JOURNAL OF AGRICULTURAL AND FOOD CHEMISTRY



# Survey of Total and Inorganic Arsenic Content in Blue Mussels (*Mytilus edulis* L.) from Norwegian Fiords: Revelation of Unusual High Levels of Inorganic Arsenic

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The present study reports the findings of unusual high levels of inorganic arsenic in samples of blue mussels (*Mytilus edulis* L.). A total of 175 pooled samples of blue mussels from various locations along the Norwegian coastline were analyzed for their content of total arsenic and inorganic arsenic. Total arsenic was determined using inductively coupled plasma mass spectrometry (ICPMS) following microwave-assisted acidic digestion of the samples. Inorganic arsenic was determined using an anion-exchange HPLC-ICPMS method following microwave-assisted alkaline solubilization of the samples. For the majority of the samples (78%) the concentration of total arsenic was below 3 mg $kg^{-1}$ wet weight (ww) and inorganic arsenic constituted <9% of the total arsenic (i.e., <0.25 mg $kg^{-1}$ ww). However, in some samples higher concentrations of total arsenic were found (up to 13.8 mg $kg^{-1}$ ww) and the inorganic arsenic content constituted up to 42% of the total arsenic (up to 5.8 mg $kg^{-1}$ ww). These are among the highest inorganic arsenic concentrations reported so far for marine animals. The findings of samples with concentrations of inorganic arsenic above 0.53 mg $kg^{-1}$ ww were restricted to sampling sites from two counties, Sogn and Fjordane and Hordaland, whereas samples from the rest of the country showed lower inorganic arsenic concentrations. Consumption of a meal containing 200 g of the blue mussels with the highest content of inorganic arsenic would for a 70 kg person lead to a 10% excess of the provisional tolerable weekly intake (PTWI) value for inorganic arsenic of 15 $\mu$g $kg^{-1}$ of body weight $week^{-1}$.

**KEYWORDS: Inorganic arsenic; total arsenic; blue mussels; HPLC-ICPMS; seafood safety**

## INTRODUCTION

Arsenic (As) is a ubiquitous element, introduced to the environment from natural sources such as volcanic activity and weathering of minerals (*1*) or from anthropogenic sources such as ore smelting, burning of coal, pesticide use, and the use of growth promoters (*2, 3*). As a result of natural metabolic processes in the biosphere As occurs as a large number of different inorganic or organic species. To date more than 40 different As species have been identified in the environment (*4*). Arsenobetaine is usually the predominant species in fish, bivalves, and crustaceans, whereas arsenosugars are the major As species in algae (*5*). Other As species, including the inorganic species arsenite, As(III), and arsenate, As(V), usually contribute with only a few percent or less in marine biota (*6, 7*). On the basis of acute toxicity studies in rodents it was found that organoarsenic compounds (e.g., arsenobetaine and arsenocholine) can be considered innocuous, simple methylated forms (e.g., monomethylarsonic acid, dimethylarsinic acid) show intermediate toxicity, and inorganic As compounds are the most toxic forms of As (*8*). In the context of human health risk assessment inorganic As may lead to serious adverse effects, including cancer. This applies to acute toxicity as well as to long-term effects resulting from low dose exposure (*9*). Seafood is the main source of As in the diet (*10*), and consequently accurate data for inorganic As in these sample types are important to make a correct dietary risk assessment analysis. In 1988 the World Health Organization (WHO) established a provisional tolerable weekly intake (PTWI) for long-term exposure to inorganic As, being a recognized human carcinogen, at 15 $\mu$g $kg^{-1}$ of body weight (*11*). The PTWI value has until now not been succeeded by any international implementation of regulatory limits in foodstuffs. Australia and New Zealand are the only two countries with national legislation to regulate the maximum level of inorganic As in seafood [crustaceans and fish, 2 mg $kg^{-1}$ wet weight (ww); molluscs and seaweed, 1 mg $kg^{-1}$ ww] (*12*).

