
Environmental impact assessment of uranium ore mining and radioactive waste around a storage centre from Mexico

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Abstract. Uranium ore tailings and radioactive waste have been stored during four decades at the Mexican Storage Centre for Radioactive Waste (SCRW). An estimation of effective doses from sources of natural and artificial origin in the general population and workers is presented. The sampling points are located at the SCRW at a 200 m neighbouring zone of the site and at distant communities around SCRW (regional background). The effective gamma dose rates were estimated from measurements of radionuclide concentration in soil samples from 1991 to 2002. Doses from the outdoor and indoor inhalation of radon were also determined. Transfer factors for ²²⁶Ra, ²¹⁰Pb, ¹³⁷Cs and ⁴⁰K from soil to wild edible species have been used to estimate the contribution of wild local foods to the dietary intake and to assess the supplementary doses received due to internal irradiation. The effective dose via the different exposure pathways estimated in the population that would live at the future in the immediate SCRW vicinity was two times higher than for the regional background.

1. INTRODUCTION

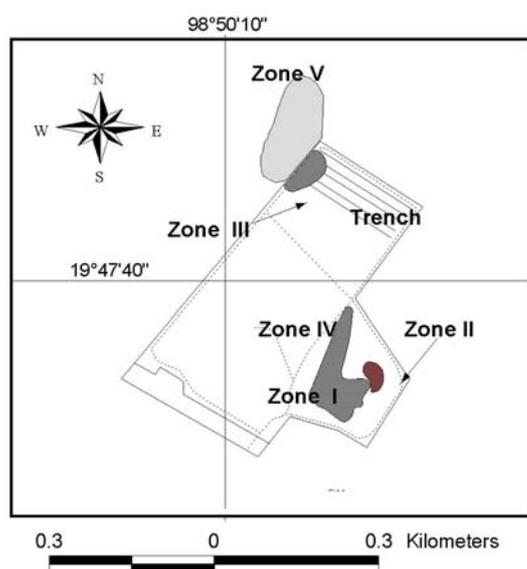
Ore extraction and processing may lead to technologically enhanced naturally occurring radionuclide materials (TENORM), in nuclear installations and the environment. Continuous releases of these materials to the environment may result in a buildup in the air, water and soil of some radionuclides, particularly ²²⁶Ra. Natural world average concentrations of ⁴⁰K, ²²⁶Ra, ²³²Th, and ²³⁸U in the soil are 400, 40, 25 and 25 Bq kg⁻¹ respectively [1]. International radioprotection norms [2] admit for an individual from the population, an additional dose “caused” by all the natural sources via all the pathways from the region in which he lives of 1 mSv y⁻¹, which has to be added to the effective dose due to the natural background of the investigated area.

At the Mexican Storage Centre for Radioactive Waste (SCRW), solid and liquid radioactive waste have been stored during four decades and uranium ore tailing piles also stood for some time before being buried in trench and specific containers. A local contamination in the soil with ¹³⁷Cs also occurred at the site as a consequence of a broken industrial source [3]. An environmental monitoring program has been systematically conducted at the SCRW and surrounding communities and intervention and decontamination actions, with removal of the upper soil layer and confining it into containers, were performed at the site from 1993 to 2000 [4]. Determinations of trace stable and radioactive elements have been also performed in the soil as well as some bioindicators [5], [6], [7].

In the present paper the effective gamma dose rates in air from terrestrial radionuclides, doses from the outdoor and indoor inhalation of radon and doses from ingestion of possibly contaminated local foods, were estimated to determine the radiological impact in agricultural soils of uranium mining past activities and SCRW operation. The sampling period considered is from 1990 to 2002. Workers are employed in waste and soil removing and they may be exposed to external radiation and outdoors radon inhalation only because there are no offices at the site. The enhancement of radioactivity levels was calculated in order to assess the additional doses received by the general public and workers due to external exposure and internal irradiation.

