

METALS AND OTHER ELEMENTS

Survey for Cadmium, Cobalt, Chromium, Copper, Nickel, Lead, Zinc, Calcium, and Magnesium in Canadian Drinking Water Supplies

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A second national survey was done to ascertain the levels of Cd, Co, Cr, Cu, Ni, Pb, Zn, Ca, and Mg in Canadian drinking water supplies. Raw, treated, and distributed water samples collected from 71 municipalities across Canada were analyzed both by atomic absorption spectrophotometry using the direct method and by an APDC-MIBK extraction procedure. As in the first national survey, the amounts of trace metals found in the 3 types of water samples were essentially the same. Contamination of the water supplies with these metals except Cu and Zn was minimal during treatment and distribution. For Canadian drinking water, the median and extreme values expressed as ng metal/mL water were: Cd ≤ 0.02 (≤ 0.02 -0.07), Co ≤ 2.0 (≤ 2.0 -6.0), Cr ≤ 2.0 (≤ 2.0 -4.1), Cu ≤ 10 (≤ 10 -900), Ni ≤ 2.0 (≤ 2.0 -69.0), Pb ≤ 1.0 (≤ 1.0 -79.7), and Zn ≤ 10 (≤ 10 -750). Hardness values as mg CaCO₃/L ranged from 6.7 in St. John's, Newfoundland, to 328.3 in Portage La Prairie, Manitoba. The median values for the Canadian drinking water supplies were well below the maximum permissible limits set by Health and Welfare Canada and the World Health Organization.

There is increasing concern about the quality of drinking water in several parts of the world. Epidemiological studies in recent years have indicated a strong association between the occurrence of certain cancers and cardiovascular diseases with the presence of trace metals such as Cd (1-3), Cr (4), or Pb (1-3, 5) and with the hardness, in terms of Ca (6, 7) or Mg (4, 6, 8), of the local drinking water supplies. The chronic and cumulative toxicities of trace metal pollutants, such as Cd and Pb, are well documented (1, 9). Lately, Ni has also been shown to be toxic (10, 11). Excessive or deficient levels of essential micronutrients such as Co, Cr, Cu, and Zn may also have detrimental effects on health (9, 12). Therefore, Health and Welfare Canada has initiated a comprehensive nationwide program to determine both the organic and inorganic contaminants in the nation's water supplies.

This paper presents the results of a second national survey for Cd, Co, Cr, Cu, Ni, Pb, Zn, Ca, and Mg in Canadian drinking water supplies. During the first national survey (13), it became evident that the concentrations of elements such as Cd, Cr, and Pb in the majority of the drinking water samples analyzed were at or below the detection limit of the direct graphite furnace atomic absorption spectrophotometric procedure. Therefore, an ammonium pyrrolidinedithiocarbamate-methyl isobutyl ketone-graphite furnace atomic absorption (APDC-MIBK-GFAA) spectrophotometric procedure, developed in our laboratory (14), was also used to determine Cd, Co, Cr, Pb, and Ni in this study.

As in the previous survey (13), samples of treated and distributed water were obtained to assess the impact of the treatment and distribution system on the quality of drinking water. The water supplies from 71 Canadian municipalities encompassing 55% of the Canadian population were included in this survey. Of these, 45 locations were not sampled in the first national survey. Distributed water samples from the remaining 26 municipalities were collected from different points in the distribution system.

Experimental

Apparatus

A Perkin-Elmer atomic absorption spectrophotometer, Model 603, equipped with a heated graphite atomizer, HGA-2100, and a deuterium background corrector was used for determining levels of Cd, Co, Cr, Ni, and Pb. Nitrogen purge gas was used for all elements except Cr, for which argon was used. Cd and Pb were determined in the gas interrupt mode. The same instrument in the flame mode (air-acetylene), equipped with a 4 in. single slot burner, permitted determinations of Cu, Zn, Ca, and Mg. Single element hollow cathode lamps were used as narrow line sources for all elements except Cd and Pb. For Cd and Pb, electrodeless discharge lamps were used to increase the sensitivity and detection limit.

All extraction studies were done in 125 mL Pyrex

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glass separatory funnels fitted with Teflon stopcocks and polyethylene stoppers.

Reagents and Solutions

For high-purity water, tap water was passed through a cellulose adsorbent and 2 mixed bed ion-exchange columns and distilled in a Corning all-glass distillation unit.

Appropriate standard solutions of Cd, Co, Cr, Cu, Ni, Pb, Zn, Ca, and Mg were prepared in 1% HNO₃ (Baker Ultrex) immediately before analysis by serial dilution of the 1000 mg/L stock solution (Fisher Scientific) stored in polyethylene bottles.

A 20% ammonium citrate buffer solution was prepared by dissolving 200 g dibasic ammonium citrate (ACS grade, Fisher Scientific) in 500 mL water and adjusting the pH to 7.2 with concentrated NH₄OH. Solutions were diluted to 1 L with water, and extracted for 3 min with 5 mL of a 1% purified APDC solution and 25 mL MIBK. This operation was repeated until the aqueous layer was virtually free of any trace metal impurities, as determined by GFAAS. The aqueous layer was stored in a 1 L pre-cleaned polyethylene bottle.

A 2% APDC solution was prepared by dissolving 20 g APDC (Baker Analyzed) in 1 L water and extracting for 3 min with 50 mL MIBK. The ketone layer was discarded, and the extraction was repeated until the organic phase became colorless. The aqueous phase, when stored in a pre-cleaned polyethylene bottle at room temperature, was stable for at least one month.

ACS grade MIBK (or 4-methyl-2-pentanone) supplied by Fisher Scientific was used without further purification.

