Sampling and initial findings for a study of fluoride in drinking water in the United States


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Abstract

The role of fluoride in reducing the risk of dental caries, especially among children, is well recognized and is the basis for current intake recommendations. The US Department of Agriculture, Nutrient Data Laboratory conducted a comprehensive study of the fluoride content of US drinking water, as part of the US National Fluoride Database and Intake Assessment Study, a collaborative effort with the University of Minnesota, the University of Iowa College of Dentistry, and Virginia Polytechnic Institute and State University. The sampling method involved: serpentine ordering of the US population by census region, division, and county; dividing the population into 72 equal population size zones; and randomly selecting one county per zone and two residences per county. Participants were recruited by phone to provide two tap water samples, 3–4 months apart; samples (n = 288) were analyzed by the direct read method. Well water averaged <20 mcg fluoride/100 g, municipal water averaged 100–110 mcg fluoride/100 g, and the national average across sources was 71 mcg fluoride/100 g. These nationally representative data for drinking water will support public health research on the impact of fluoride on bones and teeth and will provide a foundation for assessment tools in the dental and medical communities.

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Keywords: Fluoride; National Fluoride Database; Dental caries

1. Introduction

The role of fluoride in reducing the risk of dental caries, especially among children, is well recognized and is the basis for the indicators of Adequate Intake recommendations (IOM, 1997; CDC, 2001). Assessments of excessive fluoride intake from public water supplies, other beverages and foods, and non-food sources are also critical in developing an understanding of enamel and skeletal fluorosis. Existing fluoride data from small local or regional studies are inadequate to characterize the national food supply. Therefore, a nationally representative database is needed to assess the dietary intake of fluoride by the population.

The National Fluoride Database was developed as a comprehensive, nationally representative database of the fluoride concentration in foods and beverages consumed in the US. The database contains fluoride values for beverages, water, and foods that are major contributors to intake. Development of this database is integral to the National Fluoride Database and Intake Assessment Study (NFDIAS), a collaborative effort among the US Department of Agriculture (USDA) Nutrient Data Laboratory (NDL), a research unit within the Agricultural Research

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Service (ARS), the University of Minnesota, School of Public Health, the University of Iowa (UI), College of Dentistry, and Department of Biochemistry, Food Analysis Laboratory Control Center (FALCC), Virginia Polytechnic Institute and State University (VPI&SU). Water and water-based beverages are the chief sources of dietary fluoride intake (Singer et al., 1985), contributing approximately 75% of dietary fluoride. According to the 1992 Fluoridation Census (CDC, 1993), about 63% of US public water systems (PWSs) are fluoridated naturally or by added fluoride; the standard fluoride level for added fluoridation is 1 ppm (equivalent to 100 mcg/100 g water). The distribution of fluoride in drinking water may vary widely over geographical and geo-political boundaries (CDC, 1993). Variations occur with soil composition and with local political decisions to fluoridate water. The use of wells of varying depths, commercial water products, home water purifiers, and filtration systems also increase variability of fluoride in drinking water and complicate estimates of intake (Brown and Aaron, 1991; Robinson et al., 1991; van Winkle et al., 1995). Information on the variability of fluoride in water and commercial foods and beverages is crucial to understanding fluoride intake. Though total intake of fluoride must also consider fluoride contributed by toothpaste, oral rinses, and dental treatments, this paper addresses the determination of fluoride content of drinking water sampled nationally and factors related to variability.

The NDL develops reliable databases on the composition of foods available in the US and state-of-the-art methodology to evaluate and disseminate these data. NDL, in cooperation with the National Heart Lung and Blood Institute of the National Institutes of Health, conducts the National Food and Nutrient Analysis Program (NFNAP), the main goal of which is to obtain reliable estimates of means with known variability for the nutrient content of food and beverages consumed by the US population (Perry et al., 2000; Pehrsson et al., 2000). Toward this objective, highly representative probability-based food and beverage samples are selected and analyzed for over 100 nutrients and potentially bioactive components. Through a cooperative agreement with NDL, statisticians from the USDA National Agricultural Statistics Service (ARS) developed a Consolidated Metropolitan Statistical Area (CMSA) as an urban area with a population of at least one million satisfying several other requirements (US Bureau of the Census, 1999). Because not all counties are included in a CMSA, this study generalizes the CMSA concept, defining a generalized CMSA (gCMSA) as a CMSA or an individual county not contained in a CMSA. Standard Census geographic regions and districts were used.