2. MATERIALS AND METHODS

The SCRW site ($19^{\circ} 47' 40''$ N; $98^{\circ} 50' 10''$ W) is located in the State of Mexico, Mexico at altitudes between 2470 and 2490 m, in the middle part of the Mexican Neovolcanic belt (Figure 1). The climate is temperate, sub-humid with an average yearly temperature of 14°C , ranging from 6 to 32°C . During the rainy season from June to October, the mean annual precipitation is 638.5 mm. The sampling points from SCRW are: Zone I, corresponding to the area where the ore was piled and the ^{137}Cs contamination occurred; Zone II at a lower altitude than Zone I was a private property until 1993; Zone III at the bottom of the slope, the place where rain water was accumulated previously to leave the SCRW and Zone IV that include all the other sampling points inside the SCRW. Five sampling points are also located in a 200 m neighbouring zone of the site (Zone V). That external land is a private property used for local agricultural practices. Four distant communities in a 10 km radius around the site correspond to the regional background (Zone VI).



2.1 Gamma-ray spectrometry and dose evaluation from the soil activity

Several data series samples of ^{235}U , ^{226}Ra , ^{137}Cs and ^{40}K , in the soil have been collected and analyzed. The soil samples were dry at 105°C to constant weight or dry weight (d.w.) before being analyzed in the laboratory and these were sieved through a 50-60 mesh sieve. The (dry/wet) weight ratio was recorded. The samples were analyzed for ^{235}U , ^{226}Ra , ^{137}Cs and ^{40}K specific activities, with a HPGe detector having 29.4 % of relative efficiency. Aliquots of 300-350 g were kept in a 500 ml Marinelli beaker for a period of more than 21 days to allow radium and radon to reach the equilibrium.

Figure 1. Location of sampling points inside the SCRW (Zones I, II, III and IV) and outside at the border line to the SCRW (Zone V).

The effective gamma dose rate (nSv h^{-1}) from terrestrial radionuclides has been calculated with the equation deduced from [2]:

$$H_E = 0.033A_{40\text{K}} + 0.51A_{232\text{Th}} + 0.33A_{226\text{Ra}} + 0.001A_{137\text{Cs}} \quad (1)$$

where,

H_E is the effective dose in (nSv h^{-1}) and A_i is the activity concentration of the i^{th} radionuclide in soil samples (Bq kg^{-1}) (d.w.).

2.2 Radon exposure and dose evaluation

From 1990 to 1992, thirteen track detection devices (SSNTD) for determining radon in gas soil and in air were distributed at fixed stations inside the SCRW (Zones I, III and Trench). Also some SSNTDs were placed at Zone V and at the regional background (Zone VI). The SSNTDs were cellulose nitrate alpha-sensitive LR 115, Type II. They were placed at 30 cm of depth in the soil and in plastic vessels 1m from the ground. The exposure times were for 21 and 120 days respectively and they were etched and spark counted as previously reported [3].

To estimate the indoor radon concentration in (Bq m^{-3}) from outdoor measurements, the mathematical model of Knutson [8] was used. The model assume a complete mixing in the indoor air and the following relationship between indoor and outdoor radon concentrations: $C_{\text{Rn indoor}} = e'/v + C_{\text{Rn outdoor}}$; where: $C_{\text{Rn indoor}}$ and $C_{\text{Rn outdoor}}$ are the indoor and outdoor radon concentrations (Bq m^{-3}); e' =the total volume-specific entry rate of radon in ($\text{Bq m}^{-3} \text{ h}^{-1}$) and v =the ventilation rate in (h^{-1}). A mean ventilation rate of 0.5 h^{-1} in dwellings and a radon entry rate from the soil of $10 \text{ Bq m}^{-3} \text{ h}^{-1}$. were considered in our calculations [8]. For the representative radon concentrations, equilibrium and occupancy factors, and the dose coefficient in terms of Equilibrium Equivalent radon Concentration (EEC), the annual effective doses are derived [2] as:

$$H_E = C_{\text{Rn}} \text{Eq T CF}_{\text{Rn}} 10^{-6} \quad (2)$$

where,

H_E =the annual effective dose by inhalation of radon (mSv y^{-1}); C_{Rn} = the radon concentration in air (Bq m^{-3}); Eq =the equilibrium factor (0.4 and 0.6 for the indoor and outdoor environments respectively); T =the occupancy factors (7000 h y^{-1} and 1760 h y^{-1} for the indoor and outdoor environments to the general public; 2000 h y^{-1} and 6760 h y^{-1} for the outdoor and indoor environments to workers); CF_{Rn} =dose conversion factors for radon ($9 \text{ nSv Bq}^{-1} \text{ h}^{-1} \text{ m}^3$ and $6 \text{ nSv Bq}^{-1} \text{ h}^{-1} \text{ m}^3$ in terms of EEC and the equilibrium factors of 0.4 and 0.6 for the indoor and outdoor environments respectively).