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10.1021/jf073174+ CCC: $40.75 © 2008 American Chemical Society
Published on Web 02/01/2008

**Table 1.** Literature Data of Total and Inorganic As Concentrations in Mussels (Wet Weight Basis)

| origin of sample | total As, mg kg$^{-1}$ ww | inorganic As, mg kg$^{-1}$ ww | ref |
|---|---|---|---|
| Gulf of Venice, Italy | 1.1–4.6[a] | 0.01–0.05[a] | *15* |
| Venice Lagoon, Italy | 1.8–5.9[a] | <LOD–0.45[a] | *31* |
| Agean Sea, Greece | 1.3–5.1[a] | <0.008–0.15[a] | *32* |
| retail samples, U.K. | 1.3–4.2 | 0.02–0.45 | *33* |
| retail samples, Spain | 1.9–3.1 | 0.04–0.14 | *34* |
| retail samples, Belgium | 1.3–6.2 | 0.06–0.22 | *35* |
| retail samples, Australia | 2.6–8.7 | 0.05–0.56 | *36* |

[a] Concentrations given per dry matter in the original publication. A dry matter content of 150 g kg$^{-1}$ was used here to convert data to wet weight. LOD, limit of detection.


**Figure 1.** Map of Norway showing the locations of the various counties.

It is well-known that blue mussels are able to accumulate trace elements including As present in the surroundings (*13–15*). Generally As concentrations in the range of 1.1–8.7 mg kg$^{-1}$ ww have been reported for blue mussels collected in uncontaminated areas with inorganic As concentrations up to 1.5 mg kg$^{-1}$ ww (see **Table 1**). These concentration levels have led to the conclusion that the consumption of blue mussels does generally not pose a threat to human health with regard to inorganic As (*7*).

In the present study 175 blue mussel samples from Norwegian fiords were sampled from various locations along the Norwegian coastline and analyzed for their content of total and inorganic As. The obtained results are presented and used for a food safety risk assessment of the consumption of blue mussels using the PTWI value established by WHO.

## MATERIALS AND METHODS

**Standard Substances and Chemicals.** Deionized water (>17 MΩ cm$^{-1}$, Nanopure-system, Nanopure, Barnstead, U.K.) was used throughout the work. All chemicals were of pro analysi quality or better.

**Sampling and Sample Preparation.** A total of 175 pooled samples of blue mussels (*Mytilus edulis* L.) intended for human consumption were collected from various locations along the Norwegian coastline during the period 2004–2006 as part of the Norwegian monitoring program on undesirable substances. A map of Norway with the names of the various counties is given in **Figure 1**. A minimum of 50 mussel specimens were sampled at each sampling point and sent to the laboratory. The mussels were kept frozen from time of sampling until the beginning of the sample preparation. The lengths of all mussels were of eatable quality with lengths from 40 to 60 mm. The soft tissues of 25 mussel specimens from each location were pooled to form a test sample, which was homogenized, then freeze-dried, and subsequently thoroughly homogenized to a fine powder using a laboratory mill prior to analysis.

**Analysis of Samples.** For the determination of total As, subsamples (approximately 0.2 g) were submitted to microwave-assisted wet digestion using 2.0 mL of concentrated nitric acid (Merck, Darmstadt, Germany) and 0.50 mL of 30% w/w $H_2O_2$ (Merck) in an Ethos Pro microwave system (Milestone, Holger Teknologi, Oslo, Norway). An Agilent quadrupole ICPMS 7500c instrument (Yokogawa Analytical Systems Inc., Tokyo, Japan) was used as an As specific detector on mass $^{75}$As. Prior to the ICPMS determination, the sample digests were diluted to a final volume of 25 mL with water. Freshly prepared matrix-matched As standard solutions in 5% (v/v) nitric acid (Merck) were prepared by appropriate dilution of a 1000 mg L$^{-1}$ As certified As stock solution (Spectrascan, TeknoLab, Drøbak, Norway) and used to construct an external calibration curve. A diluted solution of a 1000 mg L$^{-1}$ rhodium stock solution (Spectrascan, TeknoLab) was added online and served as an internal standard to correct for instrumental drift (*16*).

