Degradation of hydrogen peroxide in seawater using the anti-sea louse formulation Interox® Paramove™50

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2014

Canadian Technical Report of Fisheries and Aquatic Sciences 3080

Fisheries and Oceans Canada | Pêches et Océans Canada

Canada

Canadian Technical Report of Fisheries and Aquatic Sciences

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Fisheries and Aquatic Sciences 3080

July 2014

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This is the three hundred and tenth Technical Report of
the Biological Station, St. Andrews, NB

Cat. No. Fs 97-6/3080E-PDF ISBN 978-1-100-23510-3 ISSN 1488-5379 (online version)

Correct citation for this publication:

Lyons, M.C., Wong, D.K.H. and Page, F.H. 2014. Degradation of hydrogen peroxide in seawater using the anti-sea louse formulation Interox® Paramove™50. Can. Tech. Rep. Fish. Aquat. Sci. 3080: v + 19p.

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ABSTRACT

Lyons, M.C., Wong, D.K.H. and Page, F.H. 2014. Degradation of hydrogen peroxide in seawater using the anti-sea louse formulation Interox® Paramove™50. Can. Tech. Rep. Fish. Aquat. Sci. 3080: v + 15p.

Interox®Paramove™50 is an anti-sea louse treatment registered in Canada to combat infestations of parasitic copepods (sea lice) at Atlantic salmon (*Salmo salar*) aquaculture sites. The active ingredient in Interox®Paramove™50 is hydrogen peroxide (H_2O_2). The formulation was used extensively at salmon aquaculture sites in southwest New Brunswick (SWNB) in 2010 to 2013. It was expected that H_2O_2 would degrade quickly during the treatment period but real time monitoring indicated that the concentration of H_2O_2 stayed at or near the treatment level. Treatment impacts on non-target organisms are dependent on the fate of anti-louse formulations in the environment. The determination of degradation times of anti-louse treatments is important information to have in assessing their fate in the environment. Short-term degradation studies in the laboratory were performed under static conditions with raw seawater at 5, 10, 15 and 20°C. Degradation was temperature dependent. Preliminary work in the laboratory with Interox® Paramove™ 50 in raw seawater has shown the half-life of 1200 $mg \cdot L^{-1}$ H_2O_2 to be 28 days at 10°C. Degradation studies in the laboratory were performed under static conditions with raw seawater at 10°C and a range of concentrations of H_2O_2 from 300 $mg \cdot L^{-1}$ to 1800 $mg \cdot L^{-1}$. The calculated half-life of H_2O_2 ranged from 8 to 19 days. The calculated half-life of 1200 $mg \cdot L^{-1}$ H_2O_2 was inconsistent with the preliminary calculated half-life (14 days versus 28 days). The influence of biotic communities and organic matter on the degradation of H_2O_2 in the Interox®Paramove™50 formulation was investigated using filtered (0.2 µm) raw seawater at 10°C and a range of concentrations of H_2O_2 from 300 to 1800 $mg \cdot L^{-1}$. Degradation of H_2O_2 was not dependent on concentration. The calculated half-life of H_2O_2 ranged from 1 to 4 days indicating a faster rate of H_2O_2 degradation in filtered seawater than in raw seawater. These results indicate that degradation of H_2O_2 in Interox®Paramove™50 was slowed down by the presence of biotic communities and/ or organic matter in raw seawater.

RÉSUMÉ

Lyons, M.C., Wong, D.K.H. and Page, F.H. 2014. Degradation of hydrogen peroxide in seawater using the anti-sea louse formulation Interox® Paramove™50. Can. Tech. Rep. Fish. Aquat. Sci. 3080: v + 15p.

L'Interox® Paramove^{MC} 50 est un traitement contre le pou du poisson homologué au Canada pour combattre les infestations de copépodes parasitaires (pou du poisson) dans les sites aquacoles de saumons de l'Atlantique (*Salmo salar*). L'ingrédient actif de l'Interox® Paramove^{MC} 50 est le peroxyde d'hydrogène (H_2O_2). La préparation a largement été utilisée dans les sites aquacoles de saumons du sud-ouest du Nouveau-Brunswick entre 2010 et 2013. On s'attendait à ce que le H_2O_2 se dégrade rapidement durant la période de traitement, mais la surveillance en temps réel a démontré que le taux de concentration du H_2O_2 demeurait le même que le taux de concentration durant le traitement, ou sinon proche de celui-ci. Les incidences du traitement sur les organismes non visés dépendent du devenir des préparations contre le pou du poisson dans l'environnement. La détermination des temps de dégradation des préparations contre le pou du poisson est importante pour obtenir des renseignements qui seront utiles durant l'évaluation du devenir de ces préparations dans l'environnement. Des études en laboratoire sur la dégradation à court terme ont été réalisées dans des conditions statiques en utilisant de l'eau de mer brute à 5, 10, 15 et 20 °C. La dégradation dépendait de la température. Les travaux préliminaires en laboratoire en utilisant l'Interox® Paramove^{MC} 50 dans de l'eau de mer brute ont démontré que la demi-vie de 1 200 mg·L^{-1} H_2O_2 est de 28 jours à 10 °C. Des études en laboratoire sur la dégradation ont été réalisées dans des conditions statiques en utilisant de l'eau de mer brute à 10 °C et dans des concentrations de H_2O_2 variant de 300 mg·L^{-1} à 1 800 mg·L^{-1}. La demi-vie calculée de H_2O_2 variait de 8 à 19 jours. La demi-vie calculée de 1 200 mg de H_2O_2 correspondait à la demi-vie préliminairement calculée (14 jours par rapport à 28 jours). L'influence des biocénoses et de la matière organique sur la dégradation du H_2O_2 dans la préparation Interox® Paramove^{MC} 50 a été étudiée en utilisant de l'eau de mer brute filtrée (0,2 µm) à 10 °C et des concentrations de H_2O_2 variant de 300 à 1 800 mg L^{-1}. La dégradation de H_2O_2 ne dépendait pas de la concentration. La demi-vie calculée de H_2O_2 variait de 1 à 4 jours, ce qui indiquait un taux plus rapide de dégradation de H_2O_2 dans l'eau de mer filtrée que dans l'eau de mer brute. Ces résultats indiquent que la dégradation de H_2O_2 dans l'Interox® Paramove^{MC} 50 était ralentie par la présence des biocénoses et (ou) de matière organique dans l'eau de mer brute.

