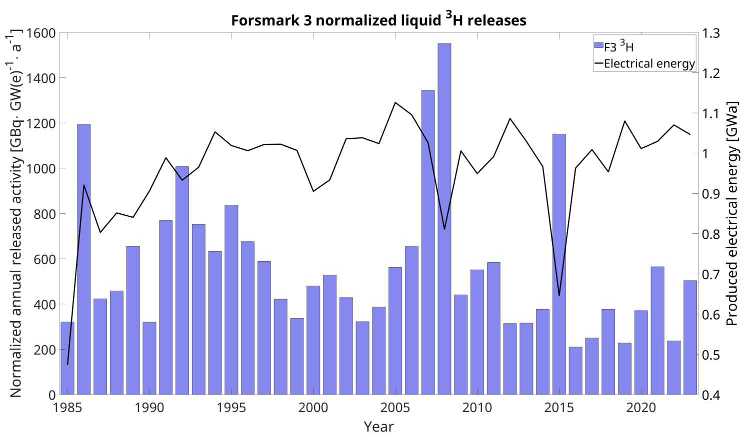


(b)

Figure 11: F3 atmospheric releases (GBq) after the year 2002 (bar graphs) a) total, and b) normalized to produced energy (GWa). Produced energy shown as black line.



(a)



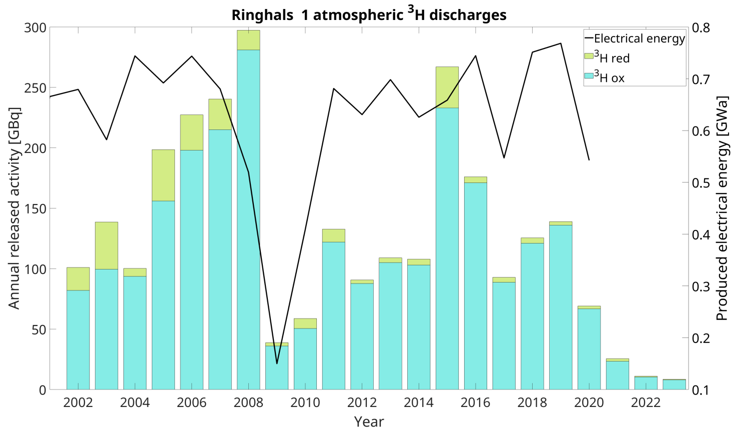
(b)

Figure 12: F3 liquid releases (GBq) a) total, and b) normalized to produced energy (GWa). Produced energy shown as black line.

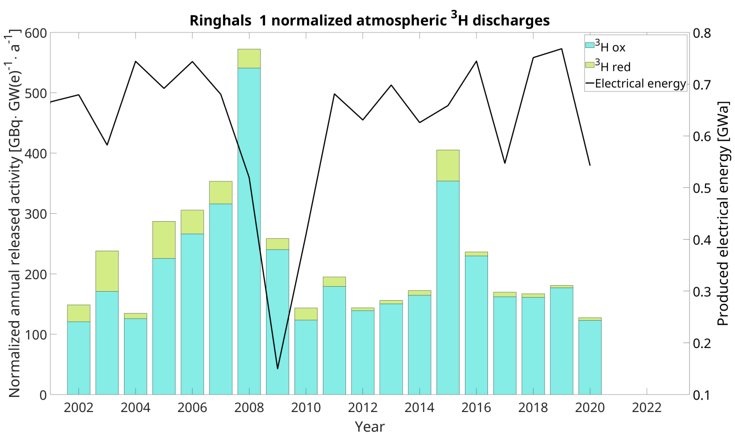
4.1.4 Ringhals

Ringhals NPP consists of 4 reactors, 1 BWR (R1) and 3 PWRs (R2-R4). R1 was produced by ASEA-ATOM and is of the same model as eg. B1 and B2 (AA-I). It started commercial production in 1976 with a power output of 760 MW_e. Due to power upgrades, the output power of R1 was 881 MW_e by the time it was shut down in 2020 [3]. R2-R4 are all 3-loop PWRs made by Westinghouse. R2 had an initial power output of 800 MW_e when it started of operation in 1975, and by the time it shut down in 2019 its power had been increased to 900 MW_e. R3 and R4 both had an initial power output of 915 MW_e when they started operation in 1981 and 1983 respectively. Both are still operational and have had their respective power outputs increased to 1074 MW_e and 1130 MW_e [3]. The measured releases of all 4 reactors was recieved directly from Vattenfall which owns and operates the NPP [17]. Figures 13 and 14 show the atmospheric ^{3}H and ^{14}C releases of R1, chemical form included. The liquid releases can be seen in Figure 15. As can be seen, most of the releases are in the oxidized form, which is to be expected from a BWR. Comparing this to the releases of R2, R3 and R4 which are shown in Figures 16 - 24, it can be seen that for the PWRs there is a much larger fraction of reduced ^{14}C.

R1 is the only BWR reactor of Ringhals NPP. Comparing the discharge values the same way as was done for the Oskarshamn NPP it is apparent that the ratio between normalized liquid and atmospheric ^{3}H discharges are as expected, but the values are much higher than for the Oskarshamn NPP. The normalized ^{3}H discharges are still about a factor 2 lower than those tabulated in Table 2 however, which indicates that also in R1 the discharge rate of ^{3}H has decreased since the values in Table 2 were calculated. The normalized ^{14}C discharges are also higher in R1 than for the Oskarshamn NPP, and comparing to Tables 2 and 3 the R1 ^{14}C discharges are approximately 50% higher.

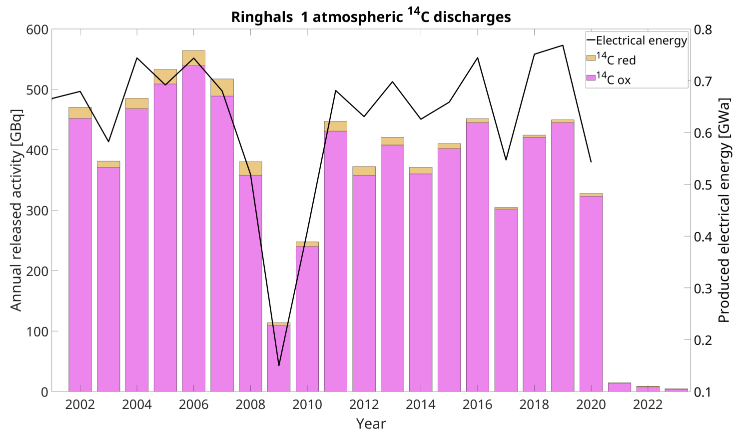


(a)

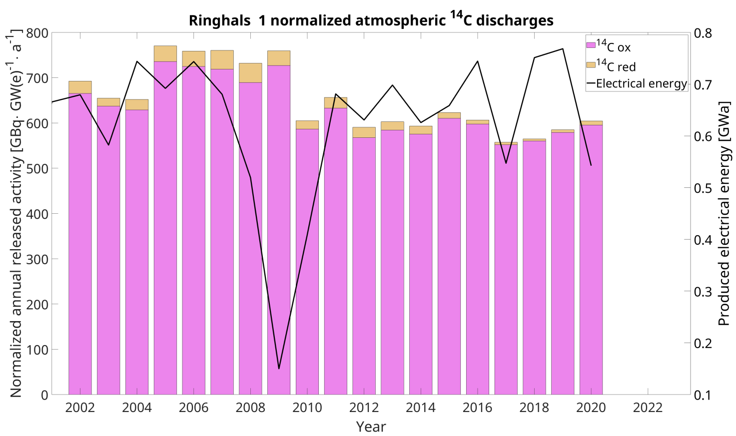


(b)

Figure 13: R1 (BWR) atmospheric ^{3}H releases (GBq) (bar graphs) a)total, and b) normalized to produced energy (GWa). Produced energy shown as black line.

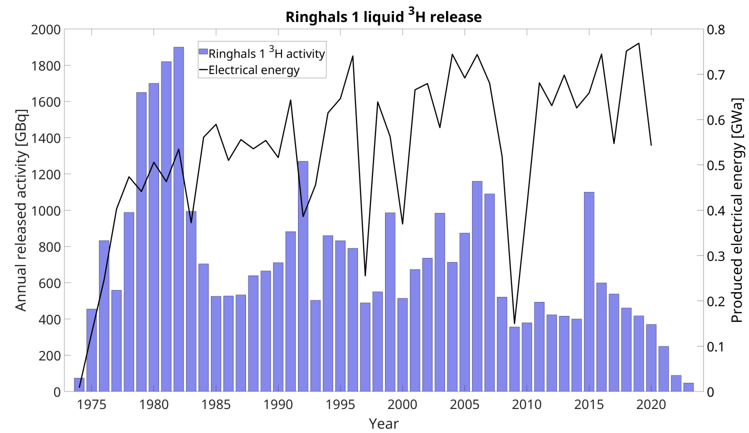


(a)

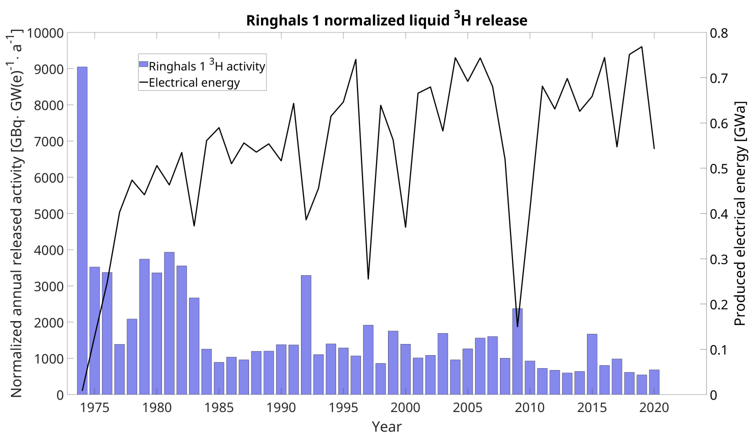


(b)

Figure 14: R1 (BWR) atmospheric ^{14}C releases (GBq) (bar graphs) a) total, and b) normalized to produced energy. Produced energy shown as black line.

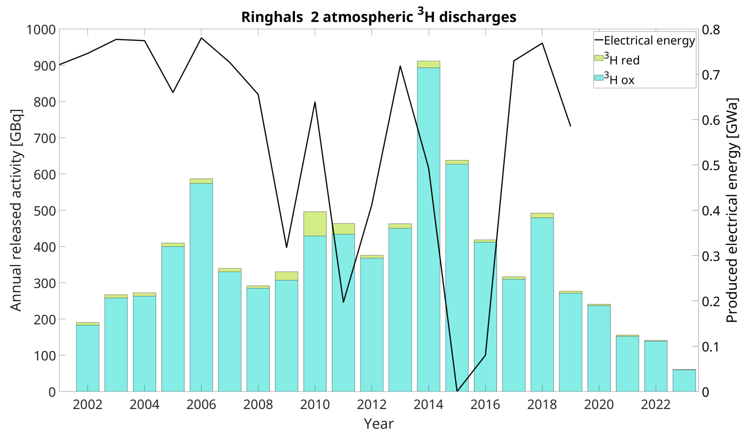


(a)

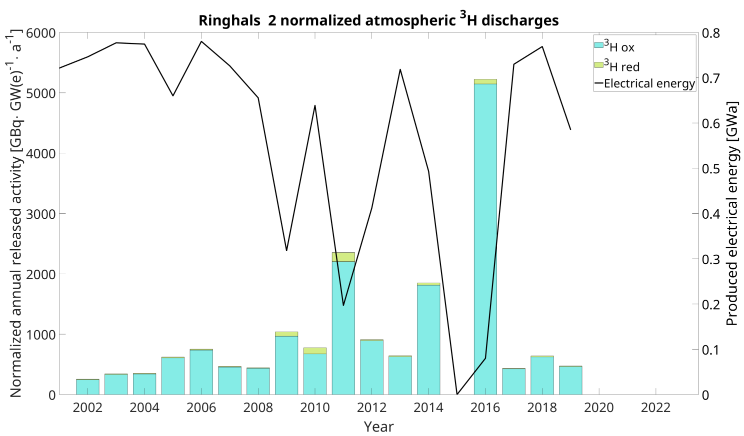


(b)

Figure 15: R1 (BWR) liquid ^{3}H releases (GBq) (bar graphs) a) total, and b) normalized to produced energy. Produced energy shown as black line.

