OVERVIEW OF THE ENVIRONMENTAL MONITORING PROGRAM FOR TRITIUM IN SPANISH RIVER WATERS

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ABSTRACT. An environmental monitoring program for tritium in Spanish river waters has been in operation since the early 1980s. Tritium activity was determined monthly at ~100 sampling points distributed throughout the main Spanish river basins (Douro, Ebro, Guadalquivir, Guadiana, and Tagus) and other small river basins. Special attention was given to river basins where nuclear power plants (NPP) are present such as Tagus (Trillo, José Cabrera, and Almaraz I and II NPPs), Ebro (Sta. Mª de Garoña, and Ascó I and II NPPs), and Júcar (Cofrentes NPP). Tritium activity was determined by mixing the water sample with an appropriate aqueous-accepting scintillation cocktail without electrolytic enrichment. Thus, minimal sample pretreatment was required, and sample preparation was rapid. For tritium determination, we used 10 mL of water sample mixed with 12 mL of the scintillation cocktail Ultima Gold™ LLT in polyethylene vials. Samples were stored in a liquid scintillation spectrometer for 1 day to decrease the chemiluminescence spectrum. The instruments used for routine measurements of tritium were a Tri-Carb 2560 TR/XL and 2200 CA (Packard, USA). The minimum detectable activity (MDA) achieved for tritium determination was ~2.5 Bq/L for a count time of 360 min. This MDA was sufficient for environmental monitoring.

The objective of this work is to give an overview of the nationwide variation of tritium levels from the early 1980s to the present throughout the main Spanish river basins and other small river basins. The results of this evaluation may also be used to improve the environmental monitoring program, because environmental monitoring occurs over long periods of time and useful information can be obtained from such long processes.

INTRODUCTION

Spain had 9 operating nuclear plants with 7896 MWe total generating capacity at the end of 2003 (Table 1). In the late 1960s, Spain started construction of the first-generation nuclear power plants (NPP) José Cabrera (Tagus River), Santa María de Garoña (Ebro River), and Vandellós I (Mediterranean Sea). These plants enabled us to gain first-hand experience in order to establish a nuclear program to cover the growing electricity demand. In the early 1970s, construction of the second-generation NPPs began at Almaraz I and II (Tagus River), Ascó I and II (Ebro River), and Cofrentes (Júcar River). In the early 1980s, construction started on the NPPs Vandellós II (Mediterranean Sea) and Trillo I (Tagus River). The Vandellós I NPP has been out of service since 1990, and in January 1998 the Ministry of Industry and Energy authorized its dismantling.

Table 1 Nuclear power plants in Spain (UNESA 2003).

Name	Rating (MWe)	Type[a]	Origin of technology	Initial connection
José Cabrera (Zorita)	160	PWR	Westinghouse (USA)	1968
Santa María de Garoña	466	BWR	General Electric (USA)	1971
Vandellós I	Shut down	GGR	HIFRENSA (Spain, France)	1972
Almaraz I	980	PWR	Westinghouse (USA)	1981
Almaraz II	982.6	PWR	Westinghouse (USA)	1983
Ascó I	1032.5	PWR	Westinghouse (USA)	1983
Ascó II	1027.2	PWR	Westinghouse (USA)	1985
Cofrentes	1095	BWR	General Electric (USA)	1984
Vandellós II	1087.1	PWR	Westinghouse (USA)	1987
Trillo	1066	PWR	KWU/Siemens (Germany)	1988

[a]PWR = pressurized water reactor; BWR = boiling water reactor; GGR = gas-graphite reactor.

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In 1972, ENUSA (formerly Empresa Nacional del Uranio, SA, now ENUSA Industrias Avanzadas, SA, a state-owned company) was set up to take charge of all nuclear fuel cycle front-end activities. Its main duties were procurement and exploitation of radioactive mineral mines, uranium enrichment, and the manufacture of nuclear fuel elements. ENUSA mining activities stopped at the end of 2000, and only the Juzbado plant (located next to the Tormes River, Duero Basin) produces fuel elements for most pressurized water reactors (PWRs) and boiling water reactors (BWRs) in Spain (Table 2).

Table 2 Fuel cycle industries in Spain.

Installation	Owner/ Company	Location	River	Year of operation	Fuel cycle operation
Juzbado Plant	ENUSA	Juzbado (Salamanca)	Tormes (Douro)	1985	Fuel manufacturing plant
El Cabril	ENRESA	Hornachuelos (Cordoba)	Bembezar (Guadalquivir)	1992	Medium and low-level radioactive waste storage
Quercus Plant	ENUSA	Saelices el Chico (Salamanca)	Agueda (Douro)	Stopped (2000)	Milling plant
Elefante Plant	ENUSA	Saelices el Chico (Salamanca)	Agueda (Douro)	Stopped (1993)	Milling plant
Lobo-G Plant	ENUSA	La Haba (Badajoz)	Ortigas (Guadiana)	Stopped (1995)	Milling plant
FUA	ENRESA	Andujar (Jaén)	Guadalquivir	Stopped (1981)	Milling plant

In 1984, ENRESA (Empresa Nacional de Residuos Radiactivos, S.A.) was established. This state-owned company is responsible for managing radioactive waste and dismantling nuclear installations in Spain. Its duties include radioactive waste treatment, managing the different operations related to the decommissioning of nuclear installations, establishing systems for collecting and transporting radioactive wastes, and the final and safe disposal of waste derived from mining and milling processes. ENRESA has a medium- and a low-level radioactive waste storage installation located in El Cabril, Córdoba (on the Bembezar River, Guadalquivir Basin) (Table 2).