Each record in the frame corresponds to a county. It contains the county name and Federal Information Processing Standards code, state name, population (2000 Census), gCMSA name and code, local (within gCMSA) urbanicity index, and Census region. The urbanicity index used in this study is a measure of urban character based on the populations of the largest cities and towns in a county (Goodall et al., 1998). This index was used to ensure that counties bordering a major city are treated more like that city than the area on the outskirts of the CMSA.

The frame was sorted first by Census region, within region by Census district, and within district by state. The gCMSAs were sorted by population size within state, and serpentine in adjacent states; i.e., in increasing order in one state then decreasing order in the next. Likewise, counties were sorted serpentine by urbanicity within

2. Methodology

Fig. 1 summarizes the sampling design. Sampling requirements for the main study were influenced by a number of factors including: (1) variability of fluoride in water; (2) desired level of confidence in the final estimates; and (3) data collection costs. NDL and the Food Composition Lab (FCL), ARS, USDA carried out two preliminary studies of municipal water supplies and carbonated beverages to examine the concentration of fluoride as well as other mineral elements (Miller-Ihli et al., 2003). Results on variability were used to establish the sample size (total number of samples needed for each beverage or food), number of locations within counties, and number of pickups over time for the larger study presented in this research.

2.1. Sampling frame development

The frame for the first stage of sampling for the fluoride survey was developed using estimated population counts for all states obtained from the US Bureau of the Census web site (www.census.gov). The Census Bureau defines a Consolidated Metropolitan Statistical Area (CMSA) as an urban area with a population of at least one million satisfying several other requirements (US Bureau of the Census, 1999). Because not all counties are included in a CMSA, this study generalizes the CMSA concept, defining a generalized CMSA (gCMSA) as a CMSA or an individual county not contained in a CMSA. Standard Census geographic regions and districts were used.

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![Figure 1: Sampling design for collecting water for fluoride analysis.](image-url)
gCMSAs. This method of sorting, combined with the sampling method described below, ensures that if multiple sample counties fell within a state, they were well dispersed with respect to geography, and that if multiple sample counties fell within a gCMSA, they were well dispersed with respect to urbanicity.

For sampling of tap water, lists of residents and their addresses and telephone numbers in the 72 sampled counties were developed (Fig. 2). The rest of the paper will refer to the 72 sampled counties; however, Los Angeles County, California was selected twice in the first stage because of its size, so, technically, the sample includes only 71 unique counties. Within each county, two residential locations were chosen at random. For sampling of other fluoride-contributing beverages (e.g., fruit juices, carbonated beverages) under NFDIAS, subsets of 36 or 18 counties were chosen by selecting one at random from consecutive pairs of counties. The mathematical approach to the sample frame development is described in greater detail in Bellow et al. (2002).

2.2. Sample selection

At the first stage of sample selection, Chromy’s (1971) zonal sampling method was used to select a sample of 72 counties. Chromy’s method effectively divides the total population of the counties along the serpentine ordering into 72 equal size zones. Then it randomly selects one county from each zone with probability proportional to size conditional on probability of minimum replacement. Selecting the counties with probability proportional to size conditional on probability of minimal replacement ensures the probability of any county being selected is proportional to its size and that no county is selected more times than the next integer larger than the ratio of its size to the size of the zones, which is 1/72 of the size of all counties.

For municipal and well waters at the second stage, a simple random sample of two households was selected from each of the selected counties. Two water samples were collected from each selected household 3–4 months apart to allow the estimation of variance due to date of collection as well as that due to location within county.

