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Tritium in fish from remote lakes in northwestern Ontario, Canada

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ABSTRACT

Tritium is most commonly generated as a by-product of nuclear reactors. As such, environmental concentrations are typically only reported near regions of interest, and background concentrations in areas unaffected by anthropogenic disturbance are not well characterized. To provide information on background levels of tritium in the natural environment, tissue-free water tritium (TFWT) and organically-bound tritium (OBT) were measured in the flesh of 106 fish collected within three lakes located at the IISD-Experimental Lakes Area (ELA) in Ontario, Canada in 2014. For the three ELA lakes studied, water tritium (HTO) activity concentration was determined to be below reliably detectable levels (0.6 Bq/L). Fish TFWT was found to be below 0.7 Bq/L, similar to the surrounding water tritium activity concentration. Fish OBT activity concentrations, at below 5 Bq/L, were also very low. Fish size was significantly related to OBT activity in Lake Whitefish and White Sucker from Lake 302, but not in other lakes. Though we observed significant differences in potential tritium exposure to humans among lakes, the levels of tritium reported here are below the Canadian natural background radiation of 1.8 mSv/y. These results provide information on background levels of tritium in freshwater fishes in Canada.

1. Introduction

Tritium is produced naturally from interactions of cosmic rays with gases in the upper atmosphere and, to a much lesser extent, by processes taking place within the earth crust. Anthropogenic sources include the production of tritium as a by-product of nuclear reactors. Because tritium is routinely released from Canada Deuterium Uranium (CANDU) reactors, tritium studies have been focused in regions close to nuclear reactors (Le Goff et al., 2016; Kořinková et al., 2016; Thompson et al., 2015; Fiévet et al., 2013).

Tritium can pose a health risk only if ingested through drinking water or food, inhaled, or absorbed through the skin. Tritium decays to helium, and in doing so, releases low energy beta radiation. Knowledge of levels of tritium in the environment is used in the assessment of population exposure to environmental tritium (Boyer et al., 2009; Fiévet et al., 2013; Melintescu and Galeriu, 2011).

Currently, baseline fresh water biota tritium data in Ontario (Canada) is very scarce as most measurements have been obtained from samples collected near nuclear facilities. Areas that are unaffected by nuclear sites can provide data on background tritium levels, but are data deficient. These background levels likely originate from nuclear weapons testing and from cosmogenic production (Kim et al., 2016). The IISD-Experimental Lakes Area (ELA) is an internationally unique research station encompassing 58 remote freshwater lakes with highly controlled access, removed from anthropogenic disturbance. The ELA is approximately 1000 km away from the closest nuclear facilities in Ontario, and over 500 km from the closest US facilities in Minnesota. As such, the analysis of biota and water samples collected from the ELA provides both opportunities to report background levels of tritium in the absence of significant nuclear operations and long term effects in fish after tritium tracer experiments. By providing a better description of background levels in the natural environment and long term effects in fish, the results of this study also contribute to a more precise understanding of the tritium footprint of nuclear facilities in Canada and elsewhere.

As part of a collaboration between Canadian Nuclear Laboratories (CNL), Health Canada and the ELA that aimed to study background tritium levels in Canadian foods, a total of 106 fish samples were obtained from three ELA lakes during the summer of 2014. Fish tissue free water tritium (TFWT) and organically bound tritium (OBT) were determined on each of the fish sampled. The TFWT activity concentration in fish reflects tritium activity concentration in water that occurred a few hours or days before sampling. In contrast, OBT activity concentration in fish can reflect environmental tritium levels accumulated

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Table 1
Inventory of ELA fish received by CNL low background laboratory.

Lake	Species	Number	Male (M), Female (F) or Unknown (U)
305	Northern Pike	11	M (8) and F (3)
	Lake Whitefish	18	M (6) and F (12)
302	White Sucker	17	M (0), F (16) and U (1)
	Lake Whitefish	30	M (18), F (10) and U (2)
226	Lake Whitefish	30	M (15), F (14) and U (1)
Total	-	**106**	-

M = males, F = females, U = gender unknown.

over months to years prior to sampling. Species of interest were Northern Pike (*Esox lucius*), Lake Whitefish (*Coregonus clupeaformis*) and White Sucker (*Catostomus commersonii*). Northern Pike and Lake Whitefish are the focus of both recreational and commercial fisheries (DFO, 2010; Kinnunen, 2003), and White Sucker are a species of importance in aboriginal culture (Cooke and Murchie, 2013). Tritium in water (as HTO) was also measured in the same three lakes, to provide context for reported fish concentrations.