The operation of these nuclear facilities causes the release of radioactive substances into the environment. All facilities discharge radioactive contaminants into Spanish river basins, and for this reason, an environmental monitoring program for tritium in Spanish rivers was set up by the Centro de Estudios y Experimentación de Obras Públicas (CEDEX) in the early 1980s (Díaz-Teijeiro et al. 1992). Samples are collected from 90 sampling stations located at the 9 major Spanish river basins (Northern coast, Douro, Tagus, Guadiana, Guadalquivir, Segura, Júcar, Ebro, and Internal Basins of Catalonia). Sampling stations were selected for different objectives:

- The establishment of a tritium background level in the main Spanish river basins;
- The study of the nationwide variation of tritium levels in the main Spanish river basins;
- The study of the radioactive impact of tritium released from nuclear power plants (NPPs).

Also, under the terms of Articles 35 and 36 of the Euratom Treaty, member states must establish the facilities necessary to carry out continuous monitoring of radioactivity levels in the air, water, and soil and provide the Commission with information on environmental radioactivity levels.

The European Union's Council Directive 98/83/CE (European Union 1998) on the quality of water intended for human consumption deals with the microbiological, chemical, and radioactive aspects of the water. As for radioactivity in water, the technical annexes provide detailed information about analyses to be taken, including tritium as an indicative parameter for radioactive releases (European Union 1998). With this in mind, we present an overview of the environmental program for the surveillance of tritium in Spanish river waters.

EXPERIMENTAL SECTION

Study Area

Hydrographically, Spain is divided into 10 drainage basins: Northern coast, Douro, Tagus, Guadiana, Guadalquivir, South, Segura, Júcar, Ebro, and Internal Basins of Catalonia (Figure 1). There are important regional differences between the North (more hydraulic resources) and the South (less resources), which cause a crucial problem with water in Spain.

Figure 1 Spanish administrative watershed distribution

Although the main river of the Northern coast Basin is the Miño River (274 km), which partly forms the northwestern border between Spain and Portugal, this basin consists of many smaller rivers such as the Navia, Nalón, Pas, Asón, Nervión, and Bidasoa, all of them flowing into the Cantabric Sea. This special feature has, for managerial reasons, forced the "basin" to be divided into 4 "sub-basins," named North I, North II, North III, and Galicia Coast.

The largest river of the Douro Basin is the Douro River (744 km long), which begins in the Sierra de Urbión in central Spain. The Douro generally flows westward across Spain and northern Portugal and into the Atlantic Ocean.

The main river of the Tagus Basin is the Tagus River, with its tributaries, the Jarama, the Alberche, the Tiétar, and the Alagón rivers. The Tagus is 906 km long and begins in east-central Spain, in the Sierra de Albarracin at an altitude of 1590 m, and flows through Portugal into the Atlantic Ocean near Lisbon. One of the most important issues for the Tagus Basin is the transfer of water via the Tajo-Segura Channel from the upper Tagus to the Segura Basin in southern Spain, where it can be used for irrigation.

The Guadiana River (824 km long) generally flows westward through south-central Spain and southeastern Portugal to the Gulf of Cádiz (Atlantic Ocean). Its flow is relatively meager because its basin drains the low-rainfall areas of the Toledo Mountains, the Sierra Morena, and the plains of La Mancha.

The Guadalquivir Basin includes the Guadalquivir River (500 km long), with its tributaries the Genil and the Bembezar rivers. The Guadalquivir begins in the Sierra de Cazorla (SE Spain) and flows generally southwest past Cordoba and Seville into the Gulf of Cádiz, near Sanlúcar de Barrameda.

The South Basin is comprised of 3 main rivers, the Guadalhorce, the Guadalfeo, and the Almanzora, with other minor rivers.

The Segura Basin's main river is the Segura (341 km long). It begins in the Segura Mountains, Jaén Province, and flows east through the driest region of the Iberian Peninsula to enter the Mediterranean Sea south of Alicante.

The Júcar Basin has 2 large rivers, the Júcar and the Turia rivers. The Júcar River begins in the Universales Mountains (Cuenca). It flows in a southerly, then easterly direction, through the Cuenca, Albacete, and Valencia provinces and into the Gulf of Valencia.