INTRODUCTION

The Atlantic salmon aquaculture industry in Atlantic Canada began in southwestern New Brunswick in 1978 and sea lice first became a problem for the industry in 1984 when a large outbreak of *Lepeophtheirus salmonis* occurred on a farm in Dark Harbour Grand Manan (Chang et al. 2011). The industry grew considerably between 1984 and 2000 and experienced a serious sea lice problem in 1994-95 when two lice species, *Lepeophtheirus salmonis* and *Caligus elongates,* infested salmon in southwest New Brunswick (SWNB) (*cf* Burka et al., 1997; Fisheries and Oceans, 2003; Burridge et al., 2010; Chang et al. 2011). The industry experienced another serious outbreak of lice in 2006 (Chang et al., 2011) and this outbreak continues at a reduced intensity to the present day.

A number of pesticides and application methods have been used to combat the above sea lice infestations. Aquaculture pesticides have been applied by one of three methods: well boats, cage tarping, and cage skirting. All involve allowing affected fish to swim in a bath of pesticide-treated water, which, once the treatment is complete, is released to the surrounding environment (Burridge, 2013). Release of the effluent water, including the pesticide formulation to the surrounding water has raised concerns that negative effects on non-target organisms might occur (Haya et al., 2005; Burridge, 2013; DFO, 2013a; DFO, 2013b). Ernst et al. (2001) studied dispersion of simulated treatments in SWNB using Rhodamine dye and found that after release the dye plume entered the intertidal zone and contacted the bottom where it could have an effect on benthic species. Of particular concern in SWNB is the potential for anti-louse treatments to negatively impact other crustaceans such as the American lobster (*Homarus americanus*).

One of the pesticides that has been used in southwest New Brunswick and elsewhere is hydrogren peroxide. Hydrogen peroxide (H_2O_2) was used to control sea lice on salmon farms in the Faroe Islands (Denmark), Norway, Scotland and Canada in the 1990s, in Scotland in 2008 and recently in Chile (Burridge et al., 2010). Throughout 2010-2013 Interox® Paramove™50 was used in SWNB in well boats as an anti-louse treatment. It is an emulsifiable concentrate containing 50% (by weight) H_2O_2 and has a Pest Control Products Act (PCPA) registration number 29783 (Health Canada, 2013). H_2O_2 acts by causing paralysis, peroxidation in organelle membranes, and inhibition of enzymes that replicate DNA (Cotran et al., 1989). Interox® Paramove™50 has emergency registration status with Health Canada until June 2014 (Health Canada, 2013). Recently studies have been conducted to determine the effects of Interox® Paramove™50 on lobster and on other non-target crustaceans (Burridge, 2013; Burridge et al., 2014; McCurdy et al., 2013).

In southwest New Brunswick Interox®Paramove™50 has been used in bath treatments at concentrations of 1.2 -1.8 $g \cdot L^{-1}$ (as H_2O_2) for up to 30 minutes (Health Canada, 2013). Its effectiveness is highly dependent on water temperature, so concentrations are sometimes increased depending on the time of year and temperature (Treasurer and Grant, 1997). Interox® Paramove™50 has a high affinity for water. The octanol-water partition coefficient for H_2O_2 is <1, indicating that it will not persist in the environment (HERA project, 2005). The half-life of stabilized H_2O_2 in seawater has been reported to be about

7 days at 10°C with aeration (Bruno and Raynard, 1994). Real time monitoring during well boat treatments with Interox®Paramove™50 in SWNB indicated that the concentration of H_2O_2 stayed at or near the treatment level during the treatment period (Michael Beattie, New Brunswick Department of Agriculture, Aquaculture and Fisheries, St. George, New Brunswick personal communication).

The pathways that have been shown to lead to the decomposition of H_2O_2 involve biological, chemical and photochemical decomposition mechanisms (Cooper and Zepp, 1990; Miller et al., 2009; Moffett and Zafiriou, 1990; Richard et al., 2007). Investigators have used filtered and unfiltered water to assess the role of algae and bacteria in the decay of H_2O_2 (Herrmann and Herrmann, 1994; Moffett and Zafiriou, 1990; Richard et al., 2007). Bruno and Raynard (1994) investigated the effects of temperature, aeration and stabilized versus unstabilized H_2O_2 on degradation. Tort et al. (2004) studied the effects of aeration and organic matter on the degradation of H_2O_2 in static and flow-through systems.

In this study four approaches were taken to examine the degradation of H_2O_2. Firstly, a short-term standardised degradation study in the laboratory was performed under static conditions with Interox®Paramove™50 in raw seawater (1200 $mg \cdot L^{-1}$ H_2O_2, nominal concentration) at 5, 10, 15 and 20°C. Water samples were collected at specific time points and water analysis conducted to measure concentration of H_2O_2 in seawater over a 3 h period. Secondly, a standardised degradation study in the laboratory was performed under static conditions with Interox®Paramove™50 in raw seawater (1200 $mg \cdot L^{-1}$ H_2O_2) at 10°C over a period of 175 days. Thirdly, a standardised degradation study in the laboratory was performed under static conditions with Interox®Paramove™50 in raw seawater at five nominal concentrations of H_2O_2 (1800 $mg \cdot L^{-1}$, 1200 $mg \cdot L^{-1}$, 800 $mg \cdot L^{-1}$, 500 $mg \cdot L^{-1}$ and 300 $mg \cdot L^{-1}$) at 10°C over a period of 35 days. The final standardised degradation study in the laboratory was performed under static conditions with Interox®Paramove™50 in filtered raw seawater at five nominal concentrations of H_2O_2 (1800 $mg \cdot L^{-1}$, 1200 $mg \cdot L^{-1}$, 800 $mg \cdot L^{-1}$, 500 $mg \cdot L^{-1}$ and 300 $mg \cdot L^{-1}$) at 10°C over a period of 8 days.

