



Savannah River

Nuclear Solutions, LLC

A Fluor Daniel Partnership_{SM}

Rapid Method for Determination of Radiostrontium in Emergency Milk Samples

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Background

- **Increasing need to develop faster analytical methods for emergency response**
 - including rapid determination of radiostrontium in milk
- **Milk ingestion**
 - Important pathway for radiostrontium intake into the human body
 - Accumulates in bone
 - 100 Bq /L Sr-90 generic emergency action level (IAEA)
- **Need for rapid method**
 - Notification of dairy farms
 - Limits radiostrontium consumption



Need for faster methods

- Classical method (Harley) of drying, furnacing, separations tedious and time-consuming
- Cation exchange + extraction chromatography (Tait 1997)
 - bicyclic polyether cryptand such as cryptand 222
 - long contact times / 24 hour method
- Microwave small volume + Sr Resin (Tovedal 2008)
 - Measure Y-90
 - 24 hours
 - LLD 50 Bq/L
- Can we analyze in less than 24 hours?



New Method

- New rapid separation method for radiostrontium in emergency milk samples - SRS Environmental lab
- Flexible with sample aliquot
 - 100 ml aliquot yields 0.5 Bq/L LLD
 - 12 samples in 8 hours
 - Much less than 100 Bq/L Sr-90 emergency action level

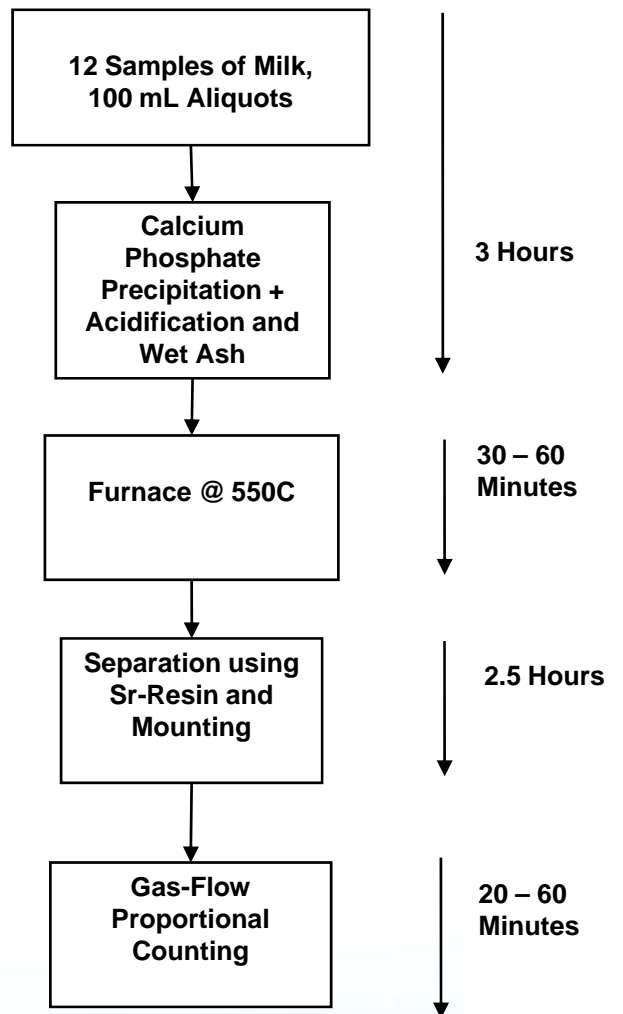
Maxwell III, SL and Culligan, B.K., "Rapid Method for Determination of Radiostrontium in Emergency Milk Samples", *Journal of Radioanalytical and Nuclear Chemistry*, in press



Method Details

- **Calcium phosphate precipitation**
- **Nitric acid dissolution of the precipitate**
 - coagulates residual fat/proteins
- **Strontium separation**
 - Sr Resin with vacuum-assisted flow rates
- **Gas proportional counting**
 - Could apply Cerenkov option for Sr-89/ Sr-90

FlowChart of New Radiostrontium Emergency Method



Sample Preparation

- 100 ml milk aliquot
- Add 50 ml water to facilitate separation
- Add 2 mL 1.25M $\text{Ca}(\text{NO}_3)_2$ and 5 mL $(\text{NH}_4)_2\text{HPO}_4$
- Add phenolphthalein indicator
- Add NH_4OH to dark pink
- Centrifuge 10 minutes