The Ebro Basin's largest river is the Ebro (928 km long), with its major tributaries, the Aragón, Gállego, Cinca, and Segre rivers on the right bank, and the Jalón and Guadalope rivers on the left. The Ebro begins in the Cantabrian Mountains of northern Spain and drains a triangular basin between the Pyrenees and Iberian mountains before flowing through a wide delta into the Mediterranean. This delta, covering 320 km^2, is one of the most important wetlands in Europe.

The Eastern Pyrenees, or Internal Basins of Catalonia, has 2 main rivers: the Ter and Llobregat.

Sampling

Water samples were collected by the River Basin Authorities (Confederaciones Hidrográficas) and taken to the CEDEX laboratory for analysis. The sampling locations are shown in Figure 2 and Table 3.

Sample codes are given as XYYZZ, where *X* corresponds to *A* (nuclear-free) or *C* (nuclearized); *YY* corresponds to the river basin (NE for north rivers, DU for Douro, TA for Tagus, GU for Guadiana, GR for Guadalquivir, SG for Segura, JU for Júcar, EB for Ebro, and PO for Pyrenees rivers); and *ZZ* is the sampling location number. The frequency of the sampling was determined with the aim of obtaining the best representative information about a specific radiological situation; this was done while taking into account the technical and material possibilities at our disposal. The southern coastal basins are not included in this study.

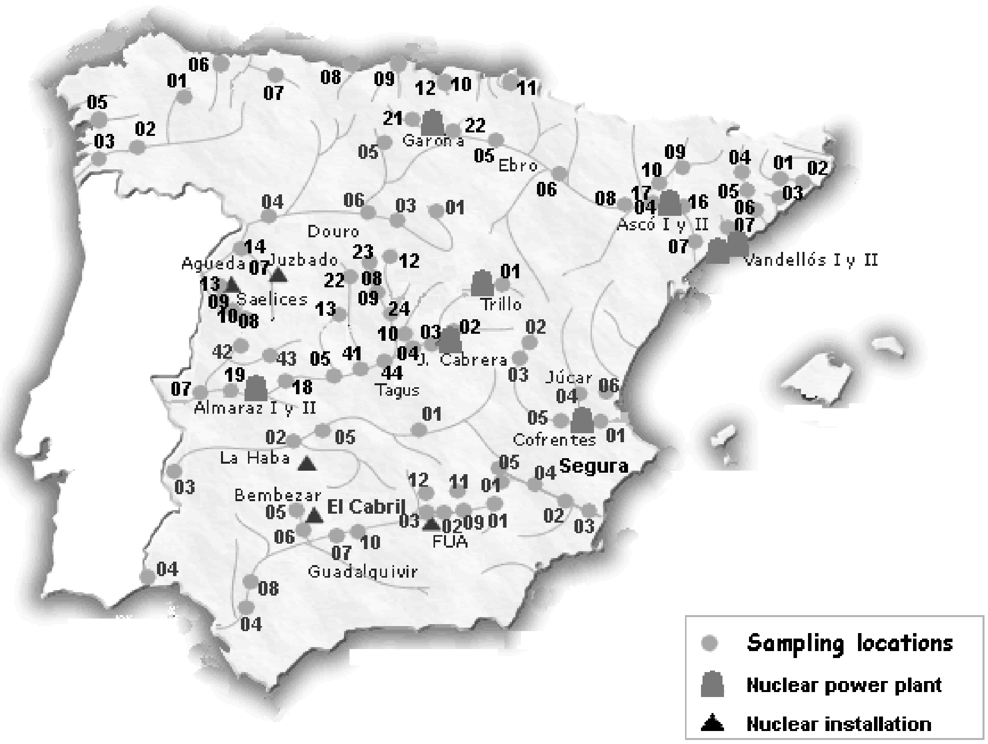

Figure 2 Sampling locations of the tritium environmental radiological network in Spain

ANALYTICAL METHOD

Water samples, not distilled, were mixed with a scintillation solution of Ultima Gold™ LLT (Packard, USA) with a sample/scintillator ratio of 10/12 in polyethylene vials (Packard). Very old water ("dead water") free of tritium was used as a background. At the same time, backgrounds and a tritium standard were prepared. Samples, backgrounds, and tritium standard were counted for at least 360 min in an ultra-low level background LS system developed by Packard Instrument Company. Our laboratory has 3 Packard scintillation spectrometers: the 2770TR/SL, the 2560 TR/XL, and the 2200CA. Typically, the minimum detectable activity was 1.5, 1.9, and 2.3 Bq/L, respectively; backgrounds ranged from 0.8–1.1, 1.5–1.8, and 2.2–2.6 cpm, respectively; and counting efficiencies were approximately 20% for the 3 LS counters (Pujol and Fé Díaz 2002).

We participated in an intersample comparison of the Tagus River water samples. The comparison included the measurements reported by 12 laboratories from Portugal and Spain. The average activity of all the measurements, excluding the results of 2 outliers, was 94.5 ± 6.2 Bq/L. Our laboratory obtained 88.1 ± 5.2 Bq/L ($k = 2$), which is within the limits of uncertainty (Figure 3).