For the determination of inorganic As a method recently applied for the analysis of various seafood samples and marine animal feedingstuffs was used (*17–19*). Briefly, an accurately weighed subsample (approximately 0.2 g) was placed in a vial of the microwave oven system (CEM Mars 5, LabConsult, Oslo, Norway), and 10 mL of a solution of 0.9 mol L$^{-1}$ sodium hydroxide (Merck) in 50% ethanol (Arcus, Oslo, Norway) was added. The samples were placed in the oven, which was adjusted to switch off the microwave power when the temperature of the mixture reached 90 °C, the approximate boiling point of the mixture. No pressure limitation was activated as the pressure reached only a few bars. The microwave treatment with the alkaline−alcoholic solution acts both as a solubilization reagent of the sample matrix and for the quantitative oxidation of As(III) to As(V), thus allowing the determination of inorganic As [As(III) + As(V)] as As(V). Prior to analysis, the samples were filtered through a 0.45 $\mu$m disposable syringe filter (Sartorius, Göttingen, Germany). Standard stock solutions of As(V) were prepared in water using a As(V) standard solution with an As concentration of 1000 ± 3 mg L$^{-1}$ as As (Spectrascan, TeknoLab). Quantification was based on peak area evaluation using external calibration with matrix-matched As(V) calibration standard solutions prepared in 0.9 mol L$^{-1}$ sodium hydroxide in 50% ethanol at three different concentration levels. The mobile phase solutions for anion exchange chromatography were prepared by dissolving an appropriate amount of ammonium carbonate (J. T. Baker, Philipsburg, NJ) in an aqueous 3% (v/v) methanol solution (Merck) followed by adjustment of pH to 10.3 with 25% (v/v) aqueous ammonia (Merck). For the liquid chromatographic separations an Agilent 1100 series quaternary pump, degasser, and autosampler (Agilent, Waldbronn, Germany) and a polymer-based strong anion exchange HPLC column, ICSep ION-120 (4.6 mm × 120 mm; 10 $\mu$m particles) (Transgenomics, San Jose, CA) were used. The outlet of the HPLC column was connected to the nebulizer of the ICPMS instrument by a short length of polyethylene tubing. **Table 2** gives the instrumental settings for the HPLC and ICPMS.

**Analytical Quality Assurance.** The trueness of the determination of total As was estimated from the concurrent analysis of the certified reference materials TORT-2, Lobster Hepatopancreas, and DORM-2 dogfish muscle (NRCC, Ontario, Canada). The obtained results for total As (average ± 2SD), 24.1 ± 1.5 mg kg$^{-1}$, $n$ = 16 (TORT-2), and 20.0 ± 1.6 mg kg$^{-1}$, $n$ = 8 (DORM-2), agreed with the target values of 21.6 ± 1.8 mg kg$^{-1}$ (TORT-2) and 18.1 ± 1.1 mg kg$^{-1}$ (DORM-2). The limit of quantification (LOQ) was estimated at 0.07 mg kg$^{-1}$ ww using the standard deviation ($s$) obtained from repeated analysis of blank samples ($N \geq 20$) (LOQ = 10$s$). The precision (i.e., repeatability) was calculated as 2% as 2 × RSD (%) from replicate analysis of samples with total As concentration at 50 mg kg$^{-1}$ ww ($N$ = 10).

For the determination of inorganic As a LOQ at 0.002 mg kg$^{-1}$ ww was estimated (10$s$). The precision (i.e., repeatability) of the method was estimated at 7.5% as × RSD (%) from repeated analysis of a sample at 0.31 mg kg$^{-1}$ ww. Currently, no reference materials with certified values for inorganic As are available. To assess the accuracy

**Table 2.** Instrumental Settings for the HPLC-ICPMS Method

| | |
|---|---|
| ICPMS Settings | |
| RF power, W | 1600 |
| carrier gas flow, L $min^{-1}$ | 1.15–1.25 |
| plasma gas flow, L $min^{-1}$ | 15 |
| auxilary gas flow, L $min^{-1}$ | 1 |
| nebulizer | Meinhard concentric |
| spray chamber | water-cooled double pass |
| spray chamber temperature, °C | 2 |
| interface cones | platinum |
| lens voltage, V | 2–3 |
| mass resolution, u | 0.8 |
| integration time, s | 1000 |
| HPLC Settings | |
| injection volume, $\mu$L | 25 |
| operating pressure, bar | 45–50 |
| mobile phase concn, mmol $L^{-1}$ $(NH_4)_2CO_3$ | 40 |
| mobile phase flow, mL $min^{-1}$ | 1 |

of the method, analysis of the inorganic As content in TORT-2 was included in each batch of samples analyzed. The obtained mean value (0.20 mg $kg^{-1}$, RSD = 12%, $N = 30$) agreed well with previous results obtained (0.19 mg $kg^{-1}$, RSD = 7.3%, $N = 20$) (*19*).