Sample Preparation

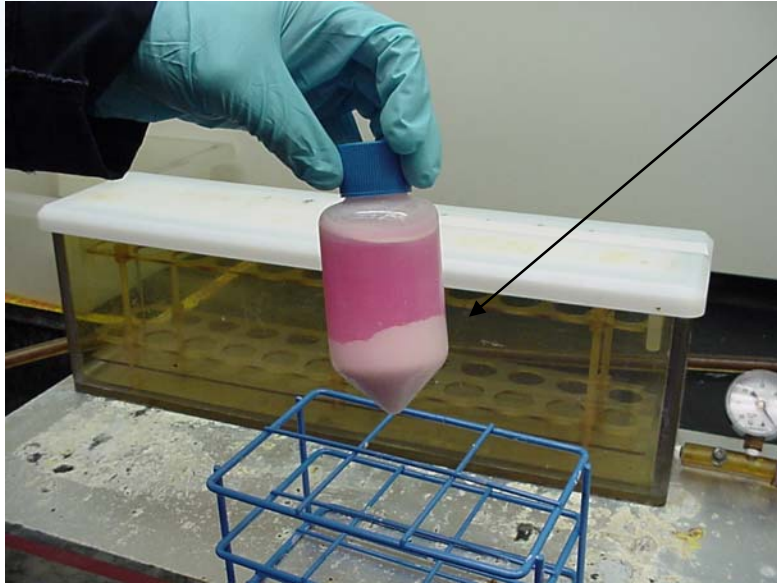
- Add 20 mL of 3M HNO₃ into each tube
- Ca₃(PO₄)₂ dissolves
- Fat/protein coagulate
- Centrifuge
- Transfer supernatant to beaker
- Rinse solids with 10-15 ml 3M HNO₃
- Transfer supernatant to beaker
- Evaporate beaker to dryness

Sample Preparation

- **Wet ash**
 - 15 ml concentrated HNO_3 and 5 ml 30 wt% H_2O_2
- **Heat beakers in a furnace**
 - at 550C for 30-60 minutes to turn the solids white
- **Wet ash**
 - 10-15 ml concentrated HNO_3 and 5 ml 30 wt% H_2O_2
- **Redissolve in 10 ml 8 M HNO_3 -1M $\text{Al}(\text{NO}_3)_3$**