RESULTS AND DISCUSSION

The main objective of this work is to provide an overview of tritium levels and variation of the tritium levels in the Spanish river basins. It should be noted that the information from this study will be used to improve the environmental monitoring program and/or to support environmental research programs.

Table 3 Sampling information of the Spanish environmental radiological network.

Basin	Sample code	Location	River	Fr[a]	Latitude (N)	Longitude (W)
North rivers	ANE01	Lugo	Miño	Q	43°00′07″	7°33′40″
	ANE02	Orense	Miño	Q	42°20′43″	7°52′02″
	ANE03	Tuy	Miño	Q	42°03′10″	8°33′55″
	ANE05	Puentevea	Ulla	M	42°45′38″	4°52′00″
	ANE06	Arbón Dam	Navia	M	43°28′38″	6°43′49″
	ANE07	Pravia	Nalón	M	43°29′33″	6°06′12″
	ANE08	Puente Viesgo	Pas	M	43°16′50″	3°50′37″
	ANE09	Ampuero	Asón	M	43°20′43″	3°25′04″
	ANE10[b]	Echevarria	Nervión	M	43°14′46″	2°53′00″
	ANE11	Lesaca	Bidasoa	M	43°17′43″	1°43′41″
	ANE12	La Peña	Nervión	M	43°14′39″	2°55′00″
Douro	ADU01	Garray	Douro	Q	41°47′43″	2°27′26″
	ADU03	Quintanilla	Douro	Q	41°37′43″	4°21′43″
	ADU04	Villalcampo	Douro	Q	41°29′40″	6°05′00″
	ADU05	Alar del Rey	Pisuerga	Q	42°39′19″	4°18′56″
	ADU06	Valladolid	Pisuerga	M	41°38′57″	4°43′55″
	ADU07	El Marín	Tormes	M	40°57′37″	5°42′17″
	ADU13	Vega de Terrón	Agueda	M	41°01′50″	6°55′45″
	ADU14	Ledesma	Tormes	M	41°05′30″	5°59′40″
	CDU08	Enusa upstream	Agueda	M	40°37′45″	6°36′30″
	CDU09	Enusa downstream	Agueda	M	40°37′25″	6°37′50″
	CDU10	Enusa release	Agueda	M	40°37′45″	6°36′30″
	CDU11	Enusa border	Agueda	M	40°37′25″	6°38′50″
	CDU12	Juzbado downstream	Tormes	M	41°04′40″	5°56′49″
Tagus	ATA04	Aranjuez	Tagus	Q	40°02′38″	3°48′31″
	ATA05	Talavera	Tagus	Q	39°57′38″	4°49′54″
	ATA07	Alcántara	Tagus	M	39°43′19″	5°53′28″
	ATA08	El Pardo	Manzanares	Q	40°31′23″	3°46′45″
	ATA09	La China	Manzanares	Q	40°21′35″	3°40′42″
	ATA10	Puente Largo	Jarama	M	40°05′08″	3°36'26″
	ATA12	Valdepeñas	Jarama	Q	40°51′53″	3°53′52″
	ATA13	Picadas	Alberche	Q	40°21′45″	4°18′30″
	ATA22	Villalba	Guadarrama	Q	40°37′44″	4°00′17″
	ATA23	Santillana	Manzanares	Q	40°42′25″	3°49′00″
	ATA24	Mejorada	Jarama	M	40°23′51″	3°51′53″
	ATA41	Castrejón Dam	Tagus	Q	39°49′48″	4°17′10″
	ATA42	Gabriel Dam	Alagón	Q	39°58′49″	6°31′27″
	ATA43	La Bazagona	Tiétar	Q	39°55′55″	5°54′13″
	ATA44	Toledo	Tagus	M	39°52′18″	3°59′18″
	CTA01	Trillo upstream	Tagus	M	40°41′36″	2°35′02″
	CTA02	Zorita upstream	Tagus	M	40°21′00″	2°49′00″
	CTA03	Zorita downstream	Tagus	M	40°20′58″	2°53′49″
	CTA18	Valdecañas	Tagus	M	39°48′45″	5°28′46″
	CTA19	Torrejón Dam	Tagus	M	39°50′02″	6°01′16″

Table 3 Sampling information of the Spanish environmental radiological network. *(Continued)*

