Appeared as: Smith, J.T., Sasina, N.V., Kryshev, A.I., Belova, N.V., Kudelsky, A.V. (2009,). A review and blind test of predictive models for the bioaccumulation of radiostrontium in fish. *Journal of Environmental Radioactivity*, 100, 950-954.

A review and test of predictive models for the bioaccumulation of radiostrontium in fish.

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Keywords: Radiostrontium, ^{90}Sr; ^{89}Sr; fish, bioaccumulation, concentration factor, model, size effect.

Abstract

Empirical relations between the ^{90}Sr concentration factor (*CF*) and the calcium concentration in freshwater aquatic systems have previously been determined in studies based on data obtained prior to the Chernobyl accident. The purpose of the present research is to review and compare these models, and to test them against a database of post-Chernobyl measurements from rivers and lakes in Ukraine, Russia, Belarus and Finland. It was found that two independently developed models, based on pre-Chernobyl empirical data, are in close agreement with each other, and with empirical data. Testing of both models against new data obtained after the Chernobyl accident confirms the models' predictive ability. An investigation of the influence of fish size on ^{90}Sr accumulation showed no significant relationship, though the data set was somewhat limited.

1. Introduction

For many radionuclides, only single "best estimate" fish-water concentration factors (*CF*s) are available for dose assessment models (e.g. IAEA, 1994). For some radionuclides, however, estimates of the *CF* may be improved using empirical models which account for different ambient concentrations of their stable isotope or stable element analogue (e.g., for radiocaesium, Blaylock, 1982; Smith et al., 2000) The fish-water concentration factor (*CF*) of ^{90}Sr has been shown to vary as an inverse function of the concentration of calcium [*Ca*] (a stable analogue of radiostrontium) in the surrounding water (Vanderploeg et al., 1975; Blaylock, 1982; Kryshev, 2003, 2006). The application of the concentration factor approach usually implies that the uptake of the radioisotope to a fish population has reached equilibrium. Due to the changing ^{90}Sr activity concentration in water (e.g. Cross et al., 2002) and the deposit of ^{90}Sr in bone, this dynamic accumulation process is to an extent dependent on fish lifespan, but equilibrium is generally considered to have been achieved 8-12 years after radioisotope fallout (Kryshev, 2003). For estimating of ^{90}Sr accumulation in fish under non-equilibrium conditions, a number of dynamic models are available, (Sazykina, 2000; Kryshev and Ryabov, 2000, Kryshev, 2003, 2006; Smith et. al., 2005a) though all of these dynamic approaches use estimates of the equilibrium *CF* as one of their input parameters.

The processes which determine the accumulation of radioisotopes in fish are dependent on environmental and biological factors such as water chemistry, trophic level of fish species (predatory or non-predatory fish), fish type and size. ^{90}Sr can be accumulated through gills of fish from water (Chowdhury and Blust, 2001) and through the food pathway (Kryshev, 2003). Under low concentrations of the isotope in water, the food pathway is believed to be the more important of the two uptake routes (Michalusev et al, 1997).

Due to its similar bioaccumulation to calcium, approximately 95% of ^{90}Sr is found in the bony parts of fish (skeleton, fins, skin) and only 5% in the soft tissues or muscles of a fish (Vanderploeg et al., 1975; Blaylock, 1982). Smith et al. (2005a) assumed that an average of 80% of the wet weight of the fish is composed of soft tissue whilst 20% of the wet weight is bony parts. Another estimation (Shekhanova, 1983) gives an average of 77% of wet weight as soft tissues and 23% as bones. Such differences can influence the accuracy of *CF* calculations where data is presented as separate measurements of ^{90}Sr concentration in soft tissues and/or bones. This issue is considered further below.

The “size effect” of radioisotope accumulation in fish can result in an increasing activity concentration (per unit weight of fish) with increasing fish size (Elliott et al., 1992; Koulikov and Ryabov, 1992; Kryshev and Ryabov, 2000). For radiocaesium, a “size effect” was observed in predatory fish such as perch and pike but no clear dependence was observed for non-predatory fish (roach) (IAEA, 2000; Smith, 2005b). There is, however, less available information on the size effect for radiostrontium: where possible, the database developed in this research will be used to address this question.