## RESULTS AND DISCUSSION

**Determination of Total As and Inorganic As in Blue Mussels.** The obtained results are presented in **Table 3** with number of samples, median, and concentration range per county where the sampling took place. The map of Norway in **Figure 1** shows the locations of the various counties. The data reveal that in most counties the concentration of total As and inorganic As for all samples is below 3.5 and 0.25 mg $kg^{-1}$ ww, respectively. This is in concordance with the literature data shown in **Table 1**. However, in some samples from the Møre and Romsdal, Sogn and Fjordane, and Hordaland counties elevated concentrations of total and inorganic As were found. The highest concentration was found in a blue mussel sample from Frønningen in the Sognefiord area (Sogn and Fjordane) with total and inorganic As concentrations of 13.8 and 5.8 mg $kg^{-1}$ ww, respectively. In this sample inorganic As constitutes 42% of the total As content and is among the highest inorganic As concentrations reported in marine animals (*20*). **Figure 2** shows as an example a chromatogram obtained from the analysis of a sample from Sogn and Fjordane with total and inorganic As concentrations of 3.6 and 0.74 mg $kg^{-1}$, respectively, corresponding to an inorganic As content of 21% of the total As. The inorganic As [As(V)] peak elutes at approximately 6.8 min and is baseline separated from other organoarsenic compounds eluting earlier in the chromatogram.

A large year-to-year variation could be found for samples from the same location. For example, the sample from Langeneset (Sogn and Fjordane) taken in August 2005 had concentrations of 8.7 and 3.7 mg $kg^{-1}$ for total and inorganic As, respectively, whereas the sample collected in August the following year had concentrations of only 1.98 mg $kg^{-1}$ (total As) and 0.012 mg $kg^{-1}$ (inorganic As). Arsenic uptake by mussels occurs primarily via the food chain (*21*). It is therefore suggested that annual variation in the amount of aquatic microbes and microalgae may play a role in the annual variation for total and inorganic arsenic in samples from the same locations as well as the large variation observed for various locations within the same county. A study on the transfer and possible biotransformation of arsenic species from the water column to phytoplankton and mussels would bring valuable information for the understanding of this phenomenon in further details.

**Figure 3** shows a plot of the inorganic As concentration versus the total As concentration for all 175 samples analyzed. For samples with total As concentrations below 3 mg $kg^{-1}$ the inorganic As concentrations were in all cases below 0.25 mg $kg^{-1}$. For total As concentrations above 3.0 mg $kg^{-1}$ a significant linear relationship between the inorganic and total As (linear correlation coefficient, $r^2 = 0.9145$) was found as illustrated by the dotted line in **Figure 3**. The data indicate the presence of a biotransformation threshold value for inorganic As in the mussels. At low inorganic As exposure the animals are capable of biotransforming the inorganic As to organoarsenic species, presumably as a detoxification process, but at higher exposure to inorganic As the biotransformation threshold is exceeded and the animals deposit and accumulate the inorganic As in their tissues. In analogy, supporting results for this hypothesis were obtained by Geiszinger et al. (*22*) in a laboratory experiment, where samples of the brown algae *Fucus serratus* were exposed to different levels of As(V), and suggested that the alkylation process at high exposure levels (100 $\mu$g $L^{-1}$) became saturated, leading to accumulation of inorganic As in the plant. At lower exposure levels (20 $\mu$g $L^{-1}$) organoarsenic compounds, dimethylarsinate (DMA), and arsenosugars were the main metabolites. In a similar experiment the marine polychaete *Arenicola marina* was exposed to different concentrations of As(V) (*23*). It was found that most of the accumulated As was present as the inorganic forms As(III) and As(V), again indicating a limited capability to biotransform the inorganic As to organoarsenic species. However, in contrast to these reports, exposure experiments with As compounds performed by Gailer et al. (*24*), including As(III) and As(V), to blue mussels, did not show any significant accumulation of inorganic As compounds in the mussel tissues, even at an exposure level of 100 $\mu$g $L^{-1}$. It is suggested that future experiments should address this issue to study the factors affecting the uptake of inorganic As by blue mussels and other marine organisms.