Basin	Sample code	Location	River	Fr[a]	Latitude (N)	Longitude (W)
Guadiana	AGU01	Balbuena	Guadiana	Q	38°58′30″	4°06′54″
	AGU02	Orellana	Guadiana	Q	38°58′45″	5°32′39″
	AGU03	Puente Palmas	Guadiana	Q	38°51′00″	6°58′39″
	AGU04	Sanlúcar	Guadiana	Q	37°28′10″	7°27′54″
	AGU05	Valdecaballeros	Guadalupejo	Q	39°18′25″	5°09′47″
Guadalquivir	AGR01	Puente Cerrada	Guadalquivir	Q	37°56′44″	3°13′08″
	AGR04	Sevilla	Guadalquivir	M	37°23′31″	6°00′18″
	AGR05	El Cabril upstream	Bembezar	Q	38°04′53″	5°25′05″
	AGR06	Hornachuelos	Bembezar	M	38°08′38″	5°14′10″
	AGR07	Posadas	Guadalquivir	Q	37°47′50″	5°06′20″
	AGR08	Alcalá	Guadalquivir	Q	37°31′05″	5°58′30″
	AGR09	Mengíbar	Guadalquivir	M	37°59′13″	3°47′15″
	AGR10	El Carpio	Guadalquivir	Q	37°58′30″	4°29′30″
	AGR11	Linares	Guadalimar	Q	38°10′08″	3°37′56″
	AGR12	Encinarejo	Jándula	Q	38°09′55″	3°59′20″
	CGR02	Andújar upstream	Guadalquivir	M	38°00′53″	4°03′00″
	CGR03	Andújar downstream	Guadalquivir	M	38°01′38″	4°04′25″
Segura	ASG01	Los gallegos	Segura	Q	38°24′58″	2°00′40″
	ASG02	Archena	Segura	Q	38°07′45″	1°18′02″
	ASG03[c]	Beniel	Segura	M	38°02′58″	1°00′17″
	ASG04	Camarillas	Mundo	Q	38°20′30″	1°39′10″
	ASG05	Tajo-Segura Channel	Tajo-Segura	M	38°33′40″	1°55′16″
Júcar	AJU02	Venta de Juan Romero	Júcar	Q	40°13′50″	1°50′30″
	AJU03	Alarcón Dam	Júcar	Q	39°33′55″	2°06′50″
	AJU06	Picassent	Canal Júcar-Turia	M	39°33′55″	2°06′50″
	CJU01	Cofrentes downstream	Júcar	M	39°33′55″	1°03′30″
	CJU04	Villatoya	Cabriel	Q	39°20′30″	1°20′30″
	CJU05	Alcalá júcar	Júcar	Q	39°11′50″	1°25′15″
Ebro	AEB05	Mendavia	Ebro	Q	42°25′00″	2°12′10″
	AEB06	Zaragoza	Ebro	M	41°39′30″	0°52′50″
	AEB07	Cherta	Ebro	M	40°55′30″	–0°29′26″
	AEB08	Vástago	Ebro	Q	41°19′30″	0°00′43″
	AEB09	Balaguer	Segre	M	41°27′13″	–0°48′30″
	AEB10	Serós	Segre	M	41°27′13″	–0°25′11″
	CEB04	Ribarroja	Ebro	M	41°15′00″	–0°28′38″
	CEB16	Ascó downstream	Ebro	B	41°10′04″	0°36′47″
	CEB17	Ascó upstream	Ebro	B	41°11′57″	0°34′34″
	CEB21	Garoña upstream	Ebro	M	42°47′11″	3°15′39″
	CEB22	Garoña downstream	Ebro	B	42°47′01″	3°04′55″

Table 3 Sampling information of the Spanish environmental radiological network. *(Continued)*

Basin	Sample code	Location	River	Fr[a]	Latitude (N)	Longitude (W)
Pyrenees rivers	APO01	Pastoral	Ter	Q	41°59′05″	–2°36′04″
	APO02	Desembocadura	Ter	Q	42°01′35″	–2°09′50″
	APO03	Nuevo Sifón	Besós	Q	41°27′26″	–2°11′26″
	APO04	Castellbell	Llobregat	Q	41°38′48″	–1°51′31″
	APO05	Abrera	Llobregat	Q	41°29′14″	–1°56′06″
	APO06	San Joan Despí	Llobregat	Q	41°20′59″	–2°02′14″
	APO07	Tarragona	Francolí	Q	41°06′58″	–1°14′15″

[a]Frequency: B = Biweekly; M = Monthly; Q = Quarterly.
[b]Excluded from radiological network in October 2000.
[c]Excluded from radiological network in March 2000.

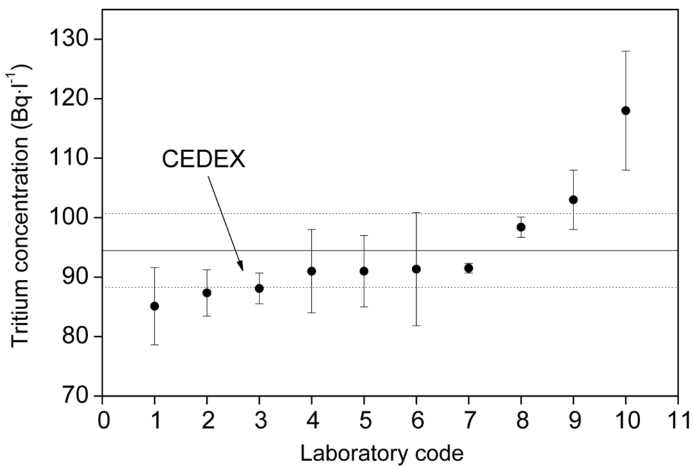

Figure 3 Reported results for tritium analyses of water in an intersample comparison exercise.