MATERIALS AND METHODS

Experimental design

Interox®Paramove™50 was provided by Mr. Ian Armstrong, Aqua Pharma Inc., Saint John, New Brunswck, Canada. Raw seawater was collected from the St. Andrews Biological Station water intake building. The seawater was collected at the start of each degradation study. Ambient water temperature was recorded when collected and salinity was recorded twice during the study. Seawater was allowed to equilibrate to the test temperature in jacketed beakers before adding the H_2O_2. Temperature was maintained at ± 0.1°C with a Polystat cooling/heating recirculating chiller (Cole-Parmer) (Fig. 1). Temperatures were confirmed using a calibrated mercury thermometer.

Short-term degradation of 1200 mg·L^{-1} H_2O_2 at four temperatures

One 5000 mL jacketed beaker (Ace Glass) was connected to the Polystat unit set to 5°C. The beaker contained a magnetic stir bar and was placed on a stir plate. Interox®Paramove™50 and raw seawater were combined to prepare 4900 mL of the test solution (1200 mg·L^{-1} H_2O_2). Water samples were collected for analysis at 0, 0.25, 0.5, 0.75, 1, 1.5, 2, 2.5 and 3h. Three replicate tests were conducted on three different days under static conditions with constant stirring. The short-term degradation studies were repeated at 10°C, 15°C, 20°C (n=3 tests at each temperature).

Degradation of 1200 mg·L^{-1} H_2O_2 over 175 days

One 5000 mL jacketed beaker (Ace Glass) was connected to the Polystat unit set to 10°C. The beaker contained a magnetic stir bar and was placed on a stir plate. Interox®Paramove™50 and raw seawater were combined to prepare 4900 mL of the test solution (1200 mg·L^{-1} H_2O_2). The beaker was covered with plastic wrap so as to avoid evaporation. A water sample was collected for analysis at T=0, once daily for days 1 through 29, day 32, day 34, day 36 and then once a week until 175 days had passed. Time of day was recorded each time water was sampled.

Effect of concentration on the degradation of H_2O_2

Five identical 5000 mL jacketed beakers (Ace Glass) were connected in series to the Polystat unit set to 10°C (Fig. 1). The beakers contained magnetic stir bars and were placed on stir plates. Interox®Paramove™50 and raw seawater were combined to prepare 4900 mL of each of the test solutions of H_2O_2 (1800 mg·L^{-1}, 1200 mg·L^{-1}, 800 mg·L^{-1}, 500 mg·L^{-1} and 300 mg·L^{-1}). Tests were conducted under static conditions with constant stirring. The beakers were covered with plastic wrap so as to avoid evaporation. Water samples were collected for analysis at 0, 1, 3, 6, 24, 48, 72 and 96 h for the first week and then on days 7, 9, 11, 14, 16, 18, 21 and 35. Time of day was recorded each time water was sampled.

Effect of filtration of raw seawater on the degradation of H_2O_2

Prior to the start of the degradation experiment raw seawater was pre-filtered through a glass fiber filter type A-E (Gelman Sciences Inc.) to remove large organic matter and then filtered through a Whatman 0.2 µm nylon filter (G.E. Healthcare Life Sciences) to remove smaller organic matter and large bacteria. Millipore vacuum pumps were used for the filtration. The 5 test solutions of H_2O_2 (1800 mg·L^{-1}, 1200 mg·L^{-1}, 800 mg·L^{-1}, 500 mg·L^{-1} and 300 mg·L^{-1}) were prepared with the filtered seawater and the experimental setup was identical to the previous setup at 10°C. Water samples were collected for analysis at 0, 3, 6, 24 h and then on days 2 through 8. Time of day was recorded each time water was sampled. The degradation study was replicated once (n=2 studies).

Water analysis

Water samples were analysed for presence and concentration of H_2O_2 using titration with a cerium sulphate/sulfuric acid mixture as prescribed by Aqua Pharma Inc. (Ian Armstrong, personal communication). Briefly, treated seawater samples were added dropwise to a mixture of 7.5 mL of 0.1 N cerium IV sulphate (Ricca Chemical Co.,

Arlington, Texas) and 5.0 mL of 5 N sulphuric acid (Ricca Chemical Co., Arlington, Texas) until all colour disappeared (yellow to clear). The volume of treated seawater added is proportional to the quantity of H_2O_2 present. The H_2O_2 concentration was calculated using the equation:

$$C_t = (7.5 \times 0.1 \times 1000 \times 34) \div 2 \times v$$

where C_t ($mg \cdot L^{-1}$) is the determined concentration of H_2O_2 of the sample, 7.5 (mL) is the volume of cerium sulfate used for the titration, 0.1 is the molarity of the cerium sulfate, 1000 $mg \cdot g^{-1}$ is a conversion factor, 34 is the molecular weight of H_2O_2 and v is the sample volume (mL) used in the titration.

Half-life determinations

The degradation of H_2O_2 appeared to be a first order reaction therefore the measured concentrations were transformed by taking the natural logarithm (Ln). The transformed data was plotted against time (days) and the equation of the line was used to determine the half-life of H_2O_2.

$$half\ life = (\ln(C_0 \div 2) - b) \div m$$

where C_0 is the measured concentration at T=0 h, b is the y intercept and m is the slope of the line. When calculating the half-life of 1200 $mg \cdot L^{-1}$ H_2O_2 for the 175 day study, the measured concentrations for the first 36 days were used in the calculation of the equation of the line so that a comparison could be made with 1200 $mg \cdot L^{-1}$ H_2O_2 in the 35 day study.

RESULTS

Short-term degradation of 1200 $mg \cdot L^{-1}$ H_2O_2 at four temperatures

Ambient water temperature was 3.4°C when collected for the 5°C study, 3.7°C for the 10°C study, 2.4°C for the 15°C study, 2.7°C for the 20°C study. Although salinity of the samples were not recorded sea water in the collection area typically has a salinity of between 28 and 32 psu. Little degradation occurred over the three hours at any of the four temperatures (Fig. 2 and Appendix 1, Table 1). The slopes of the degradation lines for H_2O_2 at the two higher temperatures (15°C and 20°C) were greater than the slopes of the degradation lines at the lower temperatures (5°C and 10°C) (Appendix 3, Table 1) indicating degradation of H_2O_2 was temperature dependent.