Sample Preparation

500 ml sample ppt. shown



$\text{Ca}_3(\text{PO}_4)_2$ and fat/protein

Add 3M HNO_3

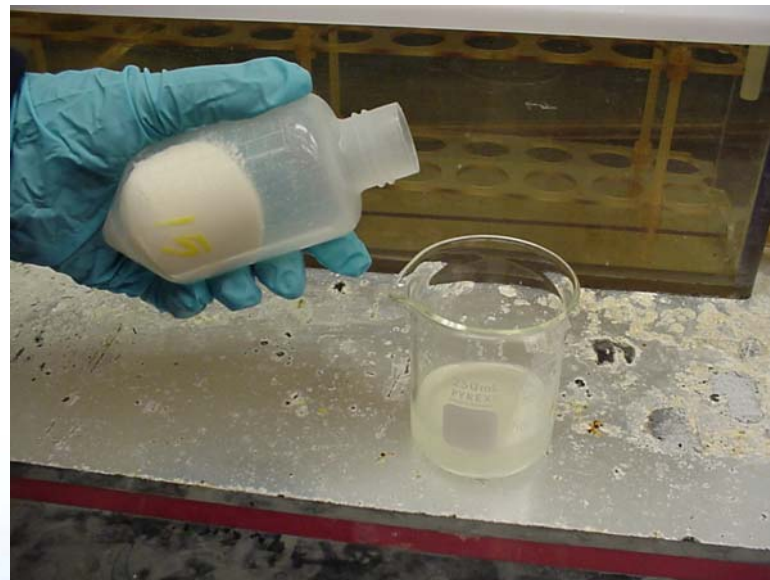




Sample Preparation



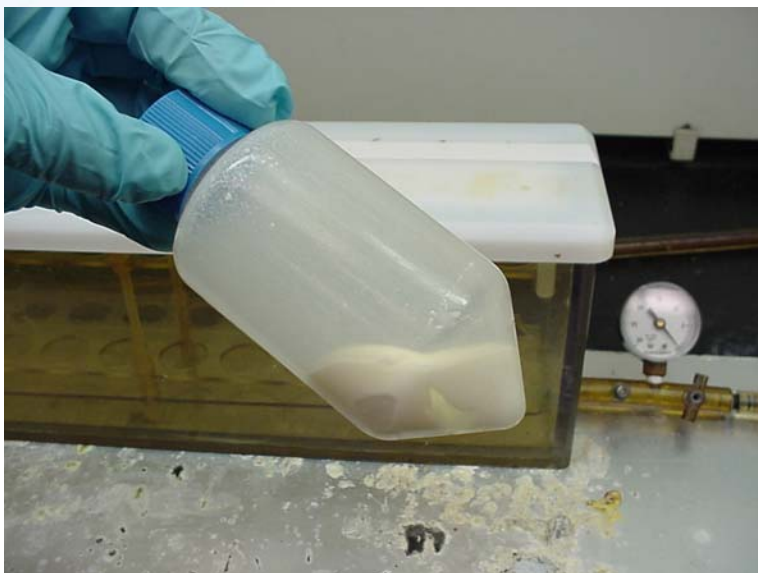
Centrifuge



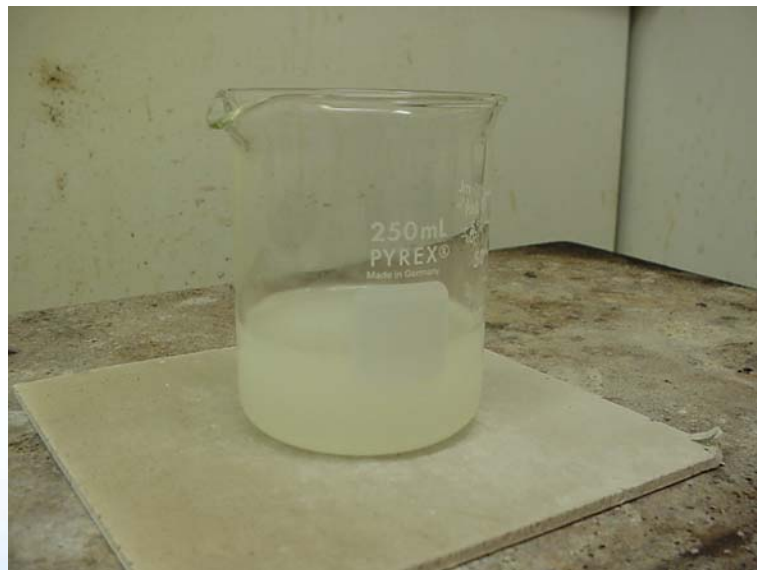
Most fat/protein is removed



Sample Preparation



Heat on hot plate



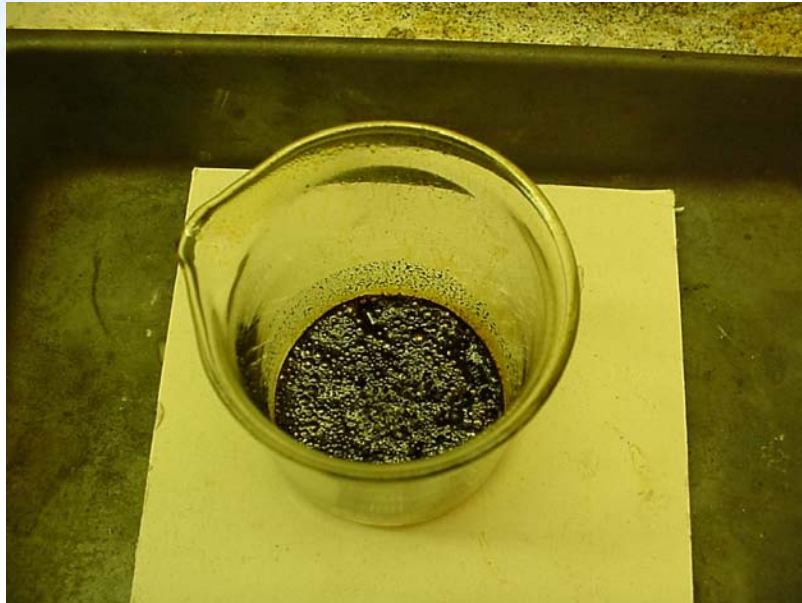
Sample Preparation



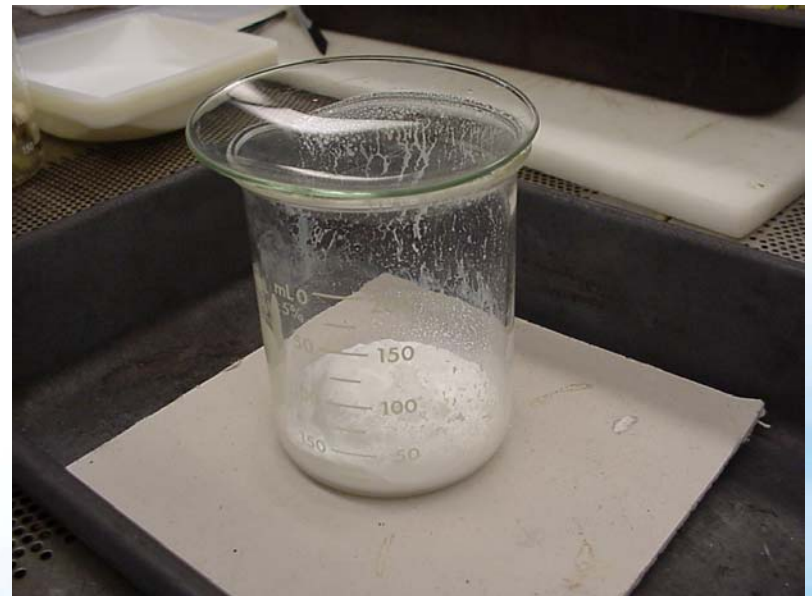
Heat to dryness



Sample Preparation



Heated at 550C 30 min.





Column Load Solution





Gravity Vs Vacuum Flow Rates



Sr-89/90 in Milk Column Extraction

- Redissolve in 10 ml 8M HNO₃-1M Al(NO₃)₃
- Perform typical Sr Resin Separation using 3 ml Sr resin
 - (2 ml +1 ml cartridges)
- Rinses:
 - 15 mL of 8M HNO₃
 - 5 ml 3M HNO₃-0.05M oxalic acid
 - 7 ml 8M HNO₃
- Sr Elution: 13 ml 0.05M HNO₃





Performance of New Radiostrontium Method - 20 minute Count

⁹⁰ Sr Added (Bq/L)	⁹⁰ Sr Measured (Bq/L)	Uncertainty (% , K=2)	Difference (%)
0	0.26	8.9	N/A
0	0.34	81.9	N/A
