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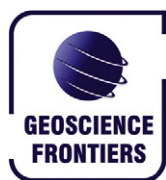
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Review paper

History and evaluation of national-scale geochemical data sets for the United States

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ABSTRACT

Six national-scale, or near national-scale, geochemical data sets for soils or stream sediments exist for the United States. The earliest of these, here termed the 'Shacklette' data set, was generated by a U.S. Geological Survey (USGS) project conducted from 1961 to 1975. This project used soil collected from a depth of about 20 cm as the sampling medium at 1323 sites throughout the conterminous U.S. The National Uranium Resource Evaluation Hydrogeochemical and Stream Sediment Reconnaissance (NURE-HSSR) Program of the U.S. Department of Energy was conducted from 1975 to 1984 and collected either stream sediments, lake sediments, or soils at more than 378,000 sites in both the conterminous U.S. and Alaska. The sampled area represented about 65% of the nation. The Natural Resources Conservation Service (NRCS), from 1978 to 1982, collected samples from multiple soil horizons at sites within the major crop-growing regions of the conterminous U.S. This data set contains analyses of more than 3000 samples. The National Geochemical Survey, a USGS project conducted from 1997 to 2009, used a subset of the NURE-HSSR archival samples as its starting point and then collected primarily stream sediments, with occasional soils, in the parts of the U.S. not covered by the NURE-HSSR Program. This data set contains chemical analyses for more than 70,000 samples. The USGS, in collaboration with the Mexican Geological Survey and the Geological Survey of Canada, initiated soil sampling for the North American Soil Geochemical Landscapes Project in 2007. Sampling of three horizons or depths at more than 4800 sites in the U.S. was completed in 2010, and chemical analyses are currently ongoing. The NRCS initiated a project in the 1990s to analyze the various soil horizons from selected pedons throughout the U.S. This data set currently contains data from more than 1400 sites. This paper (1) discusses each data set in terms of its purpose, sample collection protocols, and analytical methods; and (2) evaluates each data set in terms of its appropriateness as a national-scale geochemical database and its usefulness for national-scale geochemical mapping.

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1. Introduction

V.M. Goldschmidt, generally considered to be the father of modern-day geochemistry, stated that the primary purpose of geochemistry is to determine quantitatively the chemical composition of the Earth and its parts, and to discover the laws that

control the distribution of the individual elements (Goldschmidt, 1937, 1954). Understanding the abundance and distribution of the chemical elements in the near-surface environment of the Earth is critical for such fields as risk-based assessment of contaminated land, agriculture, animal and human health, water quality, land-use planning, mineral exploration, industrial pollution, and environmental regulation. The first national-scale geochemical surveys to address these issues began in the 1960s and expanded in number during the latter part of the twentieth century (Garrett et al., 2008). These studies resulted in a number of geochemical databases and atlases for entire countries or, in some cases, multiple countries (e.g., Webb et al., 1978; Fauth et al., 1985; Thalmann et al., 1989; McGrath and Loveland, 1992; Lahermo et al., 1996; Kadūnas et al., 1999; Ottesen et al., 2000; Reimann et al., 2003; Imai et al., 2004; Salminen et al., 2005; De Vos et al., 2006).

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To qualify as a true national-scale geochemical data set, several criteria should be met:

- Relatively uniform sample coverage of the entire nation;
- A sample density sufficient to allow robust estimates of element abundance and spatial distribution. For a large country the size of the conterminous U.S., about 1000 sites would be considered the minimum necessary;
- Consistent sample medium, or media, collected at each site;
- State-of-the-art analytical protocols with detection limits below crustal abundance concentrations.

Over the past 50 years, there have been six geochemical studies conducted in the United States (U.S.) that focused on element distribution in stream sediments or soils and meet at least some of the criteria above for a national-scale geochemical survey. The purpose of this paper is to: (1) provide a brief review of each of these studies in terms of purpose, sampling protocols, and analytical methods; and (2) evaluate the strengths and weaknesses of each generated data set in terms of its appropriateness as a national-scale geochemical database and its usefulness for national-scale geochemical mapping.

2. USGS 'Shacklette' database for soils

2.1. Background and purpose

The first effort to produce a national-scale geochemical database for the U.S. was conducted by the U.S. Geological Survey (USGS) from 1961 through 1975 using soil as the sampling medium (Boerngen and Shacklette, 1981; Shacklette and Boerngen, 1984; Gustavsson et al., 2001). The project, which focused only on the conterminous U.S. (7.9 million km²), was conceived and coordinated by H.T. Shacklette, a research botanist with the USGS. The

stated purpose of the project was to obtain estimates of the range of element concentrations in soils that were unaltered, or very little altered, from their natural condition (Shacklette and Boerngen, 1984). For this reason, this data set is considered to represent the background range for elements in soils of the conterminous U.S.

2.2. Sampling and analytical protocols

Sampling was conducted by USGS personnel at sites located along their routes of travel to project areas and within project areas in various parts of the U.S. The location of the sites, therefore, depended on both the road network and the destination of the samplers. In general, sampling sites were selected at 80-km intervals along the roads. Samples were collected at a total of 1323 sites, representing a density of approximately 1 site per 6000 km², between 1961 and 1975 (Fig. 1). Phase 1 of the study was conducted between 1961 and 1971 and resulted in the collection of 863 samples. The remaining samples were collected between 1971 and 1975 with sites chosen to complete more uniform sample coverage of the conterminous U.S. Because so many different people were involved in the sampling and because the collection of these samples was incidental to the other duties of the samplers, Shacklette kept the sampling protocol as simple as possible. He requested that, if possible, the samples should be collected from sites sustaining native vegetation. In some areas, however, only cultivated fields were available for sampling. The sampling protocol called for collection of soil samples from a depth of approximately 20 cm to minimize the effects of surface contamination, if present. Most of the samples represented true "soils" in that they were a mixture of weathered rock or unconsolidated parent material and organic matter and they supported plant growth. However, a few of the samples represented other regoliths such as sand dunes, loess, and beach deposits containing no visible organic matter.

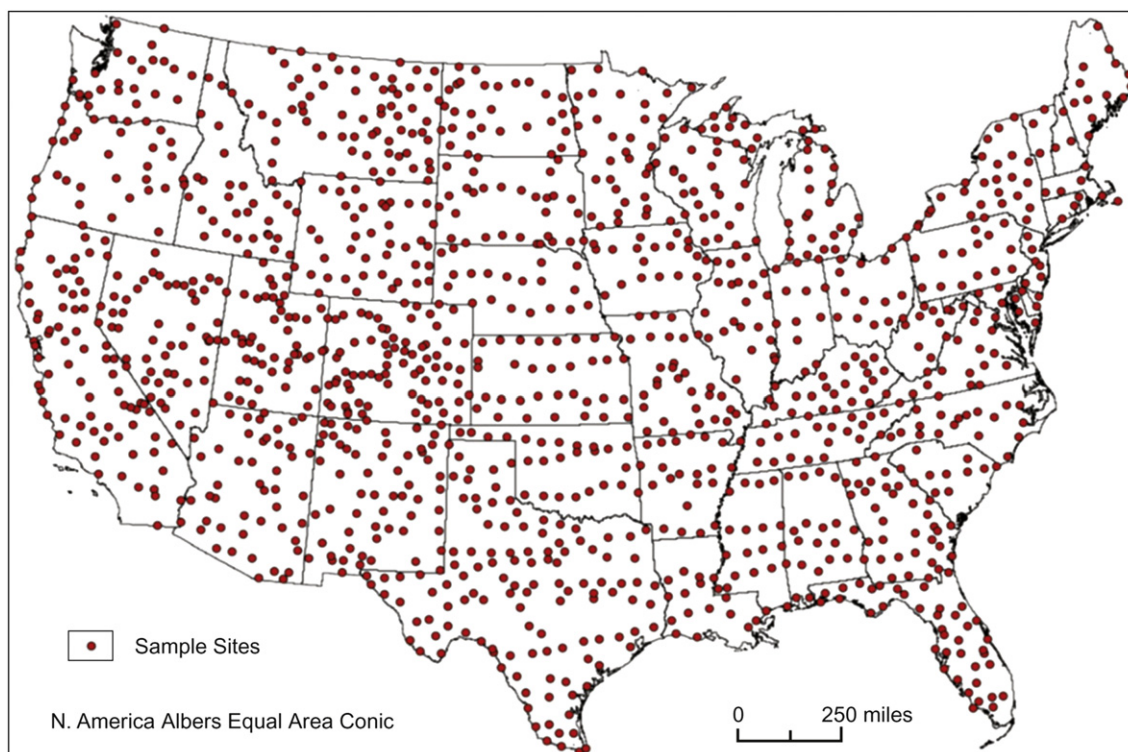


Figure 1. Location of 1323 soil samples collected by USGS from 1961 to 1975 (Boerngen and Shacklette, 1981; Shacklette and Boerngen, 1984).

Consequently, all the publications from this project refer to the samples as “soils and other surficial materials.”

The samples were shipped from the field to the USGS laboratories in Denver, Colorado, where they were oven dried at 28–30 °C, pulverized if necessary, and sieved to less than 2 mm. This fraction was further milled to less than 75 µm prior to chemical analysis. Boerngen and Shacklette (1981) report analytical concentrations for 50 elements (Ag, Al, As, B, Ba, Be, Bi, Br, C, Ca, Cd, Ce, Co, Cr, Cu, F, Fe, Ga, Ge, Hg, I, K, La, Li, Mg, Mn, Mo, Na, Nb, Nd, Ni, P, Pb, Pr, Rb, S, Sb, Sc, Se, Si, Sn, Sr, Ti, Th, U, V, Y, Yb, Zn, and Zr) analyzed by a variety of methods, all of which provided total elemental concentrations. The methods of analysis for some elements were changed during the study as new techniques became available.

Thirty of the elements (Ag, Al, B, Ba, Be, Bi, Cd, Ce, Co, Cr, Cu, Fe, Ga, La, Mg, Mn, Mo, Na, Nb, Nd, Ni, Pb, Pr, Sc, Sr, Ti, V, Y, Yb, and Zr) were analyzed by a semiquantitative six-step emission spectrographic method (Myers et al., 1961; Grimes and Marranzino, 1968; Neiman, 1976). This method reported concentrations in six geometric brackets per order of magnitude. For example, samples with concentrations of a given element between 10 and 100 mg kg⁻¹ would only be reported in the following steps: 10, 15, 20, 30, 50, 70, and 100. No values were reported between these six steps. The remaining 20 elements were analyzed by methods shown in Table 1.

