

A SYSTEMATIC INVESTIGATION ON THE EFFECTS OF THE MAJOR  
CHEMICAL AND CHEMICAL-PHYSICAL PARAMETERS ON Cs AND Sr  
DISTRIBUTION COEFFICIENTS IN A FRESH WATER SAMPLE.

I. Ciaccolini\*, G. Pampurini\*, G. Queirazza\*\*, F. Salghetti\*

\*CISE SpA, Segrate, Milan (Italy)

\*\*ENEL, Thermal and Nuclear Research Centre, Milan (Italy)

# INTRODUCTION

The aim of this work was at determining the distribution coefficients ( $K_d$ ,  $\text{cm}^3/\text{g}$ ) of Cs and Sr and the evaluation in a fresh water sample of the major chemical and chemical-physical parameters that effect them.

There are lots of  $K_d$ s in the literature which have been determined in different samples (sediments, soil, clay, silt, sand and synthetic materials), with different analytical procedures and method. Obviously, these results should not be indiscriminately used in radionuclide transport studies using mathematical models to predict the environmental impact of conventional and non-conventional discharges.

It is quite simple to think or imagine that many variables or parameters may change the  $K_d$  values so that it could be extremely useful to investigate and understand this phenomenon.

It is obvious that the best sample for the investigation is a real sample. At the same time the investigating procedure should not interfere with the natural equilibria involved.

Experiments were carried out on a fresh water sample gathered from river Po making use of scintillation counting as the analytical method.

# EXPERIMENTAL

Special care was taken in the collection, transportation and conservation of the water sample.

Because about twenty days were necessary for the lab-investigation, the stability of the sample was considered fundamental.

The sample, after the collection, was characterized from the chemical, mineralogical and granulometric viewpoints.

The stability, during the lab-activity was checked following granulometric distribution, some chemical parameters and the  $K_d$  values. The results were satisfactory so that all the  $K_d$ s can be considered reliable.

$^{137}\text{Cs}$  and  $^{85}\text{Sr}$  were employed as radioactive indicators added to the 250 ml homogeneous samples in a quantity less than the natural concentration ( $\approx 0.015$  ppb and 300 ppb respectively) just not to modify the natural equilibrium.

After addition the polyethylene capped bottles were positioned horizontally on a mechanical shaker in controlled temperature. Shaking frequency was 70±90 cycle/minute.

On the basis of preliminary kinetic investigations the time necessary for reaching the equilibrium conditions was 3h and 30' for Cs and 8h for Sr.

The analytical determination was made with the scintillation counting technique in the soluble phase, obtained through a 0.45  $\mu$ m Millipore filter.

All the procedure mentioned above was set up previously and when it was ready we made the research reported in this paper so that it will be easy to repeat the experiments and to compare the results.

The major parameters affecting Kd values and the related interactions between them was found out in a preliminary work in which we employed a factorial ANOVA (analysis of variance) model.

Kd values were determined in triplicate by batch procedure in polyethylene sealed bottles in controlled temperature.

The distribution coefficient was calculated from:

$$K_d = \frac{\frac{A_o - A_f}{P}}{A_f - B} \cdot 10^6$$

where  $A_o$  is the initial activity,  $A_f$  is the final activity in the water phase ( $< 0.45 \mu$ m), P is the particulate load (mg/l), and B is the blank activity.

The parameters investigated were:

- for Cs: carrier - on the untreated sample and on the separated granulometric fractions - temperature, pH and interaction pH-carrier, particulate load, granulometric fractions, TOC, clay concentration, UV radiations, freezing.
- for Sr: effect of pH, temperature, carrier and related interactions, granulometric fractions, TOC, clay concentration, UV radiations.

## RESULTS

The results of Kd were elaborated with ANOVA in order to find out the main effects and their interactions.

For Cs, effects: particulate, carrier, temperature and pH;  
interactions: pH-carrier and temperature-carrier.

For Sr, effects: particulate, temperature, carrier and pH;  
interactions: pH-concentration.

The relative residual variance, interpreted as an estimate of the general process error, is about 10%.

For Cs, Fig. 1 shows some typical dependence of Kd on the main factors: pH, temperature,  $Q_T$  (total amount of Cs in nano equivalents/l), P (particulate load mg/l), and in the following table some analytical dependence functions are shown.

TABLE

For Cs	For Sr
$Kd = 1.15 \cdot 10^4 \cdot Q_T^{-0.408}$	$Kd = 4.78 \cdot 10^4 \cdot P^{-0.89}$
$Kd = 2.45 \cdot 10^4 \cdot P^{-0.230}$	$Kd = 6.59 \cdot 10^3 \cdot d_m^{-1.076}$
where $Q_T$ = total concentration of the element in nano equivalents/l $P$ = particulate load in mg/l $d_m$ = mean diameter of particulate fractions in $\mu m$ .	

The other results will be reported in the final paper. Also kinetic results are available for Cs absorption obtained with a proper equipment set up for the purpose.

## COMMENTS

Considering the results obtained, the procedure adopted is recommended. At the same time as it is not complicated, it could be improved and standardized if the need for reliable Kds is considered necessary. Moreover, other interesting elements could be investigated.

Such activity is under way in our laboratories.

At the same time, because of the fact that a great quantity of Kds are available, coming from different samples and analytical procedures, we distinguish them as "natural", "isotopic", "mass" and "from leaching". This distinction is necessary for understanding the phenomena and for using Kd more correctly.

Fig. 1