Selection of Sampling Locations

Seventy-one municipalities were selected across Canada including all the large centers of population and a number of remote locations (e.g., in the North West Territories and the Yukon—included in this survey for the first time). For each Province, the percent of the population whose water supply was analyzed in this survey was: Alberta, 57; British Columbia, 60; Manitoba, 61; New Brunswick, 34; Newfoundland, 26; Northwest Territories, 26; Nova Scotia, 42; Ontario,

56; Prince Edward Island, 27; Quebec, 61; Saskatchewan, 35; and Yukon, 64. The total represents 55% of the Canadian population.

Sample Collection

The water samples were collected during the day in August and September 1977, and stored in the Nalgene® linear polyethylene 1000 mL screw-cap collection bottles. Before use, the bottles were cleaned sequentially with a detergent wash, tap water rinse, 24 h soak in 1% Baker Ultrex HNO₃, and several high-purity water rinses (15). After the cleaning operation, blanks for each bottle were prepared in high-purity water in 1% HNO₃. Bottles having detectable levels of the trace metals of interest were rejected. The bottles to be used for collection were then dried at 100°C for 1 h, cooled to room temperature, capped, and labeled. After HNO₃ (1 mL acid/100 mL water sample) was added to the partly filled bottles, each bottle was filled to the brim with the water sample to avoid any air space. Duplicate grab samples of raw (i.e., water before treatment) and treated water (i.e., water immediately after treatment) were collected from each municipality. Also, from each municipality, duplicate distributed water samples were obtained from 3 points in the distribution system: one half mile from the treatment plant; 1 mile from the plant; and 2 miles from the plant. Before distributed water samples were collected, the taps were run to waste at their maximum flow rate for 5 min to clear the lines of overnight standing water. To ensure consistency in sample collection, the number of technologists conducting the sampling was kept to a minimum, and the sampling procedures were reviewed in detail with the personnel involved in the study.

As soon as the samples were collected and preserved, they were transported by air in heavy plastic coolers containing gel-type freezer packs. Immediately upon receipt in the laboratory, samples were refrigerated at 4°C.

Analytical Procedure

Two methods, direct GFAA and APDC-MIBK-GFAA, were used to determine concentrations of Cd, Co, Cr, Ni, and Pb.

Table 1. Optimum solution conditions for the quantitative extraction of some metal-APDC chelates from aqueous solution into MIBK^a

Element	pH Range	APDC/metal ^b	Extraction time, c s	Chelate stability, d h
Cd	2.0–8.0	$\geq 5 \times 10^3$	≥ 5	2 (4–8); 0.5 (2–3)
Co	1.0–8.0	$\geq 1 \times 10^5$	≥ 5	960 (1–5); 78 (6–8)
Cr	1.8–3.0	$\geq 2 \times 10^5$	≥ 180	300 (1.8–3.0)
Ni	1.0–8.0	$\geq 1 \times 10^3$	≥ 60	48 (2–4); 20 (5–8); 3 (1)
Pb	3.5–8.0	2.5×10^2 – 2.5×10^4	≥ 5	3 (4–8); 0.5 (3.5)

^a Subramanian and Méranger (14).

^b Concentration in weight/volume.

^c Extraction time represents time required for shaking separatory funnels manually.

^d Chelate stability represents period during which absorbance values for metal remain the same as those obtained immediately after extraction. Numbers in parentheses represent pH range.

Table 2. Optimized instrumental and analytical parameters for determination of Cd, Co, Cr, Ni, and Pb by GFAAS

Parameters	Element									
	Cd		Co		Cr		Ni		Pb	
Line, nm	D ^a	E ^b	D	E	D	E	D	E	D	E
Slit, nm	228.8	228.8	240.7	240.7	357.9	357.9	232.0	232.0	283.3	283.3
Charring temp., °C	0.7	0.7	0.2	0.2	0.7	0.7	0.2	0.2	0.7	0.7
Charring times, s	300	bypass	500	500	600	500	500	500	500	400
Atomization temp., °C	30	—	20	20	20	20	20	20	10	20
Atomization time, s	1400	1600	2600	2600	2600	2600	2600	2600	2200	2200
Sensitivity, 1×10^{-12} g	5	7	10	10	10	10	10	10	6	7
Detection limit, μ g/mL	0.02 \pm 0.01	0.05 \pm 0.02	2.0 \pm 0.3	2.0 \pm 0.3	2.0 \pm 0.3	2.0 \pm 0.4	2.0 \pm 0.4	2.0 \pm 0.4	1.5 \pm 0.3	1.0 \pm 0.3
Linear range, ng/mL	0-1	0-1	0-100	0-100	0-60	0-60	0-200	0-200	0-10	0-10

^a Direct injection of 20 μ L water sample into graphite tube.

^b 25 mL water sample extracted into 5 mL MIBK after complexation with APDC; 20 μ L of extracted MIBK phase injected into graphite tube.

^c Temps. represent meter settings on control panel of HGA-2100 temp. programmer. A 30 s drying at 100°C was used for metals except Cd, for which a 50 s drying at 100°C was used when injecting organic solution.

^d Mass for 0.0044 absorbance with purge gas flow in the normal mode (50 mL/min) except Cd and Pb for which interrupt mode was used. Nitrogen was purge gas for all elements except Cr; argon was used for Cr.

^e Detection limit = 2 \times std. dev. of blank. Value for extraction refers to organic phase. Its value in aqueous phase depends on ratio of aqueous phase vol. to organic phase vol. Uncertainty in detection limit represents value at 95% confidence interval.

In the direct method, 20 μ L of each sample was injected directly into a pyrocoated graphite tube with an Eppendorf pipet fitted with a disposable polypropylene tip. Before use, the tips were decontaminated from metal traces by overnight soaking in 1% HNO₃, followed by several rinses with high-purity water.