2.3. Geochemical maps

The raw data from the study were published by Boerngen and Shacklette (1981); an interpretive report, including statistical summary tables and black-and-white point-symbol geochemical maps, was published by Shacklette and Boerngen (1984). In 2001, more modern mapping methods using a ‘Bootstrap’ estimate of the moving weighted median were utilized to produce new, full-color, geochemical maps for 22 elements (Gustavsson et al., 2001). Fig. 2 shows the map of Cu distribution from this study and illustrates the wide range of concentrations from high values in the region of serpentinite soils of northern California and southern Oregon to the

low values in the coastal sediments of Florida and other southeastern states.

Despite the extremely low sampling density and the age of the study, the Shacklette data set and reports are, by far, the most-often-quoted references for estimating background concentrations of major and trace elements in U.S. soils. Dragun and Chekiri (2005) reviewed more than 300 papers and books related to background concentrations of elements in North American soils and summarized the information by element and state for the U.S. In their table for As, the Shacklette data set was cited 67 times, whereas all other references were only cited 35 times. For Cu, the Shacklette data set was cited 70 times, compared to 32 times for all the other references reviewed. This data set for the conterminous U.S. has proved so useful that a similar study was subsequently completed in Alaska (Gough et al., 1984, 1988, 2005).

2.4. Strengths and weaknesses of the Shacklette data set

This data set was the first national-scale geochemical survey done for such a large country and included data for a large number of elements. It has been extensively used by a variety of disciplines to provide estimates of the background concentrations for those environmentally important elements that were analyzed. The Shacklette data set meets most of the four criteria for national-scale geochemical surveys listed in a section 1. The sample coverage is relatively uniform and a consistent sample medium was collected at each site. Although the sample density is considered quite low (1 site per 6000 km²), Smith and Reimann (2008) have demonstrated that the geochemical patterns produced are robust. The primary criticism of the study is in the analytical protocols. The semiquantitative, six-step emission spectrographic method used for 30 elements does not provide a continuous data distribution of element concentrations. In addition, the detection limit is considerably above crustal abundance for some important elements (e.g., Cd), resulting in a censored data set (most values below the detection limit) for these elements. Given that this study was conducted in the 1960s and 1970s, the data set has been a remarkable contribution to our understanding of the chemical composition of the Earth.

3. National Uranium Resource Evaluation Hydrogeochemical and Stream Sediment Reconnaissance (NURE-HSSR) Program

3.1. Background and purpose

Anticipating the need for additional U resources to support the burgeoning U.S. nuclear energy policies, the U.S. Atomic Energy Commission (later the Energy Research and Development Administration and now the U.S. Department of Energy, or DOE) initiated the National Uranium Resource Evaluation (NURE) Program in 1973. The Hydrogeochemical and Stream Sediment Reconnaissance (HSSR), one of nine components within the NURE Program, began in 1975 with the intention of determining the geochemical composition of sediments and soils from approximately 650,000 sites throughout the U.S. (Bolivar, 1980; ISP, 1985; Smith, 1997).

Four DOE laboratories were given regional responsibility for the collection and analysis of NURE-HSSR samples. Lawrence Livermore Laboratory (LLL) in Livermore, California was responsible for the West Coast States, Great Basin States, and Southwest States. Los Alamos Scientific Laboratory (LASL) in Los Alamos, New Mexico coordinated the Rocky Mountain States and Alaska. Oak Ridge Gaseous Diffusion Plant (ORGDP) in Oak Ridge, Tennessee oversaw the Central Plains States and Upper Midwest States. Savannah River Laboratory (SRL) in Aiken, South Carolina directed the project in the East Coast States, Appalachian States, and Southeast Gulf Coast

Table 1

Analytical methods for 20 elements reported in Boerngen and Shacklette (1981).

Element	Method
As	Arsine evolution-spectrophotometric-isotope dilution ^a
Br	Unspecified method
C	Combustion ^a
Ca	EDTA titration ^b ; X-ray fluorescence spectrometry ^c
F	Specific-ion electrode ^a
Ge	X-ray fluorescence spectrometry ^c
Hg	Flameless atomic absorption ^a
I	Unspecified method
K	Flame photometry ^b ; X-ray fluorescence spectrometry ^c
Li	Atomic absorption spectrometry ^a
P	Colorimetry ^d ; X-ray fluorescence spectrometry ^c
Rb	Atomic absorption spectrometry ^a
S	X-ray fluorescence spectrometry ^c
Sb	Unspecified method
Se	X-ray fluorescence spectrometry ^c
Si	X-ray fluorescence spectrometry ^c
Sn	Unspecified method
Th	Delayed neutron activation analysis ^{e,f}
U	Delayed neutron activation analysis ^{e,f}
Zn	Colorimetry ^d ; atomic absorption spectrometry ^a

^a Huffman and Dinnin (1976).

^b Shacklette et al. (1971).

^c Wahlberg (1976).

^d Ward et al. (1963).

^e Millard (1975).

^f Millard (1976).

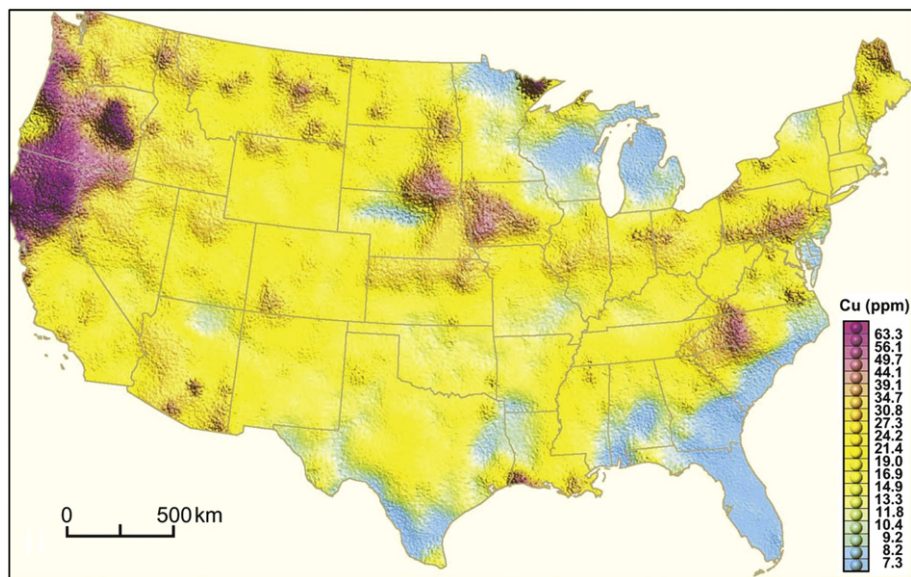


Figure 2. Map of Cu distribution based on the data from Boerngen and Shacklette (1981) and published in Gustavsson et al. (2001).

States. These areas of responsibility were modified twice during the NURE-HSSR Program. In 1977, the NURE Program was reorganized from studies based on State borders to 1° by 2° quadrangle studies (1° by 3° quadrangles in Alaska) and the boundaries of each DOE laboratory area of responsibility were adjusted. Then in 1979, SRL assumed responsibility for most of the LLL 1° by 2° quadrangles in the Western U.S. Each of these changes affected the timing and site distribution of the samples collected.

According to an annual report published in June of 1979 (Bendix Field Engineering Corp., 1979), the goal was to complete the NURE-HSSR work for all 621 quadrangles of the U.S. by 1984. However, an accident on March 28, 1979 in the Unit 2 reactor at Three Mile Island Nuclear Power Plant in Pennsylvania increased congressional opposition to the NURE Program to the point where funding was significantly reduced. Sample collection ceased as individual contracts that were already in place expired. Although the NURE-HSSR Program continued for another 4 years, the reduced effort was focused on completing the analytical work and publishing quadrangle reports for samples previously collected. In 1984, the final act of the program was to transfer all of the NURE-HSSR data, maps, field notes, and archived sample splits to the USGS (Grimes, 1984).

When the last NURE-HSSR sediment sample was collected on October 1, 1980, only 354 quadrangles had been completely sampled and another 70 quadrangles had been partially sampled (Averett, 1984), resulting in coverage of only about 65% of the U.S. (Smith, 1997).

3.2. Sampling and analytical protocols

Unlike a national-scale geochemical mapping project that has the goal of determining the abundance and spatial distribution of selected elements, the NURE-HSSR Program was designed to explore for and evaluate potential U resources. For this reason, the four DOE laboratories were encouraged to be “imaginative and innovative” in their individual approach and methodology (Shannon, 1977; Bolivar, 1980). Each laboratory determined what it considered to be the most favorable sample media within its own region of responsibility, wrote its own sampling protocol manuals (LASL: Sharp and Aamodt, 1978; LLL: Lawrence Livermore Laboratory, 1976; ORGDP: Uranium Resource Evaluation Project, 1978; Arendt et al., 1979; SRL: Ferguson et al., 1976, 1977; Price

and Jones, 1979), and subcontracted most of the actual sample collection. The sample medium most commonly collected throughout the U.S. was stream sediments, followed by soils in the Great Basin states and a significant number of lake sediments in Alaska. The majority of the sediments and soils were sieved to $<150\ \mu\text{m}$ prior to analysis. Depending upon the responsible laboratory, regional geology, topography, and access, the density for the NURE-HSSR Program ranged from 1 site per $10\ \text{km}^2$ to 1 site per $26\ \text{km}^2$. More than 378,000 sediment and soil sites were sampled before the program was terminated (Fig. 3).

Originally, the samples were only analyzed for U. However in 1977, the analytical requirements of the NURE-HSSR Program were expanded to include Th, Li, and several other elements as selected by each laboratory (Bendix Field Engineering Corp., 1978). As a result, each laboratory used their own group of analytical methods and determined a different suite of elements. Early in the program, an inter-laboratory quality control program was established to ensure that measurements of U concentration were maintained within acceptable levels. However, attempts to create similar quality control procedures for multielement analytical methods were not successful (ISP, 1985).

All of the laboratories used delayed neutron counting to analyze for U and, to varying extents, instrumental neutron activation analysis for multielement determinations. Other analytical methods included fluorimetry (ORGDP), energy-dispersive X-ray fluorescence (LASL), emission spectrography (LASL, ORGDP, SRL), atomic absorption spectrophotometry (ORGDP, SRL), and colorimetry (SRL) (Bolivar, 1980; Grimes, 1984). The NURE-HSSR database compiled by Smith (1997) gives analytical results for a total of 54 elements (Ag, Al, As, Au, B, Ba, Be, Bi, Br, Ca, Cd, Ce, Cl, Co, Cr, Cs, Cu, Dy, Eu, F, Fe, Hf, Hg, K, La, Li, Lu, Mg, Mn, Mo, Na, Nb, Ni, P, Pb, Pt, Rb, Sb, Sc, Se, Sm, Sn, Sr, Ta, Tb, Th, Ti, U, V, W, Y, Yb, Zn, and Zr). However, no sample was analyzed for more than 46 elements and some samples were never analyzed for any element other than U.