2.3 Dose evaluation from ingested radionuclides

The effective dose (H_E), incurred from the ingestion of a given quantity of food, containing a particular radionuclide was estimated from the following expression [9]:

$$H_E = \sum_i (C_i) R_i \text{Df}_{\text{ing}} 10^{-6} \quad (3)$$

$$C_i = C_s \text{TF } W_i \quad (4)$$

where,

H_E = the annual effective dose by ingestion in (mSv y^{-1}); C_i =the activity concentration (Bq kg^{-1} (w.w.)) of a particular radionuclide of i^{th} foodstuff; C_s =the activity concentration of a particular radionuclide in soil (Bq kg^{-1} (d.w.)); TF =the soil to vegetation or soil to animal food transfer factor expressed as the concentration of an isotope in foodstuff (Bq kg^{-1} (d.w.)) divided by the concentration of the isotope in soil (Bq kg^{-1} (d.w.)); W_i =the (dry/wet) weight ratio of the i^{th} foodstuff; R_i =the annual average consumption rate of i^{th} foodstuff (kg y^{-1} (w.w.)); Df_{ing} =the dose factor for ingestion (Sv per Bq) that would result from an intake of 1 Bq of this radionuclide. The corresponding dose factors (Sv per Bq) are: ^{226}Ra (2.8×10^{-7}), ^{210}Pb (6.9×10^{-7}), ^{137}Cs (1.3×10^{-8}) and ^{40}K (6.2×10^{-9}).

3. RESULTS AND DISCUSSION

The average activity concentrations and range values of ^{226}Ra , ^{137}Cs , and ^{40}K measured from 1991 to 2002 in soil samples at the regional background, at SCRW (zones (I+II), III and IV) and at Zone V, are shown in Table I. The outdoor gamma dose rates from terrestrial gamma exposure (nSv h^{-1}) estimated with equation (1) are also indicated.

Table 1. Activity concentrations in soil at the regional background (Zone VI), at SCRW (Zones I to IV) and at Zone V; external gamma effective dose estimated to the general public and workers.

Year	N	Activity concentrations in soil (average± error) in Bq kg ⁻¹ (d.w.)						Dose rate nSv h ⁻¹	Effective dose mSv y ⁻¹
		²²⁶ Ra		¹³⁷ Cs		⁴⁰ K			
		Average	Range	Average	Range	Average	Range		
Regional background (general public)									
1991-1995	10	24.8±4	17-32	4.7±0.5	1.8-8	288±15	252-347	28.91	0.051
1996-2002	48	27.4±10	12-57	2.8±0.6	1-10	305±30	187-435	30.33	0.053
SCRW (Zone I+II) (workers)									
1991-1995	36	3738±230	182-19979	3273±179	13-18148	286±55	198-560	1260	2.52
1996-2002	147	1019±109	91-7871	152±40	9-1952	286±37	165-449	360	0.72
SCRW (Zone III) (workers)									
1991-1995	11	122±17	36-218	97±6	40-158	224±18	204-258	61.52	0.12
1996-2002	37	53±14	20-115	51±7	6-188	244±27	156-299	39.36	0.08
SCRW (Zone IV) (workers)									
1991-1995	7	109±15	21-258	86±1	14-136	252±25	204-371	58.14	0.12
1996-2002	91	32±10	12-66	22±3	2-126	235±24	153-331	32.11	0.06
Zone V (general public)									
1991-1995	10	108.7±10	38-237	69.7±3.7	16-160	251±16	179-276	55.44	0.098
1996-2002	45	119.8±23	36-182	83.9±6.6	2-148	241±26	198-302	58.79	0.103