The next steps in the sample selection process included: (1) development of recruitment procedures to minimize resistance to phone solicitation and maximize first-call compliance; (2) development of a current residential phone listing by counties (randomly ordered); (3) participant survey development; and (4) successful application through the USDA survey approval office and the US Office of Management and Budget (OMB) for approval on all aspects of the study.

For this study, rigorous procedures were developed to mitigate the potential for non-response and to assure the selection of a representative sample of participants. To ensure there were adequate alternate households when the selected households declined to participate, a list of the 50 closest households to each of the selected locations was acquired. If a household declined to participate, the next household on the list was contacted. This process continued in order of nearness to the selected household until a willing participant was found. Alternates in close proximity were assumed to be as similar to the selected household as possible with respect to household water source, plumbing, and other factors affecting fluoride level. Though NDL researchers placed approximately 1500 phone calls to develop the sample of participants, the compliance rate was still high (>75%) as required by OMB criteria because alternates were in similar housing, presumably with similar plumbing, and with access to the same public water supply.

Participants completed a one-page survey that addressed details of their sources of tap water. Adult individuals, who acknowledged being in a position of responsibility for the
household and agreed to participate, completed this survey on their source of drinking/cooking water, type of plumbing (e.g., copper), and any treatment of the water (e.g., water softening or purification systems).

2.3. Water collection

Sampling of municipal (tap) water in residential homes presents different challenges than retail beverage sampling. Assurance of water sample integrity and adequacy included: (1) provision of clean, labeled bottles (pretested for fluoride contamination) and water collection kits/protocols to participants; (2) discussion of pickup strategies and delivery of collection/survey kits by USDA-contracted agents (Superior Pickup Inc.); and (3) strict shipping protocols to minimize sample loss and contamination. Three types of water-filled bottles were pilot tested for strength under shipping and very low temperatures. Bottles made from high-density polyethylene were used in the study because the material is strong and can be used over a wide temperature range. In an effort to develop and test protocols for shipping, pilot water samples were shipped in new bottles from UI, College of Dentistry to NDL and back; half of the bottles contained water with known amounts of fluoride and the other half were filled with deionized water. Samples were shipped UPS ground (several days) and held at room temperature, with deionized water. Samples were shipped in new bottles from UI, College of Dentistry to NDL at room temperature, with deionized water. Samples were shipped UPS ground (several days) and held at room temperature, with deionized water. Samples were shipped in new bottles from UI, College of Dentistry to NDL at room temperature, with deionized water. Samples were shipped UPS ground (several days) and held at room temperature, with deionized water.

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Field agents received the survey, water bottles, shipping materials, and instructions a day or two before meeting with participants. Each field agent verified the appointment the day before to maximize compliance. At each residence, two 250 mL polyethylene bottles were filled with tap water from the kitchen faucet. Participants were issued simple written instructions developed by NDL for the pilot study and modified for this sampling. The instructions included: (1) answer the survey questions read by the USDA representative; (2) sample only water which is used for drinking and/or cooking (indicate to the agent if you would normally cook with bottled water); (3) turn on the cold water tap to provide a steady, medium strength flow and run for about 3 min but do not rinse the collection bottles; (4) fill both bottles with water up to the marked lines and tightly close the caps (but not forced tight); and (5) give both water samples to the field agent. The filled bottles and completed surveys were immediately shipped by ground service and at ambient temperature by the agents to VPI&SU for sample preparation. To ensure the confidentiality of individual participants, fluoride data and household information on water source and treatment were attached to a consumer code and will be reported only in table format. The data will not be used to assess a family’s socioeconomic status or any other characteristic of that individual’s home. Each participant was awarded a non-monetary incentive at the time of the first water collection and survey.

2.4. Chemical analysis and QC

The preparation of the water samples for analysis and the analysis of fluoride were handled through NFNAP cooperative research agreements. Samples were stored at room temperature (17–25 °C) prior to subsampling for analysis; after subsampling, they were stored frozen. The UI, College of Dentistry analyzed the samples of water and other clear beverages for fluoride content using a fluoride ion-specific electrode direct read method (van Winkle et al., 1995). Three additional laboratories characterized QC materials in order to develop a consensus fluoride value that could be used for QC purposes. The analytical method for fluoride was validated using the National Institute of Standards and Technology Standard Reference Material (SRM) 2671a, Fluoride in Freeze-Dried Urine (Gaithersburg, MD) as an in-house control (Fig. 3). In addition, 5% of the samples were run in blind duplicate, pure standards were run periodically to check for instrumental drift, and readings were made only after the signal stabilized. Five standards were used to set the curve. Comparisons of results for several beverage samples analyzed by UI, College of Dentistry and FCL were performed.