Details of the APDC-MIBK-GFAA extraction procedure have been reported elsewhere (14). A 25 mL aliquot of the water sample was pipetted into the separatory funnel and extracted with 5 mL water-saturated MIBK after the addition of citrate buffer and APDC under optimum solution conditions given in Table 1.

In both methods, measurements were made using the sequential dry, char, and atomize conditions (Table 2). The peak absorbance mode was used for Co, Cr, Ni, and Pb; Cd was determined in the concentration mode at a scale expansion of ten. The amount of each metal in each type (raw, treated, and distributed) of water sample was obtained by comparison with linear working curves. Working curves were prepared for each element using fresh aqueous standards in 1% HNO₃ for the direct method, and fresh MIBK extracts of standard solutions for the extraction procedure.

A blank (a calibration standard corresponding to 10% detection limit of each element) and a repeat sample for every 10 samples were included in the study plan to check for contamination or variation in sensitivity and repeatability. It was not possible to ensure the accuracy of the 2 methods with certified reference standards because no drinking water standards are available for the elements in question. However, an indirect measure of the accuracy was obtained from recovery studies on each of 20 raw, treated, and distributed water samples spiked with 10 and 20 \times the detection limit of each element.

Cu, Zn, Ca, and Mg were determined in the flameless mode, as described in Environment Canada's *Analytical Methods Manual* (16). A 1 mL solution of 0.29% lanthanum oxide in 0.5% HCl and 0.25% HNO₃ was added to each mL of sample before measuring Ca and Mg to mask matrix interferences.

Results and Discussion

The technique of GFAA spectrometry was chosen for Cd, Co, Cr, Ni, and Pb because it had the sensitivity required for determining these metals at the ng/mL or sub-ng/mL levels. Also, the technique has the high selectivity and precision required for reliable analytical results. The coefficient of variation of each point in the calibration curve used in the direct and the APDC-MIBK extraction procedure for the determination of these metals by GFAA was 3-5%. The coefficients of variation at the 95% confidence interval for the blank and the repeat samples were 5 and 7-10%, respectively. Recoveries for the spiked samples were 100 \pm 10%.

The validity of using the direct GFAA method for determining metals in fresh waters has been

endorsed by a number of workers (15, 17-19). However, caution should be exercised when applying this method to samples of varied composition because of possible physical and chemical interferences (20-22). Recently, we have shown the suitability of using the direct method as a rapid screening technique for determining Cd (23) and Pb (unpublished results) in a large number of drinking water samples. As a further check, the APDC-MIBK extraction procedure developed in our laboratory (14) has been used in conjunction with the direct GFAA procedure for all the above metals.

The operating and analytical parameters for the determination of Cd, Co, Cr, Ni, and Pb using GFAA by both the direct and the APDC-MIBK extraction methods are given in Table 2. Although the optimum atomization temperature for the direct determination of Cd was 1600-1800°C, a temperature of 1400°C was chosen to eliminate chemical interference (23). In the extraction procedure, the ashing stage was bypassed because Cd was lost during the ashing process (14).

Tables 3 and 4 present the results for the determinations of Cd, Co, Cr, Ni, and Pb by the direct method and the APDC-MIBK extraction procedure, respectively. The median and extreme values are given for raw (lake, river, or well as the source of drinking water), treated (water immediately after treatment but before reaching the distribution system), and distributed (at the consumer's tap) water supplies for each Canadian Province. The results in Tables 3 and 4 show good agreement between the direct injection method and the APDC-MIBK extraction procedure. In general, the waters sampled contained Cd, Co, Cr, Ni, and Pb at or below the detection limit, with the following exceptions. The raw water obtained from Yellowknife, NWT, contained 0.9 and 0.8 ng Cd/mL as determined by the direct method and by the APDC-MIBK extraction procedure, respectively. However, these values decreased to ≤ 0.06 ng/mL during treatment and distribution. The Ni concentrations in the raw water from Selkirk, Man.; Baie Verte, Nfld.; Sudbury, Ont.; and Beaulac and Disraeli, PQ, were 6.8, 4.1, 73.5, 5.4, and 14.0 ng/mL, respectively, by the direct method. The corresponding values by the extraction method were 5.9, 3.2, 72.4, 4.6, and 12.0 ng/mL. The above values remained more or less the same in the treated and distributed waters, showing that the treatment methods (mainly lime, alum, or soda ash) used were not effective in removing Ni from the raw source. About 19-20 ng Pb/mL

Table 3. Median and extreme values for the direct determination of Cd, Co, Cr, Ni, and Pb in raw, treated, and distributed Canadian water supplies by GFAAs