2. Material and methods

Fish were collected from Lake 305 (Northern Pike and Lake Whitefish), Lake 302 (White Sucker and Lake Whitefish) and Lake 226 (Lake Whitefish) at the ELA using 3–4.5" mesh gillnets used for Lake 302 and Lake 226 and multimesh experimental gillnets used for Lake 305 during summer (July 4 - August 5) of 2014 (Chen et al., 2015a). Fish were sampled for length and weight, and sex was determined via dissection. Where possible, fillets of at least 100 g were taken from each fish. Fillets were dried and homogenized (Table 1; Chen et al., 2016) and delivered to the CNL Low Background Laboratory.

2.1. HTO measurements in lake water

Nine lake water samples (3 for each of the lakes) were sampled under ice in March of 2015. Tritium activity concentrations were measured following ISO 9698 (2010) using an ALOKA (LSC LB-7, Hitachi Aloka Medical Ltd., Japan) liquid scintillation counter (LSC). Prior to being placed in the counter, 70 mL of water sample was mixed with Ultima Gold LLT (PerkinElmer) cocktail using a 1:1 ratio. The samples were prepared for counting in a 145 mL low-diffusion polyethylene vial using the ALOKA system (Zinsser Analytic Ltd. Germany). The counting efficiency was determined by using a tritium standard (NIST, USA). Each water sample was counted for 400 min and the minimum detectable activity (MDA) achieved was less than 0.6 Bq/L. Although most LSC use a 20 mL vial to hold 10 mL of water sample, we used a larger water sample in order to achieve the lower detection limit used in our study.

2.2. Fish TFWT measurements

Fish samples were frozen and kept at −20 °C until analyzed. The TFWT was extracted using a laboratory designed freeze-drying system (Kim and Roche, 2013). The frozen fish flesh samples (approximately 100 g) were loaded into a vacuum flask and the extracted tissue-free water was collected under vacuum over 48 h. The extracted water was collected on a liquid nitrogen trap. For each sample, the collected water activity concentration was determined by mixing 50 mL of extracted water from fish tissues with 50 mL of Ultima Gold LLT. Each sample was counted for 400 min using the ALOKA LB-7 LSC. The MDA was approximately 0.7 Bq/L and the overall uncertainty of the measurements was estimated to be less than 10%. The TFWT per g fresh fish tissue was estimated as the measured Bq/L in extracted water multiplied by the moisture content of the fish. Where the measured activity was below detection (0.7 Bq/L), the detection limit of 0.7 Bq/L was used in TWFT calculations.

2.3. Fish OBT measurements

The freeze-dried fish tissue remaining after the extraction of the TFWT measurements was dried at 55 °C for 24 h. The completely dried samples were ground using a small mixer and combusted using a Parr apparatus (model 1121, USA) with pressurized oxygen (Kim and Roche, 2013). The combustion water was then distilled and diluted to 10 mL with tritium-free water, and mixed with 10 mL of Ultima Gold LLT. The OBT activity concentration of the samples was determined using a Quantulus LSC. The counting time was 360 min. In this study, as almost 10 mL of combustion water was collected for each fish sample, the MDA achieved was about 2.5 Bq/L. The OBT value is considered here as total OBT activity (Exchangeable OBT + Non-exchangeable OBT). The overall uncertainty of the OBT measurement was estimated to be less than 25%. The OBT of fish in fresh tissues was estimated by multiplying the measured Bq/L by the ratio of dry mass to fresh mass analyzed and a water equivalence factor of 0.6 (Kim and Stuart, 2015). In order to compare Quantulus measurements with another method, ALOKA LSC (LB-7, Hitachi Aloka Medical Ltd., Japan) were used. To do so, we created a composite large volume sample (obtained by combining 7 Quantalus vials) that was re-counted using the ALOKA LSC system.