The mean tritium activities found in Spanish river waters are shown in Table 4. The mean tritium activities were calculated using all reported values over time at each sampling location, including outliers and activities lower than the MDA. Uncertainties were derived from sample preparation and counting statistics. All reported uncertainties correspond to $k = 1$. Sample codes have been arranged to show the spatial evolution downstream (i.e. Tagus River, Tagus Basin, in Table 4 and Figure 5).

Discussion of the results for river basins is divided into nuclear-free basins (Northern rivers Douro, Guadiana, Guadalquivir, Segura, and Pyrenees rivers) and nuclearized basins (Tagus, Júcar, and Ebro).

Nuclear-free River Basins

The mean tritium activity of the Northern rivers Douro, Guadiana, Guadalquivir, and Pyrenees rivers ranged, respectively, from 0.73 ± 0.16 to 1.63 ± 0.19 Bq/L; 1.19 ± 0.13 Bq to 2.67 ± 0.18 Bq/L; 1.54 ± 0.23 to 2.05 ± 0.18 Bq/L; 1.76 ± 0.13 to 2.56 ± 0.27 Bq/L; and 1.54 ± 0.15 to 2.01 ± 0.20 Bq/L. The water collected was free of the influence of the NPPs; therefore, the concentrations found may be considered baseline values. It seems reasonable to attribute the origin of this tritium to i) natural fallout of tritium produced in the interaction of stable nuclei with cosmic radiation in the atmosphere and ii) fallout from atmospheric nuclear weapons tests.

Table 4 Mean tritium activities at various locations in Spanish river waters since the 1980s.

Basin	River	Sample code	Operation year	n^a	Mean tritium (Bq/L)
North rivers	Miño	ANE01	1982	107	1.60 ± 0.22
		ANE02	1982	87	1.40 ± 0.19
		ANE03	1982	87	1.63 ± 0.19
	Ulla	ANE05	1996	95	1.14 ± 0.16
	Navia	ANE06	1996	97	1.30 ± 0.13
	Nalón	ANE07	1996	98	1.43 ± 0.12
	Pas	ANE08	1996	96	1.27 ± 0.12
	Asón	ANE09	1996	97	1.18 ± 0.12
	Nervión	ANE10	1996–2000	47	1.56 ± 0.19
		ANE12	2001	46	0.73 ± 0.16
	Bidasoa	ANE11	1996	92	1.06 ± 0.12
Douro	Douro	ADU01	1982	81	1.91 ± 0.19
		ADU03	1982	188	1.83 ± 0.12
		ADU04	1982	197	1.75 ± 0.12
	Pisuerga	ADU05	1982	84	1.70 ± 0.19
		ADU06	1982	269	1.94 ± 0.11
	Tormes	ADU07	1982	259	1.86 ± 0.12
		CDU12	1995	110	1.36 ± 0.14
		ADU14	2000	56	1.30 ± 0.18
	Agueda	CDU08	1982	262	2.37 ± 0.13
		CDU10	1982	217	2.67 ± 0.18
		CDU09	1982	262	2.49 ± 0.13
		CDU11	1989	179	1.92 ± 0.15
		ADU13	1995	107	1.19 ± 0.13
Tagus	Tagus	CTA01	1982	188	1.76 ± 0.13
		CTA02	1982	209	20.4 ± 1.0
		CTA03	1982	208	29.1 ± 3.8
		ATA04	1982	123	24.5 ± 2.1
		ATA44	1990	160	8.12 ± 0.49
		ATA41	1991	105	8.88 ± 0.60
		ATA05	1982	125	5.94 ± 0.41
		CTA18	1982	245	4.44 ± 0.18
		CTA19	1982	206	25.0 ± 1.8
		ATA07	1982	213	7.26 ± 0.39
	Manzanares	ATA23	1982	110	1.87 ± 0.21
		ATA08	1982	108	1.76 ± 0.19
		ATA09	1982	99	2.55 ± 0.37
	Jarama	ATA12	1990	93	1.28 ± 0.15
		ATA24	1982	164	2.18 ± 0.15
		ATA10	1982	193	1.77 ± 0.12
	Guadarrama	ATA22	1982	99	2.33 ± 0.25
	Alberche	ATA13	1990	96	1.53 ± 0.16
	Tiétar	ATA43	1991	87	1.82 ± 0.26
	Alagón	ATA42	1991	83	1.70 ± 0.19
Guadiana	Guadiana	AGU01	1982	162	2.05 ± 0.18
		AGU02	1982	82	1.54 ± 0.23
		AGU03	1982	84	1.68 ± 0.18
		AGU04	1982	182	1.97 ± 0.13
	Guadalupejo	AGU05	1982	179	1.72 ± 0.15

Table 4 Mean tritium activities at various locations in Spanish river waters since the 1980s. *(Continued)*