Province ^a	Cd, ng/mL	Co, ng/mL	Cr, ng/mL	Ni, ng/mL	Pb, ng/mL
Manitoba	R (9) ^b T (10) D (11) R (10) T (10) D (24) R (8) T (4) D (7)	≤ 2.0 ($\leq 2.0-3.0$) ^d ≤ 2.0 ($\leq 2.0-5.0$) ≤ 2.0 ($\leq 2.0-6.0$) ND ND ND ND ND ND	≤ 2.0 ($\leq 2.0-3.0$) ND ND ND ≤ 2.0 ($\leq 2.0-4.0$) ND ND ND ND	4.0 ($\leq 2.0-6.8$) 2.0 ($\leq 2.0-4.0$) 2.0 ($\leq 2.0-3.3$) ≤ 2.0 ($\leq 2.0-4.1$) ≤ 2.0 ($\leq 2.0-5.3$) ≤ 2.0 ($\leq 2.0-4.6$) ND ND ND	≤ 1.0 ($\leq 1.0-1.0$) ≤ 1.0 ($\leq 1.0-1.0$) ≤ 1.0 ($\leq 1.0-5.0$) 1.0 ($\leq 1.0-2.0$) 1.0 ($\leq 1.0-16.0$) ND ≤ 1.0 ($\leq 1.0-2.9$) ND ≤ 1.0 ($\leq 1.0-4.0$) ≤ 1.0 ($\leq 1.0-2.5$) ≤ 1.0 ($\leq 1.0-2.0$) ND ≤ 1.0 ($\leq 1.0-3.0$) ≤ 1.0 ($\leq 1.0-3.0$) ≤ 1.0 ($\leq 1.0-2.0$)
Newfoundland					
Northwest Territories					
Ontario	R (31) T (31) D (52) R (36) T (32) D (58)	≤ 2.0 ($\leq 2.0-3.0$) ND ND ND ND ND	≤ 2.0 ($\leq 2.0-5.0$) ≤ 2.0 ($\leq 2.0-4.0$) ND ≤ 2.0 ($\leq 2.0-4.3$) ≤ 2.0 ($\leq 2.0-4.0$) ≤ 2.0 ($\leq 2.0-4.1$)	≤ 2.0 ($\leq 2.0-73.5$) ≤ 2.0 ($\leq 2.0-69.6$) ≤ 2.0 ($\leq 2.0-69.0$) ≤ 2.0 ($\leq 2.0-14.0$) ≤ 2.0 ($\leq 2.0-11.0$) ≤ 2.0 ($\leq 2.0-25.4$)	≤ 1.0 ($\leq 1.0-4.0$) ≤ 1.0 ($\leq 1.0-2.5$) ≤ 1.0 ($\leq 1.0-2.0$) ND ≤ 1.0 ($\leq 1.0-3.0$) ≤ 1.0 ($\leq 1.0-3.0$) ≤ 1.0 ($\leq 1.0-2.0$)
Quebec					

^a Values for Cd, Co, Cr, Ni, and Pb in the raw, treated, and distributed water supplies of Alta, BC, NB, NS, PEI, Sask., and Yukon were below the instrumental detection limits given in Table 2 except for the treated water supplies of BC and the raw water supplies of PEI where the highest values of Pb were 7.0 and 20.1 ng/mL, respectively.

^b R = raw water; T = treated water; D = distributed water. Numbers in parentheses = number of sampling locations.

^c ND = not detectable, i.e., below the instrumental detection limit in Table 2.

^d Extreme values are within the parentheses; median value is outside. Extreme values represent lowest and highest level of contaminant in water supplies.

Table 4. Median and extreme values for the APDC-MIBK-GFAA^a determination of Cd, Co, Cr, Ni, and Pb in raw, treated, and distributed Canadian water supplies

Province		Cd, ng/mL	Co, ng/mL	Cr, ng/mL	Ni, ng/mL	Pb, ng/mL
Alberta	R (8) ^b	0.03 (≤0.01–0.04) ^c	0.5 (≤0.5–1.3)	0.6 (≤0.6–1.4)	≤0.6 (≤0.6–0.8)	ND ^d
	T (8)	0.03 (≤0.01–0.05)	ND	0.6 (≤0.6–1.2)	0.8 (≤0.6–1.5)	ND
	D (9)	0.02 (≤0.01–0.05)	ND	≤0.6 (≤0.6–1.0)	1.1 (≤0.6–2.4)	0.3 (≤0.3–0.8)
	R (14)	0.03 (≤0.01–0.05)	ND	0.6 (≤0.6–1.0)	≤0.6 (≤0.6–1.4)	≤0.3 (≤0.3–0.9)
British Columbia	T (16)	0.02 (≤0.01–0.04)	ND	≤0.6 (≤0.6–0.8)	ND	0.3 (≤0.3–1.7)
	D (18)	0.02 (≤0.01–0.03)	ND	≤0.6 (≤0.6–1.0)	ND	0.3 (≤0.3–5.7)
	R (9)	0.04 (0.02–0.05)	0.9 (≤0.5–2.0)	≤0.6 (≤0.6–1.0)	2.3 (≤0.6–5.9)	0.3 (≤0.3–0.7)
	T (10)	0.04 (≤0.02–0.05)	ND	≤0.5 (≤0.6–1.0)	0.7 (≤0.6–3.4)	0.4 (≤0.3–0.5)
New Brunswick	D (11)	0.04 (≤0.02–0.05)	ND	≤0.6 (≤0.6–1.0)	≤0.6 (≤0.6–2.5)	≤0.3 (≤0.3–3.4)
	R (6)	0.02 (≤0.01–0.05)	ND	≤0.5 (≤0.6–1.0)	1.0 (≤0.6–3.0)	0.3 (≤0.3–0.7)
	T (6)	0.03 (≤0.01–0.04)	ND	≤0.6 (≤0.6–0.8)	ND	ND
	D (10)	0.04 (≤0.01–0.05)	ND	≤0.6 (≤0.6–1.6)	≤0.6 (≤0.6–0.7)	ND
Newfoundland	R (10)	0.02 (≤0.01–0.03)	ND	≤0.6 (≤0.6–1.0)	≤0.6 (≤0.6–3.2)	≤0.3 (≤0.3–0.8)
	T (10)	0.06 (≤0.01–0.12)	≤0.5 (≤0.5–0.9)	≤0.6 (≤0.6–2.0)	≤0.6 (≤0.6–4.1)	0.3 (≤0.3–0.6)
	D (24)	≤0.01 (≤0.01–0.09)	ND	≤0.6 (≤0.6–2.2)	≤0.6 (0.6–3.4)	ND
	R (8)	0.03 (0.03–0.80)	ND	≤0.6 (≤0.6–3.0)	≤0.6 (≤0.6–1.2)	≤0.3 (≤0.3–3.4)
Northwest Territories	T (4)	≤0.01 (≤0.01–0.06)	ND	ND	≤0.6 (≤0.6–1.2)	ND
	D (7)	0.03 (0.01–0.04)	ND	≤0.5 (≤0.6–2.0)	ND	0.5 (0.3–3.6)
	R (4)	0.05 (0.03–0.07)	1.3 (≤0.5–2.6)	ND	1.4 (0.6–1.9)	0.6 (0.3–1.5)
	T (4)	0.04 (0.01–0.06)	ND	≤0.6 (≤0.6–1.0)	1.0 (≤0.6–1.9)	ND
Nova Scotia	D (7)	0.06 (0.04–0.09)	ND	≤0.6 (≤0.6–1.8)	1.8 (≤0.6–2.4)	ND
	R (31)	0.04 (≤0.01–0.07)	≤0.5 (≤0.5–2.9)	≤0.6 (≤0.6–4.0)	1.0 (≤0.6–72.4)	0.3 (≤0.3–1.9)
	T (31)	0.02 (≤0.01–0.08)	≤0.5 (≤0.5–3.7)	≤0.6 (≤0.6–3.2)	0.7 (≤0.6–68.9)	0.3 (≤0.3–0.8)
	D (52)	0.04 (≤0.01–0.09)	≤0.5 (≤0.5–2.4)	≤0.6 (≤0.6–2.8)	0.8 (≤0.6–68.1)	ND
Prince Edward Island	R (7)	≤0.01 (≤0.01–0.05)	ND	1.4 (0.6–2.6)	1.5 (≤0.6–2.0)	0.3 (0.3–18.6)
	T (2)	0.04 (0.04–0.05)	ND	1.2 (1.0–1.5)	ND	0.4 (0.3–0.6)
	D (6)	≤0.01 (≤0.01–0.04)	ND	ND	ND	0.5 (0.3–1.2)
	R (36)	0.02 (0.01–0.07)	≤0.5 (≤0.5–1.8)	≤0.6 (≤0.6–3.8)	0.8 (0.6–11.9)	0.3 (0.3–1.9)
Quebec	T (32)	0.03 (0.01–0.04)	≤0.5 (≤0.5–5.0)	≤0.6 (≤0.6–5.0)	0.9 (0.6–8.3)	0.3 (0.3–1.1)
	D (58)	0.03 (0.01–0.04)	≤0.5 (≤0.5–1.0)	≤0.6 (≤0.6–3.6)	1.1 (≤0.6–22.0)	0.3 (0.3–3.4)
	R (8)	0.04 (0.01–0.05)	0.7 (≤0.5–2.4)	≤0.6 (≤0.6–1.0)	1.5 (0.9–1.8)	ND
	T (8)	0.01 (≤0.01–0.04)	≤0.5 (≤0.5–4.0)	ND	1.3 (0.8–2.0)	ND
Saskatchewan	D (11)	0.01 (≤0.01–0.05)	0.5 (≤0.5–3.9)	ND	1.1 (≤0.6–1.9)	0.3 (≤0.3–2.6)
	R (6)	0.10 (≤0.01–0.03)	ND	0.6 (≤0.6–1.0)	ND	ND
	T (2)	0.10 (≤0.01–0.02)	ND	ND	ND	ND
	D (7)	0.02 (≤0.01–0.04)	ND	ND	ND	0.3 (≤0.3–0.9)