The effective doses (mSv y⁻¹) were estimated considering an occupancy factor of 1760 h y⁻¹ and 2000 h y⁻¹ to the general public and to workers respectively. At the regional background, the ²²⁶Ra average value from 1991 to 2002 was 26 Bq kg⁻¹ (d.w.). ²³²Th appears to be essentially in secular equilibrium with its entire decay chain and gamma radiation from ²²⁸Ac, ²¹²Pb and ²⁰⁸Tl may all be used in its determination [10]. The average values from those radionuclides, measured during 1991 were: 22±3, 21±2, and 9±1 Bq kg⁻¹ (d.w.) for ²²⁸Ac, ²¹²Pb and ²⁰⁸Tl, respectively. An ²³²Th average value of 22 Bq kg⁻¹ (d.w.), was considered to estimate the gamma dose rate in air by equation (1). The concentration of ²¹⁰Pb in surface soil may be 1.2 to 10 times greater than ²²⁶Ra [11]. An average value of 35 Bq kg⁻¹ (d.w.) to ²¹⁴Pb (1.3 times greater than for ²²⁶Ra) was measured at the regional background in 1991, in agreement [11]. An ²¹⁰Pb (≈²¹⁴Pb) average value of 35 Bq kg⁻¹ (d.w.) in soil was considered to estimate the effective dose by ingestion (4). Activity concentrations of ¹³⁷Cs and ⁴⁰K average values were 4 and 297 Bq kg⁻¹ (d.w.) respectively.

The average effective dose to the general public estimated from 1991 to 2002 was 0.052 mSv y⁻¹ (0.259 mSv y⁻¹ considering 8760 hours per year). An average external effective dose of 0.60 mSv y⁻¹ at the regional background was estimated from the gamma exposure measurements (1991 to 2002) using passive integrating thermoluminescent dosimeters (TLD's) of CaSO₄: Dy [12]. That measurements include the contribution from cosmic and terrestrial radiation, so the average effective dose from cosmic radiation can be estimated by subtraction of the dose due to terrestrial gamma exposure from the total annual effective dose (0.60-0.259)=0.34 mSv y⁻¹.

At SCRW, during 2000 and 2001, 400 m³ of contaminated soil were removed from Zones (I+II). From these actions, the average values of ²²⁶Ra diminished from 3738±230 to 1019±109 Bq kg⁻¹ (d.w.); the average values of ²³⁵U were 70±8 Bq kg⁻¹ (d.w.) range (15-168) in (1991-1995) and 24±6 Bq kg⁻¹ (d.w.) range (6-65) in (1996-2002) periods. The ²³⁵U/²²⁶Ra isotopic ratio ranged between 0.012 (1.2%) and 0.043 (4.3%). At Zone II, the lowest isotopic ratio value (1.2%) was similar to ²³⁵U/²²⁸U isotopic ratio (1.29%) reported by Magnoni et al. [13] for the typical value for depleted uranium. However, the

average values of ²²⁸Ac, ²¹²Pb and ²⁰⁸Tl were: 27±5, 19±2 and 11±1 Bq kg⁻¹ (d.w.) respectively, similar to the regional background measurements. the average effective dose to workers diminished substantially (from 2.52 to 0.72 mSv y⁻¹) from (1991-1995) to (1996-2002) periods.

Derived intervention level adopted by the National Regulatory Commission (CNSNS) for ²²⁶Ra at the SCRW is 130 Bq kg⁻¹. At Zone V, the maximum value (237 Bq kg⁻¹) obtained in the period 1991-1995, was higher than the limits adopted internationally to exempt solid materials or as screening level for the unrestricted use (185 and 200 Bq kg⁻¹ respectively). The activity concentrations of ¹³⁷Cs in the surface soil were 19 times higher than for the regional background, while average values of 246±22 Bq kg⁻¹ (d.w.) range (179-302) and 57±12 Bq kg⁻¹ (d.w.) range (26-85) were obtained for ⁴⁰K and ²¹⁴Pb respectively. Once the average value (1991-2002) of the regional background (0.052 mSv y⁻¹) is subtracted (0.101-0.052= 0.048 mSv y⁻¹), the additional external dose is obtained for the general public (1760 h y⁻¹ outdoors). The external effective dose for a person living in the future at Zone V, considering 8760 hours per year and the cosmic radiation (0.34 mSv y⁻¹) will be 0.84 mSv y⁻¹, one and a half times higher than for the regional background.