3.3. Geochemical maps

One of the last publications produced by LASL for the NURE-HSSR Program was a geochemical atlas for the state of Alaska (Weaver et al., 1983). The stream- and lake-sediment data from 61,923 sample sites were interpolated by universal kriging

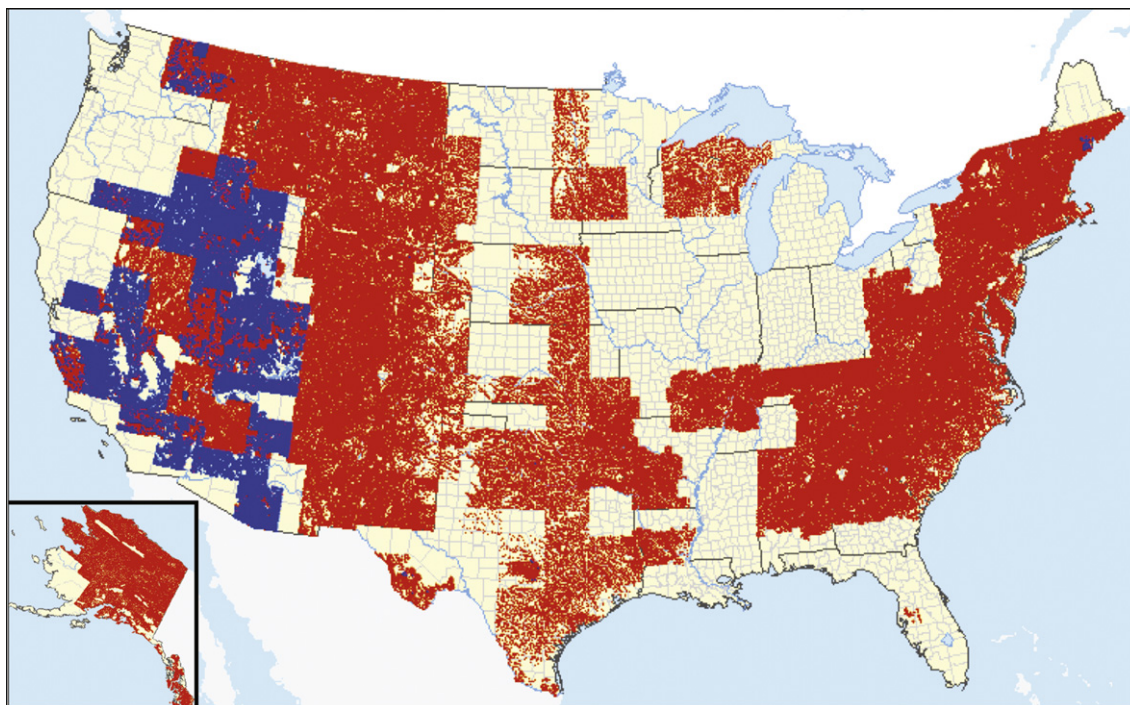


Figure 3. Location of 378,789 samples collected for the NURE-HSSR Program between 1975 and 1980 (based on data from Smith, 1997). Sediment samples are shown in red; soil samples are in blue.

techniques into 46,276 grid cells of 5 km on each side. The results for 41 elements were displayed at a 1:6,000,000 scale with 256 levels of color and color intensities representing the elemental concentration of each grid cell. As an example, Fig. 4 shows the results that Weaver et al. (1983) obtained for the distribution of Co.

Most of the elemental variation illustrated in the geochemical maps produced by Weaver et al. (1983) can be attributed to underlying geologic factors. However, these maps also reveal another source of variation in the NURE-HSSR data. Large blocks of data, readily identifiable by their straight edge boundaries, show the effects of uncorrected bias in the analytical results. For example, the Co map (Fig. 4) shows a rectangular block (four 1° by 3° quadrangles) in the center of the state where the results are elevated with respect to the surrounding areas despite the fact that the surrounding quadrangles were analyzed by the same laboratory and method. This problem is even more pronounced in areas where different laboratories and methods were employed.

Neither LASL nor any of the other DOE laboratories published geochemical maps for their areas of responsibility in the conterminous U.S. Several years after NURE had ended, Grossman (1998) produced geochemical maps from the HSSR data for 11 elements (As, Ce, Cu, Fe, Hf, Na, Pb, Th, Ti, U, and Zn). Data from sediment and soil samples were mathematically leveled to remove analytical bias and then gridded into 2-km by 2-km cells using a minimum curvature interpolation algorithm to create a smoothed surface. The resultant grids were mapped in colors representing 7 data classes with breaks at the 40th, 80th, 90th, 95th, 98th, and 99th percentiles. The distribution of U (Fig. 5) highlights regional geologic structures as well as known U-producing districts. The areal coverage shown in Fig. 5 also illustrates the gaps resulting from the unforeseen early termination of the NURE-HSSR Program. Unlike U, which was determined in almost every sample, the geochemical map for As (Fig. 6) illustrates that this element was analyzed only in a relatively small percentage of the HSSR samples because of variation in the elements analyzed by the different laboratories.

3.4. Strengths and weaknesses of the NURE-HSSR data set

One of the greatest strengths of the NURE-HSSR data set is the incredible sample density ranging from 1 site per 10 km² to 1 site per 26 km². No other national-scale geochemical sampling program in the U.S. approaches this density. As a result, the NURE-HSSR data are often used at local and regional scales that would be inappropriate for a lower-density data set. Another strength is the limited time frame of the study. The NURE-HSSR data represent a geochemical snapshot of the U.S. between 1975 and 1980. Archived splits of the samples have been reanalyzed by more modern analytical methods to supplement research into mineral resource assessments, environmental changes, and medical geology.

Unfortunately, because the NURE-HSSR Program was designed as a U exploration program and not as a geochemical mapping project, the data do not satisfy three of the four criteria listed in section 1 for a national-scale geochemical data set:

- (1) The NURE-HSSR data do not provide a relatively uniform coverage of the entire nation. About one-third of the U.S., including entire states and large regional provinces, was not sampled (Fig. 3).
- (2) The sample medium was not consistent between sites. Although the majority of the samples are <150-μm stream sediments, large areas of the country were sampled for soils or lake sediments. Mixed in with these are also a few pond, spring, playa, and stream-bank sediments as well as samples sieved to different size fractions. The samples were collected by subcontractors for four laboratories using seven different sampling protocol manuals. The accompanying field observations vary in type, coded responses, and quality.
- (3) The analytical protocols were not consistent. Care was taken to produce reliable U determinations at concentrations below crustal abundance. However, the different multielement methods utilized varied between laboratories, often

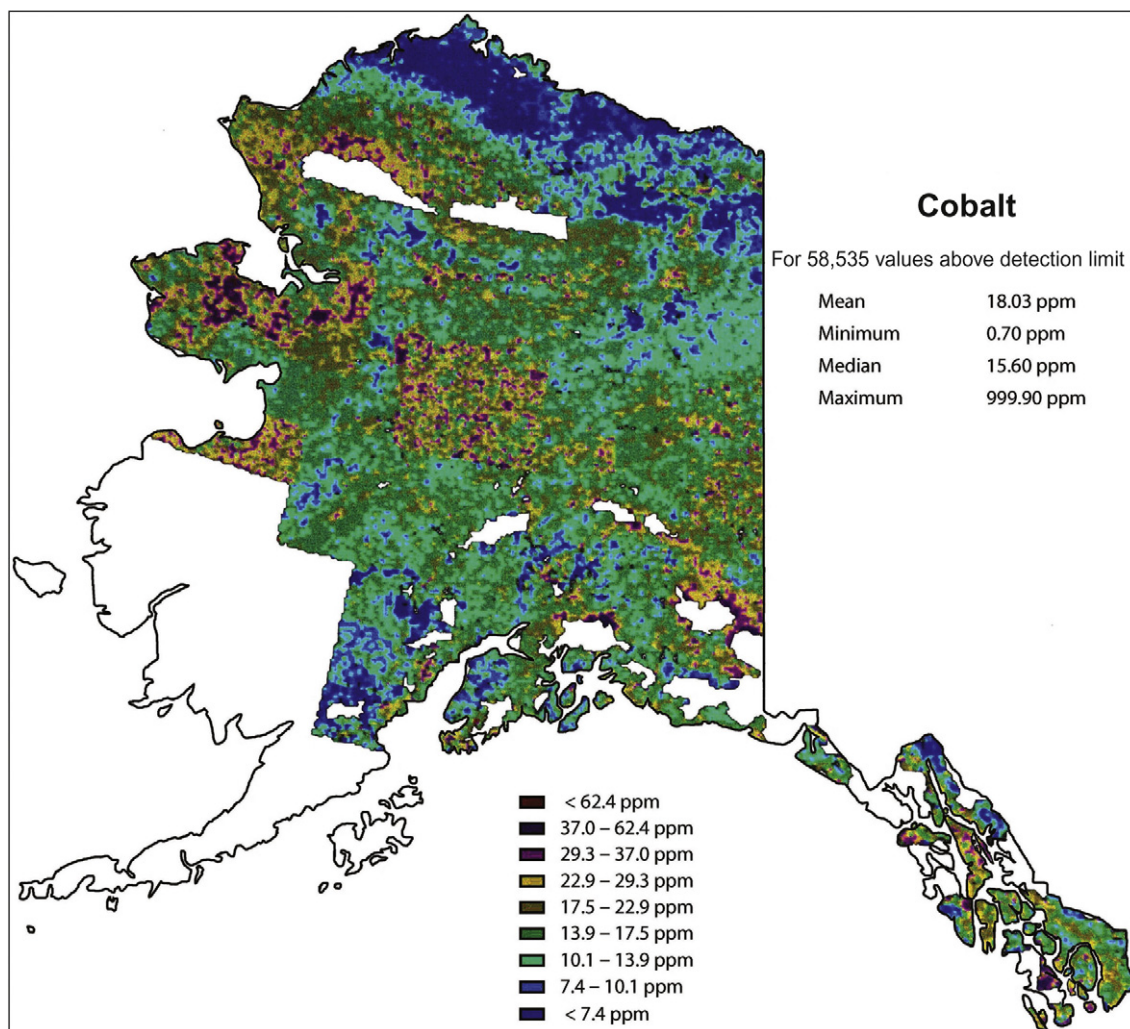


Figure 4. Map of Co distribution for NURE-HSSR stream- and lake-sediment samples collected in Alaska by the Los Alamos Scientific Laboratory (modified from Weaver et al., 1983).

had detection limits above crustal abundance for several elements, and suffered from problems with analytical bias (Fig. 4).