^a APDC-MIBK-GFAAS = Ammonium pyrrolidinedithiocarbamate-methyl isobutyl ketone-graphite furnace atomic absorption.
^b R = raw water; T = treated water; D = distributed water. Numbers in parentheses = number of sampling locations.
^c Extreme values are within parentheses; median value is outside. Extreme values represent lowest and highest level of contaminant in water supplies.
^d ND = not detectable, i.e., below instrumental limit in Table 2.

was found in one of the wells used as a raw water source in Charlottetown, PEI, but the value decreased to ≤ 1.0 ng Pb/mL in both the treated and distributed waters. The sodium fluorosilicate used as a treatment chemical in this location appears to have quantitatively removed Pb. Treated water from St. George's, Nfld, contained 14–16 ng Pb/mL compared with ≤ 1.0 ng Pb/mL in the raw water. Also, distributed water from Victoria, BC; Winnipeg, Man.; St. John, NB; Halifax, NS; and Hearst, Ont. showed 5.7, 5.0, 16.0, 19.0, and 79.7 ng Pb/mL by the direct method, and 7.0, 4.4, 14.7, 20.6, and 76.8 ng Pb/mL by the extraction method, respectively. The corresponding raw and treated water values were ≤ 2.0 ng Pb/mL. In addition, 3–4 ng Pb/mL was picked up during distribution in a number of other locations compared with the raw and treated water values of ≤ 1.0 ng Pb/mL. Table 5 lists the percentage of distributed water samples showing pick-up of Pb for the various

Table 5. Percentage of water samples showing increases of Pb, Cu, and Zn concentrations during distribution

Province	Pb	Cu	Zn
Alberta	0	0	22
British Columbia	11	50	50
Manitoba	10	60	30
New Brunswick	30	90	0
Newfoundland	26	58	22
Northwest Territories	14	57	29
Nova Scotia	56	29	29
Ontario	14	48	40
Prince Edward Island	0	84	33
Quebec	10	64	53
Saskatchewan	0	62	0
Yukon	0	86	14

Canadian Provinces. Nova Scotia had the highest percentage (56%), and no pick-up occurred in Prince Edward Island, Saskatchewan, and Yukon. As explained in the first survey (13), the source of this apparent contamination during distribution may have been lead pipes, galvan-

Table 6. Median and extreme values for Cu, Zn, Ca, and Mg in raw, treated, and distributed Canadian water supplies as determined by GFAAS