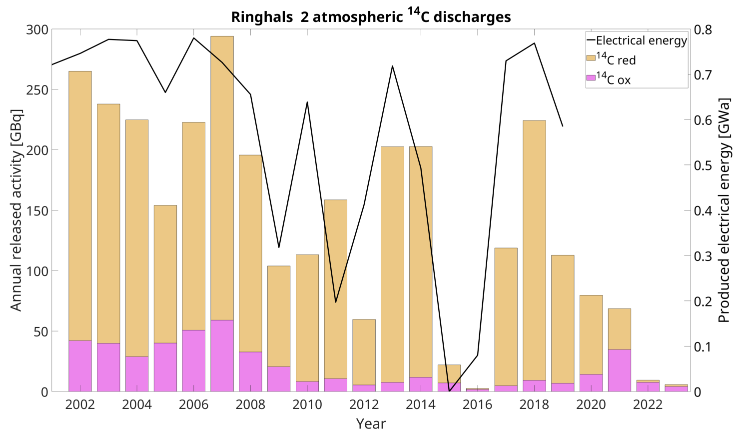


(a)

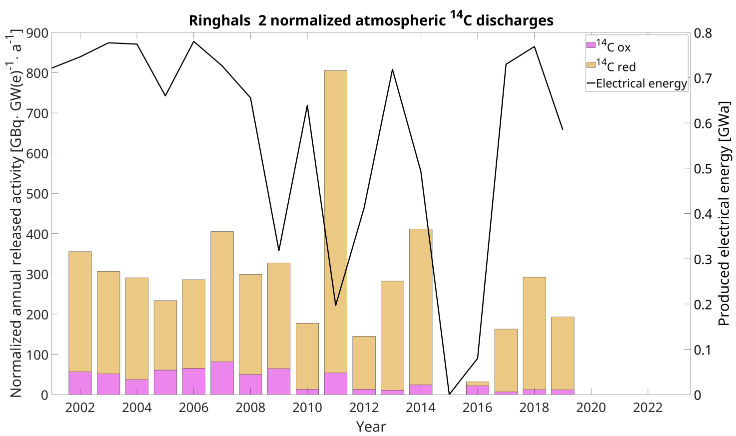


(b)

Figure 16: R2 (PWR) atmospheric ^{3}H releases (GBq) (bar graphs) a) total, and b) normalized to produced energy (GWa). Produced energy shown as black line.

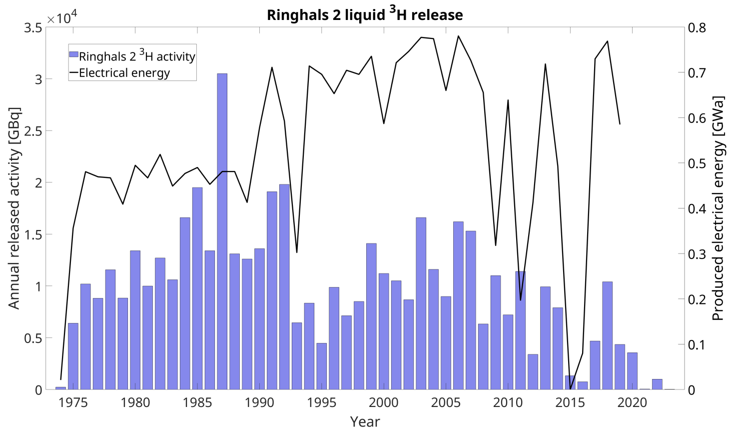


(a)

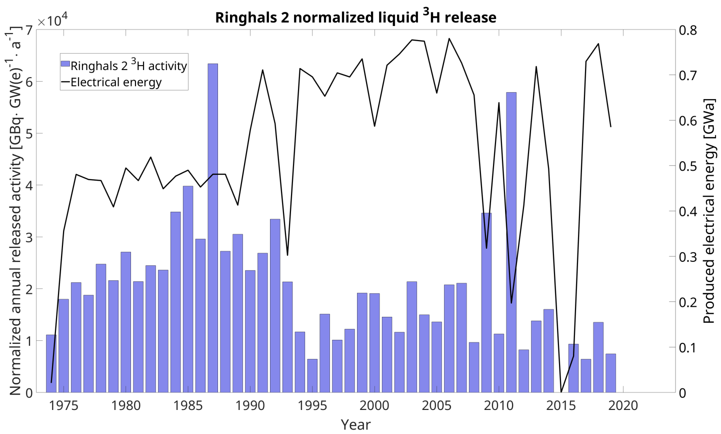


(b)

Figure 17: R2 (PWR) atmospheric ^{14}C releases (GBq) (bar graphs) a) total, and b) normalized to produced energy. Produced energy shown as black line.

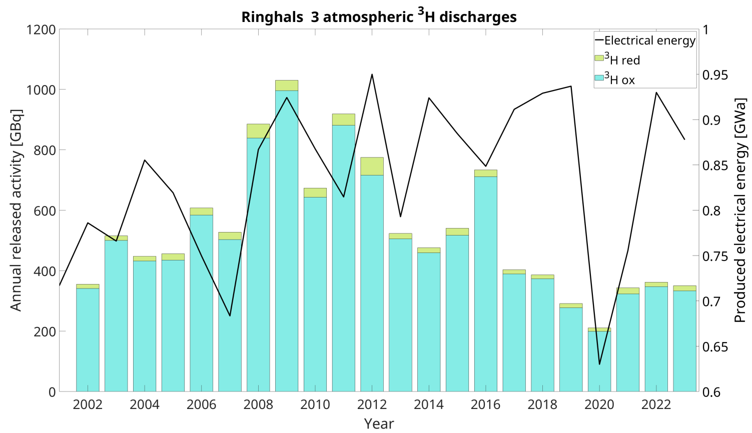


(a)

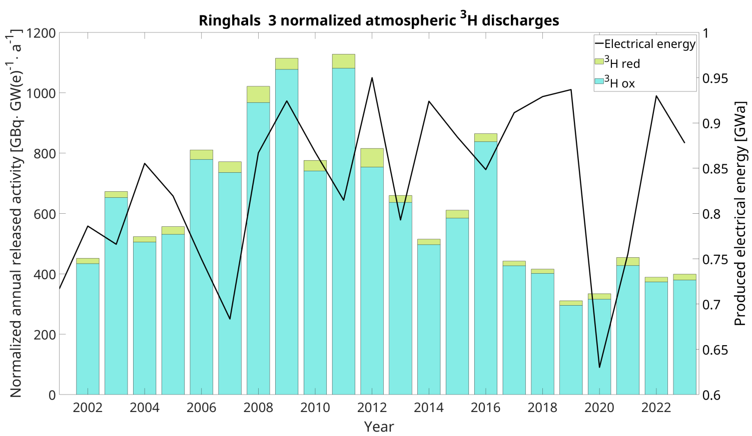


(b)

Figure 18: R2 (PWR) liquid ^{3}H releases (GBq) (bar graphs) a) total, and b) normalized to produced energy. Produced energy shown as black line.

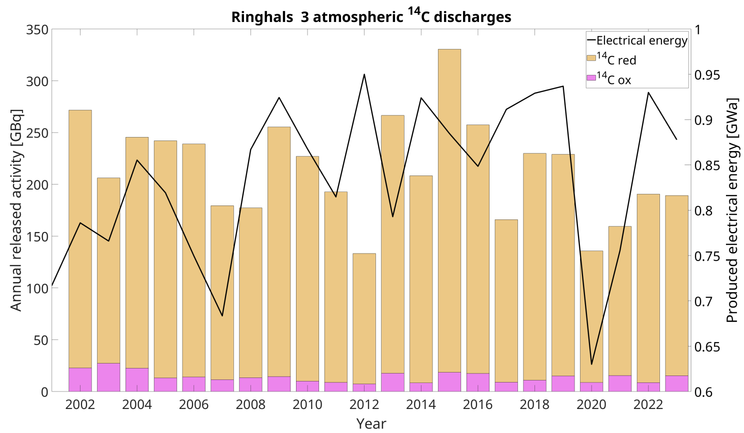


(a)

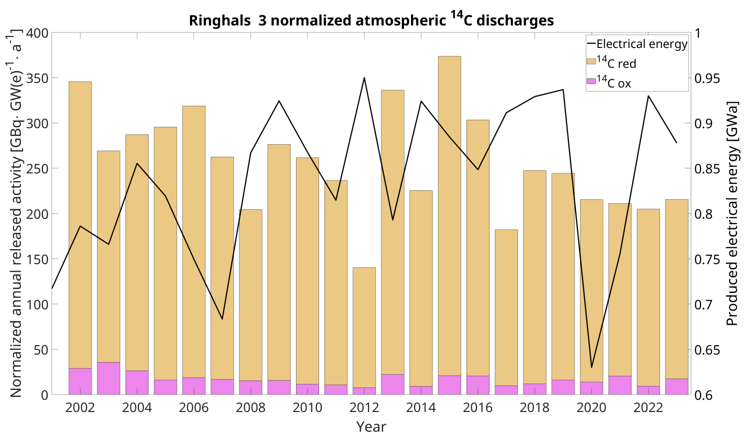


(b)

Figure 19: R3 (PWR) atmospheric ^{3}H releases (GBq) (bar graphs) a) total, and b) normalized to produced energy (GWa). Produced energy shown as black line.

