Prompt Determination of ^{137}Cs in Large Volume seawater using Cu-hexacyanaferrates cartridges

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1. Introduction

Office of Atoms for Peace, Radiation monitoring group has carried out radiation monitoring program in Thailand for establishment of radiation baseline information and assessment of the radiological impact on the environment in case of any incidents. Cs-137 is one of the fission products which has a long half life of 30 years. It can be released from nuclear accident, nuclear waste discharge and atomic boom test to atmosphere and through ocean. Therefore ^{137}Cs is one of our monitored radionuclides in environmental samples especially in seawater. The determination of ^{137}Cs in ocean can be applied to monitor a release of any radiological incident in the pacific region. Routinely the classical ammonium molybdophosphate (AMP) pre-concentration method has been used to determine ^{137}Cs in seawater. This analysis passed the proficiency test exercise under the project "IAEA proficiency test exercise on Marine benchmark study on the possible impact of the Fukushima radioactive releases in the Asia-Pacific Region for Caesium Determination in Sea Water (RCA RAS/7/021)". However this technique is time consuming and quite laborious. Therefore $CuFeCN_6$ cartridge technique has been developed. The results of the developed method were compared with the traditional AMP pre-concentration technique. This developed method is more cost-efficient and less time consuming. In addition it can be easily performed in sampling fields

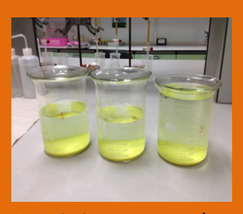
2. Experimental

Local sea water at Sriracha Fishery Research Station, Thailand (13°11' 03.8"N, 100°55' 43.7"E) was used for both of AMP pre-concentration and $CuFeCN_6$ cartridge methods.

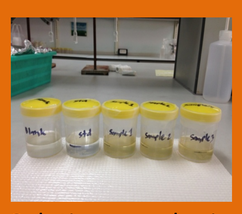
2.1 AMP pre-concentration method

Seawater samples

Precipitating samples



Precipitate samples



Solution samples in calibrated containers

- Three 200 litter seawater samples in separated containers were acidified with HCl to pH 1.6.
- 2.6 g of CsCl + 40 g of AMP were added and the mixture were stirred and left the precipitate to settle down.
- The precipitate samples were centrifuged , washed with 1 M HCl and dissolved with 10 M NaOH.
- The solution were heated to eliminate ammonia and the precipitation step was repeated and the precipitate samples were dissolved with 10 M NaOH.
- The solution were transferred to calibrated containers and made up volume to 20 ml

2.2 $CuFeCN_6$ cartridges method

$CuFeCN_6$ cartridges

Cartridges set

Ashed Cartridge samples

Ashed samples in Calibrated containers

- The $CuFeCN_6$ cartridges set was fabricated : pump > cotton cartridge > first $CuFeCN_6$ cartridge (A) > second $CuFeCN_6$ cartridge (B) > flow meter > outlet tube.
- Two 200 , 300 and 600 litter seawater samples were pumped through the cartridges with flow rate at 4 L/min.
- The cartridges samples were ashed at 400°C for 15 hr
- The ashed samples were transferred to calibrated containers

3. Results

3.1 The AMP pre-concentration method

AMP sample	Sample volume (L)	Activity (mBq/L)	Uncertainty (mBq/L)
200L/AMP/sample 1	200	0.782	0.059
200L/AMP/sample 2	200	0.748	0.058
200L/AMP/sample 3	200	0.749	0.059
200L/AMP/average	200	0.760	0.059

The AMP pre-concentration technique :

- The result was precise with standard deviation of 0.019. This technique passed the proficiency test exercise, "IAEA proficiency test exercise on Marine benchmark study on the possible impact of the Fukushima radioactive releases in the Asia-Pacific Region for Caesium Determination in Sea Water (RCA RAS/7/021)", for accuracy, precision and trueness criteria with -1.00 % bias.

3.2 The $CuFeCN_6$ cartridge method

Cartridge sample	Sample volume (L)	Efficiency of Cs collection	Activity (mBq/L)	Uncertainty (mBq/L)
200 L/cartridge/sample 1	200	79.05	0.837	0.080
200 L/cartridge/sample 2	200	52.08	0.698	0.101
200L/cartridge/average	200	65.57	0.768	0.091
300 L/cartridge/sample 1	300	57.58	0.901	0.077
300 L/cartridge/sample 2	300	89.86	0.831	0.060
300 L/cartridge/average	300	73.72	0.866	0.069
600 L/cartridge/sample 1	600	68.33	0.889	0.057
600 L/cartridge/sample 2	600	74.31	0.762	0.050
600 L/cartridge/average	600	71.32	0.826	0.079

The AMP pre-concentration vs. $CuFeCN_6$ cartridge methods :

- Comparing between the two methods at 200 L samples, both result gave insignificant difference i.e.+ 1.05 % (0.760 mBq/L for the AMP technique and 0.768 mBq/L for the cartridge method). Assuming the $CuFeCN_6$ cartridges would have + 0.05 % bias approximately referring to the IAEA proficiency test RCA RAS/7/021. Consequently the $CuFeCN_6$ cartridge method gave the accurate result as the PT exercise judged ± 20% bias was accepted for accuracy.

The $CuFeCN_6$ cartridge method :

- Comparing between the different sample volumes, the activity concentrations were close together i.e. 0.768 mBq/L, 0.866 mBq/L and 0.826 mBq/L for 200 L, 300 L and 600 L respectively. Assuming the minimum 200 L seawater was adequate to perform the $CuFeCN_6$ cartridge method.
- The Cs collection efficiencies were not stable, but the repeated samples with different Cs collection efficiencies gave the similar final ^{137}Cs activity, assuming the two $CuFeCN_6$ cartridges in series were sufficient to absorb Cs in the seawater samples.

4. Conclusion

-The developed $CuFeCN_6$ cartridge technique gave accurate results as the activity concentration of ^{137}Cs in 200 L seawater samples was similar to those of the AMP pre-concentration technique i.e. 0.760 mBq/L for the AMP technique and 0.768 mBq/L for the cartridge method with + 0.05 % bias approximately as referred to the IAEA proficiency test exercise RCA RAS/7/021.

-The minimum 200 L seawater samples can be adequate to perform the $CuFeCN_6$ cartridge technique.

- The $CuFeCN_6$ technique should be concerned about quality of unstable prepared $CuFeCN_6$ cartridges since the repeated samples had significantly different Cs collection efficiencies. Fortunately the two $CuFeCN_6$ cartridge series was efficient to adsorb Cs in the seawater samples as the final activity concentrations of the repeated samples were proved to be similar.
- The $CuFeCN_6$ cartridge technique was more cost efficient and less time consuming.