Province		Cu, ng/mL	Zn, ng/mL	Ca, μ g/mL	Mg, μ g/mL
Alberta	R (8) ^a	≤ 10 (≤ 10 –50)	ND ^b	39.8 (30.9–45.1)	15.2 (12.5–18.7)
	T (8)	ND	≤ 10 (≤ 10 –20)	33.1 (14.9–49.6)	15.0 (7.2–18.0)
	D (9)	ND	≤ 10 (≤ 10 –90)	30.7 (15.3–53.0)	15.1 (7.1–17.6)
British Columbia	R (14)	≤ 10 (≤ 10 –70)	10 (≤ 10 –60)	4.0 (1.9–13.5)	0.4 (0.3–2.0)
	T (16)	70 (≤ 10 –230)	10 (≤ 10 –50)	5.5 (1.8–55.8)	1.2 (0.2–15.5)
	D (18)	80 (≤ 10 –790)	20 (≤ 10 –250)	3.3 (1.9–40.8)	0.3 (0.2–9.6)
Manitoba	R (9)	ND	10 (≤ 10 –170)	18.7 (14.6–65.5)	5.5 (3.9–40.8)
	T (10)	ND	10 (≤ 10 –190)	23.4 (14.7–40.2)	5.6 (3.7–13.8)
	D (11)	30 (≤ 10 –90)	10 (≤ 10 –210)	21.3 (14.8–39.1)	5.7 (3.6–14.2)
New Brunswick	R (6)	≤ 10 (≤ 10 –230)	ND	6.2 (4.6–28.1)	1.1 (0.8–3.7)
	T (6)	≤ 10 (≤ 10 –120)	ND	6.6 (4.5–27.9)	1.1 (0.7–3.6)
	D (10)	120 (30–280)	ND	17.1 (4.6–27.9)	2.2 (0.7–3.5)
Newfoundland	R (10)	≤ 10 (≤ 10 –20)	ND	3.5 (1.3–11.2)	1.0 (0.6–8.2)
	T (10)	20 (≤ 10 –360)	≤ 10 (≤ 10 –50)	3.6 (1.6–10.8)	1.1 (0.7–13.8)
	D (24)	100 (≤ 10 –900)	≤ 10 (≤ 10 –410)	4.5 (1.8–11.7)	5.7 (3.6–14.2)
Northwest Territories	R (8)	≤ 10 (≤ 10 –510)	≤ 10 (≤ 10 –350)	10.6 (4.6–16.4)	3.1 (1.6–4.4)
	T (4)	ND	ND	10.5 (4.5–16.6)	3.0 (1.6–4.4)
	D (7)	30 (≤ 10 –330)	10 (≤ 10 –750)	10.8 (4.5–16.8)	3.0 (1.6–4.2)
Nova Scotia	R (6)	390 (230–540)	20 (≤ 10 –30)	6.4 (2.0–12.1)	0.8 (0.7–0.9)
	T (6)	230 (220–240)	ND	10.5 (9.9–11.5)	0.8 (0.7–0.9)
	D (8)	350 (10–470)	10 (10–70)	11.6 (11.4–12.9)	0.8 (0.7–0.9)
Ontario	R (31)	≤ 10 (≤ 10 –160)	10 (≤ 10 –60)	28.9 (6.7–63.3)	4.9 (1.7–20.2)
	T (31)	≤ 10 (≤ 10 –210)	10 (≤ 10 –50)	29.9 (6.6–40.2)	5.1 (1.9–20.3)
	D (52)	50 (≤ 10 –550)	20 (≤ 10 –170)	29.6 (6.4–41.1)	5.8 (1.6–20.3)
Prince Edward Island	R (7)	≤ 10 (≤ 10 –50)	10 (≤ 10 –30)	53.4 (14.0–77.3)	2.3 (1.0–21.4)
	T (2)	20 (10–30)	30 (≤ 10 –60)	31.6 (25.5–37.6)	18.0 (15.0–20.9)
	D (6)	40 (20–60)	≤ 10 (≤ 10 –130)	40.4 (26.4–67.7)	8.4 (1.3–16.4)
Quebec	R (36)	10 (≤ 10 –220)	10 (≤ 10 –100)	16.6 (3.3–65.9)	3.4 (0.7–16.9)
	T (32)	≤ 10 (≤ 10 –200)	10 (≤ 10 –510)	16.5 (4.2–38.9)	3.1 (0.6–16.3)
	D (58)	30 (≤ 10 –470)	20 (≤ 10 –410)	18.3 (4.5–70.5)	3.6 (0.6–25.5)
Saskatchewan	R (8)	ND	ND	34.4 (31.9–40.7)	18.1 (12.1–32.6)
	T (8)	ND	ND	33.5 (20.0–39.0)	15.3 (11.9–30.0)
	D (11)	20 (≤ 10 –520)	≤ 10 (≤ 10 –20)	33.2 (1.1–112.8)	12.5 (0.3–59.5)
Yukon	R (6)	ND	ND	13.2 (13.1–30.3)	2.2 (2.2–2.3)
	T (2)	ND	ND	13.6 (13.3–13.8)	2.8 (2.8–2.8)
	D (7)	70 (10–110)	≤ 10 (≤ 10 –140)	15.1 (15.0–15.3)	2.1 (2.0–2.2)

^a R = raw water; T = treated water; D = distributed water. Numbers in parentheses = number of sampling locations.

^b ND = not detectable, i.e., below instrumental limits in Table 2.