One of the two 250 mL bottles of water from each site was frozen and the other bottle was subsampled for fluoride analysis. After mixing the non-frozen water sample several times by inversion, 10 mL aliquots were transferred using a disposable polyethylene transfer pipette into 15 mL polypropylene screw-cap test tubes (Sarstedt #60540;

![Fig. 3. Quality control results. The dotted lines for the SRM show the certified range, and the dotted lines for the quality control material show the acceptable range that was established based on analysis of the material by three independent laboratories prior to beginning the study.](image-url)
Three aliquots were taken from each water sample, and from every twentieth bottle a fourth 10 mL aliquot was taken as a blinded analytical duplicate. All of the subsamples were capped under nitrogen and frozen (−60 ± 5 °C). Except for two of the aliquots from each water sample, which were held in frozen storage, all aliquots were shipped along with QC samples (described below) on dry ice via overnight express shipping to UI, College of Dentistry for analysis of fluoride.

QC samples included a commercial reference material with certified fluoride concentration [Calgary 93 Drinking Water; LGC Promochem (Teddington, Middlesex, UK)], which was repackaged in 15 mL test tubes as described above. Four blinded aliquots of the reference material were shipped with the test samples for fluoride analysis; they were dispersed among four batches of samples (i.e., run at different times). Additionally, two QC solutions were prepared specifically for the NDL, one at high concentration (+150 μg/100 g) and one at low concentration (+80 μg/100 g) of fluoride. These solutions were formulated gravimetrically and produced using a fluoride standard obtained from High-Purity Standards (Charleston, SC) and deionized water. Sucrose at 10% w/v was added to the high-concentration material so it would also serve as a control material for other types of beverages. The bulk materials (approximately 3 L each) were shipped at ambient temperature to the FALCC, VPI&SU where they were dispensed into test tubes as described above. Blinded aliquots of the control solutions were shipped with test samples at a rate of one high-concentration control samples and one low-concentration control sample for every 40-test sample. Three independent laboratories experienced in analyzing water and other clear beverages for fluoride analyzed duplicate samples of each reference material twice using ion-specific electrodes and read the samples directly. From these data a target value and uncertainty were established.

A QC oversight program was established by the NFDIAS Laboratory Methods/Quality Control Working Group with representation from NDL, UI, College of Dentistry, the University of Minnesota, and VPI&SU. The program included provision of the NFDIAS QC materials prepared by NDL (above) and validation of the analytical method for fluoride. A data quality review group evaluated the QC data prior to acceptance of the analytical results. Prior to beginning the study, a comparison of results for several beverage samples analyzed by UI, College of Dentistry and FCL was made. The blind control, in-house control, and duplicate results were examined and decisions made as to whether the data for each batch of samples was acceptable or if repeat analyses were necessary.

Data analysis

SAS statistical software (Littell et al., 1996) was used to determine mean and standard deviations for fluoride in the water samples by fluoridation status as well as the national average. Mixed model analyses were performed to determine the components of variability. One model treated source (private well or PWS) and fluoridation status as separate effects. Since no private wells were reported to contain added fluoride, this model essentially treated the data as coming from an incomplete block design. A second treatment model for the data was to combine source and fluoridation status into a single water-type variable with three values, municipal fluoridated, municipal non-fluoridated, and well. Result from these two data analysis approaches differed considerably.