Previous modelling approaches

Two models (Vanderploeg et al., 1975; Kryshev, 2006), based on different empirical data sets have been developed relating the ^{90}Sr *CF* (in l kg^{-1}, fresh weight) to the water calcium content [*Ca*] (mg l^{-1}). The inter-comparison and testing (against new empirical data) of these independently-developed models, carried out here, represents a strong test of both models.

Based on 34 measurements of fish bone-water *CF* and 19 measurements of fish muscle-water *CF*, Vanderploeg et al. (1975) (also presented in IAEA, 1994) determined the following relations to estimate the *CF* of fish (numbers in brackets show uncertainty range):

$$CF(Muscle) = \frac{181\ (59 - 540)}{[Ca]^{1.2(0.8-1.6)}} \qquad (1)$$

$$CF(Bone) = \frac{16317}{[Ca]^{1.2(0.8-1.6)}} \qquad (2)$$

Assuming that 23% of wet weight of a fish is composed of bony parts (Shekhanova, 1983), then the whole fish *CF* is given by:

$$\mathrm{CF(Whole\ Fish)} = \frac{3850}{[\mathrm{Ca}]^{1.2}} \qquad (3)$$

Using a similar linear regression approach Kryshev (2006) analysed 115 values of the *CF* at different environmental concentrations of calcium [Ca^{2+}], to obtain the following relationship for whole fish:

$$CF(Whole\ Fish) = \frac{3940(1770 - 6110)}{[Ca]} \quad (4)$$

where numbers in brackets show the uncertainty range. An assessment was made (Kryshev, 2006) for predatory and non-predatory fish separately and different parameter values were obtained for the two types. The average parameter value was found to be 40 % higher for non-predatory fish than for predatory species, although the confidence intervals overlapped.

The two models for whole fish *CF* are very similar: the main difference being in the slope of the inverse power law relationship.

2. Methods

Modelling

As in the previous studies, we will model the fish-water *CF* of radiostrontium as an inverse function of the calcium concentration of the water body (e.g. Blaylock, 1982):

$$CF = A_1[Ca]^{-B} \quad (5)$$

where A_1 and B are parameters to be determined empirically. This model is here called Model 1 and the simpler special case of Eq (5) in which B equals 1:

$$CF = A_2/[Ca] \quad (6)$$

will be called Model 2.

Equations 5 and 6 were fitted to the empirical data using the SAS statistical analysis package (SAS 2002). Prior to fitting, *CF* and [*Ca*] data were log-transformed (to give a distribution closer to the normal distribution, and to linearise the relationships) and results back-transformed for presentation. The SAS software (SAS, 2002) gives as output the best-fit model parameters and estimates of 95% confidence interval in those parameter values.

Use of previously developed databases

The measurements used by Vanderploeg et al. (1975) consisted of 34 measurements of fish bone-water *CF* and 19 measurements of fish muscle-water, together with measurements of [*Ca*]. The later study of Kryshev (2006) consisted of 115 measurements of *CF* in whole fish, but because of the risk of overlap between data sets, 16 measurements obtained by Kryshev (2006) were not used, reducing the data set to 99 measurements. Obviously, all of the Vanderploeg et al. (1975) measurements were pre-Chernobyl and the Kryshev (2006) study used only two data points from freshwater systems contaminated by the Chernobyl accident. For re-analysis, in order to separate the pre- and post-Chernobyl data, these two data points were removed from the Kryshev (2006) data set, leading to a total of 97 data points.

Post-Chernobyl CF database for ^{90}Sr

Post-Chernobyl datasets, from the period 1994-2004, were collected from a literature review (Table 1). They contain observations of ^{90}Sr activity concentration in various species of whole fish as well as soft tissues and bones but sometimes only separate measurements in muscle or bony tissue were available. In this case, following Kryshev (2006), whole fish activity concentrations were estimated assuming that 77 % of the weight is soft tissues and 23% in bony tissue. The dataset includes both predatory (pike *(Esox lucius)*, perch (*Perca fluviatilis*), pike-perch (*Sander lucioperca*), cat-fish (*Ictalurus punctatus*)) and non-predatory species (roach (*Rutilus rutilus*), tench (*Tinca tinca*), bream (*Abramis brama*), carp (*Cyprinus carpio*), goldfish (*Carassius auratus gibelio*), ruffe (*Gymnocephalus cernuus*)).

The post-Chernobyl datasets were used as predictive tests of the previously developed models. Due to the time required for equilibration of the ^{90}Sr uptake process, only post-1994 data were used to test the *CF* models.