Degradation of 1200 $mg \cdot L^{-1}$ H_2O_2 over 175 days at 10°C

Ambient water temperature was 2.4°C on the day of collection. Although salinity of the sample was not recorded sea water in the collection area typically has a salinity of between 28 and 32 psu. Measured concentrations of H_2O_2 are shown in Table 2a. H_2O_2 degraded more quickly in the first few weeks of the study (Appendix 1, Table 2b and Fig. 3). The half-life was determined to be ~28 days (Appendix 2, Table 1).

Effect of concentration on the degradation of H_2O_2 at 10°C

Ambient water temperature was 8.5°C and salinity was recorded as 32 psu on the day of collection. Measured concentrations of H_2O_2 are shown in Appendix 1, Table 3a. H_2O_2 degradation was not concentration dependent (Appendix 1, Table 3b and Fig. 4). The half-lives were calculated and are presented in Appendix 2, Table 1. The rate of decomposition of the 1200 $mg \cdot L^{-1}$ H_2O_2 was faster than in the previous 175 day study of the same concentration. The half-life of 1200 $mg \cdot L^{-1}$ H_2O_2 was calculated as ~14 days as compared to ~28 days in the 175 day study (Appendix 2, Table 1).

Effect of filtration of raw seawater on the degradation of H_2O_2 at 10°C

Ambient water temperature was 4.3°C for replicate 1 and 3.5°C for replicate 2 on the day of collection. Salinity of raw seawater was recorded as 32 psu. Measured concentrations of H_2O_2 are shown in Appendix 1, Table 4. H_2O_2 degraded faster in filtered seawater versus raw seawater for all test concentrations (Fig. 5) with the calculated half-lives between 1 and 4 days (Appendix 2, Table 1).

Half-life determinations

The half-life of 1200 $mg \cdot L^{-1}$ H_2O_2 prepared in raw seawater was not consistent between the two studies (Appendix 2, Table 1). The slope of the degradation line for 1200 $mg \cdot L^{-1}$ H_2O_2 was lower in the 175 day trial compared to the 35 day trial indicating a slower degradation rate (Appendix 3, Table 2A). Degradation was not concentration dependent regardless of whether the test solutions of H_2O_2 were prepared in filtered or unfiltered raw seawater. H_2O_2 degraded (between 3X and 8X) faster in filtered seawater than in unfiltered seawater (Appendix 2, Table 1). The slopes of the degradation lines for H_2O_2 prepared in filtered raw seawater were much higher than the slopes of the lines for H_2O_2 prepared in unfiltered raw seawater (Appendix 3, Table 2A & B) indicating much faster degradation of H_2O_2 in filtered seawater.

DISCUSSION

In commercial fish farm sea lice treatments the dilution of hydrogen peroxide in the receiving environment is controlled by mechanical and ambient physical mixing as well as by chemically and biologically mediated degradation. The factors influencing the physical dispersion of the pesticide include advective (horizontal) current velocities; rate of mixing in the horizontal and vertical dimensions; weather, wind and waves; and proximity of vertical boundaries, including vertical stratification, the sea bottom and inter-tidal zones; proximity of horizontal boundaries such as the shoreline, bottom and pycnocline (DFO, 2013a). The chemical and biological degradation of the pesticide is also influenced by many factors. Pure hydrogen peroxide (H_2O_2) is a natural chemical that can be produced by chemical and biological processes (Moffett and Zafiriou, 1990) and it decomposes into water and oxygen at rates which depend on the abiotic and biotic characteristics of the water, including temperature, salinity, light intensity and wavelength, organic load, catalytic materials (metals, activated carbon, enzymes) into which it is introduced. With the exception of fluorine, the halogens catalyse the decomposition of H_2O_2 by a cyclic oxidation-reduction mechanism (EUR 20844 EN,

2003). H_2O_2 is quite a reactive substance in the presence of other substances, elements, radiation, materials or cells. Both biotic and abiotic degradation processes are important routes in the removal of hydrogen peroxide in the environment. Biological degradation of H_2O_2 is an enzyme-mediated process. Abiotic degradation of H_2O_2 is due to reaction with itself (disproportionation), reaction with transition metals, organic compounds capable of reacting with H_2O_2, reaction with free radicals, heat or light. H_2O_2 is normally a short-lived substance in the environment. Rapid degradation will occur due to many alternative and competitive degradation pathways. However, like most substances, in special circumstances when degradation processes are inactive, hydrogen peroxide can be an extremely persistent substance in the environment. Degradation in the aquatic environment takes place in the presence of a catalyst. Most transition metals, and especially Fe, Mn and Cu may have significant influence on degradation rates of hydrogen peroxide in natural waters (EUR 20844 EN, 2003).The product formulation in the case of synthetic peroxide may also play a role in the degradation rate of H_2O_2. Pesticide formulations are prepared to optimise the probability that the active ingredient reaches and affects the target organism, in this case sea lice. The formulation, therefore, is prepared with a number of chemicals in addition to the active ingredient, which may include solvents, surfactants, and stabilisers. The ingredients in the formulation will affect how the active compound behaves in the environment (Burridge, 2013).