3. Results and discussion

3.1. Survey results and considerations

Results of the survey of participants showed well water was reported as used for drinking by 18% (26 of 144) of the participants. Ten percent of the participants used a water treatment system, but reporting on use of reverse osmosis systems was assumed to be irregular; no follow-up calls were made to those reporting use of water treatment systems. Reverse osmosis is known to remove from two-thirds to almost all of the fluoride in tap water (van Winkle et al., 1995). Fifty-nine percent (85 of 144) of the participants reported using water from a fluoridated public water supply; this was supported by values and/or follow-up phone calls to the local water treatment plants to verify reported fluoridation status, if not clear from the survey. Survey responses for non-fluoridated public and well water users were verified only if the actual fluoride values contradicted the responses.

3.2. Initial findings

The national mean fluoride content for drinking water was 71 ± 48 (s.e.) mcg/100 g water across source (municipal and well) and fluoridation status (Table 1). The national mean is most useful when dietary assessment surveys cannot ascertain the different individual locations or incidence of fluoridation or type of plumbing used, that is for default purposes in the database; e.g., when “preparing” foods with water in a recipe program. The bimodal distribution of fluoride in all drinking water and for municipal and well waters is shown in Fig. 4. Predictably, the first peak for all water is at non-fluoridated levels (<20 mcg/100 g water). The second peak is at the 100–120 mcg/100 g (1 ppm) level. This is also true when municipal tap water is separated out. The distribution for well water peaked at <20 mcg/100 g level. However, a few well water samples fell within the 140–180 mcg/100 g range for both pickups, perhaps reflecting pockets of naturally occurring fluoride. Table 1 shows mean fluoride concentration of all drinking water (municipal and well combined) was highest in the Midwest (88 ± 42 mcg/100 g, n = 68) and lowest in the West (47 ± 38 mcg/100 g, n = 64). Water samples in the South had a mean fluoride concentration...
of 76 ± 46 mcg/100 g (n = 100) and those in the Northeast had a mean concentration of 69 ± 56 mcg/100 g (n = 56). Municipal and well waters varied considerably in mean fluoride content (Table 1): 51 vs. 24 mcg/100 g in the West, 93 vs. 10 mcg/100 g in the South, 102 vs. 48 mcg/100 g in the Midwest, and 74 vs. 9 mcg/100 g in the Northeast. Fig. 5 shows the inconsistent distribution patterns by region, with the least differences among samples in the West. Fluoride concentration peaks in the Northeast at the 0 or non-fluoridated level. Municipal water has a predictable distribution between fluoridated and non-fluoridated water, except for the few pockets in some areas around the US (e.g., the Midwest) of high naturally occurring fluoride (Fig. 6). A few samples from fluoridated supplies had very low fluoride levels, most likely due to home water treatments or to the failure of water supply plants to fluoridate (see later discussion). The overall mean for drinking water in this study exceeds that determined in the 1999 pilot study. Comparisons of results with other data collected at an earlier point in time or from a specific region were not pursued because of changes in fluoridation over time and geographic variability observed in this study.

The mixed model analysis using water source and fluoridation status as separate variables found that region, pickup time, water type (source and fluoridation status) were treated as fixed effects; pickup time was treated as a repeated measure. County and location were treated as random effects.

With source and fluoridation status treated as a single variable, the model showed significant differences and resulted in a remarkably simple, practical model for use by dental practitioners, with only region (P = 0.035) and water type (P < 0.001) being significant effects. This model treated region, pickup time, and water type as fixed effects, and pickup time as a repeated measure. County and location were treated as random effects. The model showed significant differences between the Midwest and Northeast (P = 0.008), Midwest and South (P = 0.021), Midwest and West (P < 0.001), and the South and West (P = 0.087). Pickup time was also significant in the Northeast (P < 0.001), although this may be explained by an observation from a fluoridated PWS that reported that it failed to add fluoride on at least one occasion near a pickup time. It was no surprise that fluoridation affected fluoride levels (P < 0.001), or that the source of the water was also a significant predictor of fluoride content even when controlling for fluoridation (P < 0.022).