3. Results and discussion

Comparison of models for ^{90}Sr CF in fish

Both of the models (Model 1 and its special case, Model 2) were fitted to the available data of *CF* vs. [Ca] and estimated parameter values are shown in Table 2. In interpreting these parameter values, note that the value of the *A* parameters depends on the endpoint measured (bone, muscle or tissue), but the value of the *B* parameter is expected to be independent of endpoint measured. Model fits to the empirical data are shown in Figure 1.

Both models explained a large proportion of the variation in *CF* values (R^2 values were from 65 – 89%). There is some evidence in Table 2 for an inverse power law relationship of slope ("*B"* in Eq. 5) greater than 1, since the analysis of the whole data set estimated *B* to be 1.11 with confidence intervals in the range 1.02 – 1.20. However, the R^2 values of the simpler inverse model (Model 2: $B = 1$) are very close to those of Model 1, so the model which allows "*B*" to be varied offers no major improvement over the simple inverse relationship of Model 2.

Model testing

The two models (Vanderploeg et al., 1975; Kryshev, 2006) were used to predict the whole fish - water *CF* and muscle-water *CF* of ^{90}Sr in rivers and lakes impacted by the Chernobyl accident. As shown in Figure 2 (a) and (b) both models generally performed well, showing good agreement with the empirical data. In two out of 12 cases (Braginka River – whole fish; Sozh River – fish muscle), the model predictions were significantly outside the error bars (± 2 S.D.) in the empirical data. This may in part have been due to poor estimation of the uncertainty in empirical data since there were in some cases relatively few measurements. In addition, it was not possible, with the available data, to estimate the uncertainty in measurements of the water ^{90}Sr activity concentration.

It can be seen (Figure 2) that the low calcium Lake Saamia in Finland has significantly higher bioaccumulation of ^{90}Sr than the significantly higher calcium waterbodies in Belarus, Russia and Ukraine.

Ratio of ^{90}Sr in bone:muscle tissue

The re-analysis of the Vanderploeg et al. (1975) data set gives a best-estimate ratio of 73.3 (bone÷muscle activity concentration). This was calculated from the ratio of A_2 values (bone÷muscle) in Table 2. A previous study by Saxén and Koskelainen (2002) measured a significantly higher bone-muscle ratio of 248 (±59; 1 S.D.). In this latter study, all bones, large and small were separated very carefully from the muscle. The lower ratio of Vanderploeg et al. (1975) may be due to inclusion of small bones, skin and/or fins in the "muscle" sample. The higher ratio observed by Saxén and Koskelainen (2002) is likely to be more accurate, but for practical purposes of radiation protection, this lower ratio (i.e. higher predicted activity concentration in "muscle") may better reflect the "edible" parts of the fish which are typically consumed.

"Size effect" on ^{90}Sr accumulation by fish

The influence of fish size on ^{90}Sr accumulation in fish has been studied for the Pripyat and Sozh Rivers and for the Chernobyl Cooling Pond. ^{90}Sr activity concentrations in whole fish were plotted as a function of wet weight of fish for each of these systems (Figure 3). Contrary to observations for radiocaesium (Hadderingh et al., 1997; Smith et al., 2002), there was no evidence of a clear "size effect" of increasing ^{90}Sr activity concentration with increasing fish weight. None of the relationships observed in Figure 3 showed a statistically significant correlation between ^{90}Sr activity concentration and fish weight. It should be noted, however, that the sample sizes were not large, so the ability of the data set to test for a weak size effect relationship is limited. Further, since the measurements we have analysed were made some years after the Chernobyl accident, any effects of differential uptake rates in small and large fish may have been missed. Rapidly changing water activity concentrations in the months and years after the accident could have led at that time to different observed *CF* values in different fish sizes if, for example, equilibrium was more rapidly achieved in small fish than large.

3. Conclusions

1. The previously determined empirical models between *CF* for ^{90}Sr accumulation in fish and [Ca^{2+}] (mg l^{-1}) in the water are shown to be in good agreement with new measurements for the water bodies affected by the Chernobyl contamination;
2. On the basis of the available data, no significant relationships between fish size and ^{90}Sr activity concentration in fish (the "size effect") were determined;
3. Remaining variation in *CF* not explained by an inverse relationship with [Ca^{2+}] is significant, but is likely to be due to a number of factors such as fish feeding behaviour, recruitment and population age which may be difficult to predict using general (as opposed to lake- or river- specific) models.

