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QUANTIFICATION OF ¹²⁹I IN MILK AT LOW LEVEL FOR ENVIRONNEMENTAL MONITORING

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Iodine in milk : Origin and interest





lodine 129 can be transferred to grass and then in milk after ingestion by mammals (*e.g.* cows, goats)



Iodine is an essential constituent of the thyroid gland [3]

¹²⁹I could therefore bind to the thyroid gland and cause thyroid cancer

lodine 129 quantification is important

[1] E. García-Toraño et al., 'The half-life of 129I', Appl. Radiat. Isot., vol. 140, pp. 157–162, Oct. 2018, doi: 10.1016/j.apradiso.2018.06.007.

[2] K. Markou, N. Georgopoulos, V. Kyriazopoulou, and A. g. Vagenakis, 'Iodine-Induced Hypothyroidism', Thyroid, vol. 11, no. 5, pp. 501–510, May 2001, doi: 10.1089/105072501300176462.
[3] M. Kadowaki et al., 'Impacts of anthropogenic source from the nuclear fuel reprocessing plants on global atmospheric iodine-129 cycle: A model analysis', Atmos. Environ., vol. 184, pp. 278–291, Jul. 2018, doi: 10.1016/j.atmosenv.2018.04.044.



Usual protocol



Efficient technic \rightarrow LOD* = 0.01Bq/L



chemical yield (127I)

Use of CCl_4 :

- CMR**solvant
- No longer marketed in France under the REACH regulation

Replace liquid-liquid extraction by SPE*** and gravimetry by ICP-MS measurement

*LOD: Limit of Detection **CMR: Carcinogenic, Mutagenic, or toxic for Reproduction ***SPE: Solid Phase Extraction



New protocol development



[1] C. Carrier et al., 'Determination of environmental gaseous 1291 trapped in charcoal cartridges by ICP-MS/MS', J. Radioanal. Nucl. Chem., Mar. 2023, doi: 10.1007/s10967-023-08845-0.



NaOCl compatibility with CL resin



Solutions : 10mL of iodine 50µg/L 1mL of Cl resin Measurement by ICP-MS

- → NaOCI 1.8% not compatible with CI resin
- ➔ 0.54% improves the chemical yield

Extraction yield on AG®1-X8 with NaOCl 0.54%?

Elution of iodine from AG®1-X8 resin



Solutions : 10mL of iodine 50µg/L 0.54% NaOCI Measurement by ICP-MS

- 230mL : Extraction yield of iodine from AG[®]1-X8 resin > 90%
- Dilution by 3.33 factor (1.8/0.54) of 1.8% solution after elution ?

End-of-fixing volume of Cl resin?

End-of-fixing volume of Cl resin



Solutions : lodine 100µg/L 0.54% NaOCl Measurement by ICP-MS

50% iodine loss between 10mL and 50mL

- → Dilution not compatible with end-of-fixing volume
- → 1.8% NaOCl concentration is maintained to elute iodine from the AG[®]1-X8 resin

Improvement of percolation step?



Improvement of the purification yield



Adding a reducing agent After NaOCI elution : iodine is in I_3^- form Hydroxylamine hydrochloride (E°= -1.87V) I_3^-/I^- (0.54V) $I_3^- + 2 \text{ HONH}_3^+ \rightarrow N_2 + 3 I^- + 4 H^+ + 2 H_2O$

Purification yield from 20 to 75%
Selected concentration : 40g/L

Percolation medium in Cl resin retained : NaOCl 1.8% + H₃NO.HCl 40g/L



Developed protocol



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Validation of the new method



Spiked milk samples

Milk samples : comparaison of the old and the new method in milk collected in the environment

→ Excellent results



Conclusion

Elimination of liquid-liquid extraction with CCl_4 \rightarrow SPE on Cl resin with recovery yield > 80%

Conclusive work to make AG®1-X8 resin compatible with CI resin

- \rightarrow Elution in AG®1-X8 with 100 mL of NaOCl 1.8%
- \rightarrow Percolation on Cl resin with NaOCl 1.8% + H₃NO.HCl 40g/L

Improvement of the LOD with well-type detector : 6mBq/L

Accurate chemical yield by ¹²⁷I ICP-MS analysis

What's next?

¹²⁹I determination by ICP-MS/MS
→ Application of the protocol for milk :

C. Carrier et al., 'Determination of environmental gaseous ¹²⁹I trapped in charcoal cartridges by ICP-MS/MS', J. Radioanal. Nucl. Chem., Mar. 2023, doi: 10.1007/s10967-023-08845-0.

Work on freeze-drying to increase the sample size Iodine in water? Organically bound?

Better knowledge of ¹²⁹I level in environmental samples under and ouside nuclear site influence_

Next Step

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THANK YOU FOR YOUR ATTENTION

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