3.5. Geochemical maps based on NURE airborne radiometric data

A discussion of national-scale geochemistry of the U.S. would be incomplete without mentioning the maps produced from the airborne radiometric surveys conducted during the NURE Program in the late 1970s and early 1980s. These surveys measured natural gamma radiation generated by surface materials to a depth of ca. 30 cm and provided data on the concentration of the three naturally occurring radioactive elements K, U, and Th. The surveys were flown using both fixed-wing and helicopter systems with 33–50 L of thallium-activated sodium iodide crystals. The nominal survey altitude was 122 m with survey lines flown generally east-west at spacings of 1.6–10 km. Tie lines were flown perpendicular to the flight lines at intervals of 16–30 km (Duval, 1990). The NURE radiometric data for these three elements have been used to generate the highest resolution geochemical maps that exist, or likely will ever exist, for the U.S. (Duval, 1990; Duval et al., 1990; Phillips et al., 1993). It would require hundreds of thousands to millions of soil or sediment samples to produce maps at an equivalent resolution. Fig. 7 shows the K map generated from these radiometric surveys.

4. NRCS ‘Holmgren’ data set for soils of the conterminous U.S.

4.1. Background and purpose

The U.S. Department of Agriculture’s Natural Resources Conservation Service (NRCS), under the leadership of G.G.S. Holmgren, initiated a project in 1978 whose stated purpose was to determine background concentrations of Pb and Cd in the major food crops of the U.S. and in the soils on which these crops grow (Holmgren et al., 1993). Such information was needed by regulatory agencies charged with developing action levels for maximum allowable Pb and Cd in foods. The study also provided data on Zn, Cu, and Ni in these soils. The information on site selection, sampling protocols, and analytical methods given below is taken from Holmgren et al. (1993).

4.2. Sampling and analytical protocols

Sampling of soils and crops from 3045 sites, representing 307 different soil series, was conducted from 1978 to 1982. These sites were located only in the major crop producing areas of the U.S. that contribute a significant portion of Cd to the diet of U.S. citizens (Fig. 8). As a result, the distribution of samples is very uneven with some multi-state regions of the conterminous U.S. having no samples. The sites shown in Fig. 8 represent individual counties in which samples

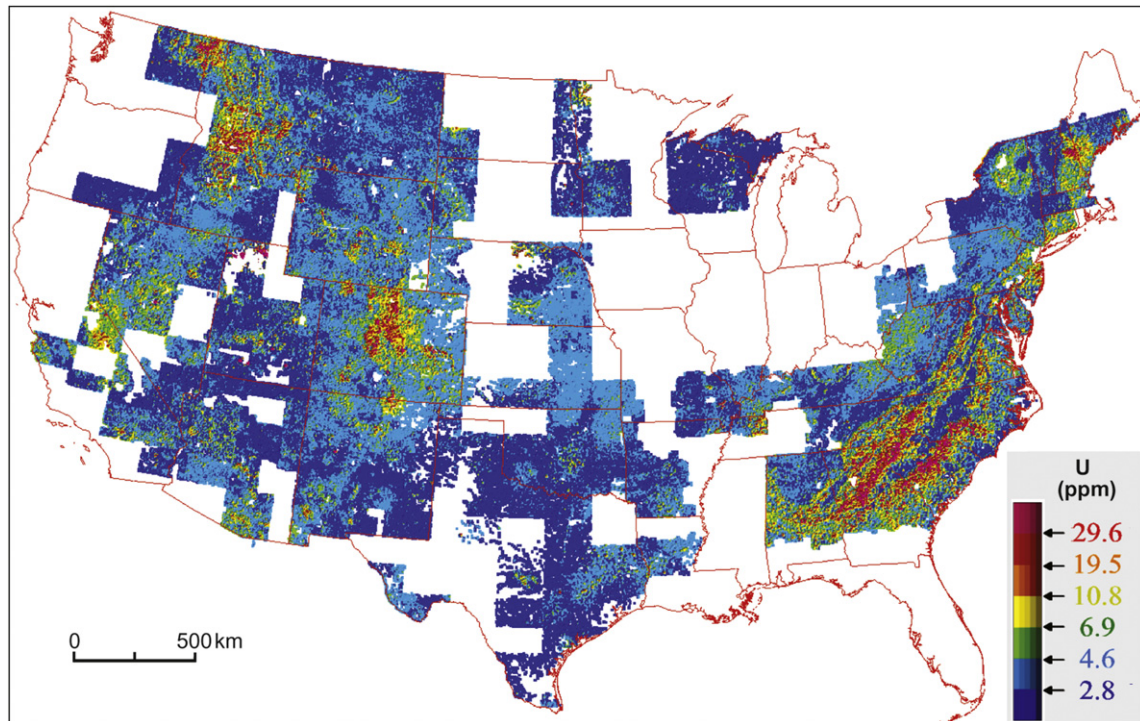


Figure 5. Map of U distribution for NURE-HSSR sediments and soils of the conterminous U.S. (from Grossman, 1998).

were collected, and each of the sites may represent many samples. For example, Fig. 8 shows 3 sites in Oregon representing 3 counties in which a total of 106 samples were collected. Care was taken that each site was at least 8 km from any stack emitter such as a coal-fired power plant or smelter, 200 m from major highways, 100 m from rural roads, 100 m from current or abandoned building sites, and 50 m

from field boundaries. Wolnik et al. (1983a, b; 1985) reported on metal concentrations in the crop samples and Holmgren et al. (1993) discussed Cd, Pb, Zn, Cu, and Ni in the soils.

Sample sites were chosen where a mature crop was growing on a previously selected soil series. Surface soil samples and one or two subhorizons to a depth of 50 cm were collected. The samples

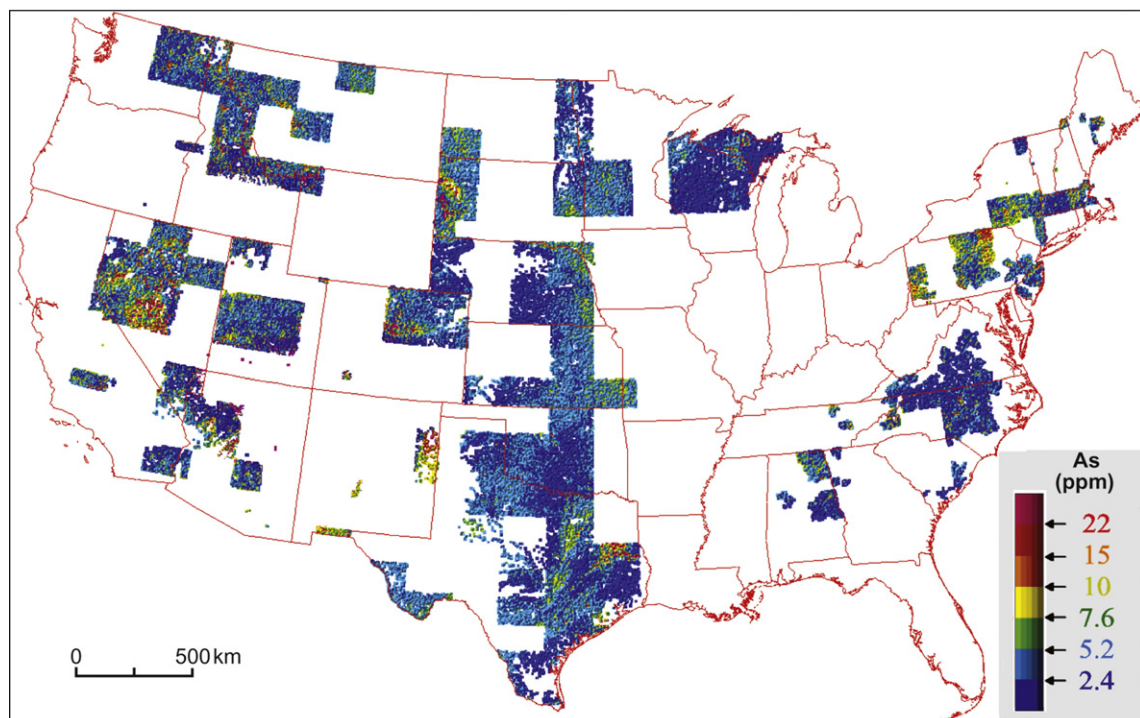


Figure 6. Map of As distribution for NURE-HSSR sediments and soils of the conterminous U.S. (from Grossman, 1998).

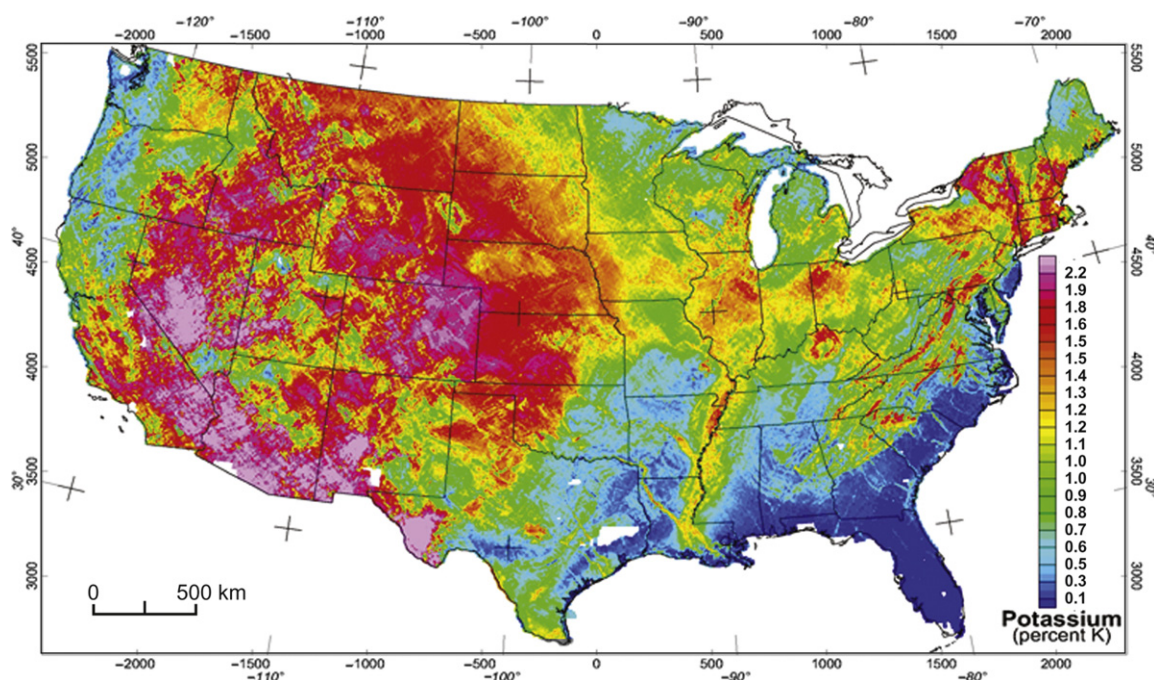


Figure 7. Map of K distribution for the conterminous U.S. based on airborne radiometric surveys (from Duval, 1990).

were mixed by kneading the soil within the plastic collection bags. The unsieved samples were air-dried at 35 °C and then crushed with a stainless steel roller. A subsample of the crushed material was digested in concentrated HNO₃ for approximately 15 h at 150 °C. Lead was then analyzed by anodic stripping voltammetry, Cd by graphite-furnace atomic absorption spectrometry, and Cu, Ni, and Zn by direct-current plasma spectrometry.