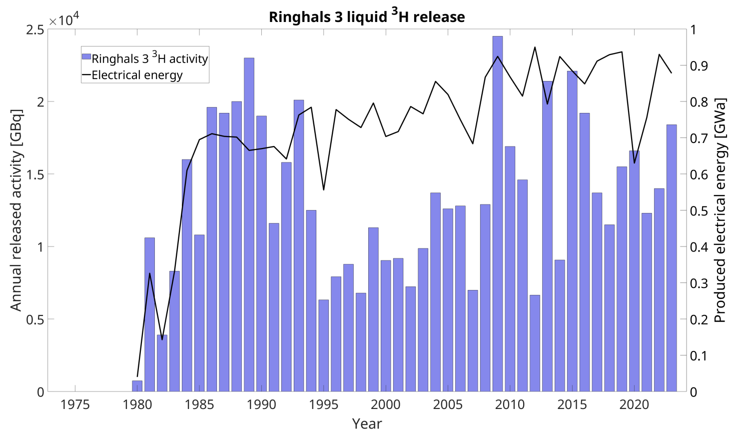


(a)

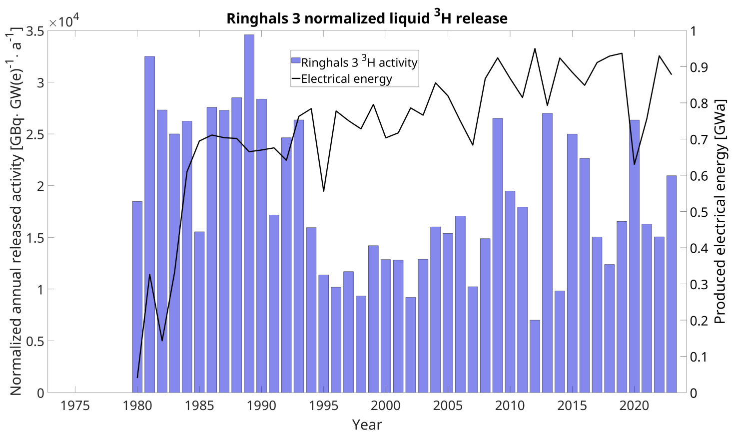


(b)

Figure 20: R3 (PWR) atmospheric ^{14}C releases (GBq) (bar graphs) a) total, and b) normalized to produced energy. Produced energy shown as black line.

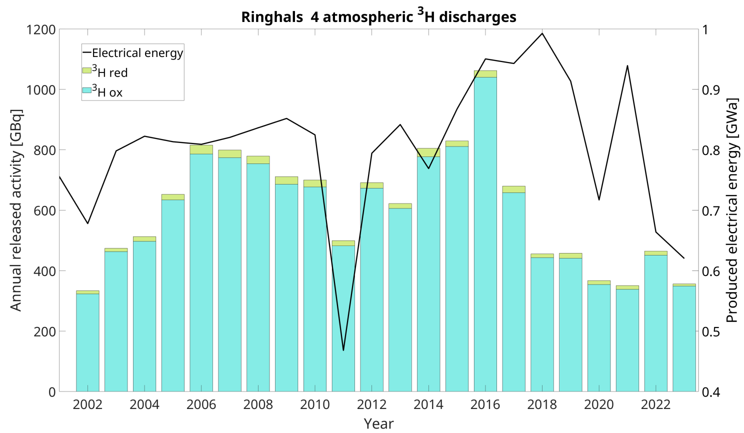


(a)

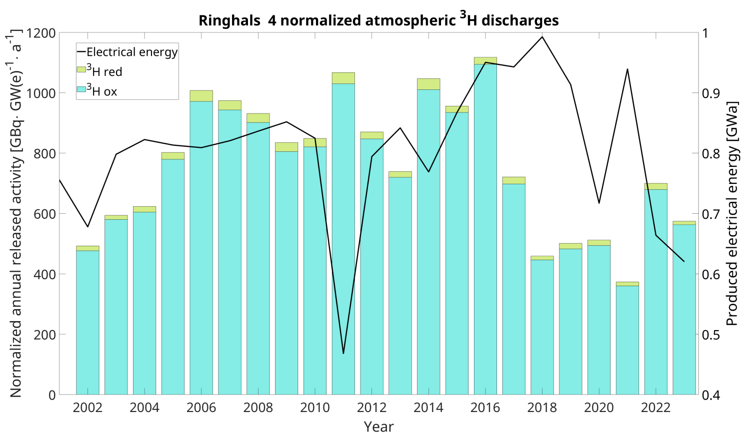


(b)

Figure 21: R3 (PWR) liquid ^{3}H releases (GBq) (bar graphs) a) total, and b) normalized to produced energy. Produced energy shown as black line.

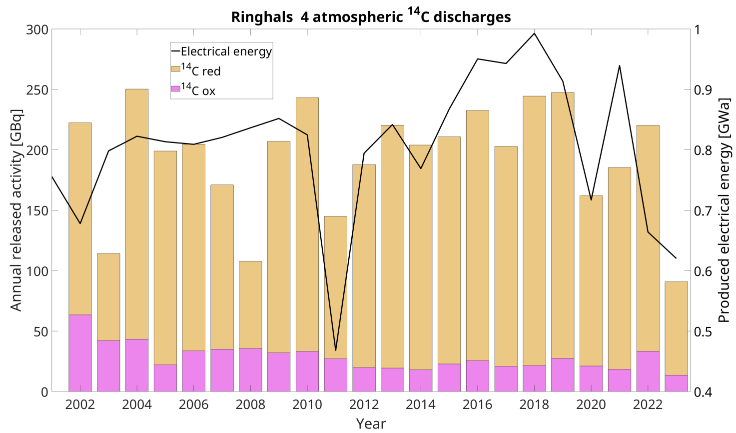


(a)

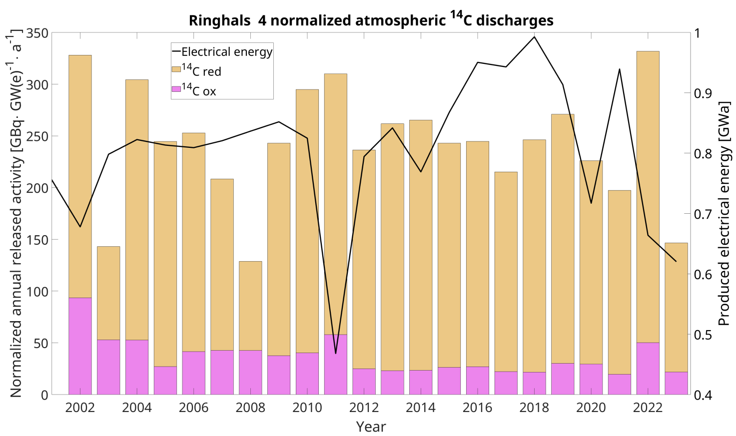


(b)

Figure 22: R4 (PWR) atmospheric ^{3}H releases (GBq) (bar graphs) a) total, and b) normalized to produced energy (GWa). Produced energy shown as black line.

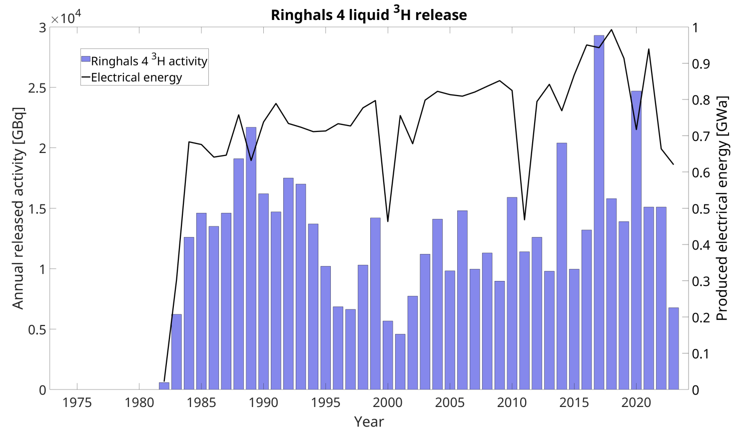


(a)

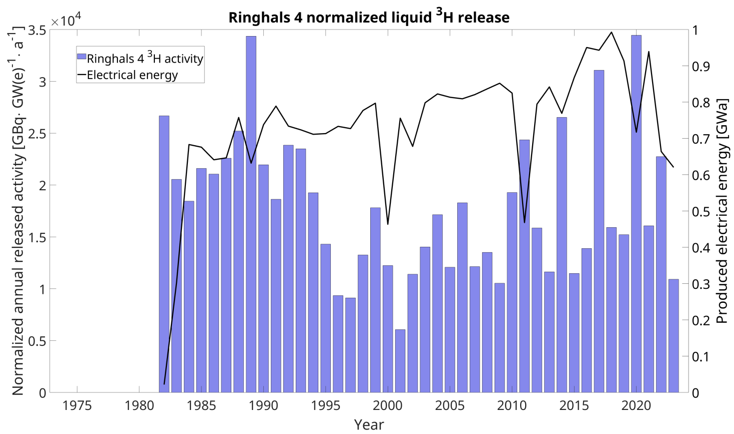


(b)

Figure 23: R4 (PWR) atmospheric ^{14}C releases (GBq) (bar graphs) a) total, and b) normalized to produced energy. Produced energy shown as black line.



(a)



(b)

Figure 24: R4 (PWR) liquid ^{3}H releases (GBq) (bar graphs) a) total, and b) normalized to produced energy. Produced energy shown as black line.

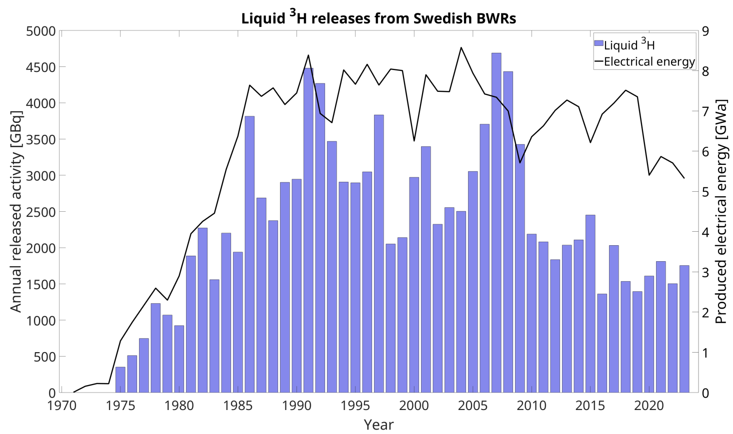
The mean atmospheric ^{3}H release rates of R2-R4 were 1034, 638 and 761 GBq/GWa respectively, while the atmospheric ^{14}C release rates were 294, 257 and 243 GBq/GWa. Lastly, the mean liquid ^{3}H release rates were 20944, 18897 and 18056 GBq/GWa respectively for R2-R4. Comparing these values for example to the Korean PWRs in Table 5, the normalized discharges of ^{14}C are in general a bit lower in the Korean PWRs than in the Swedish PWRs, as are the normalized liquid ^{3}H release rates. The normalized atmospheric ^{3}H release rates of the Ringhals PWRs are however lower than in the Koran PWRs. Comparing the R2-R4 values to Table 2, the normalized atmospheric ^{14}C release rate is similar , as is the liquid ^{3}H release rate. The normalized atmospheric ^{3}H release rates are however significantly lower in the R2-R4 PWRs, about a factor

3 comparing the highest mean value from R2-R4 (1034 GBq/GWa in R2) to the lowest value from 2 (2800 GBq/GWa). This indicates a further decrease of these discharges than what could be seen in Table 2. Comparing the normalized ^{14}C release rates of R2-R4 to the one found in Table 3, these are similar.