Measurements in the soil (30 cm depth) and (1m above ground) had shown the presence of enhanced concentrations of up to 1705 (660-2596) Bq m⁻³ of ²²²Rn in gas soil at Zone I and of 4747 Bq m⁻³ of ²²²Rn in gas soil at the trench zone (Table 2). The average concentration values of ²²²Rn in air at the same zones were 57 and 36 Bq m⁻³ respectively, while the average values of ²²²Rn in soil and in air at the regional background were 31 Bq m⁻³ and 7 Bq m⁻³ respectively.

Table 2. ²²²Rn concentrations (average ± error) in Bq m⁻³; internal effective dose to the general public and workers during 1990 to 1992.

Zone	N	²²² Rn concentration Bq m ⁻³		Effective dose (workers) mSv y ⁻¹	Effective dose (general public) mSv y ⁻¹	
		30 cm depth	1 m above ground	Outdoor	Outdoor	Indoor
Background (VI)	3	31±2 (26-35)	7±1		0.044	0.68
Zone V	5	134±6 (77-265)	20±1 (8-32)		0.13 (0.05-0.20)	1.01 (0.40-1.61)
SCRW	I	1705±85 (660-2596)	57±2 (41-78)	0.41 (0.29-0.56)		
	III	275± 11 (41-753)	45±2 (25-73)	0.32 (0.18-0.53)		
	Trenchs	805±40 (44-4747)	36±1 (15-63)	0.26 (0.11-0.45)		
SCRW (average)	20	928±46	48±2	0.33		

The average indoor radon concentration values estimated at regional background (Zone VI) and at Zone V are:

$$C_{Rn \text{ indoor (Zone VI)}} = 10 \text{ (Bq m}^{-3} \text{ h}^{-1}) / 0.5 \text{ (h}^{-1}) + 7 \text{ (Bq m}^{-3}) = 27 \text{ (Bq m}^{-3})$$

$$C_{Rn \text{ indoor (Zone V)}} = 10 \text{ (Bq m}^{-3} \text{ h}^{-1}) / 0.5 \text{ (h}^{-1}) + 20 \text{ (Bq m}^{-3}) = 40 \text{ (Bq m}^{-3})$$

In Mexico City, relatively near of the SCRW, the average radon concentration values from 50 dwellings [14], showed an average indoor radon concentration of 19.2 Bq m⁻³ (range 4-115) and outdoors: 13 Bq m⁻³ (range 4-57). The city of Zacatecas, with a climate cooler than Mexico City, shows an average indoor radon value of 46.2 (14-86) Bq m⁻³ and 23.1 (4-56) Bq m⁻³ outdoors [14].

For the representative background radon concentrations, equilibrium and occupancy factors, and the dose coefficient, the following annual effective doses are derived from the regional background:

Indoors: $27 \text{ Bq m}^{-3} \times 0.4 \times 9 \text{ nSv Bq}^{-1} \text{ h}^{-1} \text{ m}^3 \times 7000 \text{ h y}^{-1} \times 10^{-6} \text{ mSv nSv}^{-1} = 0.68 \text{ mSv y}^{-1}$ and

Outdoors: $7 \text{ Bq m}^{-3} \times 0.6 \times 6 \text{ nSv Bq}^{-1} \text{ h}^{-1} \text{ m}^3 \times 1760 \text{ h y}^{-1} \times 10^{-6} \text{ mSv nSv}^{-1} = 0.044 \text{ mSv y}^{-1}$

If we add the minor contribution from radon dissolved in blood (0.06) and from thoron and their short-lived decay products (0.1) [2], at the total value obtained (0.724 mSv y⁻¹), the average exposure to radon at the regional background (Zone VI) would be: 0.884 mSv y⁻¹.

The world average concentration values for ²²²Rn are: 40 (2-50) Bq m⁻³ indoor and 3-7 Bq m⁻³ outdoor and for ²²⁰Rn: 0.3 Bq m⁻³ indoor and 0.1 Bq m⁻³ outdoor. The corresponding average doses from ²²²Rn are: 0.67 mSv y⁻¹ and 0.02 mSv y⁻¹ for the indoor and outdoor environments respectively; the contribution by thoron and its decay products is about 0.1 mSv [2].

Monitoring of conventional agricultural and wild animal products for the local market (Zone VI) and of some wild products at SCRW and at Zone V are undertaken to determine levels of activity and doses estimations to the consuming populace. The food group and consumption rates included in the routine surveillance programme are shown in Table 3. For dose estimations, the average values of radioactivity in the soil (1991-2002) at Zone VI were considered while at Zone V, the average values from (1996-2002) were taken.