Acknowledgments

Nataliya Sasina would like to acknowledge the support of INTAS – 2005 Young Scientist Fellowship (Ref. number 05-109-4221). The study was also supported in part by project RFBR no. 07-04-00002.

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Figure captions

Figure 1. Fit of Model 2 to (a) Vanderploeg et al. (1975) data; (b) Kryshev (2006) data; (c) combined data set.

Figure 2. Test of Vanderploeg et al. (1975) and Kryshev (2006) models against post-Chernobyl data for (a) whole fish (Vanderploeg model: $CF=3850\times[Ca]^{-1.2}$; Kryshev model: $CF=3610\times[Ca]^{-1}$) and; (b) fish muscle (Vanderploeg model: $CF= 181\times[Ca]^{-1.2}$.

Figure 3. Relationships between ^{90}Sr concentration factor and fish weight for various predatory and non-predatory species in (a) Pripyat River (Choiniki); (b) Sozh River (Gomel); (c) Chernobyl Cooling Pond.

Table 1. Measurements of ^{90}Sr concentration factors in whole fish after Chernobyl

Water body	Number of observations	Sampling date	Calcium concentration in water, mg l^{-1}	*CF* Muscle	*CF* Whole fish	References
River Dnieper (Bragin)	4	1994-95	40.3	-	36.3	(1)
River Sozh (Gomel)	17	1994-95	48.9	2.59	159	(1)
River Braginka (Bragin)	12	1994-95	55.7	5.11	7.96	(1)
Lake Perstok	6	2002	26.45	-	250	(2)
Lake Kozhanovskoe	*	1994	33	-	173	(3)
Chernobyl Cooling Pond	27	2002-04	50.5	2.88	38.6	(3)
Lake Glubokoye	10	2003-04	27.2	5.52	202	(3)
Lake Saimaa, Finland	4	1994	3	-	455	(4)

1. Michalusev et al., 1997; 2. Smith et al., 2005b; 3. Belova N.V., Severtsov Institute, Moscow, unpubl. res.; 4. Saxén R., Koskelainen U., 2001. * This given as a mean value, but the no. of observations is not known.

Table 2. Parameter values determined by fitting Models 1 and 2 to the data sets of Vanderploeg et al. (1975) and Kryshev (2006).

Data set	No. of obs.	Model 1 CF = $A_1[Ca]^{-B}$			Model 2 CF = $A_2[Ca]^{-1}$	
		A_1	B	R^2	A_2	R^2
Vanderploeg (bone)	35	13430 (8913 – 20000)	1.12 (1.26 – 0.99)	89%	9750 (8110 – 11700)	88%
Vanderploeg (muscle)	19	204 (57.1 – 727)	1.16 (1.59 – 0.73)	66%	133 (77.6 – 231)	65%
Kryshev (whole fish)	97	6412 (4450 – 9238)	1.18 (1.29 – 1.07)	82%	3610 (3231 – 4034)	80%
All data (whole fish)	132	4511 (3385 – 6013)	1.11 (1.20 – 1.02)	82%	3224 (2923 – 3557)	81%

Table 3. Test for relationships between ^{90}Sr activity concentration (whole fish, f.w.) and fish weight. None of the correlations was statistically significant.

Water body (period of study)	Fish Type	R^2 value
River Pripyat (1994-5)	Predatory	0.016 (n=13)
River Sozh (1994-5)	Predatory	0.042 (n = 7)
Chernobyl Cooling Pond (2002-4)	Predatory	0.24 (n = 8)
River Pripyat (1994-5)	Non-Predatory	0.094 (n = 11)
River Sozh (1994-5)	Non-Predatory	0.121 (n = 10)
Chernobyl Cooling Pond (2002-4)	Non-Predatory	0.026 (n = 19)