2.4. Dose calculations

We used ERICA, a software program that allows the assessment of environmental risks arising from radionuclide exposure (Larsson, 2008; Brown et al., 2016). Based on the measured tritium activity concentration in ELA fishes, the total tritium dose rate was calculated for each fish. In addition, the potential fish tritium dose concentration to humans consuming these fish was calculated based on the Canadian fish products consumption (Fisheries and Oceans Canada, 2013).

2.5. Data analysis

We evaluated the effect of body size (mass, g) on TFWT measurements (Bq/kg fresh weight of fish) of fish using linear regression. We similarly evaluated OBT measurements against body size for all species in all lakes. To determine the effect of lake and sex (male, female, undetermined) on estimated ^{3}H dose exposure, we examined differences among Lake Whitefish (common species among all lakes) using 2-way ANOVA. Differences among species within lakes were compared using *t*-tests with a Welch correction for heterogeneous variance. Data were log-transformed where necessary to normalize data and residuals. Data were analyzed using R version 3.4.2 (R Core Team, 2017).

3. Results and discussion

3.1. HTO activity concentrations

The measured HTO activity concentrations for lakes 305, 302 and 226 from the ELA in 2015 were similarly below reliably detectable levels (0.6 Bq/L) in all lakes (Table 2). The levels measured were found to be much lower than the levels found in the Ottawa River water, which in 2016 were determined to be approximately 3–5 Bq/L near

Table 2
Water tritium (HTO) activity concentration of lake water.

Sampling Date	Lake	Number of Samples	Activity Concentrations
2014 October 26	302	3	< MDA[a] (0.6 Bq/L)
2014 October 27	305	3	< MDA[a] (0.6 Bq/L)
2014 October 27	226	3	< MDA[a] (0.6 Bq/L)

[a] Less than the Minimum Detectable Activity of 0.6 Bq/L.

Table 3
Tissue-free water tritium (TFWT) activity concentrations from ELA fish flesh.

Lake	Number of Samples	Measured TFWT Range (Bq/L)	Average TFWT (Bq/L)
305	29	< MDA[a]	< 0.7
302	33	< MDA[a]	< 0.7
226	28	< MDA[a] – 1.6	0.85 ± 0.26

[a] Less than the Minimum Detectable Activity of 0.7 Bq/L.

Pembroke, Ontario (CNL, 2016). Both ELA and Ottawa River water were lower than the tritium activity concentrations measured in Lake Ontario in 1965 during the nuclear bomb testing era (43 Bq/L) and the 7 Bq/L measured in Lake Ontario in 1997 (Klukas and LaMarre, 2000).

The tritium measured in nature prior to nuclear weapons testing was approximately 0.6 Bq/L in precipitation, 0.3–0.8 Bq/L in river water and 0.1 Bq/L in seawater (Boyer et al., 2009), comparable to the background HTO activities reported here. Recently, they measured tritium activity concentrations around 1 Bq/L in precipitation, less than 10 Bq/L in river water and less than 0.3 Bq/L in sea water in France (Jean-Baptiste et al., 2018).

3.2. Fish TFWT activity concentrations

All fish samples analyzed for TFWT were lower than 2.0 Bq/L (Table 3), which are lower than reported TFWT measurements from other background-level sites near Saskatoon, Canada (Thompson et al., 2015). All fish from lakes 305 and 302 were below the MDA of 0.7 Bq/L, while 62% of fish samples obtained from Lake 226 were actually above the MDA value. Because the HTO activity concentration of the lake water was not found to be elevated, we did not expect to observe elevated TFWT in the fish collected from Lake 226.

On August 2, 1989, 370 GBq of ^{3}H (added as ^{3}HHO) was added to the hypolimnion of Lake 226 (Bird et al., 1998). The mean residence time of Lake 226 was estimated as 6.5 years (± 1.8 years), suggesting that any of the ^{3}H added in 1989 would have left the system by 1997. Thus, any fish in our collections from 2014 that were 17 years or older may have received direct exposure to ^{3}H in the system, and may explain elevated levels in fish there. Alternatively, incorporation of ^{3}H into sediment porewaters may have also created a legacy effect in this lake. With a half-life of 12.3 years, even without any significant turnover, additions of ^{3}H from 1989 would be at low levels in the lake by 2014 regardless due to radioactive decay. Based on fin scarring from previous captures, the earliest time period during which fish were previously captured in this lake was 2000, but this fish was not among the highest ^{3}H concentrations, suggesting legacy effects of previous experimental additions of ^{3}H are not the cause of elevated ^{3}H in Lake 226. There was no evidence of any relationship of TFWT with fish body size (length or body weight) from Lake 226 (linear regression, $P > 0.05$); both the highest and lowest TFWT values we observed were from whitefish approximately 400 g in weight.