**Food Safety Aspects.** A PTWI value of 15 $\mu$g $kg^{-1}$ of body weight was established for inorganic As by WHO in 1988 (*11*). The consumption of a meal consisting of 200 g of the mussels with the highest inorganic As concentration (i.e., 5.8 mg $kg^{-1}$ ww) would lead to an intake of 1160 $\mu$g of inorganic As, thus exceeding the PTWI value for a person of 70 kg body weight by approximately 10%. In this case contribution from other dietary sources of inorganic As has not been taken into account. Recently, Karouna-Renier et al. (*25*) established a screening value (SV) for inorganic arsenic in a study of contaminants in shellfish in Florida. The SVs are concentrations of chemicals that are of potential public health concern (*26*) based on the cancer slope factor (CSF) for inorganic arsenic of 1.5 (mg $kg^{-1}$ $day^{-1})^{-1}$ (*27*). In the present study 50% of the samples have inorganic arsenic concentration exceeding this SV. The CSF can also be used to estimate the carcinogenic risk associated with continuous consumption of blue mussels with the highest inorganic arsenic levels. Accordingly, the carcinogenic risk for a 70 kg person with a weekly consumption of 200 g of blue mussels with 5.8 mg $kg^{-1}$ inorganic arsenic would be as high as 0.0036, thus clearly exceeding the generally accepted safe level of $10^{-6}$ (*27*). This value is 20 times higher than the carcinogenic risk reported for male adults in Catalonia, Spain, estimated from their dietary arsenic exposure from various types of seafood (*28*). However, the degree of bioaccessibility of the inorganic As present in the mussels has also to be taken into account to perform a more complete food safety risk assessment. Future studies, using an in vitro simulation of the gastrointestinal tract, will address the question of bioaccessibility of inorganic

**Table 3.** Results (Median and Range) for Total As, Inorganic As, and the Fraction of Inorganic As for Blue Mussel Samples from Various Counties in Norway[a]

| | | total As, mg kg$^{-1}$ ww | | inorganic As, mg kg$^{-1}$ ww | | inorg As, % of total As |
|---|---|---|---|---|---|---|
| county | *N* | median | range | median | range | range |
| Troms | 7 | 2.0 | 1.6–2.7 | 0.007 | 0.003–0.017 | 0.16–0.75 |
| Finnmark | 2 | 2.1 | 1.9–2.3 | 0.010 | 0.010–0.010 | 0.10–3.5 |
| Nordland | 43 | 2.0 | 1.3–3.5 | 0.008 | 0.001–0.046 | 0.10–4.4 |
| Sør-Trøndelag and Nord-Trøndelag | 31 | 1.9 | 1.3–4.4 | 0.010 | 0.001–0.24 | 0.10–8.9 |
| Møre og Romsdal | 12 | 2.5 | 1.5–6.1 | 0.037 | 0.007–0.53 | 0.50–6.9 |
| Sogn og Fjordane | 24 | 2.9 | 1.4–13.8 | 0.088 | 0.002−5.8 | 0.90–42 |
| Hordaland | 28 | 3.3 | 1.6–10.0 | 0.31 | 0.002–3.9 | 1.8–39 |
| Rogaland | 10 | 2.1 | 1.4–3.2 | 0.010 | 0.001–0.060 | 0.40–2.5 |
| Aust-Agder and Vest-Agder | 13 | 1.9 | 1.3–2.6 | 0.008 | 0.002–0.020 | 0.40–2.3 |
| Østfold | 5 | 2.0 | 1.2–2.3 | 0.006 | 0.001–0.040 | 0.15–2.0 |
| all counties | 175 | 1.7 | 1.2–13.8 | 0.011 | 0.001−5.8 | 0.10–42 |

[a] Please see **Figure 1** for the location of the various counties.