Table 7. Comparison of the Cu and Zn content in treated and distributed waters from selected Canadian cities

Province	City	pH	Hardness, mg CaCO ₃ /L	Median Cu content, ng/mL		Median Zn content, ng/mL	
				Treated	Distributed	Treated	Distributed
British Columbia	Cassiar	6.5	11.4	≤10	≤10	94	233
	Vancouver	6.3	4.0	122	383	≤10	18
	Victoria	6.7	19.1	36	221	13	254
Manitoba	Flin Flon	7.6	48.4	≤10	87	193	211
New Brunswick	Fredericton	7.4	83.5	≤10	221	≤10	≤10
	St. John	6.8	15.2	≤10	275	≤10	11
Newfoundland	Baie Verte	5.3	16.3	≤10	376	26	105
	(Rattling Brook)						
	Gander	5.4	6.7	≤10	320	≤10	10
	Happy Valley	5.3	10.0	33	313	≤10	413
	St. Georges	5.6	32.8	362	900	≤10	193
Northwest Territories	Inuvik	6.5	59.1	≤10	329	≤10	751
Nova Scotia	Sydney	6.3	31.4	239	472	≤10	10
Ontario	Hearst	6.8	64.3	134	553	≤10	101
	Kirkland Lake	—	42.2	21	54	10	169
	North Bay	6.8	24.5	77	325	53	54
	Peterborough	7.9	97.6	≤10	231	10	45
	Sudbury	6.6	85.7	86	175	18	26
	Thunder Bay	7.5	45.5	≤10	271	14	16
	Quebec	7.1	55.6	≤10	188	≤10	230
	Asbestos	7.8	62.3	39	117	≤10	35
	Athabaska	—	29.6	<10	18	≤10	411
	Beaulac	8.1	53.5	<10	152	77	240
Quebec	Drummondville	—	30.6	23	200	<10	12
	Hull	7.5	52.5	197	342	13	155
	Sherbrooke	7.3	28.4	<10	465	≤10	17
	Thetford Mines	6.7	212.7	<10	520	10	16
	Swift Current	—	42.2	<10	66	≤10	≤10
Saskatchewan	Whitehorse						

ized pipes, and lead-tin solders in the distribution system, probably by interaction with the humic substances found in water (24).

Table 6 shows the results of flame atomic absorption analyses. The median concentration of Cu in the raw waters sampled rarely exceeded 10 ng/mL and few samples exceeded 100 ng/mL. These few included: Drummondville, Hull, and Windsor, PQ; Yellowknife, NWT; Halifax and Sydney, NS; Fredericton, NB; and Kingston, Ont. with values of 188, 167, 202, 505, 537, 235, 225, and 156 ng Cu/mL, respectively. The Cu concentrations of treated waters were generally <10 ng/mL. However, values of 194, 225, 239, 176, 362, 119, 134, 207, 104, and 156 ng Cu/mL were found in the treated waters from Schefferville, PQ; Halifax and Sydney, NS; Baie Verte and St. George's, Nfld; Moncton, NB; Hearst and Toronto, Ont.; and Prince Georges and Prince Rupert, BC, respectively. The level of Cu was significantly higher in a number of distributed water samples than the corresponding treated water samples (Table 7). The increase was higher in those areas where the water was soft and corrosive. In these cases, both the low pH and low hardness of the water enhances cupro-solvency, thereby promoting leaching of Cu

from the distribution system. Although not shown in Table 7, Cu concentrations increased during distribution in a number of other locations. The percentage of samples showing such increases is given in Table 5 for each of the Canadian Provinces. More than 50% of the water supplies of all Provinces except Alberta and Nova Scotia contained more Cu in the distributed water than in the treated water.

The median Zn content in the raw, treated, and distributed water samples rarely exceeded 10 ng/mL. Major exceptions were: Yellowknife, NWT and Flin Flon, Man. where the raw water sample values were 354 and 173 ng/mL, respectively. The value decreased below 10 ng/mL in the treated and distributed waters in Yellowknife, but in Flin Flon, the value increased to 193 and 208 ng/mL in the treated and distributed waters, respectively. Plessisville and Windsor, PQ; Charlottetown, PEI; and Baie Verte, Nfld had values of 505, 90, 61, and 46 ng Zn/mL in treated water compared with <10, 13, 27, and 10 ng/mL, respectively, in raw water. As shown in Table 7, samples from a number of locations had significant concentrations of Zn in the distributed water compared to the treated water values. The percentage of samples showing increases in Zn

Table 8. Median and extreme values^a for some elements in Canadian drinking water supplies

Element ^a	Raw water			Treated water			Distributed water		
	Direct ^b	Extraction ^c		Direct	Extraction		Direct	Extraction	
Cd (5) ^d	≤0.02 (≤0.02–0.90) ^e	0.03 (≤0.01–0.80)		≤0.02 (≤0.02–0.02)	0.03 (≤0.01–0.12)		≤0.02 (≤0.02–0.07)	0.03 (≤0.01–0.09)	
Co ^f	≤2.0 (≤2.0–3.0)	<0.5 (≤0.5–2.9)		≤2.0 (≤2.0–5.0)	≤0.5 (≤0.5–4.0)		≤2.0 (≤2.0–6.0)	≤0.5 (≤0.5–3.9)	
Cr (50)	≤2.0 (≤2.0–5.0)	≤0.6 (≤0.6–4.0)		≤2.0 (≤2.0–4.0)	≤0.6 (≤0.6–5.0)		≤2.0 (≤2.0–4.1)	≤0.6 (≤0.6–3.6)	
Cu (1000)	≤10.0 (≤10.0–540.0)	—		≤10.0 (≤10.0–360.0)	—		77.5 (≤10.0–900.0)	—	
Ni ^g	≤2.0 (≤2.0–73.5)	≤0.6 (≤0.6–72.4)		≤2.0 (≤2.0–69.6)	≤0.6 (≤0.6–68.9)		≤2.0 (≤2.0–69.0)	≤0.6 (≤0.6–68.1)	
Pb (50)	≤1.0 (≤1.0–20.1)	≤0.3 (≤0.3–18.6)		≤1.0 (≤1.0–16.0)	≤0.3 (≤0.3–14.4)		≤1.0 (≤1.0–7.0)	≤0.5 (≤0.3–5.7)	
Zn (5000)	≤10.0 (≤10.0–350.0)	—		≤10.0 (≤10.0–510.0)	—		≤10.0 (≤10.0–750.0)	—	
Ca (200)	21.8 (1.3–77.3)	—		18.2 (1.6–55.8)	—		21.4 (≤1.1–112.8)	—	
Mg (150)	5.4 (0.3–40.8)	—		6.0 (0.2–30.0)	—		6.0 (0.2–59.5)	—	