4.3. Geochemical maps

To generate maps showing soil element concentrations, Holmgren et al. (1993) averaged all data within half degree cells of

latitude and longitude. The presence of more than one sampling site per cell reduced the number of mapped locations from 470 to approximately 150 map points (Holmgren et al., 1993). They then used a regression tree approach to generate the areal divisions shown on the maps. This procedure considers the cell average and its corresponding centroid of latitude and longitude. The data set is examined in terms of all possible divisions by latitude or longitude, and the areal division is selected which results in the greatest decrease in variance from the original data set. Subsequent divisions are generated by this same procedure until an arbitrarily selected threshold is reached. The resulting regions shown on the maps produced by Holmgren et al. (1993) are a nonparametric

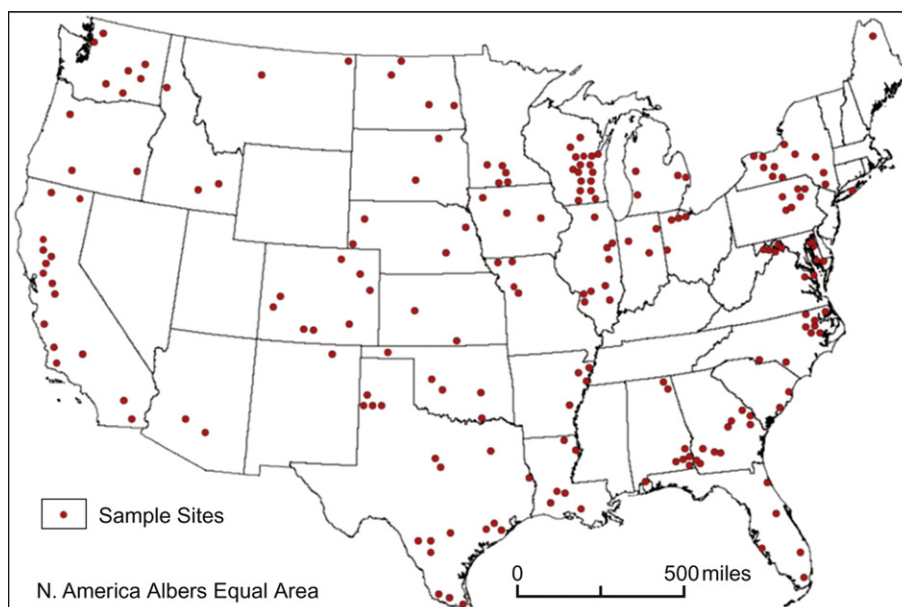


Figure 8. Location of soil sampling sites from Holmgren et al. (1993). Each site represents a county in which several samples were collected.

representation of the areas of maximum uniformity as represented by the data set.

Fig. 9 shows their map of Cu distribution based on these regions. Despite the sporadic distribution of sample sites, there are a number of similarities with the Cu map based on the Shacklette data (Fig. 2). Northern California has enriched concentrations of Cu in both maps because of the serpentinite soils in this area. One obvious dissimilarity is the high Cu concentration shown in southern Florida in Fig. 9. Holmgren et al. (1993) attributed these high values to agricultural practices for these soils, which included application of Cu fertilizers to correct Cu deficiency and Cu-containing fungicidal sprays.

Table 2 compares the geometric means and ranges for Cu, Zn, Ni, and Pb from the 'Shacklette' and 'Holmgren' data sets. Zinc and Cu show somewhat higher maxima in the 'Shacklette' data set. This is most likely because the 'Holmgren' data set does not include samples from some areas of the U.S. (e.g., Nevada, central Colorado) that contain major metal ore deposits. Soils in these regions may have formed on parent material that was naturally enriched in Cu, Zn, and other ore-related elements. Another contributing factor may be that the 'Holmgren' data were generated by a partial extraction that only involved concentrated HNO_3 , whereas the 'Shacklette' data were generated by methods yielding total elemental concentrations.

4.4. Strengths and weaknesses of the 'Holmgren' data set

The primary strength of this data set is that both crop and soil samples were collected from each site. This provides the opportunity to study element uptake by major agricultural crops that does not exist for the other five data sets. In addition, consistent sample collection and analytical protocols were used throughout the study. The weaknesses of the data set as a true indication of national-scale geochemical variation in soils are (1) the uneven distribution of

Table 2

A comparison of geometric means for Cu, Zn, Ni, and Pb from Shacklette and Boerngen (1984) and Holmgren et al. (1993).

Element	Geometric mean ^a (mg kg ⁻¹)	Geometric mean ^b (mg kg ⁻¹)	Range ^a (mg kg ⁻¹)	Range ^b (mg kg ⁻¹)
Cu	17	18	<1–700	<0.6–495
Zn	48	42.9	<5–2900	<3–264
Ni	13	16.5	<5–700	0.7–269
Pb	16	10.6	<10–300	<1–135

^a Shacklette and Boerngen (1984).

^b Holmgren et al. (1993).

sample sites throughout the conterminous U.S. and the low number (150) of resulting map points; (2) the samples were not sieved to a uniform grain size prior to analysis; (3) the soil samples were only analyzed for five elements (Cd, Pb, Zn, Cu, and Ni); and (4) the partial dissolution technique used (concentrated HNO_3) may cause difficulties in comparing data with studies that determined total element content.

5. USGS National Geochemical Survey

5.1. Background and purpose

Although the NURE-HSSR Program (section 3) only sampled about 65% of the U.S. and lacked consistency in terms of sample media, elements analyzed, and analytical methods used, the original data and, particularly data from reanalysis of archived NURE-HSSR samples, proved to be quite useful for a variety of studies ranging from regional-scale mineral resource appraisals to environmental investigations (e.g., John et al., 1993; Erdman et al., 1995; Smith, 1995; Nowlan, 1996; Goldhaber et al., 2001; Cannon et al., 2004). In 1997, the USGS established a project called the National Geochemical Survey (NGS), whose purpose was to provide

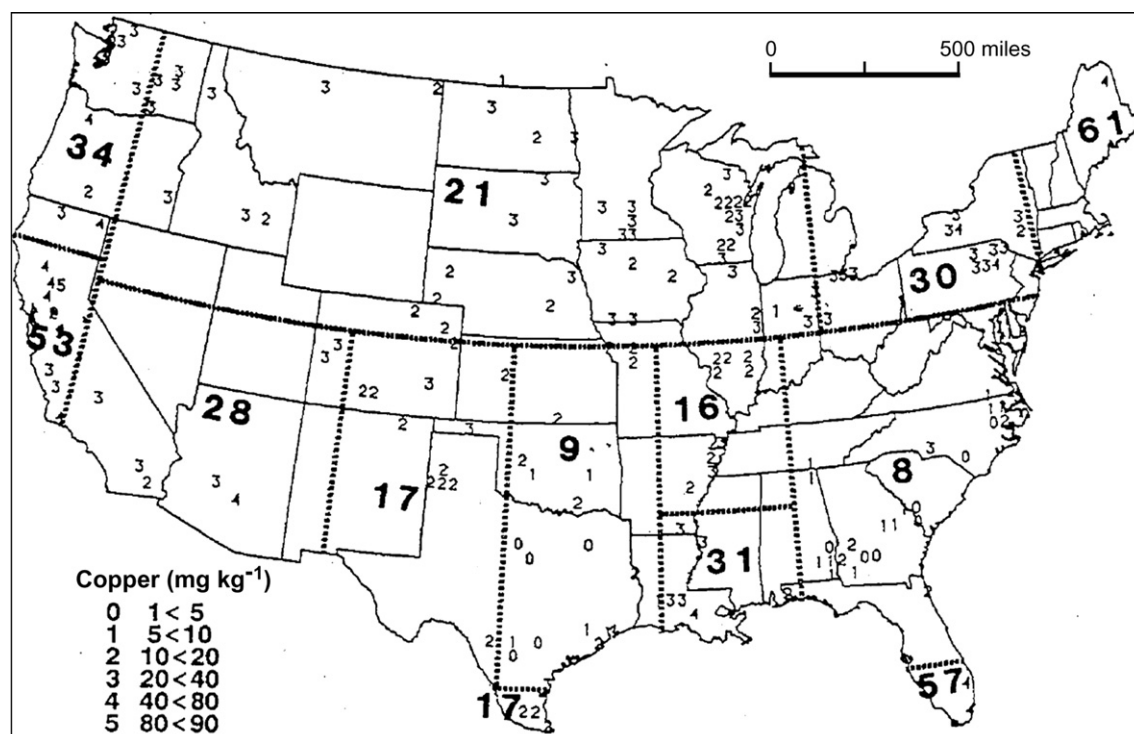


Figure 9. Geographic distribution of Cu in soils of the conterminous U.S. (from Holmgren et al., 1993). The bold numbers represent the means of the data within the selected areas. The small numbers represent codes for average concentrations within the counties sampled.

complete national-scale geochemical coverage of the U.S. using the NURE-HSSR sample archives as its starting point. The complete history of this effort is detailed in [USGS \(2004\)](#) and summarized below.