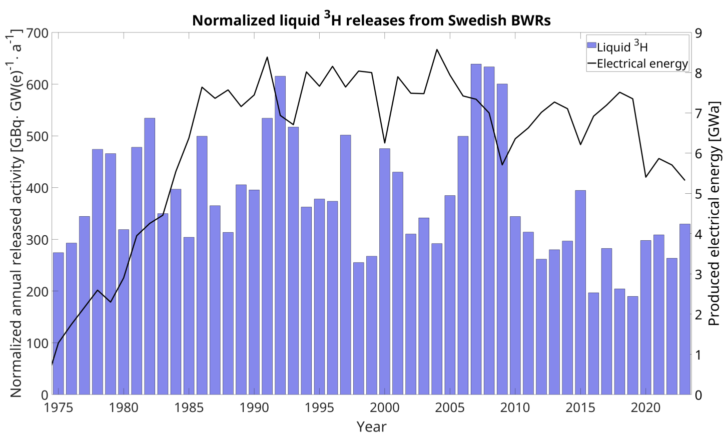
4.1.5 Swedish BWRs

Aggregating the releases and electrical energy production for all Swedish BWRs (9 total, however the contribution of B1 and B2 to the atmospheric releases is minor) yields Figures 25a, 25b, 26a and 26b. It's not obvious from these figures if there has been any change in the normalized discharges. Taking the median normalized discharge rates, these are 457.5 GBq/GWa for the atmospheric ^{14}C (2002-2023), 347.1 GBq/GWa for the liquid ^{3}H (1975-2023) and 181.6 GBq/GWa for the atmospheric ^{3}H (2002-2023). This data is also compiled in Table 6 along with the data for Swedish PWRs.

A linear fit of the data in Figures 25b and 26b yields instead Figures 27a, 27b and 27c. From the negative slopes in these figures it seems like all of the discharges are slowly decreasing with time. The p-value for each of linear fits is 0.04654, 0.87911 and 0.03562 respectively, meaning that the negative slope of the normalized liquid ^{3}H and atmospheric ^{14}C release rates are significantly different from zero. It is difficult however to draw any general conclusions about BWRs just from this data set as the sample size is small. This is also apparent from the large 95 % prediction intervals also shown in the figures. As stated in the Forsmark report "Utsläpp av radioaktiva ämnen 2023" [20] ("Discharges of radioactive substances 2023"), there is a regulation in place that Swedish NPPs should have a program which aims to decrease discharges long-term [21]. In the report it is stated that the airborne discharges are difficult to affect as they are tied to the energy production, but efforts have been made to decrease the liquid discharges. The negative slopes of Figures 27a, 27b and 27c could therefore also indicate that these efforts have had an effect at least in Swedish BWRs.

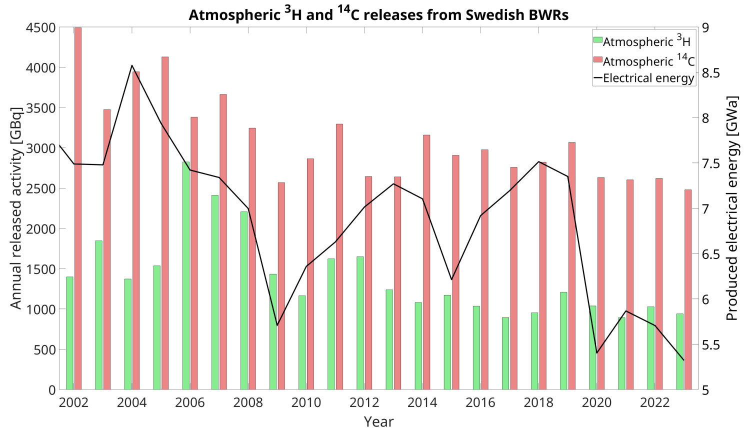


(a)

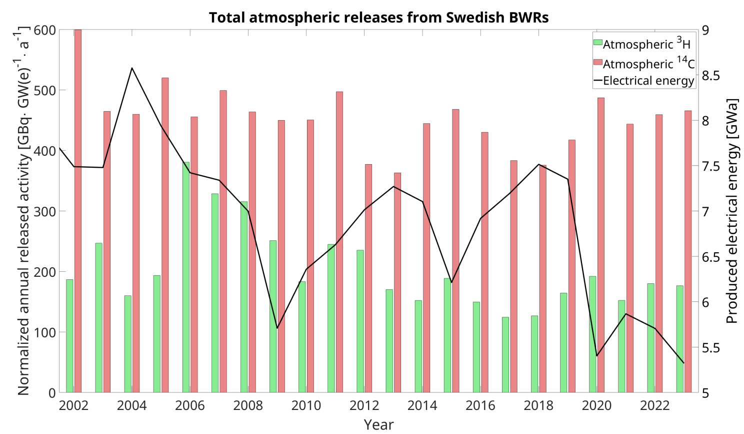


(b)

Figure 25: Liquid releases of ^{3}H from Swedish BWRs a) total, and b) normalized to energy production (GBq/GWa). Produced electricity shown as black line (GWa).

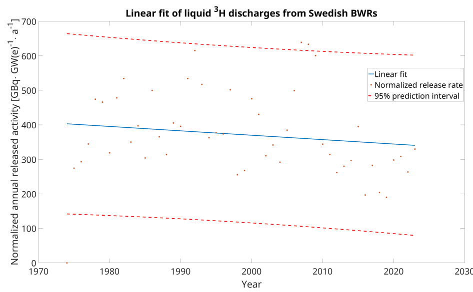


(a)

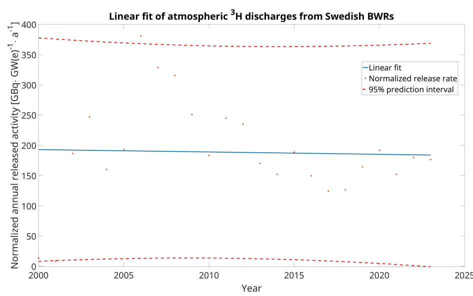


(b)

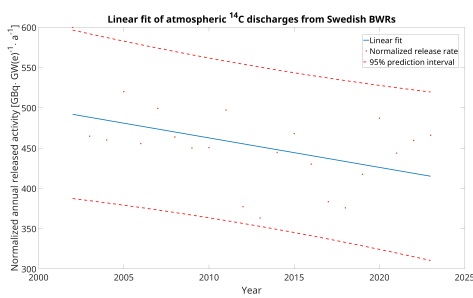
Figure 26: Atmospheric releases from Swedish BWRs a) total, and b) normalized to produced electricity (GBq/GWa). Produced electricity shown as black line (GWa).



(a)



(b)



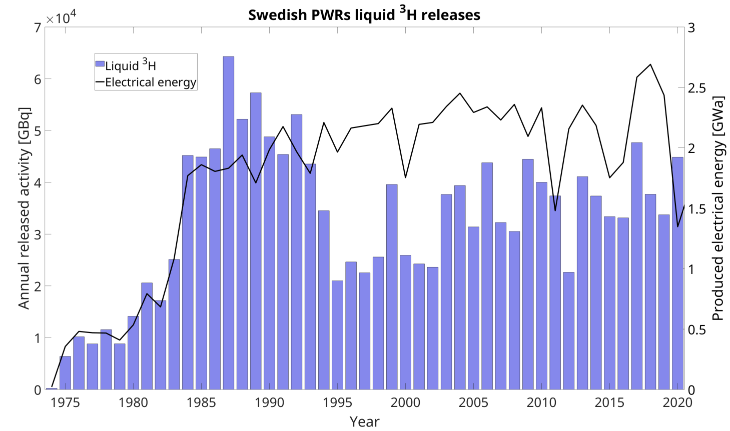
(c)

Figure 27: Linear fits of normalized release rates with time for a) liquid ^{3}H, b) atmospheric ^{3}H and c) atmospheric ^{14}C discharges from Swedish BWRs. The normalized release rates are shown as red stars while the 95 % prediction interval is shown as red dashed lines.

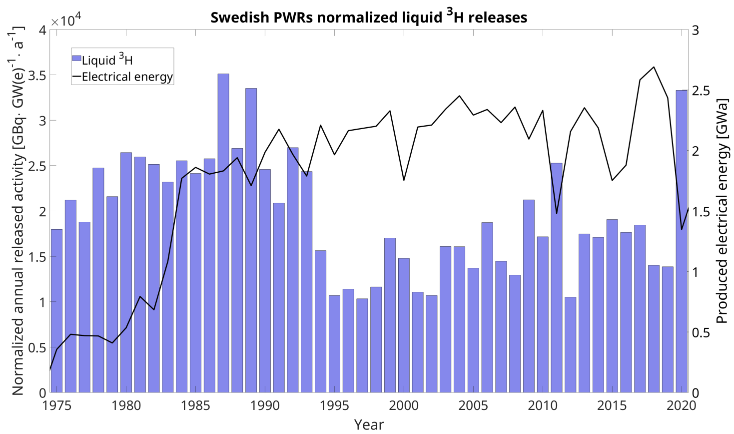
4.1.6 Swedish PWRs

Aggregating the results of all Swedish PWRs yields the graphs in Figures 28 and 29. As there are only 3 PWRs in Sweden it is not possible to say anything general about the normalized emissions from PWRs. It does however seem that the normalized liquid ^{3}H emissions decreased in the beginning of the 1990s. There is an indication that the normalized atmospheric ^{3}H emissions have started to decrease since 2017, but it may just be a random variation. The normalized atmospheric ^{14}C discharges do not seem to have changed significantly during the 21st century for this data set. The median normalized discharge rates for this data set are 262.9 GBq/GWa for the atmospheric ^{14}C (2002-2023), 18214 GBq/GWa for the liquid ^{3}H (1975-2023) and 672.9 GBq/GWa for the atmospheric ^{3}H (2002-2023). This data is compiled in Table 6 along with the data for Swedish BWRs.

Performing a linear fit to the normalized discharges yields the plots shown in Figures 30a, 30b and 30c. As can be seen, there seems to be a negative trend for all of the discharges in the Swedish PWRs. These linear fits have p-values of 0.02491, 0.69927 and 0.13381, which means that only the linear fit of the normalized liquid ^{3}H release rate is significantly different from zero. It is also important to reiterate that this data only includes 3 reactors which is not enough to achieve statistical significance. Just as for the BWRs, this is also apparent from the large 95 % prediction intervals also shown in the figures. Any trend in the data could just as well be a statistical variation, although as stated in the previous chapter, efforts have been made to decrease the discharges from Swedish PWRs. Any statements about future discharges in general will however have to wait until the next sub-chapter about European NPPs.

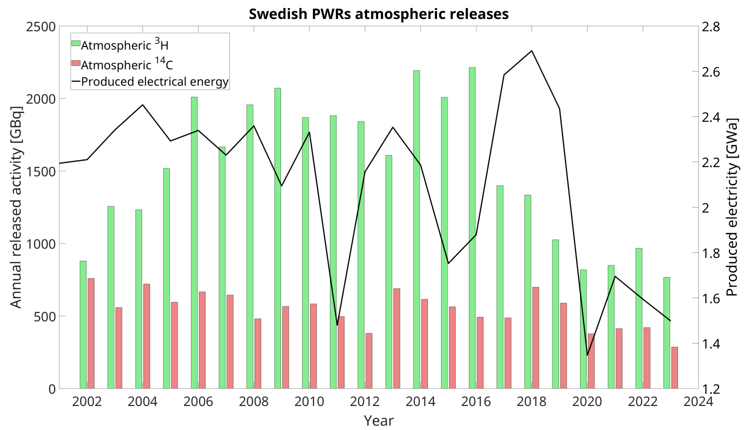


(a)

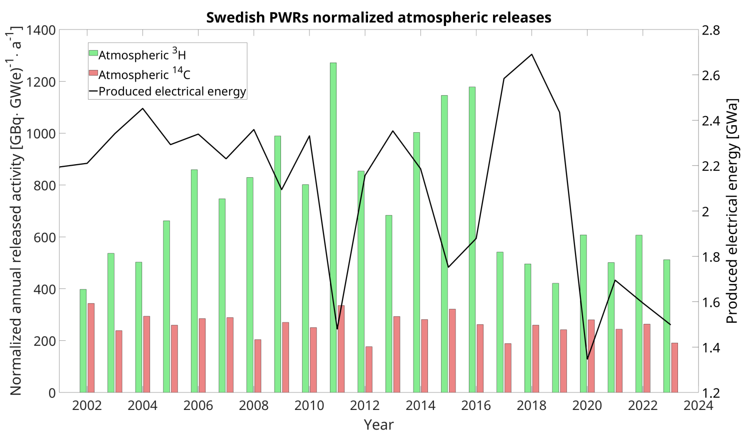


(b)

Figure 28: Liquid releases of ^{3}H from Swedish PWRs a) total, and b) normalized to energy production (GBq/GWa). Produced electricity shown as black line (GWa).