2.86*	2.66	24.1	-7.0
2.86*	3.96	24.7	+38
2.86*	3.31	20.2	+15.7
2.86*	2.67	18.7	-6.6
5.70	6.11	16.7	+7.2
5.70	5.71	13.1	+0.2
5.70	5.16	13.9	-9.5
14.3	12.8	9.1	-11
14.3	15.2	8.5	+6.3
14.3	14.1	8.6	-1.4
		Average	+3.19

* Added using NRIP water standard



Performance of New Radiostrontium Method - 60 minute Count

⁹⁰ Sr Added (Bq/L)	⁹⁰ Sr Measured (Bq/L)	Uncertainty (% , K=2)	Difference (%)
0	0.11	130	N/A
0	0.27	59	N/A
2.86*	3.09	13.2	+8.0
2.86*	3.11	16.7	+8.7
2.86*	2.67	13.6	-6.6
2.86*	2.67	11.3	-6.6
5.70	5.85	10.4	+2.6
5.70	5.75	8.3	+0.9
5.70	6.04	8.2	+5.9
14.3	13.6	6.1	-4.9
14.3	14.0	6.1	-2.1
14.3	14.2	6.1	-0.7

* Added using NRIP water standard

Average +0.52



Summary

- New rapid emergency method for Sr in milk developed at SRS
- Faster than cation exchange collection
 - 12 samples in 8 hours
- Calcium phosphate precipitation and nitric acid dissolution to coagulate proteins/fats
- LLD 0.5 Bq/L for 100 ml aliquot counted 20 minutes
- Can also be used for larger aliquot routine environmental monitoring