There is strong evidence by investigators that H_2O_2 degradation is dominated by biological processes and hence the rates observed in unfiltered water were expected to be greater than those from filtered water. Filtering natural water samples does to some extent allow differentiation between the biodegradation and abiotic elimination processes. Moffett and Zafiriou (1990) used ^{18}O-labelled H_2O_2 and found that in experiments with seawater there was no decomposition over 48 h in filtered seawater but there was decomposition in unfiltered seawater. The formation of O_2 and H_2O and their product distribution indicated that 65-80% of the decay was due to catalase and 20-30% due to peroxidase activity in that study. Photochemical decomposition of H_2O_2 was also observed in the Moffett and Zafiriou (1990) study. They suggested other powerful, currently unknown, oxidizing species are present in irradiated seawater probably generated by the photoxidation of dissolved organic matter. Penru et al. (2012) studied the application of UV/ H_2O_2 to seawater for disinfection and natural organic matter removal. They noted that the specific chemistry of seawater has to be considered due to the high reactivity of bromide and chloride with oxidant species (ozone, hydroxyl radicals). They were able to disinfect seawater and at the same time natural organic matter underwent oxidation. Pedersen et al. (2006) used small-scale biofilters in closed recirculation systems to find decomposition rate constants were significantly related to the amount of organic matter and initial dosage of H_2O_2. Tort et al. (2004) found that under static conditions in the absence of aeration and organic matter, low dose H_2O_2 concentration decreased to approximately 50% by day 6 and decreased to 0 mg L^{-1} by day 10 in freshwater aquaria. In the presence of aeration and/ or organic matter, H_2O_2 concentration decreased to below the level of detection within 48-72 hours. Pedersen et al. (2006) used small-scale biofilters in closed recirculation systems to find decomposition rate constants were significantly related to the amount of organic matter and initial dosage of H_2O_2.

In the new laboratory studies described here only static degradation was considered; the factors influencing dispersion were not considered. Our studies involved only Interox®Paramove®50 and degradation experiments were conducted at temperatures ranging from 5 to 20°C and with seawater having a salinity of between 28 and 32 psu. Interox®Paramove®50 was chosen since it is the product presently being used by the salmon farming industry in SWNB and the temperature and salinity conditions were chosen because they bracket values at which the commercial treatments in SWNB are conducted with most of the commercial treatments being conducted between 5 and 15°C. All of our half-life results were determined after following the same experimental protocol- continuously stirring the peroxide solution, maintaining the water temperature at 10°C±0.1°C, and the salinities at about 32 psu. The degradation studies were conducted under ambient day-night laboratory fluorescent lighting.

Literature reported degradation half-lives for H_2O_2 range from <1 h to 7 days (Appendix 4, Table 1). This range is associated with a variety of peroxide formulations and experimental conditions (temperature, salinity, presence of aeration and organic matter). Our half-life results ranged from 1 day (using filtered seawater) to 28 days (using natural unfiltered seawater) and are inconsistent with the manufacturers reported half-life of 5 days. Our finding that H_2O_2 degraded faster in filtered water than in unfiltered water is inconsistent with the literature expectations. Moffett and Zafiriou (1990) measured degradation of 100 nM H_2O_2, a concentration typically found in daytime surface waters and reported no decay after 48 h in filtered seawater compared to that found in natural seawater where half-lives of 8 to 11.5 h were reported whereas we saw shorter half-lives in filtered seawater (1 to 4 days) compared to 9 to 19 days in natural unfiltered seawater. We expected that the filtration process should have removed most of the organic matter and hence reduce degradation induced by this material. Metals and enzymes dissolved in the water should have been consistent between the filtered and unfiltered samples. The manufacturer of Interox® Paramove™50 provides degradation information on its material safety data sheet (MSDS). Abiotic degradation in water is reported as a redox reaction with a half-life of 5 days. The conditions reported are mineral and enzymatic catalysis in freshwater and salt water (Solvay Chemicals Inc., 2012). Miller et al. (2009) investigated the impact of natural organic matter on H_2O_2 – mediated oxidation of Fe(II) in a simulated freshwater system. They found that natural organic matter may stabilize Fe(II) in the presence of elevated H_2O_2 concentrations, significantly increasing the lifetime of ferrous iron and reducing the flux of hydroxyl radicals produced through this oxidation pathway. By removing the natural organic matter by filtration of the seawater in the present study, perhaps Fe(II) was less stable and more reactive with H_2O_2 and therefore faster degradation occurred.

Our half-life results (28 days versus 14 days) between trials for 1200 $mg \cdot L^{-1}$ H_2O_2 in raw seawater may be a result of differences in the water matrix at the time of collection. Ambient water temperature when collected in February for the 175 day trial was 2.4°C and 8.5°C when collected at the beginning of December for the 35 day trial. Seasonal differences occur in biota and organic matter in water and these could affect the pathways of H_2O_2 degradation. Bruno and Raynard (1994) reported half-lives for H_2O_2 at 10°C that

ranged from 5 to 8 days depending on whether it was stabilized or aerated. At 15°C degradation was faster and they reported half-lives that ranged from 3 to 7 days. Tort et al. (2004) reported half-lives ranging from 20 h to several days for low doses of H_2O_2 in freshwater depending on whether organic matter was present or the water was aerated. The presence of organic matter accelerated the degradation of H_2O_2. Similarly Pedersen et al. (2012) found the decay rate of H_2O_2 was orders of magnitude faster in freshwater systems with a higher organic load. The average half-lives were found to be 36 minutes in the system with the higher organic matter load and approximately 6 hours in the system with a low organic load. In the study reported here the opposite effect on degradation was seen using filtered seawater. H_2O_2 degraded 3X to 8X faster in filtered seawater than in unfiltered seawater indicating that by removing biota and organic matter the degradation process was faster. Many H_2O_2 formulations contain stabilizers. The results presented here indicate that H_2O_2 in the formulation Interox®Paramove™50 was more stable in water containing biota and organic material.

In most of our studies, the Interox®Paramove®50 was observed to degrade over time. In the short-term study reported here it was found that under static laboratory conditions the working concentration of 1200 $mg \cdot L^{-1}$ H_2O_2 in the formulation Interox®Paramove™50 was stable over 3 h at lower temperatures (5°C and 10°C) but not at the higher temperatures (15°C and 20°C). Bruno and Raynard (1994) used stabilized and unstabilized H_2O_2 (30% w/v, Merck product code 28519 7Y) in experiments and reported that aeration and higher temperatures (10°C or 15°C versus 4°C) increased the long-term breakdown of a working concentration (1400 $mg \cdot L^{-1}$) of H_2O_2 in seawater. Tort et al. (2004) used two concentrations (either 10 or 100 $mg \cdot L^{-1}$) of H_2O_2 (Akzo Nobel. Marietta, Georgia) and found there was no significant difference in the decline of H_2O_2 concentration at 15°C and 20°C in a static freshwater system with aeration and organic matter. The H_2O_2 concentrations were below the level of detection by 1 h post-treatment.