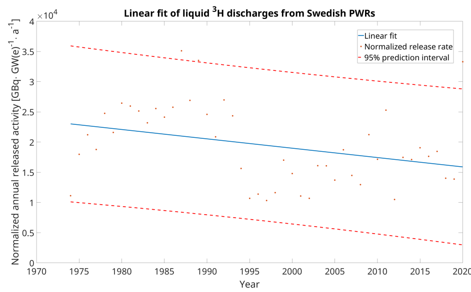


(a)

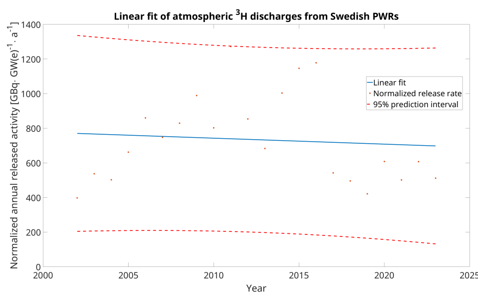


(b)

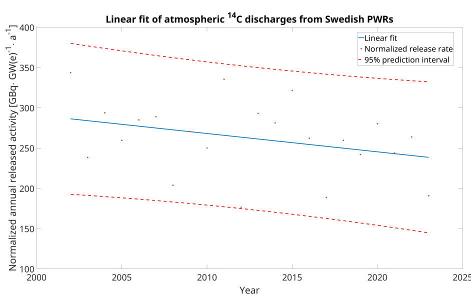
Figure 29: Atmospheric releases from Swedish PWRs a) total, and b) normalized to produced electricity (GBq/GWa). Produced electricity shown as black line (GWa).



(a)



(b)



(c)

Figure 30: Linear fits of normalized release rates with time for a) liquid ^{3}H, b) atmospheric ^{3}H and c) atmospheric ^{14}C discharges from Swedish PWRs. The normalized release rates are shown as red stars while the 95 % prediction interval is shown as red dashed lines.

Table 6: Median release rates liquid/atmospheric ^{3}H as well as atmospheric ^{14}C of Swedish BWRs and PWRs [GBq·GW(e)$^{-1}$·a^{-1}].

Release type	BWR	PWR
Liquid ^{3}H [GBq·GW(e)$^{-1}$·a^{-1}]	347.1 [190 - 639]	18214 [10329 - 35114]
Atmospheric ^{3}H [GBq·GW(e)$^{-1}$·a^{-1}]	181.6 [9 - 381]	672.9 [398 - 1272]
Atmospheric ^{14}C [GBq·GW(e)$^{-1}$·a^{-1}]	457.5 [363 - 600]	262.9 [177 - 344]

4.2 European NPPs

In this chapter, the emission data for the entire European nuclear fleet is presented. The total emissions reported by the NPPs, as well as the calculated normalized release rates are presented. As far as it is possible and appropriate, only measurement data (i.e. excluding any non-measured, estimated reported data) are included. The goal here is to study the trends over time of the atmospheric releases of ^{14}C, and the atmospheric and liquid releases of ^{3}H, from both PWRs and BWRs across all of Europe, including the previously studied Swedish NPPs. First, the releases of the atmospheric ^{14}C are presented, starting with the total European activity over time followed by the calculated European normalized release rate over time, provided with the number of reactors and data points that were included in the data composition. During the whole period 1995-2023, 129 reactors at a total of 57 different sites were included (out of a total of 61 sites found in RADD). Secondly, the atmospheric and liquid releases of ^{3}H are presented, starting with the total European activities over time, followed by the calculated European normalized release rates over time, again provided together with the number of reactors and data points that were included in the data composition.

As has been mentioned earlier, in order to study the release data for the European fleet of LWRs, data has been gathered from the RADD database. This database provides release data on a yearly basis. Most of the data points are provided for some collection of units, primarily grouped by reactor type (PWRs or BWRs) and for each site. For instance, for the Spanish site Almarez, which has two PWRs, only the aggregate yearly activity of both units is available. The corresponding energy production for this datapoint is thus the sum of the energy production from the same year for both Almarez 1 and Almarez 2. Such energy production values are composed using the PRIS database, which provides the needed data for all individual units. This methodology has been used for all of the following presented data for the European nuclear fleet. Please note that this methodology for composing data therefore requires that both RADD data and PRIS data are available for any one data point. Any data not meeting these criteria is excluded when calculating the normalized release rates.

Notice 1: During compilation of the RADD data it became clear that all the French reactors have an abnormally small spread in their reported normalized release rates for ^{14}C before 2017, indicating that these reported values are estimates using the energy production, not measurement data. To include only measurement data in calculating the mean normalized release rates across Europe, the French data before 2017 is excluded from the analysis. This exclusion is always done unless otherwise stated. The ^{3}H discharge data is included like usual.

Notice 2: When comparing the RADD data for the Swedish NPPs with the data acquired directly from the facilities themselves, it was noticed that the data for Ringhals 2 differed significantly, often being three times higher in RADD. The other NPPs showed at worst differences consistent with rounding. Issues with the data for Ringhals 2 in RADD was discussed already by Zizzeri et al [4]. For these reasons, Ringhals 2 is excluded from all analysis of the European nuclear fleet, without exception.

Notice 3: Some reactors are not present in RADD at all, such as Ringhals 3 and Ringhals 4.

European ^{14}C Emissions

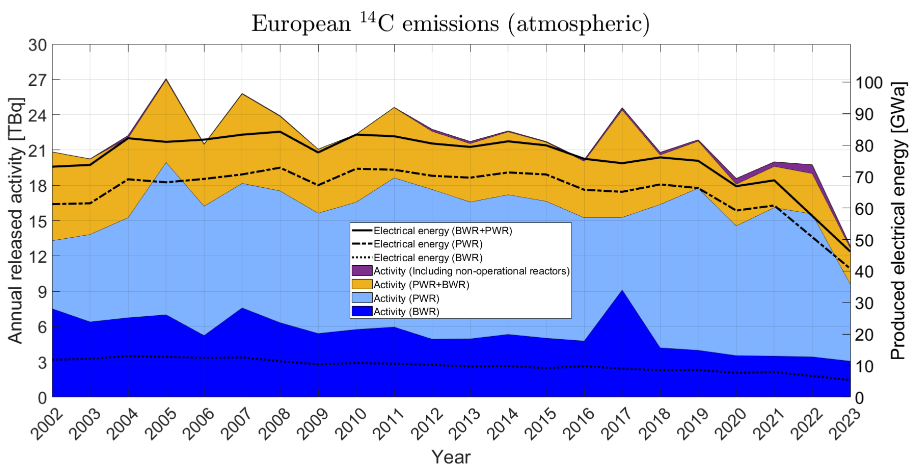


Figure 31: The total reported atmospheric released activity of ^{14}C from all European BWRs and PWRs present in RADD over time since 2002. The contribution from the PWRs and the BWRs are also shown. The reported emissions from non-operational reactors are also included[2]. Note that the French data is included for all years in the graph, including before 2017. For reference, the total amount of electrical energy produced by BWRs, PWRs and PWRs+BWRs, are also shown over time.

[2]The category of non-operational reactors include all units which, for that particular year,

When studying Figure 31, it can be noticed that the total emissions vary significantly from year to year. Looking at the total activity from the BWRs, it does decrease over time, but it seems to follow the decreased electrical energy production in a proportionate manner. Furthermore, the decreasing trend in the activity in the past few years from PWRs also seems to match the decreased energy production over the same time. This decreased energy production, in turn, mostly overlaps with the number of included PWRs over time (not shown in the figure). Thus Figure 31 while certainly informative about the trends in the absolute releases of atmospheric ^{14}C from the European nuclear fleet since 2002, does not contain enough information to, by itself, strongly indicate anything in particular about the qualitative trends in the normalized release rates. This is true for both of the LWRs of interest. This is remedied by the subsequently presented data.

Table 7: Median ^{14}C normalized release rates calculated using measurement data of the European nuclear facilities, composed using the RADD Database (2024) and their electricity production using PRIS (2024). The interquartile range is in square brackets. (NRs) and (DPs) are the number of Nuclear Reactors and the number Data Points included in the data composition, respectively.

Reactor Type	^{14}C normalized release rate [GBq·GW(e)$^{-1}$·a^{-1}] Median [Q_1-Q_3] (NRs) (DPs) 1995-2015	1995-2023
PWR	263.0 [167.7-366.7] (39) (370)	259.7 [167.8-363.4] (99) (629)
BWR	475.8 [375.6-643.5] (18) (164)	480.8 [369.8-625.2] (18) (206)

Now we turn our attention to the calculated normalized release rates of atmospheric ^{14}C. In Table 7 the calculated median normalized release rates along with the interquartile range for PWRs and BWRs are presented for the periods 1995-2015 (same as in Table 3) as well as 1995-2023 (the entire available dataset). As can be seen upon comparison to Table 3, the values for the period 1995-2015 are not equal. The most probable cause for this is likely that a different number of reactors have been included in the calculation (see notices 1-3 in the beginning of this subchapter). Another probable cause is that the data in either RADD or PRIS has been updated during the period 2017 (when [4] was published) to 2024 to remediate potential errors in previously reported values. Upon comparison of the two time intervals in Table 7, there seems to be no apparent change in normalized release rate after 2015, except for statistical variation.

lack PRIS data about its energy production. It is certainly possible that a thorough study of all these data points would reveal that the label "non-operational" is inappropriate/incorrect for some reactors for some years. Such a thorough investigation has not been performed, since the fraction of the total releases contributed by the non-operational category is so small.

Table 8: Median ^{14}C normalized release rates calculated and presented as in Table 7, but split into seven periods of four years each, starting from 1996-1999 and ending in 2020-2023. The top left table entry shows the units and the format of the data presented in the table, where Q_1 and Q_3 indicate lower and upper quartile, respectively, and (NRs)(DPs) indicate (Number of included Reactors)(Number of included Data Points).