In the 1970's, Lake 227 was injected with a tritium tracer to measure vertical diffusion rates (Quay et al., 1980). At the time, tritium activity concentrations were approximately 70,000 Bq/L in Lake 227. Lake 305 receives water from Lake 227, but is much larger than Lake 227 and any ^{3}H transported through the Lake 227 outflow would experience significant dilution. This is supported by the low TFWT values observed in Lake 305 fishes (all below MDA, Table 3). Lake 226 has no hydrologic connection to Lake 227.

3.3. Fish OBT activity concentrations

Measured OBT activity concentrations ranged from below reliable MDA (2.5 Bq/L) to 6.8 Bq/L in ELA fish. Seven percent of Lake 305 fish, 39% of Lake 302 fish and 93% of Lake 226 fish had measured OBT activity concentrations above MDA. OBT activity concentrations were an order of magnitude greater than the TFWT activity concentrations, on average. The differences between TFWT and OBT are thought to be due to differences in analytical processes between the two methods and also differences in the volumes of combustion water used for counting (much smaller sample volumes involved in OBT combustion methods). In addition, OBT formation and accumulation in fish is different to TFWT because OBT formation is a long process, integrating all tritium accumulated prior sampling (Yankovich et al., 2011). Also, the OBT activity concentration in fish is affected not only the body mass, but also by the fish species, fish age and food type (Melintescu et al., 2011; Melintescu and Galeriu, 2011).

Table 4
Organically-bound tritium (OBT) activity concentrations from ELA fish flesh.

Lake	Number of Samples	Measured OBT Range (Bq/L)	Average OBT (Bq/L)
305	29	< MDA[a] – 4.4	1.8 ± 0.8
302	33	< MDA[a] – 5.5	2.4 ± 1.2
226	28	< MDA[a] – 6.8	3.8 ± 1.0

[a] Less than the Minimum Detectable Activity of 2.5 Bq/L.

Our results suggest that the background fish flesh OBT activity concentration in this region would typically be below 5 Bq/L (Thompson et al., 2015), though 5 Lake Whitefish from Lake 226 and one White Sucker from Lake 302 actually exceeded this value. OBT results from Lake 305 and 302 were less than MDA on average, but OBT values of Lake 226 were above the detection limit on average (Table 4).

We found significant negative relationships between fish size and OBT activity in Lake 302 fishes only (Fig. 1). The observed OBT activity declined with body size in both Lake Whitefish (linear regression, $F_{1,27} = 9.0$, $P = 0.006$) and White Sucker (linear regression, $F_{1,14} = 11.0$, $P = 0.005$). Only Lake Whitefish with mass less than 500 g were observed to have OBT values greater than MDA in Lake 302. There

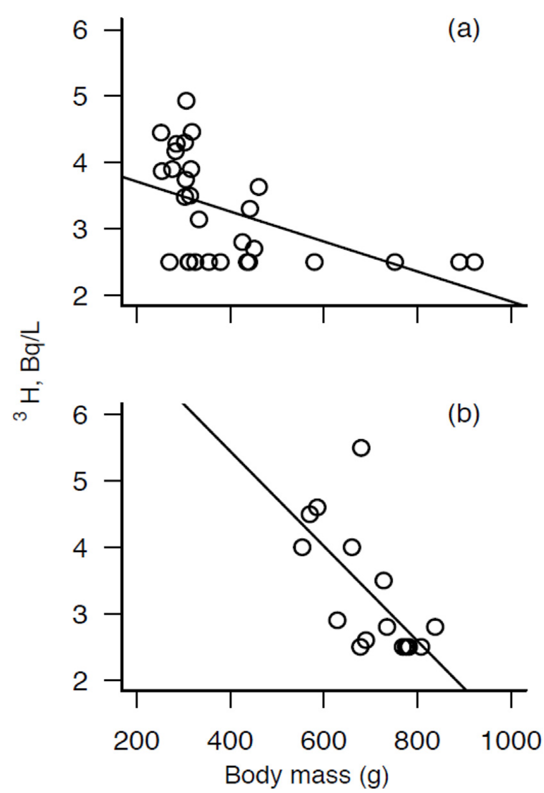

Fig. 1. Significant negative relationships between measured organically-bound tritium (OBT) and body size in Lake Whitefish (a) and White Sucker (b) from Lake 302, IISD-Experimental Lakes Area.