In conclusion, H_2O_2 degradation in the formulation Interox®Paramove™50 was slower in natural unfiltered seawater than in filtered seawater contrary to what has been previously reported in the literature. The calculated half-life of H_2O_2 was variable between studies which may be attributed to different water matrix at time of collection. The results presented here are preliminary and replication of the experiments should occur. We believe further degradation studies either monthly or seasonally might be useful to determine if water matrix is a factor in degradation. Water analysis for metals could be performed in conjunction with further studies.

ACKNOWLEDGEMENTS

The authors wish to thank Ken MacKeigan for analytical support. Funding was provided by Fisheries and Oceans Canada, Program for Aquaculture Regulatory Research (PARR) through the National Contaminants Advisory Group.

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FIGURES

Figure 1. Polystat chiller unit and experimental setup of jacketed beakers connected to chiller unit for hydrogen peroxide degradation study.

Figure 2. Degradation of 1200 mg·L^{-1} H_2O_2 over three hours.

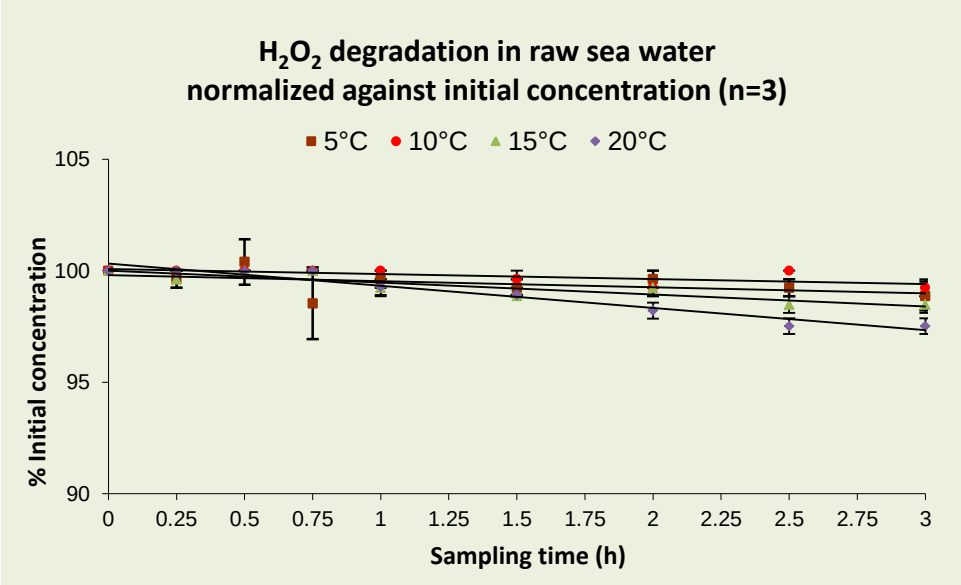


Figure 3. Degradation of 1200 mg·L^{-1} H_2O_2 over 175 days.

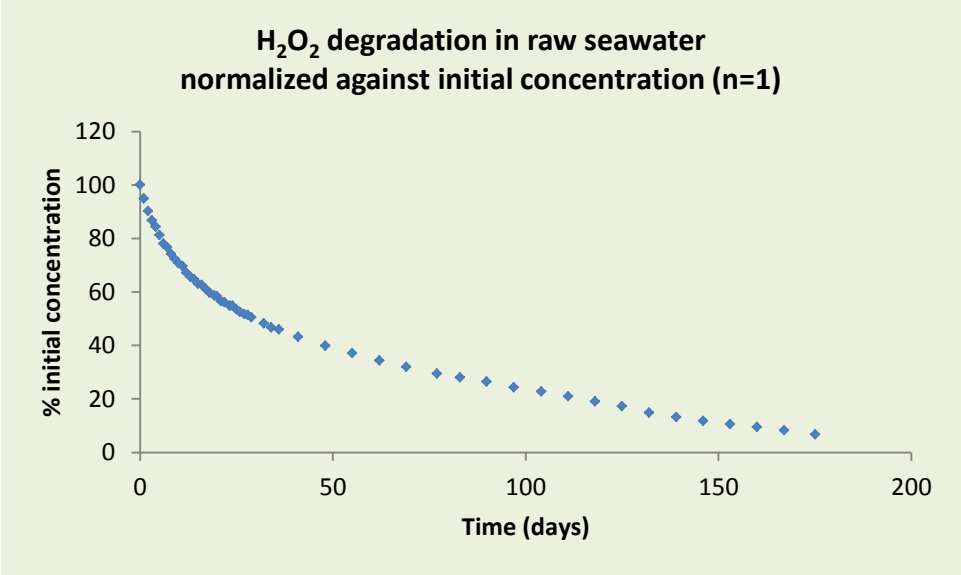


Figure 4. Degradation of H_2O_2 over 35 days.