Years	^{14}C normalized release rate [GBq·GW(e)$^{-1}$·a^{-1}] Median [Q_1-Q_3] (NRs) (DPs)	
	PWR	BWR
1996-1999	171.4 [85.7-301.2] (15)(54)	422.4 [311.8-593.4] (8)(24)
2000-2003	257.4 [157.5-316.4] (15)(56)	480.0 [402.5-656.8] (16)(32)
2004-2007	258.3 [196.8-363.2] (36)(77)	459.3 [371.9-753.3] (18)(40)
2008-2011	308.8 [188.6-389.3] (37)(90)	519.8 [423.8-626.8] (16)(37)
2012-2015	284.6 [212.9-430.9] (33)(80)	532.6 [421.4-618.9] (13)(25)
2016-2019	249.7 [183.5-338.2] (90)(133)	558.4 [315.8-599.3] (12)(24)
2020-2023	258.4 [154.2-378.2] (90)(126)	458.1 [233.2-604.7] (9)(18)

Moving on to Table 8, here the same calculations of ^{14}C normalized release rates as those shown in Table 7 are repeated, but this time after splitting the data set into four year ranges. Note that both the number of reactors and the number of data points included in the data composition vary significantly across time for both reactor types. Of course, the very sharp increase in the number of PWRs included in the 2016-2019 period is due to the exclusion of the French NPPs before 2017 (see notice 1 at the start of this chapter). The intended purpose of presenting the data in Table 8 is to try to show any trends in the normalized release rates, while simultaneously using large enough bins of time that the amount of included data per bin should have statistical value. For the BWRs, the statistical significance is questionable due the often very low number of available data. However, since this is a direct consequence of the low number of BWRs that exist in Europe, and since the normalized release rates presented here represent the entirety of (or at least the vast majority of) the European fleet of BWRs, these are proper population means and statistical significance is not necessarily relevant.

As the final part of this sub-chapter, the calculated ^{14}C normalized release rates for PWRs and BWRs are graphed for each year of the dataset in Figures 32 and 33, respectively. The thick black line represents the median normalized release rate, while the filled region (green and purple respectively) represents the interquartile range. Also plotted in the graphs are the number of reactors included in the data each year (blue line), as well as the number of data points (red line). As can be seen, the number of reactors is in many cases higher than the number of data points. As was discussed in the beginning of this chapter, this is due to the fact that several reactors may be grouped into a single data point representing the emissions for an entire NPP in the RADD emissions dataset.

To give a concrete and detailed example of how this works in practice, consider the Swedish NPP Oskarshamn, which has three units, Oskarshamn 1, 2 and 3, where Oskarshamn 1 and 2 shut down around 2016-2017. However, the RADD entry for Oskarshamn is just one number for each year in 1995-2023, all of which are listed as the aggregated emissions of all the units in Oskarshamn during that year. When performing the composition of the RADD data with the PRIS operational data, the single yearly data point from 1995-2016 in the RADD data includes information from three reactor units, namely O1, O2 and O3. Then, during 2017, only O1 and O3 are included. Then, from 2018-2023, only O3 is included.

The data for all European PWRs and BWRs presented above suggest only minor changes in the normalized discharge rates of ^{14}C over time. It is also apparent from the large interquartile ranges that there is a significant variation in discharge rates both from year to year, but also from reactor to reactor. This is in line with what was seen in the Swedish NPPs in chapter 4.1.

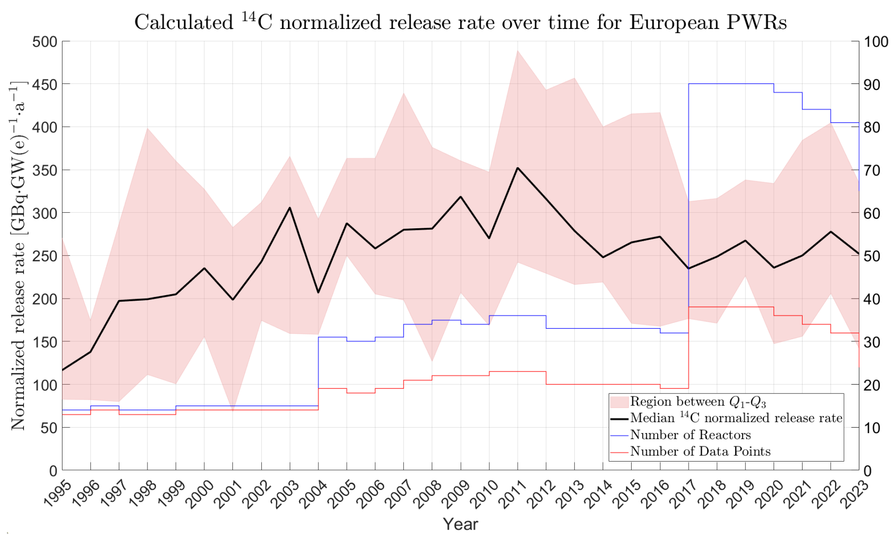


Figure 32: The European PWR calculated yearly ^{14}C normalized release rate. The region within the lower and upper medians Q1 and Q3, calculated yearly, is also shown. Furthermore, the yearly number of reactors, as well as the yearly number of included RADD ^{14}C release data points, are both also shown over time.

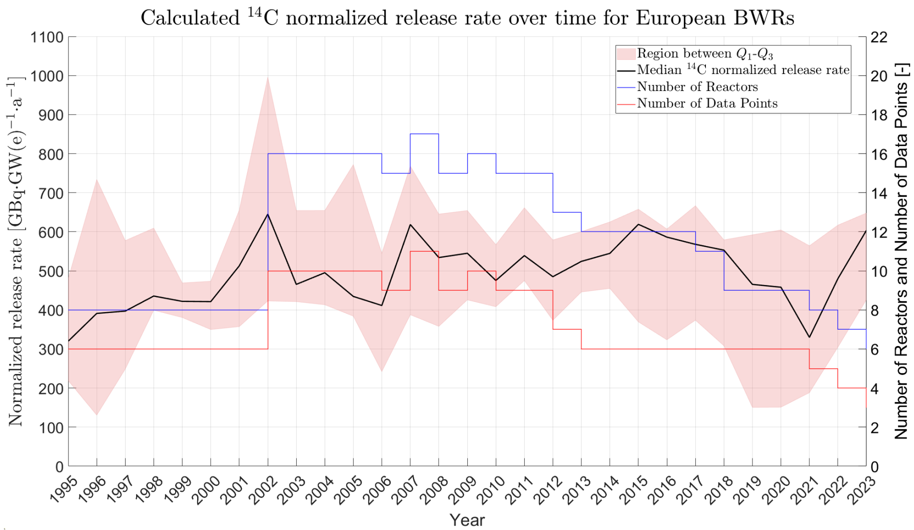


Figure 33: The European BWR calculated yearly ^{14}C normalized release rate. The region within the lower and upper medians Q1 and Q3, calculated yearly, is also shown. Furthermore, the yearly number of reactors, as well as the yearly number of included RADD ^{14}C release data points, are both also shown over time.

European ^{3}H Emissions

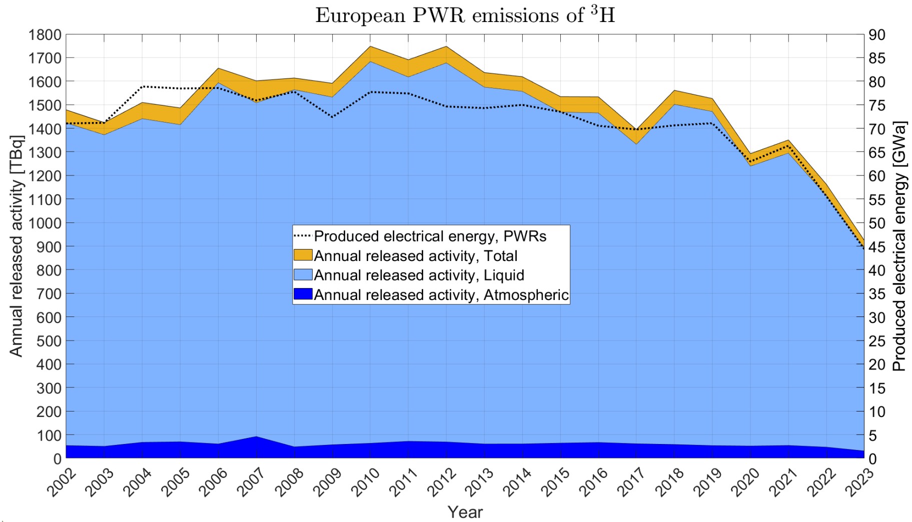


Figure 34: The European fleet of PWRs total emissions of ^{3}H, presented for both releases i.e. atmospheric and liquid. For reference, the corresponding total produced electrical energy from the PWRs is also shown. Note that the liquid effluents constitute the vast majority of all released ^{3}H from PWRs.

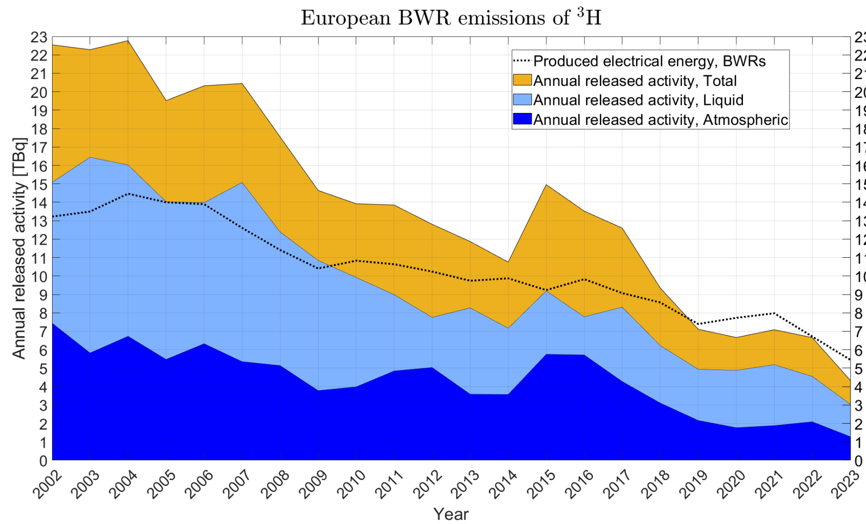


Figure 35: The European fleet of BWRs total emissions of ^{3}H, presented for both releases i.e. atmospheric and liquid. For reference, the corresponding total produced electrical energy from the BWRs is also shown.

In Figures 34 and 35, the total released activity of ^{3}H are presented, from PWRs and from BWRs, respectively. The total released activites into the atmosphere and as liquid discharges are shown as well. By comparing the two graphs, it is clearly the case that the liquid releases of ^{3}H from PWRs constitute the vast majority of all releases, at least for the data presented here. From the two graphs it is also apparent that the ^{3}H discharges from PWRs seem to follow the energy production somewhat well, while for the BWRs there is an apparent decrease in the discharges compared to energy production. This was also seen for the Swedish reactors in chapter 4.1. It should also be noted that the discrepancy between liquid and gaseous ^{3}H releases shown in Figure 34 is vastly larger than between the values of Table 2.

Table 9: Median ^{3}H normalized release rates in units of [GBq·GW(e)$^{-1}$·a^{-1}], calculated using measurement data of the European nuclear facilities, composed using the RADD Database (2024) and their electricity production using PRIS (2024). The interquartile range is in square brackets. The number of reactors included in the data composition is in the first parenthesis. The second parenthesis shows the total number of data points included in the data composition.