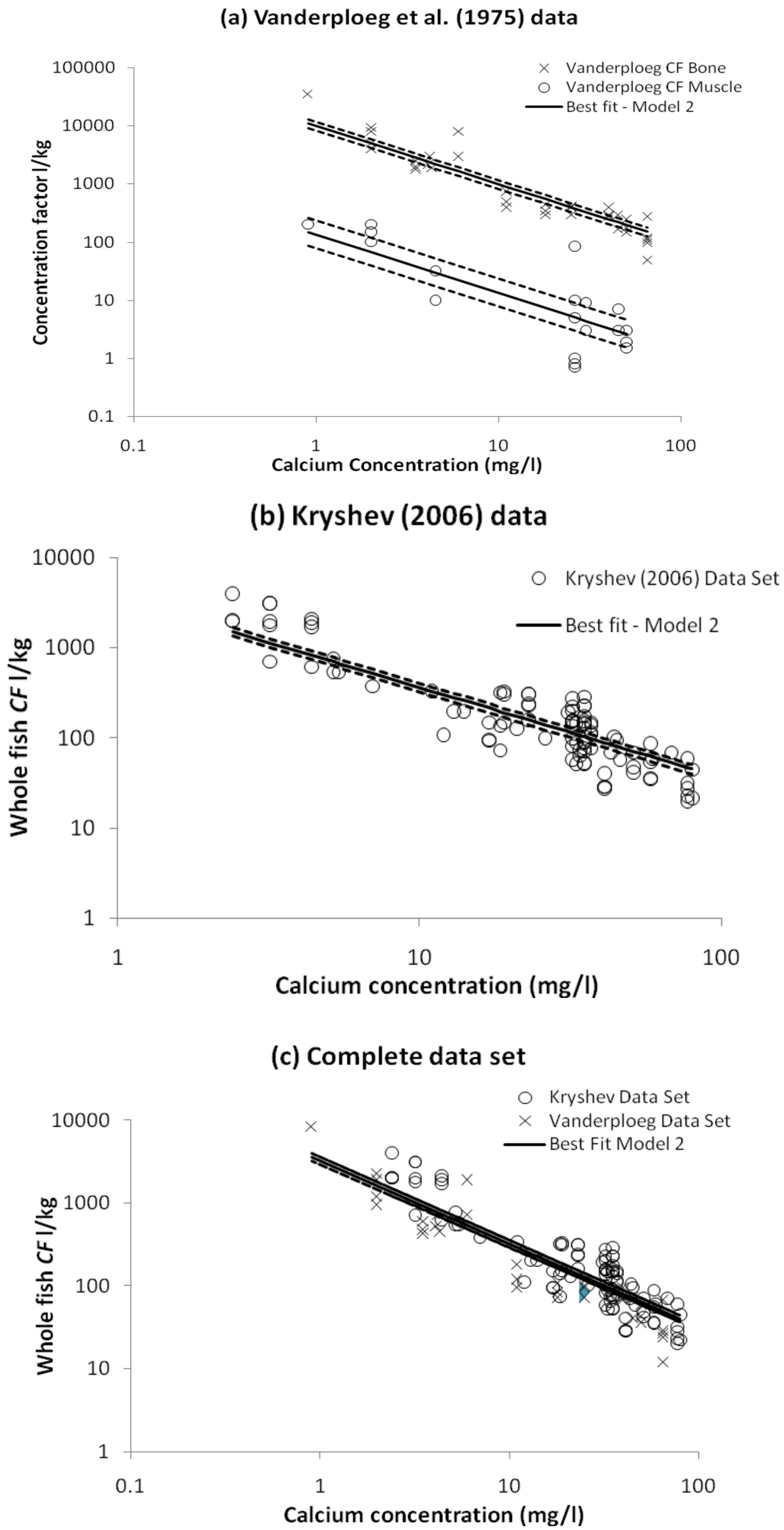

(a) Vanderploeg et al. (1975) data
Vanderploeg CF Bone
Vanderploeg CF Muscle
Best fit - Model 2
Concentration factor l/kg
100000
10000
1000
100
10
1
0.1
0.1
1
10
100
Calcium Concentration (mg/l)
(b) Kryshev (2006) data
Kryshev (2006) Data Set
Best fit - Model 2
Whole fish CF l/kg
10000
1000
100
10
1
1
10
100
Calcium concentration (mg/l)
(c) Complete data set
Kryshev Data Set
Vanderploeg Data Set
Best Fit Model 2
Whole fish CF l/kg
10000
1000
100
10
1
0.1
1
10
100
Calcium concentration (mg/l)