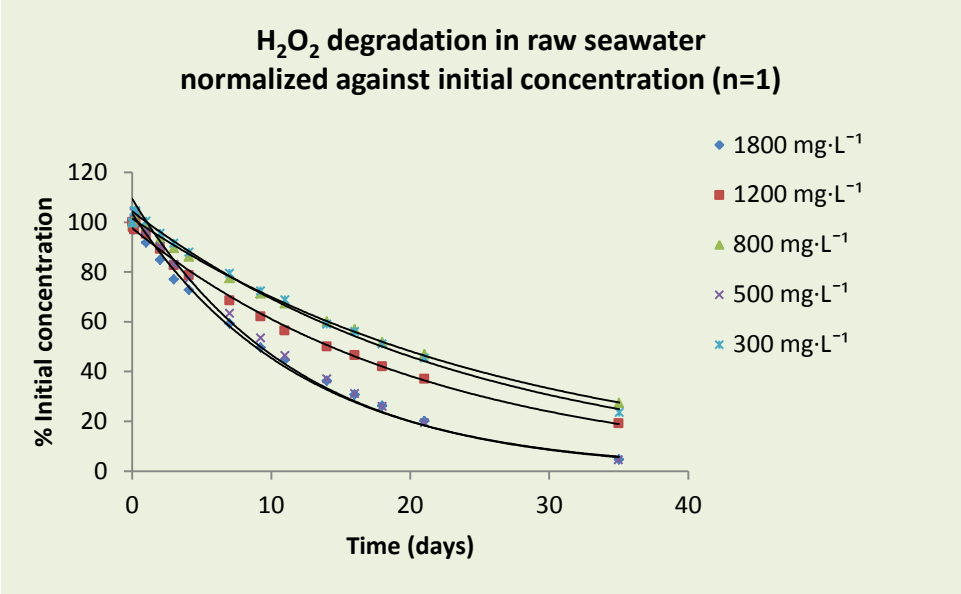
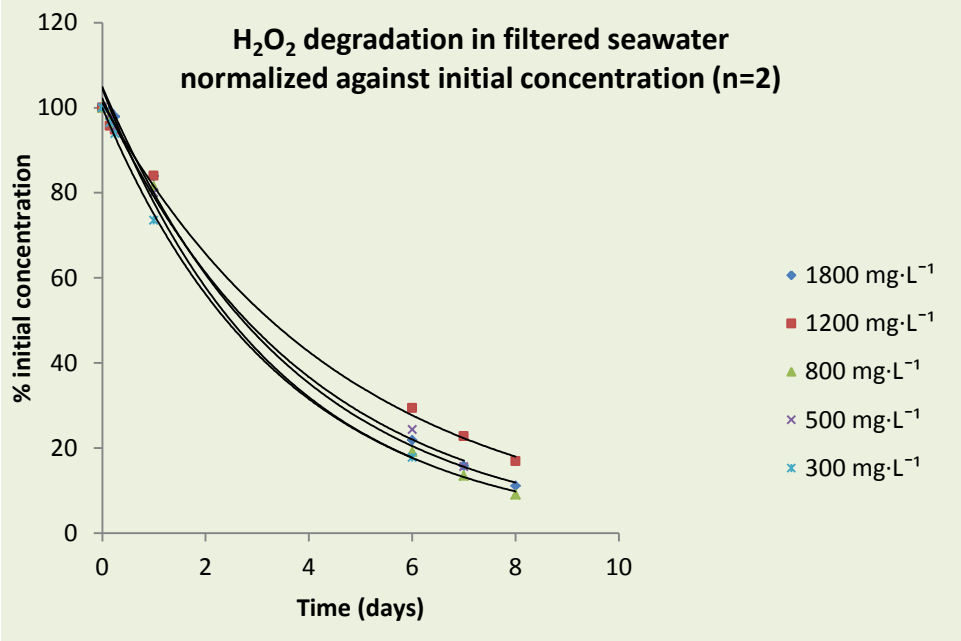


Figure 5. Degradation of H_2O_2 in filtered seawater over 8 days.



APPENDICES

Appendix 1

Table 1. Measured concentration ($mg \cdot L^{-1}$) of H_2O_2 over three days. Data are for three replicates.

Time-point (h)	Concentration at 5°C	Concentration at 10°C	Concentration at 15°C	Concentration at 20°C
0	1466	1466	1466	1401
0.25	1466	1466	1466	1401
0.5	1466	1466	1466	1401
0.75	1401	1466	1466	1401
1	1449	1466	1466	1386
1.5	1449	1466	1449	1386
2	1466	1466	1449	1371
2.5	1449	1466	1449	1356
3	1449	1449	1449	1356
0	1449	1466	1466	1386
0.25	1449	1466	1466	1386
0.5	1483	1466	1466	1386
0.75	1466	1466	1466	1386
1	1449	1466	1449	1386
1.5	1449	1466	1449	1371
2	1449	1449	1466	1356
2.5	1449	1466	1449	1356
3	1449	1466	1449	1356
0	1466	1466	1483	1386
0.25	1449	1466	1466	1386
0.5	1449	1466	1483	1386
0.75	1449	1466	1483	1386
1	1466	1466	1466	1371
1.5	1449	1449	1466	1371
2	1449	1449	1466	1371
2.5	1449	1466	1449	1356
3	1433	1449	1449	1356

Table 2a. Measured concentration ($mg \cdot L^{-1}$) of H_2O_2 over 175 days at 10°C.

Time (day)	0	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Conc.	1386	1314	1250	1203	1170	1125	1081	1062	1028	1004	981	966	931	911	898
Time (day)	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29
Conc.	873	867	844	828	812	807	782	777	759	759	741	729	716	712	701
Time (day)	32	34	36	41	48	55	62	69	77	83	90	97	104	111	118
Conc..	668	647	638	599	552	514	476	443	409	389	366	337	315	290	264
Time (day)	125	132	139	146	153	160	167	175							
Conc.	238	206	182	163	146	130	114	92							

Table 2b. Weekly drop in H_2O_2 measured concentration (% of initial concentration) over 175 days at 10°C.

Day	7	14	21	28	35	41	48	55	62	69	77	90
	23%	35%	44%	48%	53%	57%	60%	63%	66%	68%	71%	74%
Day	97	104	11	118	125	132	139	146	153	160	167	175
	76%	77%	79%	81%	83%	85%	87%	88%	90%	91%	92%	93%

Table 3a. Measured concentration ($mg \cdot L^{-1}$) of H_2O_2 in raw seawater over 35 days at 10°C.

Nominal concentration ($mg \cdot L^{-1}$)	Measured concentration															
	Time (hours)				Time (days)											
	0	1	3	6	1	2	3	4	7	9	11	14	16	18	21	35
300	312	310	326	326	313	298	285	274	248	226	215	184	175	159	142	73
500	529	527	550	550	510	478	440	409	336	283	246	196	164	137	103	24
800	823	823	839	862	812	768	737	708	638	588	554	494	469	426	386	225
1200	1275	1250	1238	1288	1214	1138	1054	1004	873	792	720	638	593	536	472	245
1800	1903	1875	1848	1875	1747	1614	1466	1386	1128	944	850	689	585	500	383	89

Table 3b. Weekly drop in H_2O_2 measured concentration (% of initial concentration) over 35 days at 10°C.