Reactor type	Liquid ^{3}H normalized release rate (1995-2023)	Atmospheric ^{3}H normalized release rate (1995-2023)
PWR	17715 [14266-24948] (107) (1030)	583 [316-983] (107) (1030)
BWR	765 [524-1264] (18) (235)	249 [129-547] (18) (235)

The normalized release rates seen in Table 9 show that the liquid and atmospheric normalized release rate of ^{3}H is in line with what was seen for the Swedish PWRs and BWRs, and which was seen for the Korean PWRs in Table 5. Since these values are lower than those presented in Table 2, there seems to have been a definitive reduction in ^{3}H releases compared to the values used in the TRS-421 report[2]. This is however not especially visible when looking at the values of Tables 10 and 11, which seem to be somewhat constant. This can also be seen in Figures 36-39. This means that the majority of the decrease in the ^{3}H normalized release rates occured somewhere during the time period 1980-1995, as the values in Table 2 are based on a report from 1980 [2]. This is also somewhat confirmed in the TRS-421 report on page 31 [2] were it was stated that the atmospheric ^{3}H decreased by a significant amount due to better maintenance and management of the reactor component system [2].

Table 10: Median PWR ^{3}H normalized release rates for liquid and for atmospheric releases, in units of [GBq·GW(e)$^{-1}$·a^{-1}], calculated using measurement data of the European nuclear facilities, composed using the RADD Database (2024) and their electricity production using PRIS (2024). The interquartile range is in square brackets. The number of reactors included in the data composition is in the first parenthesis. The second parenthesis shows the total number of data points included in the data composition. The top left entry of the table shows the data format of the table.

European PWRs ^{3}H normalized release rates Median [Q_1-Q_3] (NPPs) (DPs)	Liquid	Atmospheric
1996-1999	14845 [12346-23172] (28)(78)	568 [253-1014] (28)(78)
2000-2003	16775 [13930-23998] (86)(124)	461 [272-1053] (86)(124)
2004-2007	17072 [14188-24615] (102)(175)	469 [234-1171] (102)(175)
2008-2011	18749 [14854-25689] (102)(174)	621 [310-981] (102)(174)
2012-2015	18530 [14977-25669] (98)(164)	638 [438-859] (98)(164)
2016-2019	17936 [14579-25397] (97)(160)	579 [429-852] (97)(160)
2020-2023	18325 [14892-24721] (97)(135)	546 [401-850] (97)(135)

Table 11: Median BWR ^{3}H normalized release rates for liquid and for atmospheric releases, in units of [GBq·GW(e)$^{-1}$·a^{-1}], calculated using measurement data of the European nuclear facilities, composed using the RADD Database (2024) and their electricity production using PRIS (2024). The interquartile range is in square brackets. The number of reactors included in the data composition is in the first parenthesis. The second parenthesis shows the total number of data points included in the data composition. The top left entry of the table shows the data format of the table.

European BWRs ^{3}H normalized release rates Median [Q_1-Q_3] (NPPs) (DPs)	Liquid normalized release rate	Atmospheric normalized release rate
1996-1999	681 [448-1348] (10)(32)	183 [78-633] (10)(32)
2000-2003	662 [509-1287] (18)(40)	240 [91-568] (18)(40)
2004-2007	726 [522-1339] (18)(46)	242 [123-486] (18)(46)
2008-2011	1111 [650-1606] (17)(43)	284 [174-1069] (17)(43)
2012-2015	666 [560-1105] (13)(25)	319 [215-426] (13)(25)
2016-2019	854 [469-1195] (12)(23)	221 [171-646] (12)(23)
2020-2023	644 [383-921] (9)(18)	247 [209-321] (9)(18)

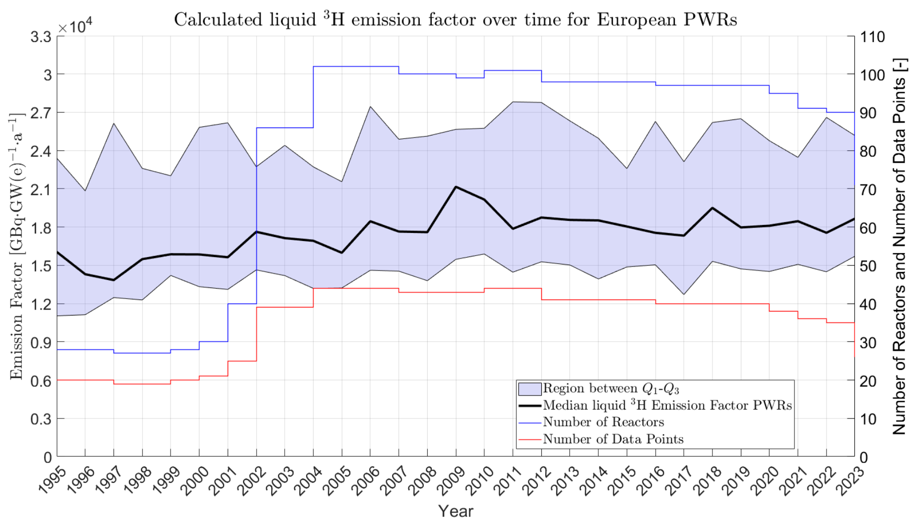


Figure 36: The yearly European PWR calculated normalized release rate of ^{3}H. The region within the lower and upper medians Q1 and Q3, calculated yearly, is also shown. Furthermore, the yearly number of reactors, as well as the yearly number of included RADD liquid ^{3}H release data points, are both also shown over time.

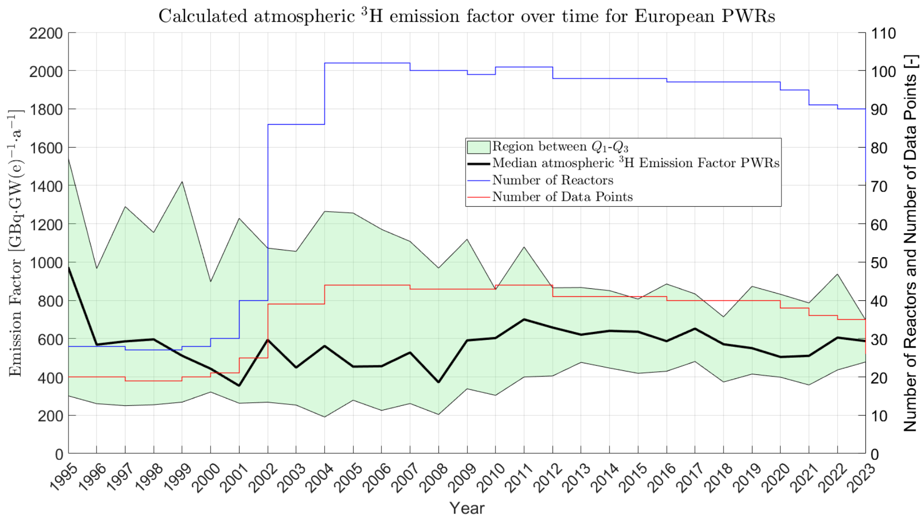


Figure 37: The yearly European PWR calculated normalized release rate of ^{3}H. The region within the lower and upper medians Q1 and Q3, calculated yearly, is also shown. Furthermore, the yearly number of reactors, as well as the yearly number of included RADD atmospheric ^{3}H release data points, are both also shown over time.

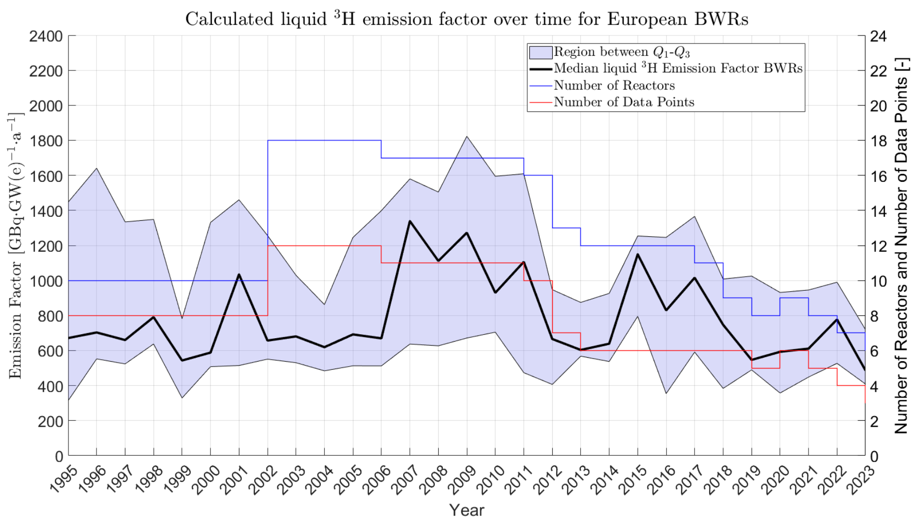


Figure 38: The yearly European BWR calculated normalized release rate of ^{3}H. The region within the lower and upper medians Q1 and Q3, calculated yearly, is also shown. Furthermore, the yearly number of reactors, as well as the yearly number of included RADD liquid ^{3}H release data points, are both also shown over time.

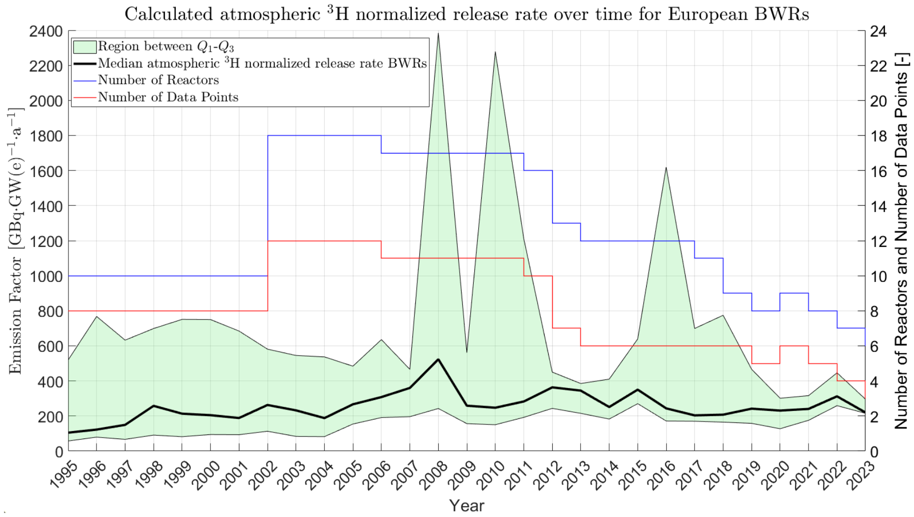


Figure 39: The yearly European BWR calculated normalized release rate of ^{3}H. The region within the lower and upper medians Q1 and Q3, calculated yearly, is also shown. Furthermore, the yearly number of reactors, as well as the yearly number of included RADD atmospheric ^{3}H release data points, are both also shown over time. The very sharp peaks in Q3 in the years 2008 and 2010 come from the Spanish facilitiy Santa Maria de Garona, which has very large reported emissions of ^{3}H for those two years. The sharp peak in Q3 in the year 2016 is due to two sites, the Spanish facility Confrentes and the Finnish facility Olkiluoto. All of these outliers seems to be correctly reported values.

5 Issues with the DIRATA Database

This chapter is intended to show and to discuss issues with IAEA's database on Discharges of Radionuclides to the Atmosphere and the Aquatic Environment, or DIRATA for short [22]. For this report it was originally planned to use the DIRATA dataset to obtain values for the released quantities of both atmospheric and liquid ^{14}C and ^{3}H. However, while working with the DIRATA dataset, several clearly erroneous data entries in the dataset were encountered[3]. This is the main reason (albeit not the only reason, it should be said) that the DIRATA database was never used for the production of this report.