Table 5
OBT activity concentration re-counting using ALOKA LSC after combining 7 samples counted using the Quantulus LSC. This was done on two sets of samples.

Set 1 Sample ID from Lake 226	Activity Concentration obtained using the Quantulus (Bq/L)	Set 2 Sample ID from Lake 302	Activity Concentration obtained using the Quantulus (Bq/L)
A091	2.6	A109	4.5
A092	2.8	A110	3.7
A093	3.3	A111	4.2
A094	3.4	A113	3.1
A095	3.1	A114	3.9
A096	3.0	A115	< 2.2
A097	3.4	A116	< 1.7
Average	**3.1 ± 0.3**	**Average**	**3.3 ± 1.0**
Activity Concentration obtained using the ALOKA (Bq/L)	4.9 ± 1.3	Activity Concentration obtained using the ALOKA (Bq/L)	5.5 ± 1.2

was no significant relationship between body size and OBT activity in Lake 226 whitefish ($P > 0.05$). Nearly all OBT measurements in Lake 305 were below MDA. Though a positive relationship was initially observed in Lake 305 for Lake Whitefish, this was largely due to the influence of one outlier value (the largest fish at 1400 g, OBT = 3.6 Bq/L); without this single value there was no significant relationship with body size. The only detectable OBT value for Northern Pike in Lake 305 was a mid-sized (5 kg) individual (OBT = 4.4 Bq/L). It is well known that OBT activity in animals such as fish, mammals decrease with body size due to growth dilution (Melintescu and Galeriu, 2011) but it depends on fish species, bioenergetics, age, food type, the type of aquatic environment itself. In this study, we are focusing the OBT measurement and it is hard to predict the environmental conditions in ELA except fish biomass. Other radioactive tracers (^{210}Po) have been shown to vary negatively with body size in freshwater fish from other regions (Chen et al., 2015b) and these fish in particular (Chen et al., 2016).

Because of the discrepancy between the two methods, some of the samples obtained using the Parr method (originally counted using the Quantalus) were combined (to increase the volume) and re-counted using the ALOKA system (Table 5). The results obtained with the ALOKA was 1.7 times higher than the values obtained from the Quantulus LSC, perhaps reflecting the larger sample sizes using this method. This showed that sample sizes are likely important factors to take into account when LSC are used to evaluate very low levels of tritium. Such a discrepancy highlights the influence of sample size and LSC sensitivity as well as the difficulties caused by not having a standard method for OBT environmental sample analysis and lack of suitable reference materials (Baglan et al., 2015). The OBT inter-laboratory comparisons were conducted recently (Baglan et al., 2018; Kim et al., 2015) and the results of the participating laboratories were within 20% or less. For those exercises, a few hundred or over 40 Bq/L OBT level of environmental samples were tested (10–100 times greater than the activities involved in the current study). Particularly, OBT measurements in background areas (less than 5 Bq/L) using ALOKA LSC is far more sensitive than the conventional LSC but may require alternative methods such as the well-established ^{3}He ingrowth mass spectrometry (Jean-Baptiste et al., 2010). Combustion water from fish tissues was strongly acidic and contained high concentrations of nitrates, sulfates, which are contributing factors to increase quench for LSC counting. Further study will be required to improve the current OBT skills for ultra-low OBT level measurement in fish sample.