Figure 1

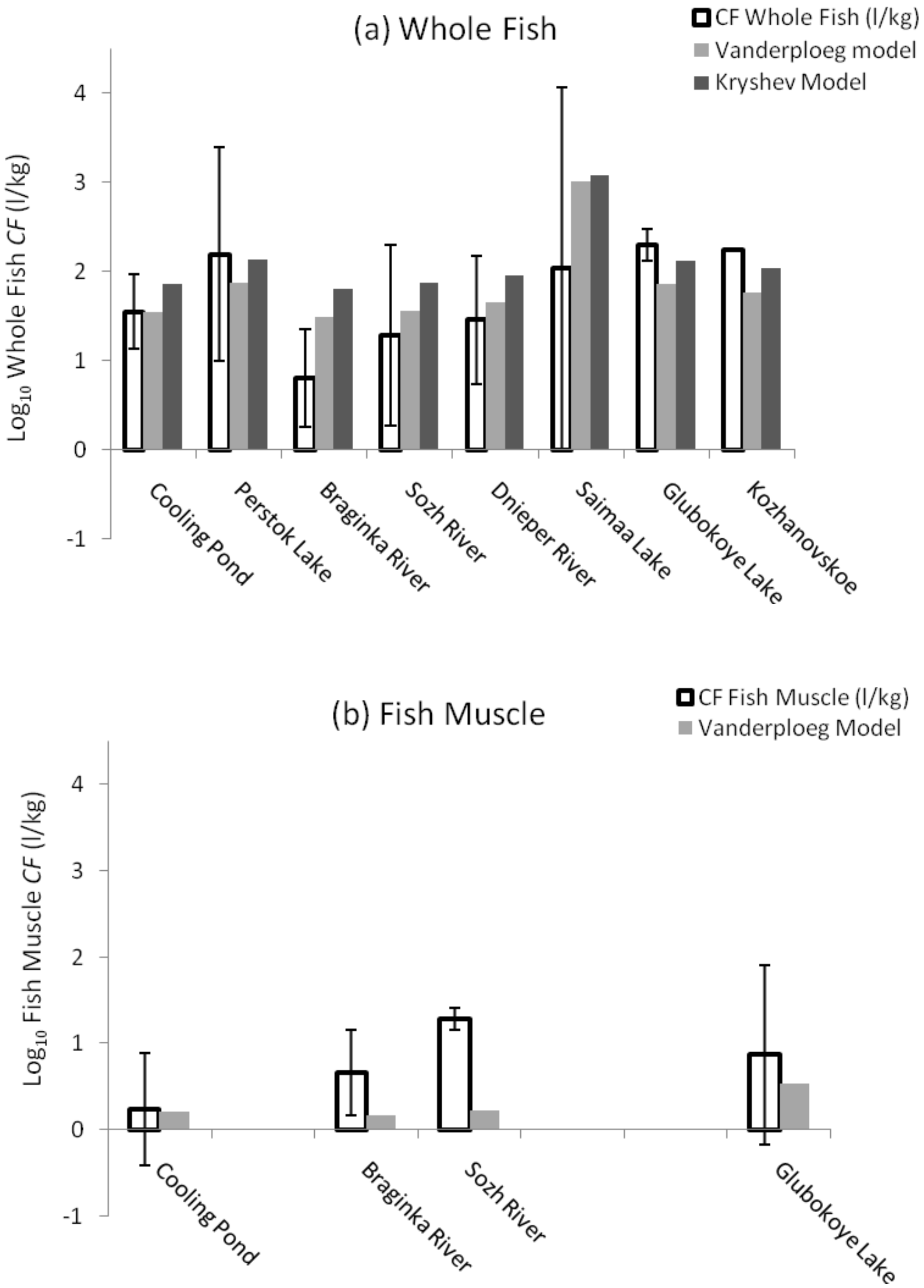
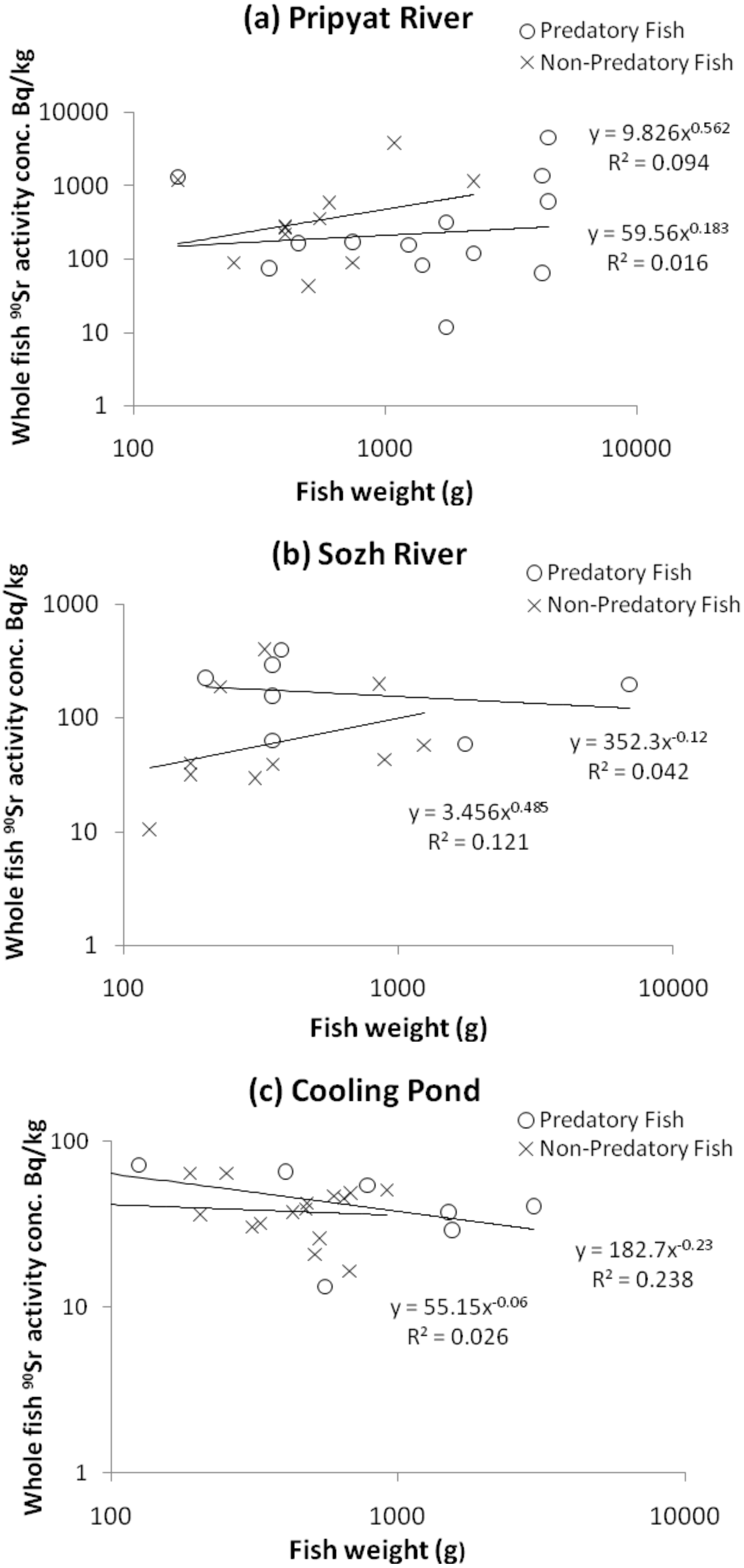