When the data were treated in this way, only region (P = 0.035) and water type (P < 0.001) were significant effects (Table 3). The effect of pickup time disappeared, and interactions between region and water type were not statistically significant. The fit of this model was only slightly worse than the more complicated (and difficult to use) model with fluoridation status and source as separate variables. Residual and normal QQ plots were examined to compare fits of these models. Residuals from the simple models were only slightly larger than, and had a similar dispersion to, those from the more complicated (and difficult to use in practice) model with fluoridation status and source as separate variables. Table 4 presents the predicted values and standard errors for each region and source as separate variables.
contain the mineral deposits found in older plumbing. In general, since every region has pockets of high levels of naturally occurring fluoride in the ground water, this approach is more appropriate for research than in patient care.

Each value was reviewed several times for accuracy but there were a few unusual observations. For example, in the New York locations, two sites in close proximity and receiving water from the same PWS had completely different fluoride concentrations for the two pickups. A follow-up phone call to the PWS revealed there had been a day around the first pickup when the addition of fluoride was overlooked, as reported by the contact. This was reflected in an increase from less than 30 mcg/100 g to over 100 mcg/100 g (pickup 2) in both sites. Since the initial pickups in sites 1 and 2 were within a few weeks of each other but not on the same day (as were the second pickups in the two sites), routine fluoridation at the PWS may have been overlooked more than once. In another county in Texas, two locations used water from the same PWS but one household used a reverse osmosis water treatment system. The home with the water treatment system had significantly less fluoride in the water samples than the other location. In this case, the survey proved useful in explaining some of the inconsistencies in the data.

These data will be added to a computer-based survey instrument for assessment of food and beverage intake currently being developed by the Nutrition Coordinating Center at University of Minnesota. They have been included in the National Fluoride Database, released in 2004 on NDL’s web site (http://www.ars.usda.gov/ba/bhnrc/ndl) and in Release 18 of the USDA National Nutrient Database for Standard Reference, released in late 2005.

Table 2
Test of fixed effects for model including region, pickup time, water source, and fluoridation status on variability of fluoride in drinking water

<table>
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<tr>
<th>Effect</th>
<th>df</th>
<th>F-value</th>
<th>P</th>
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</thead>
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<tr>
<td>Region</td>
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<td>4.82</td>
<td>0.0050</td>
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<tr>
<td>Pickup (time)</td>
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<td>5.52</td>
<td>0.0202</td>
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<td>Well water</td>
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<td>0.0215</td>
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<td>53.71</td>
<td>&lt;0.0001</td>
</tr>
<tr>
<td>Region × pickup</td>
<td>3</td>
<td>3.09</td>
<td>0.0294</td>
</tr>
<tr>
<td>Region × fluoridated</td>
<td>3</td>
<td>2.57</td>
<td>0.0590</td>
</tr>
<tr>
<td>Pickup × fluoridated</td>
<td>1</td>
<td>7.19</td>
<td>0.0062</td>
</tr>
<tr>
<td>Region × pickup × fluoridated</td>
<td>3</td>
<td>4.62</td>
<td>0.0041</td>
</tr>
</tbody>
</table>

Table 3
Effect of region and water type on variability of fluoride in drinking water

<table>
<thead>
<tr>
<th>Effect</th>
<th>df</th>
<th>F-value</th>
<th>P</th>
</tr>
</thead>
<tbody>
<tr>
<td>Region</td>
<td>3</td>
<td>3.06</td>
<td>0.0347</td>
</tr>
<tr>
<td>Water type</td>
<td>2</td>
<td>61.09</td>
<td>&lt;0.0001</td>
</tr>
</tbody>
</table>

aWell water, municipal fluoridated, or municipal non-fluoridated.
4. Conclusions

The fluoride database resulting from the national study will provide values on the fluoride content of tap water and other fluoride-contributing beverages and foods, support important research on public health research, and be of considerable value to USDA and other investigators in the US dental and medical research communities. This sampling approach for drinking water gives a good proportioning of the sample of counties to the states, Census Divisions and Census Regions and a good geographical distribution of the sampled counties to the States, Census Divisions and Census Regions, appropriate for the development of national fluoride values. These properties were considered most important to the appropriateness and usefulness of the data for the National Fluoride Database as well as dental and health research.

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References