The sample design was based on a 17-km by 17-km sampling grid resulting in a planned sample density of 1 site per 289 km². To obtain national coverage, the NGS used data from the following sources ([USGS, 2004](#)): (1) NURE-HSSR samples that had already been reanalyzed by other USGS projects using appropriate methods; (2) A subset of the existing NURE-HSSR stream sediments, soils, and lake sediments that were reanalyzed by the NGS analytical methods, which are discussed in section 5.2; (3) Data from other USGS projects involving regional-scale geochemical sampling for stream sediments or soils, if the collection and analytical methods were compatible with those of the NGS; (4) When possible, samples taken from the archives of stream sediments collected by earlier USGS projects that were reanalyzed by the NGS to fill in any unsampled areas; (5) New sample collection and chemical analyses that were conducted by the NGS in areas where no coverage was available from either the NURE-HSSR Program or previous USGS projects. Stream sediment was the primary sample medium and was collected wherever possible. Data from all of these sources representing at least three broad categories of sample media (stream sediments, soils, and lake sediments) were then combined into a single national database.

5.2. Sampling and analytical protocols

Stream sediments were chosen as the principal sample medium because they were the primary sample medium used by the NURE-HSSR Program and stream sediments tend to integrate all sources of sediment (primarily rock and soil) in the stream's drainage basin. Soils, the other commonly used sample medium for regional- and national-scale geochemical surveys, are likely to represent smaller areas of influence. This stream-sediment-based sampling protocol potentially allows detection of geochemical anomalies due to mineralization or anthropogenic sources of contamination using

a much lower sampling density of stream sediments as compared to soils. However, good stream-sediment samples are not available in all locations. In the NURE-HSSR Program, stream sediments were collected at about 80% of the sites, soils at about 12% of the sites, and lake or pond sediments at about 8% of the sites. Because the NURE-HSSR samples provide the starting point for the NGS, all of these media were accepted for inclusion into the database and for reanalysis. In those parts of the U.S. where new sampling was conducted, soils were substituted for stream sediments only where necessary. This included areas of very low relief and poor drainage and farm lands where local streams had been largely channelized and diverted for agricultural purposes. [Fig. 10](#) shows the distribution of sample types for the NGS.

The sample preparation protocol for the NURE-HSSR samples was documented in section 3.2. Similar protocols were followed for samples that were collected for the NGS. After drying, both stream sediments and soils were sieved to <150 µm prior to chemical analysis. Aluminum, Ca, Fe, K, Mg, Na, P, Ti, Ag, As, Au, Ba, Be, Bi, Cd, Ce, Co, Cr, Cu, Eu, Ga, Ho, La, Li, Mn, Mo, Nb, Nd, Ni, Pb, Sc, Sn, Sr, Ta, Th, U, V, Y, Yb, and Zn were determined by inductively coupled plasma-atomic emission spectrometry (ICP-AES) using a method similar to [Briggs \(2002\)](#). The samples were digested in a mixture of HCl, HNO₃, HClO₄, and HF at low temperature (~125–150 °C). The 4-acid digestion is effectively a total dissolution for most mineral constituents of the sediment and soil samples. However, it does not fully dissolve some of the more refractory or resistant minerals such as barite, chromite, rutile, and cassiterite. In addition to the 40 elements determined by ICP-AES, As, Se, and Hg were determined by separate, single-element methods. Arsenic and Se were determined by hydride-generation atomic absorption spectrometry ([Hageman and Brown, 2002](#)). For determination of As, the sample was dissolved by fusion with sodium peroxide at 750 °C for 4 min and then, following cooling, taken into solution with HCl. For determination of Se, the sample was digested in a mixture of HNO₃, HF, and HClO₄. Mercury was determined by cold-vapor atomic absorption spectrometry using a modification of the method published by the U.S. Environmental Protection Agency ([USEPA, 2007](#)).

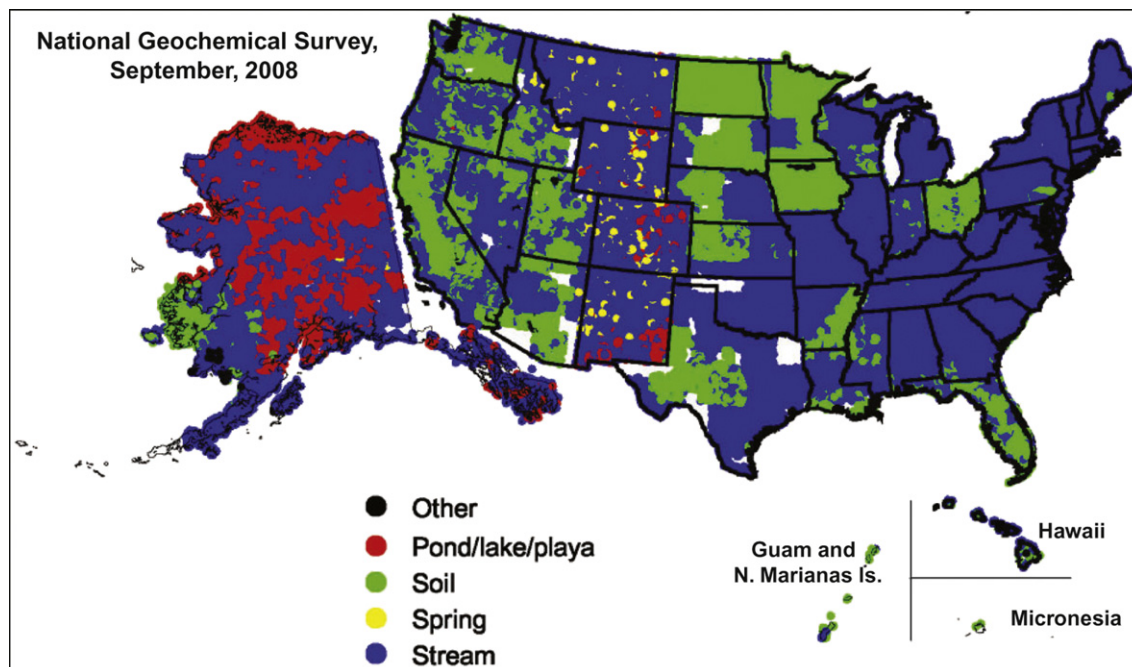


Figure 10. Distribution of sediment types and soils collected for the USGS National Geochemical Survey ([USGS, 2004](#)).

5.3. Geochemical maps

Geochemical maps for the analyzed elements are published and available on-line (USGS, 2004). Figs. 11 and 12 show geochemical maps for Cu and Pb, respectively. The higher resolution of these maps generated from a sample density of 1 site per 289 km² as compared to the maps based on the Shacklette data at a density of 1 site per 6000 km² is obvious from a comparison of the Cu maps in Figs. 2 and 11. The Pb map based on the NGS data set (Fig. 12) demonstrates some of the problems encountered when trying to combine data from different sample media and different analytical methods. Five rectangular geochemical features are pointed out in Fig. 12. These occur in northeast Washington (a), southwest North Dakota (b), the border area between North Dakota and Minnesota and between South Dakota and Minnesota (c), northwest Kansas (d), and south-central Oregon (e). Inspection of Figs. 3 and 10 shows that these features are generally caused by the juxtaposition of data from stream sediments collected during the NURE-HSSR Program with soils collected during the NGS. Differing analytical methods, with possible differences in detection limits, may also contribute to these features.

5.4. Strengths and weaknesses of the USGS NGS data set

The obvious strength of the NGS data set is its high sample density (1 site per 289 km²). True national-scale coverage was achieved through a combination of the original NURE-HSSR samples collected between 1975 and 1980, samples from other USGS projects involving regional-scale soil or stream-sediment geochemistry, and new sampling conducted from about 1997 to 2008. A large number of elements were analyzed by a standardized set of analytical methods.

The primary weakness of the NGS data set is the same as one of the weaknesses for the NURE-HSSR data set; namely, its use of multiple sample media (i.e., stream sediments, lake sediments, and soils). For some elements, this has resulted in geochemical patterns, such as the rectangular features discussed previously, which are not related to geogenic processes or anthropogenic activities but are fully explained by only this difference in sample media. Having no

consistent sample medium covering the entire country also makes it impossible to establish the true national-scale geochemical variation for any of the specific sample media used. The sample preparation protocol for soils, as established during the NURE-HSSR Program and used by the NGS, requires sieving each soil sample to <150 μ m. Unfortunately, the choice of this size fraction does not allow for comparison of the generated soil geochemical data with data generated from almost all other published broad-scale soil geochemical surveys conducted throughout the world, which use the <2-mm fraction for analysis.

6. North American Soil Geochemical Landscapes Project

6.1. Background and purpose

Although the NGS provided relatively high-density geochemical coverage for the entire U.S., soils were only collected for about 30% of the country. Therefore, the NGS data set is not useful to those agencies and organizations needing national-scale soil geochemical data. These generally include public health specialists concerned with soil pathways for potentially toxic elements and environmental regulators involved in risk-based assessment of contaminated land and in establishing soil screening levels and action levels. Therefore, in 2002, the USGS, the Mexican Geological Survey (Servicio Geológico Mexicano), and the Geological Survey of Canada initiated the North American Soil Geochemical Landscapes Project (NASGLP) with the final objective of full continental-scale soil geochemical coverage at a sample density of 1 site per 1600 km² (ca. 13,500 sites for the continent). The specific goals of the project are to: (1) develop a continental-scale design and protocols for generating soil geochemical and mineralogical data; (2) provide baseline soil geochemical and mineralogical data that are useful for a wide range of applications and disciplines; (3) interpret the resulting geochemical and mineralogical patterns in terms of processes acting at the broad scale of the project; and (4) establish an archive of soil samples for future investigators. The pilot phase of the project was conducted from 2004 to 2007 with results published in a series of scientific papers in a special issue of *Applied Geochemistry* (Bern, 2009; Cannon and Horton, 2009; Chipr  s et al.,

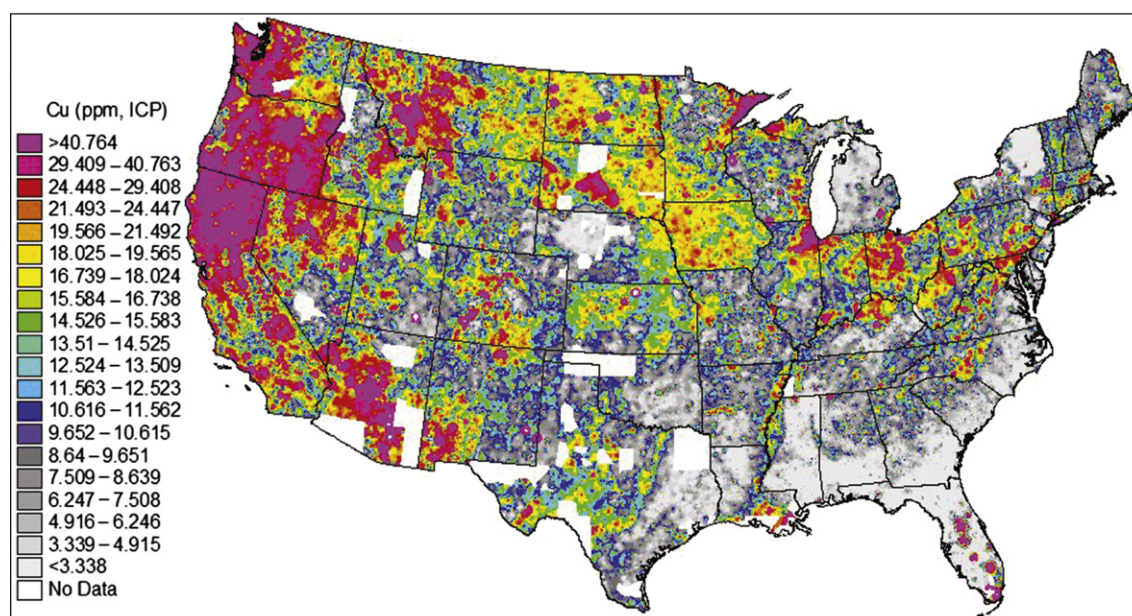


Figure 11. Geochemical map of Cu based on the USGS National Geochemical Survey (USGS, 2004).