(a) Whole Fish
CF Whole Fish (l/kg)
Vanderploeg model
Kryshev Model
Log10 Whole Fish CF (l/kg)
4
3
2
1
0
-1
Cooling Pond
Perstok Lake
Braginka River
Sozh River
Dnieper River
Saimaa Lake
Glubokoye Lake
Kozhanovskoe
(b) Fish Muscle
CF Fish Muscle (l/kg)
Vanderploeg Model
Log10 Fish Muscle CF (l/kg)
4
3
2
1
0
-1
Cooling Pond
Braginka River
Sozh River
Glubokoye Lake

Figure 2



(a) Pripyat River
Predatory Fish
Non-Predatory Fish
Whole fish 90Sr activity conc. Bq/kg
10000
1000
100
10
1
100
1000
10000
Fish weight (g)
y = 9.826x0.562
R² = 0.094
y = 59.56x0.183
R² = 0.016
(b) Sozh River
Predatory Fish
Non-Predatory Fish
Whole fish 90Sr activity conc. Bq/kg
1000
100
10
1
100
1000
10000
Fish weight (g)
y = 352.3x-0.12
R² = 0.042
y = 3.456x0.485
R² = 0.121
(c) Cooling Pond
Predatory Fish
Non-Predatory Fish
Whole fish 90Sr activity conc. Bq/kg
100
10
1
100
1000
10000
Fish weight (g)
y = 182.7x-0.23
R² = 0.238
y = 55.15x-0.06
R² = 0.026

Figure 3