Table 3. Average consumption rate, (dry/wet) weight ratios and transfer factors soil-local foods; effective dose by ingestion estimated in wet weight for the general public at background (Zone VI) and at Zone V.

Food group	Consumption rate kg y ⁻¹ (w.w)	dry/wet weight ratio	Transfer factor (TF) (d.w.)				Effective dose (mSv y ⁻¹)	
			²²⁶ Ra	²¹⁰ Pb	¹³⁷ Cs	⁴⁰ K	Zone VI	Zone V
<i>Helix aspersa</i>	10	0.2	0.69	0.90	0.25	0.57	0.056	0.119
<i>Lyometopum appiculatum</i>	5	0.3	0.30	0.30	0.10	0.67	0.016	0.035
<i>Opuntia megacantha</i>	25	0.2	0.30	0.09	0.30	3	0.049	0.092
<i>Agave atrovirens</i>	15	0.4	0.80	0.09	0.30	2	0.070	0.202
<i>Avena sativa</i>	10	0.3	0.04	0.02	0.03	0.90	0.0073	0.011
<i>Zea mays</i>	40	0.8	0.04	0.02	0.001	0.90	0.078	0.111
<i>Phaseolus vulgaris</i>	40	0.8	0.04	0.02	0.01	0.90	0.078	0.112
<i>Other vegetables</i>	70	0.2	0.20	0.09	0.20	3	0.128	0.209
Effective dose (mSv y ⁻¹) Zone VI (ZoneV)	215		0.10 (0.46)	0.14 (0.23)	0.0004 (0.008)	0.24 (0.19)	0.482	0.891

The local wild foods sampled were: land snails (*Helix aspersa*), “escamoles” known as eggs (immature stages of the ant specie *Lyometopum appiculatum*), “nopal” (*Opuntia megacantha*), “maguey” (*Agave atrovirens*) and oat (*Avena sativa*), corn (*Zea mays* Sacharata) and beans (*Phaseolus vulgaris* Jamapa), that were cultivated “in situ” reproducing the local agricultural practices [7]. For other vegetables not locally produced and which do not have radiation measurements, we take the TF from [9]. If the average value of the dose by ingestion estimated for regional background (0.482 mSv y⁻¹) is subtracted (0.891-0.482=0.409 mSv y⁻¹), the additional dose by ingestion is obtained for the general public at Zone V. At Zone VI the main contributions (50%) of the total ingestion correspond to ⁴⁰K, followed by ²¹⁰Pb (29%) and ²²⁶Ra (21%). At Zone V, the estimated supplementary dose via ingestion was 0.41 mSv y⁻¹.

4. CONCLUSIONS

As a result of the activity deposited on soil at SCRW, we found at Zone V, in the upper 10 cm layer of soils, four times higher concentrations of ²²⁶Ra and 19 times higher concentrations of ¹³⁷Cs than for the regional background (considering the average values). The excess in specific activities results in an increase in exposure to gamma rays. The supplementary average effective dose defined as the addition of all the possible pathways (external, inhalation and ingestion) for a person living at Zone V

will be 0.87 mSv y^{-1} . If the calculation is made considering 8760 h y^{-1} , the average annual effective dose (external, inhalation and ingestion) for the population at Zone VI (background) is 1.97 mSv y^{-1} and 3.03 mSv y^{-1} at Zone V (1.1 mSv y^{-1} additional). At SCRW (Zones I+II) the average external effective dose (1991-2002) from terrestrial gamma field was estimated to be: 1.62 mSv h^{-1} , 31 times higher than for the regional background (0.052 mSv h^{-1}). For workers the additional average dose (external and radon outdoors) was 1.9 mSv y^{-1} . If the maximum values (1991-1995) are considered, the dose would reach 3 mSv y^{-1} . At present time there is no population inhabiting Zone V and adverse health effects from increased natural background radioactivity may seem unlikely for the near term. However for a future scenario, where people would live at this zone adverse health effects for the long-term accumulation of radioactive materials from the SCRW could occur. Restriction of land use, with special emphasis on the control of ^{226}Ra , or even remediation would be advisable.

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