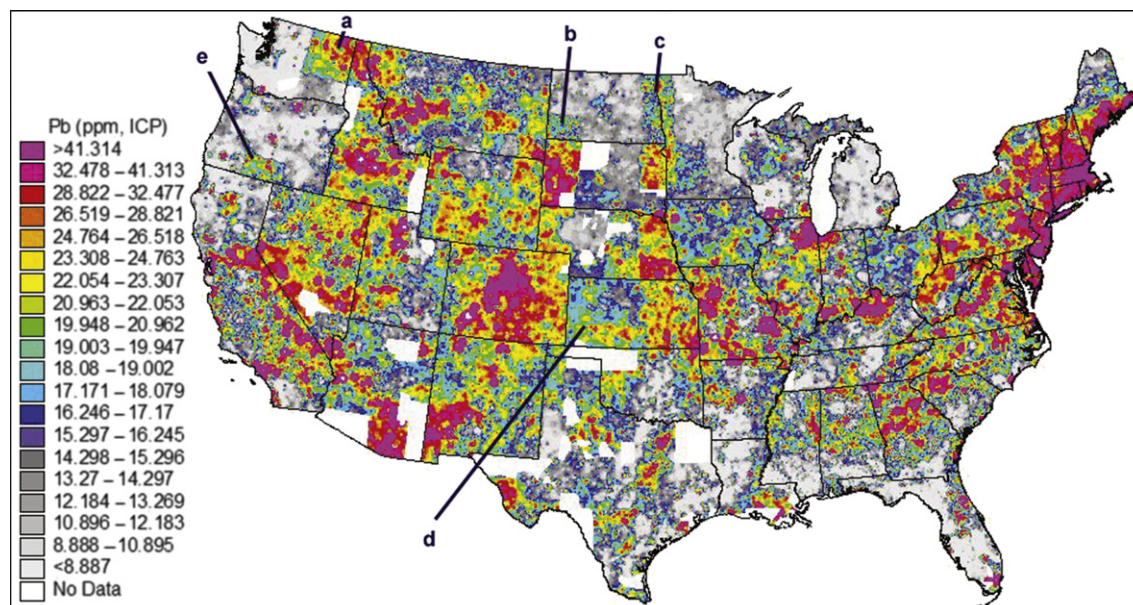


Figure 12. Geochemical map of Pb based on the USGS National Geochemical Survey (USGS, 2004) [a, b, c, d, and e point out the 'rectangular' geochemical features as discussed in the text].

2009a, 2009b; Eberl and Smith, 2009; Garrett, 2009; Garrett et al., 2009; Goldhaber et al., 2009; Griffin et al., 2009; Grunsky et al., 2009; Holloway et al., 2009; Klassen, 2009; McCafferty and Van Gosen, 2009; Morman et al., 2009; Morrison et al., 2009; Reeves and Smith, 2009; Smith, 2009; Smith et al., 2009; Tuttle and Breit, 2009; Tuttle et al., 2009; Wanty et al., 2009; Woodruff et al., 2009). The full-scale sampling of the continent began in 2007, and sampling was completed for the conterminous U.S. (ca. 4800 sites) in 2010 (Fig. 13). A complete history of the project is given by Smith et al. (2011) and summarized below.

6.2. Sampling and analytical protocols

Target sites were selected in the U.S. by a Generalized Random Tessellation Stratified Design (Olsen, 2005) at a nominal density of 1 site per 1600 km² for a total of 5813 sites, of which, 4871 are in the conterminous U.S. (Fig. 13). If a target site was inaccessible for any reason, an alternative site was chosen as close as possible to the original site, matching as closely as possible the landscape and soil characteristics of the original site. The following guidelines were also used in the site selection process to ensure that obviously polluted sites were avoided:

- (1) No sample should be collected within 200 m of a major highway.
- (2) No sample should be collected within 50 m of a rural road.
- (3) No sample should be collected within 100 m of a building or structure.
- (4) No sample should be collected within 5 km downwind of active, major industrial facilities such as power plants or smelters.

Sample-collection protocols were developed in a series of workshops with project stakeholders during 2003–2004. These protocols represent a combination of depth-based and horizon-based sampling. Ideally, at each site the following samples are collected: (1) soil from a depth of 0–5 cm, regardless of what soil horizon(s) this might represent; (2) a composite of the soil A

horizon; and (3) a sample from the soil C horizon or, in cases where the top of the C horizon was more than 1 m below the surface, a sample from a depth of approximately 80–100 cm.

Each sample was air-dried at ambient temperature, disaggregated, and sieved through a 2-mm stainless steel screen. The <2-mm material was crushed to <150 µm in a ceramic mill prior to chemical analysis. The analytical protocols were very similar to those used for the NGS discussed in section 5.2. Aluminum, Ca, Fe, K, Mg, Na, S, Ti, Ag, Ba, Be, Bi, Cd, Ce, Co, Cr, Cs, Cu, Ga, In, La, Li, Mn, Mo, Nb, Ni, P, Pb, Rb, Sb, Sc, Sn, Sr, Te, Th, Tl, U, V, W, Y, and Zn were analyzed by ICP-AES and inductively coupled plasma-mass spectrometry (ICP-MS) following a near-total digestion in a mixture of HCl, HNO₃, HClO₄, and HF. Mercury was determined by cold-vapor atomic absorption spectrometry, As and Se by hydride-generation atomic absorption spectrometry, and total C by combustion. The details of the analytical protocols are given by Smith et al. (2009).

One of the unique aspects of this study is the determination of the major mineralogical components in the soil A and C horizon samples. Splits of the <2-mm material were mixed with a ZnO internal standard, ground in a micronizing mill, lightly pressed into back-loaded mounts, and analyzed by X-ray diffractometry (Cu K α radiation). Mineral percentages were quantified using a Rietveld refinement technique. About 20 minerals or mineral groups can be quantified by this technique.

6.3. Geochemical maps

As of May 2012, about 84% of the samples collected within the conterminous U.S. have been chemically analyzed and the mineralogy has been determined on about 88% of the samples. The distribution of Na is shown in Fig. 14 for that portion of the U.S. where samples have been analyzed and the distribution of plagioclase is shown in Fig. 15.

6.4. Strengths and weaknesses of the NASGLP data set

The primary strengths of this data set include (1) the use of a formal process for engaging the community of stakeholders to

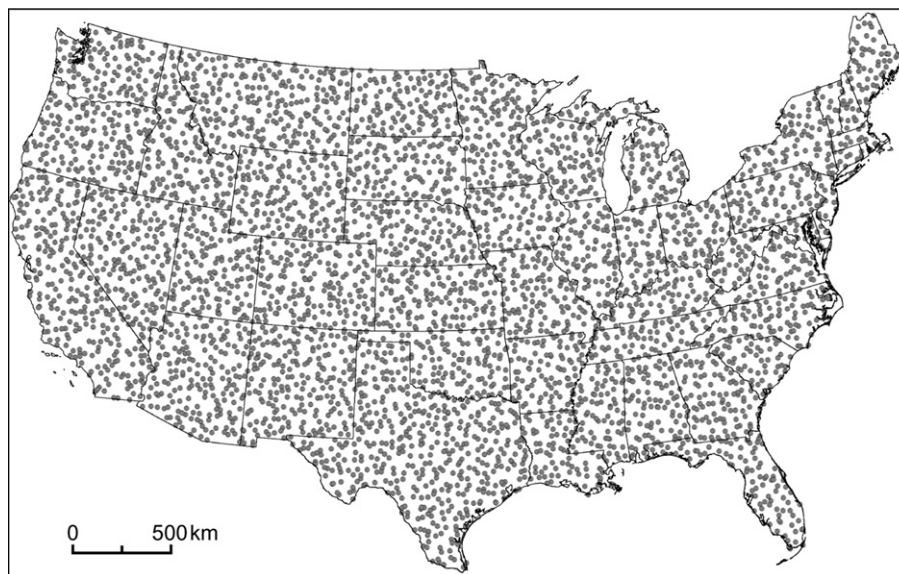


Figure 13. Map showing the location of ca. 4800 soil sampling sites in the conterminous U.S. for the North American Soil Geochemical Landscapes Project.

establish the sampling and analytical protocols; (2) utilization of the same sample media throughout the entire conterminous U.S.; (3) the limited amount of time (4 years) for collection of the samples provided a geochemical snapshot of the conterminous U.S. between 2007 and 2010; (4) the use of standardized analytical protocols that included virtually all the elements of environmental concern; and (5) the quantification of the major mineralogical components of the soil, which greatly aids in the interpretation of the observed geochemical patterns. One of the major outcomes of the project is also the establishment of a sample archive consisting of several hundred grams of each collected sample. This archive is available to researchers for future investigations.

The weakness of the data set is its relatively low sample density. While about 3.5 times the density of the Shacklette data set, the density of 1 site per 1600 km² is still too low to address issues at a more local scale (e.g., county- or state-scales). For studies at these scales, more intense sampling is required.