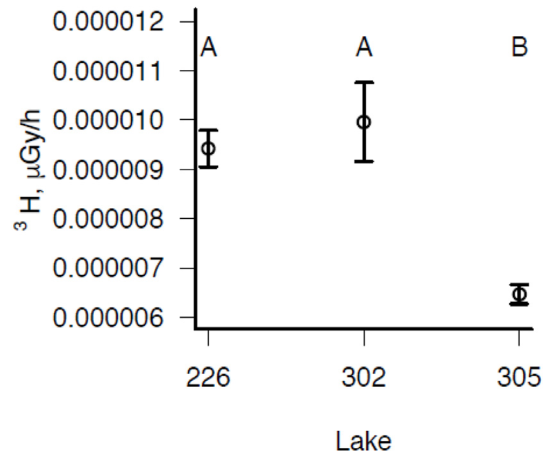

Fig. 2. Mean tritium dose rate plus or minus one standard error to fish in three IISD-Experimental Lakes Area lakes. Common capital letters denote similarity among groups.

3.4. Calculated total tritium dose rate to freshwater fish from the ELA

The calculated tritium dose rates ranged from 4.9×10^{-6} µGy/h (Lake 305 Northern Pike) to 2.2×10^{-5} µGy/h (Lake 302 Lake Whitefish). Examining Lake Whitefish, which were found across all three lakes, we found significant differences between lakes (ANOVA, $F_{2,66} = 11.6$, $P < 0.0001$; Fig. 2). There was no significant interaction between the source lake and sex of fish ($F_{3,66} = 0.8$, $P = 0.5$) and no additive effect of sex on tritium dose levels (F2,66 = 2.0, P = 0.14).

Comparing species within lakes, we found that Lake Whitefish in Lake 302 had significantly higher dose rates than White Sucker (Lake Whitefish = 9.9×10^{-6} µGy/h, White Sucker = 7.3×10^{-6} µGy/h, $t_{3.01} = 35.3$, $P = 0.004$). Comparing species in Lake 305, however, revealed no significant difference between Lake Whitefish and Northern Pike dose rates (*t*-test, $P > 0.05$).

Based on the HTO and OBT activity concentration measurements, the calculated average tritium dose rates to fish were very low and no radiological effects on non-human biota are expected at such levels (Chouhan et al., 2008).

3.5. Estimated human radiation dose from fish consumption

Based on the measured HTO and OBT concentrations, the tritium ingestion dose to human was also estimated. The dose conversion factors used were 1.8×10^{-11} Sv/Bq for HTO and 4.2×10^{-11} Sv/Bq for OBT (ICRP, 2013). Fish consumption in Canada fluctuates over the years but the average Canadian consumption rate for fish products is 8.8 kg (edible weight) per person yearly (Fisheries and Oceans Canada, 2013). Assuming that an adult consumed 8.8 kg of fish in a year and that all of the consumed fish contain the maximum levels of HTO and OBT measured in this study, the resulting radiation dose would be 4.72×10^{-4} mSv, a small fraction of the annual public dose limit of 1 mSv, while the numerical value provides a benchmark for comparison, the limit applied only to doses from licensed activities. The natural background radiation in general (all radionuclides included) is in the order of 2.4 mSv (UNSCEAR, 2000).

Kim et al. (2014) reported that the total effective tritium dose after ingestion of fish subjected to the Canadian Drinking Water Guideline (7000 Bq/L) was approximately 2.1×10^{-3} mSv/year, which is 4.4 times higher than exposures reported this study and is significantly lower than the public radiation dose limit (1 mSv/year) which is applied to doses from licensed facilities.

4. Conclusion

TFWT and OBT activity concentrations of ELA fish were low (many samples yielded values that were below the MDA). For the three lakes studied, the measured water tritium (HTO) activity concentration was below 0.6 Bq/L. Though TFWT values were elevated in Lake 226, concentrations were generally found to be below 0.7 Bq/L, which was consistent with the surrounding water tritium activity concentration. Fish OBT activity concentrations, at below 5 Bq/L, were also low (although higher than the TFWT), but varied among lakes and increased with fish size in Lake 302. The levels of tritium reported here reflect the Canadian tritium background and no radiological health concerns (both for fish and people consuming those fish) are expected for humans consuming fish exposed to these levels of tritium. Finally, the discrepancies observed between tritium measurements in low-level OBT samples were noted as part of this study. This stresses the need for standardization of methods and for the development of relevant reference material with low-level tritium contained environmental samples.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2018.10.003.

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