^a Values are expressed in ng/mL except Ca and Mg which are in µg/mL.
^b Direct injection of 20 µL sample into graphite tube.
^c 25 mL sample extracted into 5 mL MIBK after complexation with APDC; 20 µL of MIBK phase injected into graphite tube.
^d Max. permissible levels in drinking water. Values are in ng/mL except Ca and Mg which are in µg/mL. Values for Cd, Cr, Cu, Pb, and Zn are from Ref. 24; those for Ca and Mg are from Ref. 25.
^e Extreme values are within parentheses; median value is outside. Extreme values represent lowest and highest level of contaminant in Canadian water supplies.
^f No limits have been set for Co and Ni by Health and Welfare Canada, WHO, or EPA.

values during distribution is shown in Table 5 for each Province. Zinc was introduced into the water from the municipal distribution lines of all Provinces except New Brunswick and Saskatchewan. In these cases, Zn probably entered the distributed waters from the deterioration of galvanized iron pipes and brass fittings.

The median Ca and Mg contents of raw, treated, and distributed waters were approximately the same for most samples. Exceptions occurred in raw water softened by treatment in Saskatoon, Sask.; Edmonton, Alta; Portage La Prairie and Selkirk, Man.; Sudbury and Tilbury, Ont. where the Ca values decreased from 40.5, 44.5, 65.3, 58.0, 26.4, and 59.5 mg/L to 20.0, 15.3, 24.0, 38.0, 18.6, and 35.1 mg/L, respectively, after treatment. The effect of treatment on Mg was noticed only in Edmonton, Portage La Prairie, and Selkirk where the treated water values were only 7.5, 10.0 and 13.6 mg/L, corresponding to the raw water values of 12.7, 40.0, and 37.5 mg/L, respectively. The raw water supplies which received lime treatment showed significant increases in the amount of Ca in the treated water. These included Chicoutoumi, Hull, Quebec, Trois Rivières, and Windsor in PQ; Halifax, NS; Thompson, Man.; and Ottawa, Ont. with Ca values of 9.0, 14.0, 10.0, 8.5, 20.5, 10.0, 23.0, and 16.3 mg/L in the treated waters compared with the raw water values of 4.6, 8.2, 5.3, 3.4, 11.5, 2.0, 17.5, and 8.6 mg/L, respectively. The hardest waters sampled were from the Prairies, particularly Manitoba and Saskatchewan, and Southern Ontario. British Columbia, the Maritimes, and Northern Ontario were soft water areas and Quebec was in between.

Table 8 contains a summary of the concentrations of Cd, Co, Cr, Cu, Ni, Pb, Zn, Ca, and Mg in Canadian water supplies. These elements are present in the distributed drinking water at well below the maximum permissible limits set by Health and Welfare Canada (25), WHO (26), and the U.S. Environmental Protection Agency (27). A survey conducted in 1978 by V. C. Armstrong and R. S. McCullough of the Environmental Health Directorate, Health and Welfare Canada, showed that the average daily consumption of water, including beverages, by a Canadian adult is 1.3 L, with certain individuals consuming ≥ 4.0 L daily. Therefore, the daily intake of each of the trace elements by an average Canadian adult in μg would be: Cd ≤ 0.03 , Co ≤ 2.6 , Cr ≤ 2.6 , Cu ≤ 13.0 , Ni ≤ 2.6 , Pb ≤ 1.3 , and Zn ≤ 13.0 . These levels could increase to Cd 0.32, Co 19.8, Cr 15.4, Cu 3600.0, Ni 274.2, Pb 25.4, and Zn 3000.0 μg for individuals who consume 4 L of water daily and

who live in areas with the highest trace element content (Table 8). The amount of tap water consumed by children under 3 years of age has been estimated at up to 500 mL/day (1). This would result in the intake of up to 0.01, 1.0, 5.0, 1.0, 0.5, and 5.0 μg for Cd, Co, Cr, Cu, Ni, Pb, and Zn, respectively. Thus the quantity of toxic elements, specifically Cd and Pb, present in the Canadian water supplies is usually negligible compared to the amount reported to be present in food (28). However, in the case of individuals consuming 4 L of tap water in areas with the highest levels of water-borne lead, the contribution of tap water to the total daily intake cannot be neglected since it can account for about 25% of the average levels reported for food. For these individuals, the daily intake of 3600 μg of Cu from tap water exceeds the reported 2217 μg Cu intake from foods (28). In addition, the amounts of essential elements such as Co, Cr, Cu, and Zn are too low to represent a significant part of the total daily requirements (29). Recent studies (30–35) have shown that trace element levels from household tap water sampled after standing overnight can be appreciable. Therefore, detailed studies are under way in this laboratory to determine the impact of distribution lines and household tap fixtures on the levels of trace elements present in drinking water.

Conclusions

Both the present survey and the previous survey (13) have shown that, in general, the surface and ground waters used as sources of drinking water in Canada contain negligible amounts of Cd, Co, Cr, Cu, Ni, Pb, and Zn. During treatment and distribution this level is not altered significantly except for Cu and Zn which increased in a few locations. The surveys taken together, encompassed 115 municipalities across Canada—municipalities containing nearly 63% of the Canadian population. The quality of drinking water supplied to Canadians more than satisfactorily meets the maximum acceptable recommended limits given in the "Guidelines for Canadian Drinking Water Quality 1978" (25) with respect to the above trace elements. The median levels for all the elements except Cr are below the objective limits (24) of our ultimate water quality goal.

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