7. NRCS soil geochemistry spatial data set

7.1. Background and purpose

This ongoing project, led by M.A. Wilson and R. Burt of the NRCS National Soil Survey Center, Soil Survey Laboratory, in Lincoln, Nebraska, analyzes selected pedons, sampled by horizon, as part of the National Cooperative Soil Survey (NCSS) Program. The resulting data will define element ranges for different soils, parent materials, and regions and will provide the basis for improved understanding of soil geography, pedogenic processes, and basic soil characterization data generated by the NCSS Program (Wilson et al., 2008). The data set currently comprises more than 4000 samples from approximately 1150 sites (Fig. 16). Summaries of the sample collection protocols and analytical methods used in the project are given in Burt et al. (2003) and Wilson et al. (2008) and can be found on-line at <http://soils.usda.gov/survey/geochemistry>.

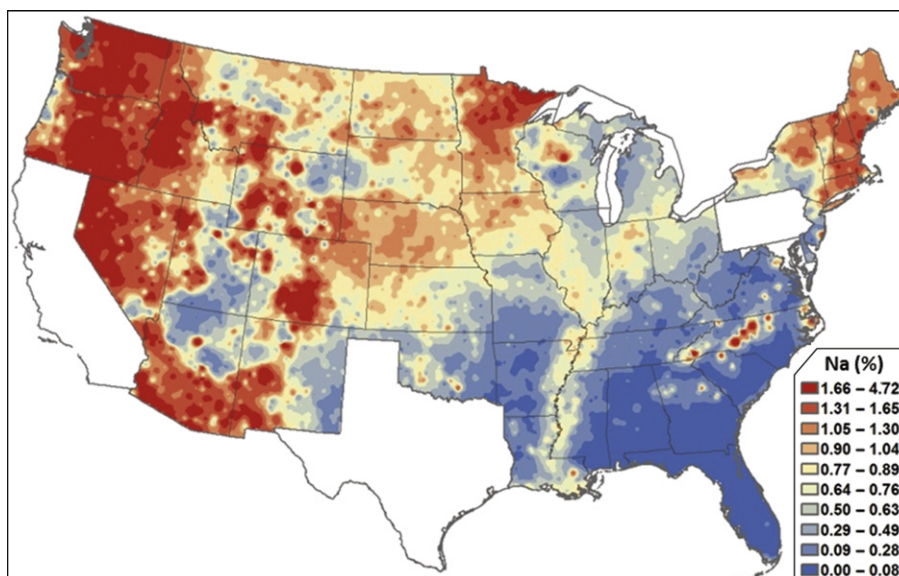


Figure 14. Map showing the distribution Na in soil C horizon samples from the conterminous U.S. as determined by the North American Soil Geochemical Landscapes Project.

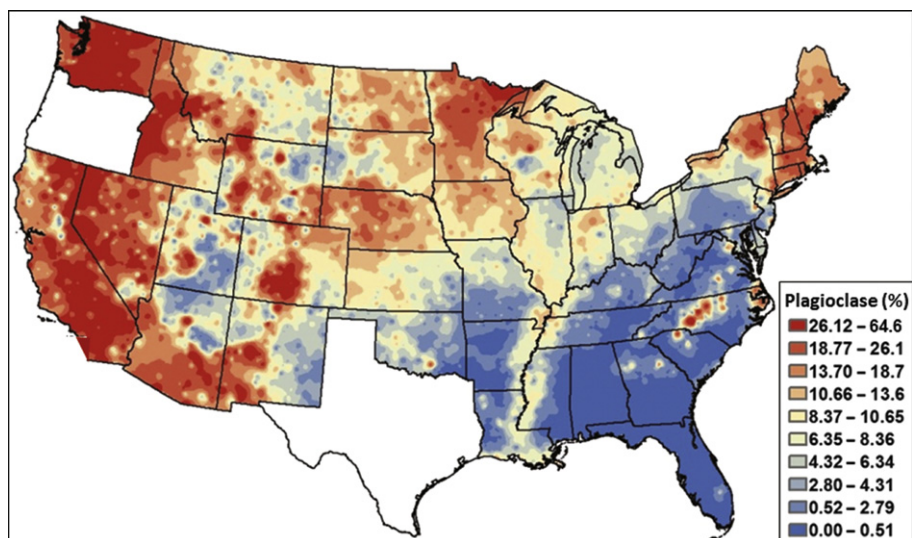


Figure 15. Map showing the distribution of plagioclase in soil C horizon samples from the conterminous U.S. as determined by the North American Soil Geochemical Landscapes Project.

7.2. Sampling and analytical protocols

Pedons were described and samples were collected according to a detailed, horizon-based protocol employing standard soil survey procedures (Soil Survey Division Staff, 1993; Burt, 2006). Samples were sieved to <2 mm and ground to <150 μm prior to chemical analysis. Aluminum, Ca, Fe, K, Mg, Mn, Na, P, Si, Ti, and Zr were analyzed by ICP-AES following a microwave digestion (180 °C for 9.5 min) in a mixture of HF, HNO₃, and HCl. Trace elements reported in the data set include Ag, As, Ba, Be, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, Sb, Se, Sn, Sr, V, W, and Zn. These elements were analyzed by ICP-AES following a digestion in a mixture of HNO₃ and HCl at 175 °C for 4.5 min except for Hg, which was analyzed by cold vapor hydride atomic absorption (Burt et al., 2003; Wilson et al., 2008).

7.3. Geochemical maps

Because of the relatively low sample density and uneven distribution of sampled pedons to date, geochemical maps have not been published for the NRCS Soil Geochemistry Spatial Data Set.

7.4. Strengths and weaknesses of the NRCS soil geochemistry spatial data set

The primary strength of this data set is that all the samples were collected as part of the NCSS Program. The samples came from pedons that were carefully selected by landscape evaluation to be representative of the soil mapping unit. These pedons have been described in great detail; the attributes include horizon

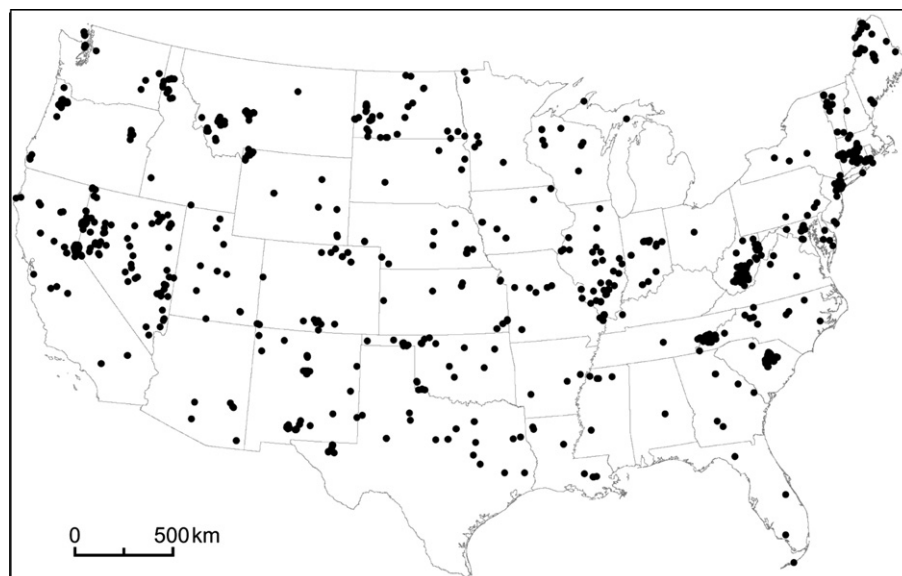


Figure 16. Distribution of sample sites containing trace element data from the NRCS soil geochemistry spatial data set.

designation, depth and thickness of the horizons, color, texture, structure, soil consistency at several water states, and presence of roots, rock fragments, pores, nodules, and salts (Soil Survey Division Staff, 1993). Samples are collected on the basis of soil horizons for laboratory measurements of various physical and chemical parameters. These parameters include major and trace element concentrations, particle size, bulk density, soil pH, cation exchange capacity, and exchangeable cations. The availability of this wide array of soil characterization data is very helpful in interpreting the geochemical data both in terms of areal distribution of the elements and distribution with depth.

Currently, the primary weakness of this data set for mapping national-scale elemental variation is the relatively low density of sites and the non-uniform distribution of these sites (Fig. 16). However, the project is ongoing and these problems should diminish with time. Also, elemental data are incomplete on some samples, with samples from certain projects only having major element data and others only having trace element data.

8. Discussion

Each of the six data sets discussed in sections 2 through 7 was designed to meet the needs of a specific clientele and, collectively, demonstrate the need for such national-scale, or near national-scale, geochemical data. The clientele for these data sets include, among others, specialists in agriculture, environmental regulation, risk assessment, public health, mineral resources, geoscience, soil science, and land planning. Each of the data sets has been, or certainly will be upon completion, very useful to one or more of these groups.

However, each of the six data sets has its own shortcomings. Most of these concern the sample collection protocols or analytical methods used. Ideally, a national-scale geochemical data set should be based upon a consistent sample medium or media collected at all sites in the study area. Of the six data sets, the NURE-HSSR data set and the USGS NGS data set do not meet this important criterion. However, it must be recognized that the NURE-HSSR Program never had a stated goal of producing a consistent national-scale geochemical data set. Rather, the goal of this program was to identify areas of the U.S. where undiscovered U resources are likely to be found. Thus, the four laboratories leading the program had no requirements to standardize sampling protocols, and as a result, the samples in the data set are a mixture of stream sediments, soils, and lake sediments. The USGS NGS, having the majority of its samples coming from the NURE-HSSR archives, inherited this problem of multiple sample types.

Each of the six data sets has its own set of analytical issues. The NURE-HSSR Program did not use consistent analytical methods throughout the study and did not analyze all the samples for a consistent suite of elements. The 'Shacklette' data set includes several elements whose concentrations were determined by semi-quantitative methods that were commonly used by the USGS during the time of this study but are inadequate by today's standards. The 'Holmgren' data set only contains concentration determinations for five elements. The other data sets, while analyzing a large number of elements by consistent methodologies, still have a number of elements that were not analyzed and some elements with detection limits that are higher than preferred, resulting in highly censored data for these elements. Usually, the choice of elements analyzed and analytical methods is a function of the techniques available through the laboratories of the agency or institution conducting the study. This generally leads to some combination of the problems observed with the data sets discussed here.

The two ongoing projects, NASGLP and NRCS Soil Geochemistry, use very similar analytical protocols and have a similar philosophy regarding sample collection. There is a good opportunity, at some point, to investigate combining these data sets to provide higher resolution soil geochemical data for the U.S.

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