The main type of problematic data entry that has been encountered in DIRATA, at least for the purposes of producing this report, is that of several different and incompatible reported values for a single release data point. A few data entries with unrealistically large values have been encountered as well, and they are also presented here. Another large category of erroneous reported data are all of the US reactors, where all the reported released activities are in Curie's, not in Giga-Bequerels, even though it clearly says it should be [GBq] on the DIRATA website. This last category of error is of course easily handled, unlike the first category where several numbers are reported for a single release data point.

Detailed description of how DIRATA has been used

In order to ensure full transparency as well as full reproducibility, a detailed description of how the DIRATA database has been used is now given. The DIRATA database has been downloaded in full from the DIRATA website, which can be found at `https://dirata.iaea.org/index.html`.

The way the entire database has been downloaded is as follows: when accessing the website, if one goes to the tab "Map Data", and then clicks on any nuclear reactor site, lets say the Forsmark NPP in Sweden, and then click the button "Download discharge totals for all sites", the complete DIRATA database that has been used here is downloaded as the file `all_sites_totals.csv`.

This file contains 28922 rows of data (as of November 2024), where each individual row constitutes a single data entry in the dataset. The rows are labelled by
`Year, Relase Type`[4]`, Site, Installation`[5]`, Nuclide Type, Activity,`

[3]It is fair to say that some of these erroneous entries are worse offenders than other. It is also the case that for some of these entries the value that was originally intended is easily deducible. In this chapter, all erroneous data entries that have been encountered are shown without any such considerations or distinctions, as they should all be fixed just the same.

[4]Note that this is misspelled in the original file. In the following, "Release Type" will be referred to instead

[5]For clarity, in the context of nuclear power plants, this denotes what units of the site is

where Activity is in [GBq]. This dataset is filtered for only those data entries where Nuclide Type equals one of {H-3, C-14}. In the resulting filtered dataset, the Release Type already equals only one of {Liquid, Atmospheric}, so there is no need for further filtering. In this filtered database, only the data entries corresponding to PWRs or BWRs are of interest here[6].

5.1 List of Erroneous Data Entries

Now all the erroneous data entries that have been encountered are listed, in no particular order. Since there are quite a few entries shown, only minimal descriptions are given when it's necessary. Note that it is certainly possible that this list is not exhaustive.

Multiple values provided for a single release data point

As was mentioned above already, most of the encountered erroneous data points are problematic simply because there are more than one number provided for a specific release data point, so the issue then is of course that it's unclear which is the actual value. Please note that there are no instructions given anywhere on the DIRATA website nor anywhere else on how to interpret several provided values for a single release data point[7]. On the DIRATA website, when looking at the "Discharges Data" tab, it seems to be the case that the website simply adds up all values it finds for any one single release data point[8]. However, in some cases this is clearly a mistake. The fact that this is a mistake can be realized from the following arguments: as is plainly obvious from looking at some of the numbers shown here, some of them are the same number, provided twice, once without rounding, and once after rounding. Other numbers are different by exactly a factor of 10, otherwise being exactly the same. Some of the data points have three values, where one of the numbers is the sum of the other two. Lastly, some of these multiple-value data points are different in a way where what the problem is is not clear at all.

Site	Installation	Year	Atmospheric ^{14}C [GBq]
Almaraz	All	2007	29.865
Almaraz	All	2007	29.9

included in the data point. In the vast majority of the cases, this is reported as "Aggregated record", which means all reactors of the site, and this is renamed here to "All"

[6]Examples of data entries that are irrelevant to this report are those from reprocessing plants such as Sellafield and Cap de la Hague, since reprocessing plants are not studied here.

[7]At least as far as the authors of this report is aware. If such instructions exist, their location on the internet is not easily found

[8]However, there are a few exceptions to this as well.

Site	Installation	Year	Atmospheric ^{3}H [GBq]
Almaraz	All	2007	4357.5
Almaraz	All	2007	4360

Site	Installation	Year	Liquid ^{3}H [GBq]
Almaraz	All	2007	38000
Almaraz	All	2007	38047

Site	Installation	Year	Liquid ^{14}C [GBq]
Belleville	All	2007	21.7
Belleville	All	2007	32.2

Site	Installation	Year	Atmospheric ^{3}H [GBq]
Belleville	All	2007	196
Belleville	All	2007	1960

Site	Installation	Year	Liquid ^{14}C [GBq]
Cattenom	All	2008	27.5
Cattenom	All	2008	62.9

Site	Installation	Year	Atmospheric ^{3}H [GBq]
Cattenom	All	2008	289
Cattenom	All	2008	2890

Site	Installation	Year	Liquid ^{14}C [GBq]
Chinon	All	2007	34.5
Chinon	All	2007	41.8

Site	Installation	Year	Liquid ^{14}C [GBq]
Chinon	All	2008	22
Chinon	All	2008	43.9

Site	Installation	Year	Atmospheric ^{3}H [GBq]
Chooz	All	2007	566
Chooz	All	2007	569

Site	Installation	Year	Atmospheric ^{3}H [GBq]
Chooz	All	2008	542
Chooz	All	2008	545

Site	Installation	Year	Atmospheric ^{14}C [GBq]
Confrentes	All	2007	291.0734
Confrentes	All	2007	595

Site	Installation	Year	Atmospheric ^{3}H [GBq]
Confrentes	All	2007	291
Confrentes	All	2007	594.7

Site	Installation	Year	Liquid ^{3}H [GBq]
Confrentes	All	2007	534
Confrentes	All	2007	534.315

Site	Installation	Year	Atmospheric ^{14}C [GBq]
Gundremmingen	All	2020	0.055
Gundremmingen	All	2020	150
Gundremmingen	All	2020	150.055

Site	Installation	Year	Atmospheric ^{3}H [GBq]
Gundremmingen	All	2020	1.88
Gundremmingen	All	2020	85
Gundremmingen	All	2020	86.88

Site	Installation	Year	Atmospheric ^{14}C [GBq]
Gundremmingen	All	2021	0.011
Gundremmingen	All	2021	260
Gundremmingen	All	2021	260.011

Site	Installation	Year	Atmospheric ^{3}H [GBq]
Gundremmingen	All	2021	0.947
Gundremmingen	All	2021	149
Gundremmingen	All	2021	149.947

Site	Installation	Year	Atmospheric ^{14}C [GBq]
Nogent	All	2007	192
Nogent	All	2007	492

Site	Installation	Year	Atmospheric ^{3}H [GBq]
Paluel	All	2006	5110
Paluel	All	2006	109000

Site	Installation	Year	Atmospheric ^{14}C [GBq]
Santa Maria de Garona	All	2007	192.76
Santa Maria de Garona	All	2007	199

Site	Installation	Year	Atmospheric ^{3}H [GBq]
Santa Maria de Garona	All	2007	1150
Santa Maria de Garona	All	2007	1151.9

Site	Installation	Year	Liquid ^{3}H [GBq]
Santa Maria de Garona	All	2007	712.5851
Santa Maria de Garona	All	2007	713

Site	Installation	Year	Atmospheric ^{14}C [GBq]
St Alban	All	2008	248
St Alban	All	2008	284

Site	Installation	Year	Liquid ^{14}C [GBq]
St Laurent	St Laurent B1-B2	2008	11.4
St Laurent	St Laurent B1-B2	2008	24.2

Site	Installation	Year	Atmospheric ^{14}C [GBq]
Tricastin	All	2008	269
Tricastin	All	2008	569

Site	Installation	Year	Atmospheric ^{14}C [GBq]
Trillo	All	2007	44.477324
Trillo	All	2007	44.5

Site	Installation	Year	Atmospheric ^{3}H [GBq]
Trillo	All	2007	745.63542
Trillo	All	2007	746

Site	Installation	Year	Liquid ^{3}H [GBq]
Trillo	All	2007	21700
Trillo	All	2007	21717.389

Extreme outliers among the reported data

This is a short list of a few clearly erroneous reported values. Please note that these reported values are not necessarily only for BWRs or PWRs, unlike the previous chapter. The two final tables in this chapter have values which are probably reported in Bequerels, not Giga-Bequerels, as it they should.

Site	Installation	Year	Atmospheric ^{14}C [GBq]
Penly	All	2006	462000

Site	Installation	Year	Liquid ^{3}H [GBq]
Bohunice	?	2016	8.98 Trillion
Bohunice	?	2017	10.13 Trillion

Site	Installation	Year	Liquid ^{3}H [GBq]
Dounreay	?	1985	0.890 Trillion
Dounreay	?	1986	0.992 Trillion

Activities reported using wrong units for US BWRs

It was noticed during data-processing of the DIRATA dataset that all American based BWRs have reported their emissions in Curie's, not in Giga-Bequerels. This should of course be fixed as soon as possible.

6 Conclusions

It was found in the previous chapter that the median (atmopsheric) ^{14}C normalized release rates were approximately 260 GBq·GW(e)$^{-1}$·a^{-1} for the Euopean PWRs, 263 GBq·GW(e)$^{-1}$·a^{-1} for the Swedish PWRs, 481 GBq·GW(e)$^{-1}$·a^{-1} for the European BWRs and 457.5 for the Swedish BWRs. Both the Swedish and the European reactors exhibited ^{14}C discharges that were similar to those presented in previous publications, although some of the Swedish reactors were on the higher side.

As for the ^{3}H discharges, for the evaluated data set the atmospheric and liquid ^{3}H normalized release rates were found to be 583 GBq·GW(e)$^{-1}$·a^{-1} and 17715 GBq·GW(e)$^{-1}$·a^{-1} respectively for the European PWRs, 673 GBq·GW(e)$^{-1}$·a^{-1} and 18214 GBq·GW(e)$^{-1}$·a^{-1} respectively for the Swedish PWRs. For the BWRs the corresponding normalized release rates were 249 GBq·GW(e)$^{-1}$·a^{-1} and 765 GBq·GW(e)$^{-1}$·a^{-1} respectively for the European BWRs and 181.6 and 347.1 for the Swedish BWRs. It could be seen that the emissions from the PWRs followed the energy production quite well over the entire investigated time period and were quite similar to the values calculated for the Korean PWRs, yet the calculated values were much lower than those presented in [2]. As was also stated in [2] this was likely due to a reduction in discharges during the period 1980-1995 caused by better maintenance and management of the reactor component system [2]. For the BWRs a drastic decrease in normalized release rate for ^{3}H discharges could be seen compared to the values in [2]. This was also visible in Figure 35, but could not be seen in Figures 38 and 39. This could be caused by many BWRs being included to make Figure 35 not having operational data available in PRIS, yet still having discharge data available in RADD. This means the apparent decreased emissions compared to energy production visible in Figure 35 is not true to reality. Most likely the decreased ^{3}H normalized release rates that were apparent by comparing to Table 2 occured before 1995. The limited number of European BWRs makes it difficult however to draw any definitive conclusions.

What can be said about both BWRs and PWRs is that there exists a large variation in normalized release rate between different reactors, and even for the same reactor on a yearly basis. This was apparent from the large spread in the interquartile range. While it may be possible to calculate a "general" normalized release rate which, if used for a large number of reactors over a significant timespan, would give a somewhat accurate estimation of the emissions, it is likely not accurate on an individual basis. We are therefore of the opinion that normalized release rates, if used, should be calculated for each reactor.

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