Nominal concentration ($mg \cdot L^{-1}$)	Day			
	7	14	21	35
300	21%	41%	54%	77%
500	36%	63%	81%	96%
800	22%	40%	53%	73%
1200	32%	50%	63%	81%
1800	41%	64%	80%	95%

Table 4. Measured concentration ($mg \cdot L^{-1}$) of H_2O_2 in filtered raw seawater over 8 days at 10°C.

Replicate 1

Nominal concentration ($mg \cdot L^{-1}$)	Measured concentration								
	0 h	3 h	6 h	Day 1	Day 4	Day 5	Day 6	Day 7	Day 8
300	325	310	299	203	32	16	8		
500	536	527	502	397	131	83	51	29	
800	850	850	828	701	335	250	176	126	86
1200	1275	1238	1238	1090	647	520	417	327	254
1800	1848	1875	1875	1678	988	802	631	488	366

Replicate 2

Nominal concentration ($mg \cdot L^{-1}$)	Measured concentration								
	0 h	3 h	6 h	Day 1	Day 2	Day 3	Day 6	Day 7	Day 8
300	324	316	310	274	229	196	106	88	71
500	543	534	527	465	405	350	212	140	129
800	856	828	812	689	540	419	149	103	68
1200	1289	1214	1192	1062	885	720	335	256	178
1800	1903	1848	1796	1466	1118	817	181	97	44

Appendix 2

Table 1. Half-lives of H_2O_2 in seawater.

Nominal concentration ($mg \cdot L^{-1}$)	Half-life (days) of H_2O_2				
	Raw seawater	Raw seawater	Filtered raw seawater 1	Filtered raw seawater 2	Mean of filtered raw seawater
1800		8.8	3.9	1.9	2.9
1200	28.3	14.3	3.7	3.0	3.3
800		19.0	2.7	2.4	2.5
500		9.2	1.8	4.0	2.9
300		18.0	1.2	3.7	2.5

Appendix 3

Table 1. H_2O_2 degradation line equations over three days. Concentration data Ln-transformed. Mean of three replicate concentrations at each time point.

Test temperature	H_2O_2 degradation over 3 days
5°C	$y = -0.0027x + 7.2841$ $R^2 = 0.2444$
10°C	$y = -0.0022x + 7.2907$ $R^2 = 0.4837$
15°C	$y = -0.0054x + 7.2939$ $R^2 = 0.8086$
20°C	$y = -0.0101x + 7.241$ $R^2 = 0.9474$

Table 2. H_2O_2 degradation line equations. Concentration data Ln-transformed.

A. Raw seawater.

Nominal concentration ($mg \cdot L^{-1}$)	Raw seawater - 175 day trial (degradation over 36 days)	Raw seawater - 35 day trial (degradation over 35 days)
1800		$y = -0.0822x + 7.5832$ $R^2 = 0.9894$
1200	$y = -0.0206x + 7.1239$ $R^2 = 0.9665$	$y = -0.0469x + 7.126$ $R^2 = 0.9975$
800		$y = -0.0372x + 6.7263$ $R^2 = 0.9982$
500		$y = -0.0849x + 6.3585$ $R^2 = 0.9897$
300		$y = -0.0409x + 5.784$ $R^2 = 0.9954$

B. Filtered seawater.

Nominal concentration ($mg \cdot L^{-1}$)	Filtered raw seawater 1 (degradation over 8 days)	Filtered raw seawater 2 (degradation over 8 days)
1800	$y = -0.1979x + 7.5933$ $R^2 = 0.9894$	$y = -0.4527x + 7.7141$ $R^2 = 0.9795$
1200	$y = -0.1966x + 7.1797$ $R^2 = 0.9947$	$y = -0.2387x + 7.1885$ $R^2 = 0.9927$
800	$y = -0.2811x + 6.8022$ $R^2 = 0.9936$	$y = -0.3134x + 6.8207$ $R^2 = 0.9982$
500	$y = -0.4094x + 6.3365$ $R^2 = 0.9665$	$y = -0.182x + 6.327$ $R^2 = 0.9864$
300	$y = -0.6172x + 5.8275$ $R^2 = 0.9993$	$y = -0.1892x + 5.7923$ $R^2 = 0.999$

Appendix 4

Table 1. Comparison of half-lives of H_2O_2 reported in the literature (NA-data not available).

Half-life	Formulation	H_2O_2 initial concentration	Experimental conditions	Temperature	Reference
~4.8 to ~7.0 days	30% w/v Merck product code 8596 (unstabilized & stabilized)	1400 $mg \cdot L^{-1}$	static/ aerated or not aerated/seawater	10°C	Bruno and Raynard (1994)
~3.3 to ~7.0 days	30% w/v Merck product code 8596 (unstabilized & stabilized)	1400 $mg \cdot L^{-1}$	static/ aerated or not aerated/ seawater	15°C	Bruno and Raynard (1994)
5 days	Interox®Paramove™50	NA	freshwater & saltwater	NA	Solvay Chemicals, Inc. (2012)
~8 h to ~11.5 h	^{18}O-labelled H_2O_2	100 nM	seawater in light and dark	NA	Moffett and Zafiriou (1990)
>48 hours (no decay after 48 h)	^{18}O-labelled H_2O_2	100 nM	0.2 µm filtered seawater	NA	Moffett and Zafiriou (1990)
~12 h to ~30 h	NA	10 $mg \cdot L^{-1}$	freshwater static/ aerated; static/ aerated/ organic matter; static/ organic matter	15°C or 20°C	Tort et al. (2004)
>6 days	NA	10 $mg \cdot L^{-1}$ or 100 $mg \cdot L^{-1}$	freshwater/ static	15°C or 20°C	Tort et al. (2004)
45 min to 1.5 h	2 $Na_2CO_3 \cdot 3H_2O_2$ (BioCare SPC; CAS#15630-89-4)	13 $mg \cdot L^{-1}$, 26 $mg \cdot L^{-1}$ or 39 $mg \cdot L^{-1}$	freshwater/ biofilter; low or high organic matter	~15.5°C	Pedersen et al. (2006)