Basin	River	Sample code	Operation year	n^a	Mean tritium (Bq/L)
Guadalquivir	Guadalquivir	AGR01	1982	76	2.05 ± 0.28
		AGR09	1982	206	1.94 ± 0.15
		CGR02	1982	220	1.76 ± 0.13
		CGR03	1982	228	1.93 ± 0.16
		AGR10	1982	174	2.08 ± 0.17
		AGR07	1982	178	2.01 ± 0.23
		AGR08	1982	176	2.10 ± 0.20
		AGR04	1982	240	1.68 ± 0.13
	Guadalimar	AGR11	1982	169	2.14 ± 0.19
	Jándula	AGR12	1982	177	1.79 ± 0.18
	Bembezar	AGR05	1982	84	2.56 ± 0.27
		AGR06	1982	236	1.77 ± 0.15
Segura	Segura	ASG01	1991	92	1.96 ± 0.31
		ASG02	1991	93	6.15 ± 0.35
		ASG03	1991–2000	108	4.13 ± 0.25
	Mundo-Channel	ASG05	1991	164	7.66 ± 0.53
	Mundo	ASG04	1991	91	8.46 ± 0.52
Júcar	Júcar	AJU02	1993	84	1.45 ± 0.15
		AJU03	1993	85	14.0 ± 0.8
		CJU05	1993	92	5.06 ± 0.44
		CJU01	1982	256	4.08 ± 0.28
	Júcar-Turia Channel	AJU06	1994	128	4.94 ± 0.23
	Cabriel	CJU04	1993	92	1.30 ± 0.16
Ebro	Ebro	CEB21	1992	163	1.55 ± 0.13
		CEB22	1992	310	1.87 ± 0.12
		AEB05	1982	89	1.82 ± 0.21
		AEB06	1979	257	1.88 ± 0.12
		AEB08	1982	178	2.15 ± 0.17
		CEB04	1982	259	2.11 ± 0.13
		CEB17	1995	247	1.78 ± 0.11
		CEB16	1987	439	8.61 ± 0.47
		AEB07	1982	185	6.40 ± 1.00
	Segre	AEB09	1982	248	2.31 ± 0.14
		AEB10	1982	253	2.41 ± 0.14
Pyrenees rivers	Ter	APO01	1993	76	1.54 ± 0.15
		APO02	1993	75	1.78 ± 0.23
	Besós	APO03	1993	74	2.01 ± 0.20
	Llobregat	APO04	1993	76	1.85 ± 0.15
		APO05	1993	76	1.76 ± 0.16
		APO06	1993	76	1.90 ± 0.18
	Francolí	APO07	1993	64	1.55 ± 0.19

[a] n = number of samples.

Some nuclear fuel cycle facilities are present in Douro Basin (Saelices el Chico on the Agueda River and Juzbado on the Tormes River) and in the Guadalquivir Basin (El Cabril on the Bembezar River and FUA on the Guadalquivir River), but no significant increase in tritium activity was detected.

One interesting feature is the mean tritium activity found in Segura Basin (Figure 4). The mean tritium value observed in ASG01 may be considered a baseline value (1.96 ± 0.31 Bq/L) similar to that of nuclear-free river basins. Nevertheless, an increase above the baseline value is observed from ASG05, Tajo-Segura Channel, to the mouth. This may be due to the transport of water from the upper Tagus (nuclearized river basin) to the Segura Basin (Payeras et al. 1994).

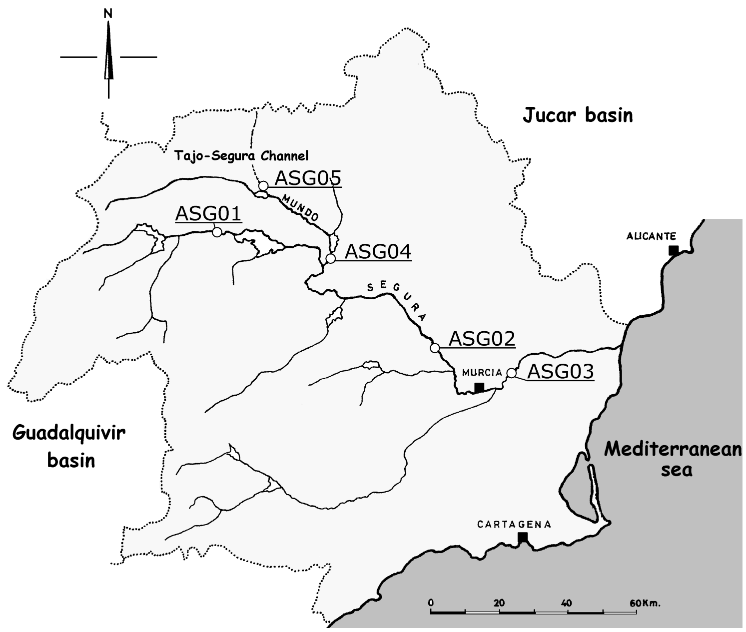

Figure 4 Sampling locations in Segura Basin (ASG03 currently excluded from the radiological network).