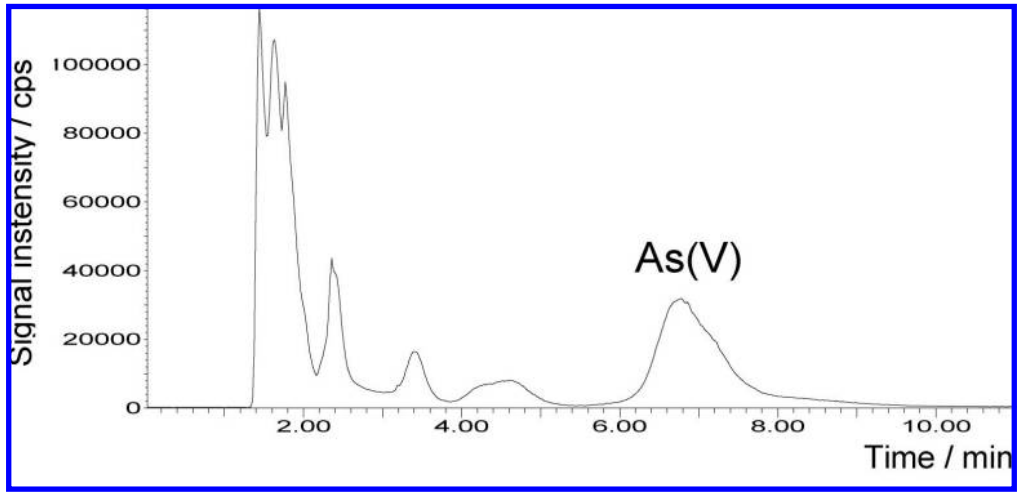


**Figure 2.** Chromatogram of blue mussel sample with 3.6 mg kg$^{-1}$ ww total As and 0.74 mg kg$^{-1}$ ww inorganic As.

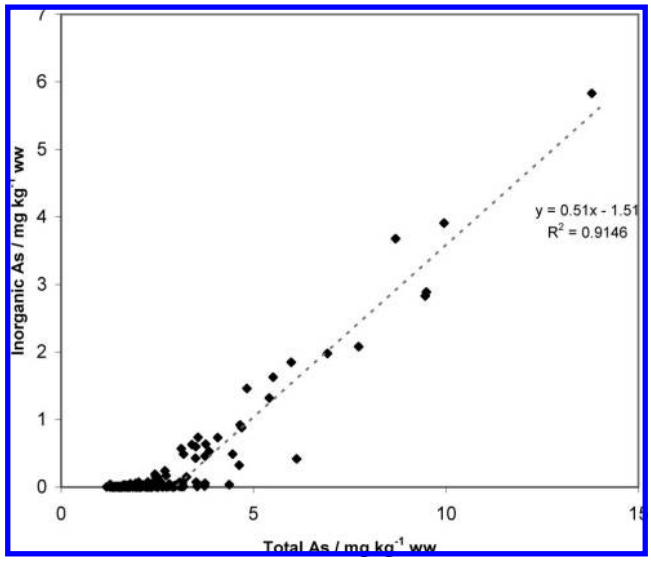


**Figure 3.** Plot of total As concentration versus inorganic As concentration in the 175 blue mussel samples from the present study (one dot per sample). The dotted line is the linear regression of samples with total As concentrations above 3 mg kg$^{-1}$ ww.

As from mussels. Previously, Laparra et al. (*29, 30*) reported a bioaccessibility of inorganic As of up to 88% in various forms of seaweed samples. If this rather high rate of bioaccessibility is true for inorganic As in blue mussels as well, excessive consumption of blue mussel with high levels of inorganic As may pose a potential threat to human health.

In conclusion, the analysis of 175 pooled samples of blue mussels from Norway showed a large variation in the contents of total and inorganic As, respectively. For mussels with total As concentrations below 3.0 mg kg$^{-1}$ ww, the inorganic As concentration was below 0.25 mg kg$^{-1}$ ww. However, for samples with total As concentrations above 3.0 mg kg$^{-1}$ ww increasing inorganic As concentrations was observed, and a linear relationship was found for inorganic versus total As concentration. The results suggest the presence of a biotransformation threshold for inorganic As in the mussels. However, further investigations are necessary to verify this and to elucidate the main factors responsible for this phenomenon. Additionally, the need for further investigations to study the bioavailability of the inorganic As is called upon to enable a better risk assessment analysis of consumption of blue mussels with high levels of inorganic arsenic.

## ACKNOWLEDGMENT

Siri Bargård, NIFES, is thanked for skillful help with the analysis of the mussel samples.

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**Received for review October 30, 2007. Accepted December 21, 2007. The Norwegian Food Authority is thanked for funding the survey projects 2004–2006.**

JF073174+