Tagus Basin

Figure 5 shows the spatial distribution of mean tritium activities in the Tagus River. The mean tritium concentrations ranged over 1 order of magnitude (1.76 ± 0.13 to 29.1 ± 3.8 Bq/L). Water collected in CTA-01, upstream from Trillo NPP, was free of the influence of the nuclear power plant and, therefore, the concentration may be considered as a baseline value for those values observed downstream. Significant mean tritium concentrations can be observed after the Trillo, José Zorita, and Almaraz NPPs. There was significant dilution of these levels along the river downstream from the NNPs. In the Alcántara Dam, near the Portuguese border, the mean tritium activity concentration is low, 7.26 ± 0.39 Bq/L. It should be stressed, however, that the concentrations observed do not represent any radiological risk for the population.

Mean tritium activity in the main Tagus tributaries (Manzanares, Jarama, Guadarrama, Alberche, Tiétar, and Alagón) ranged from 1.28 ± 0.15 to 2.55 ± 0.37 Bq/L. These values are similar to those found in nuclear-free river basins.

Júcar Basin

The mean tritium activity concentration in the Alarcón Dam (AJU03), upstream from Cofrentes NPP, was 14.0 ± 0.8 Bq/L. This value is above the baseline level of 1.45 ± 0.15 Bq/L found upstream in AJU02. The Alarcón Dam forms part of the Tajo-Segura Channel; thus, Tagus River water

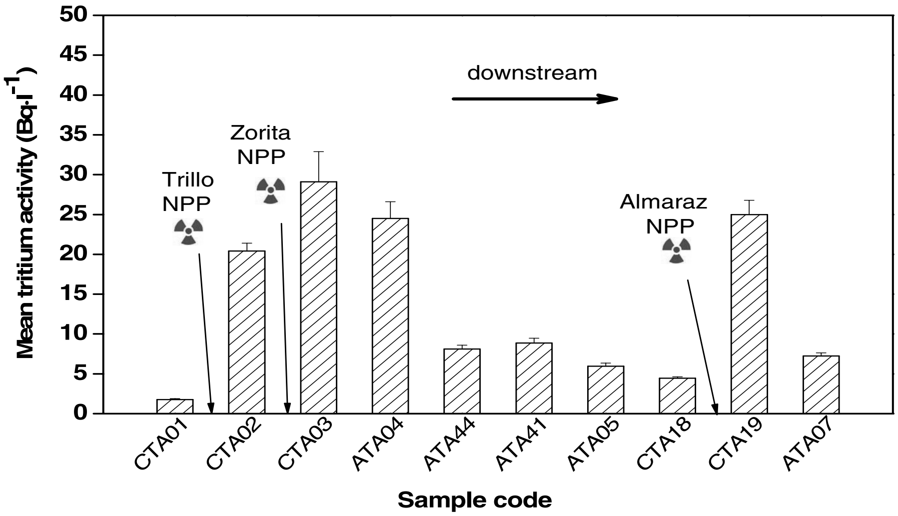

Figure 5 Mean tritium concentration in Tagus river from its rise to the Portuguese border

(downstream Trillo NPP and upstream Zorita NPP) is transferred to this dam before reaching the Segura Basin. Therefore, all sampling locations downstream from Alarcón Dam (CJU05, CJU01, and AJU06) in the Júcar River present mean tritium concentrations above the baseline value. As a result, the mean tritium activity in Cabriel River (CJU04), tributary of the Júcar, is 1.30 ± 0.16 Bq/L because this river does not receive water from the Tajo-Segura Channel. On the other hand, it seems that the mean tritium levels are not increased by the influence of the Cofrentes NPP.

Ebro Basin

The mean tritium concentrations upstream (CEB21) and downstream (CEB22) from Santa María de Garoña NPP were statistically indistinguishable from the mean tritium activity of nuclear-free river waters. From this location to CEB17, the tritium concentration ranged from 1.78 ± 0.11 to 2.15 ± 0.17 Bq/L. Tritium above these baseline levels was detected downstream of Ascó I and II NPPs (CEB16 and AEB07). During normal operation, these 2 units generate low-activity radioactive liquid waste, including tritium, which is released into the river under regulatory control.

CONCLUSIONS

This study provides an overview of the tritium levels in Spanish river waters since the 1980s. We see a clear influence of the relatively high levels of tritium existing in the water of the Tagus River, which has several nuclear power plants (Trillo, José Cabrera, and Almaraz I and II). There was also a significant dilution downstream from the NPPs. It is important to note the influence of these tritium releases on the Júcar and Segura basins due to the Tajo-Segura Channel.

Tritium was also detected in the Ebro River, where the NPPs Ascó I and Ascó II are located. During normal operation, these 2 units generate low-activity liquid wastes, including tritium, which is released into the river under regulatory control. However, the mean tritium activities were similar to those in nuclear-free river basins upstream and downstream of the Santa María de Garoña NNP. No tritium was detected in non-nuclearized Spanish river basins, e.g. Northern rivers Douro, Guadiana, Guadalquivir, and Pyrenees rivers, with the exception of Segura Basin.

It should be noted that information provided in this study may be used to construct a dispersion model to reproduce the influence of tritium from NPPs from the Tagus River to the Tajo-Segura Channel.

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