The distribution of meteoric $^{36}Cl/Cl$ in the United States: a comparison of models

Stephen Moysey · Stanley N. Davis · Marek Zreda · L. DeWayne Cecil

Abstract The natural distribution of $^{36}Cl/Cl$ in groundwater across the continental United States has recently been reported by Davis et al. (2003). In this paper, the large-scale processes and atmospheric sources of ^{36}Cl and chloride responsible for controlling the observed $^{36}Cl/Cl$ distribution are discussed.

The dominant process that affects $^{36}Cl/Cl$ in meteoric groundwater at the continental scale is the fallout of stable chloride from the atmosphere, which is mainly derived from oceanic sources. Atmospheric circulation transports marine chloride to the continental interior, where distance from the coast, topography, and wind patterns define the chloride distribution. The only major deviation from this pattern is observed in northern Utah and southern Idaho where it is inferred that a continental source of chloride exists in the Bonneville Salt Flats, Utah.

In contrast to previous studies, the atmospheric flux of ^{36}Cl to the land surface was found to be approximately constant over the United States, without a strong correlation between local ^{36}Cl fallout and annual precipitation. However, the correlation between these variables was significantly improved (R^2=0.15 to R^2=0.55) when data from the southeastern USA, which presumably have lower than average atmospheric ^{36}Cl concentrations, were excluded. The total mean flux of ^{36}Cl over the continental United States and total global mean flux of ^{36}Cl are calculated to be 30.5±7.0 and 19.6±4.5 atoms m^{-2} s^{-1}, respectively.

The $^{36}Cl/Cl$ distribution calculated by Bentley et al. (1986) underestimates the magnitude and variability observed for the measured $^{36}Cl/Cl$ distribution across the continental United States. The model proposed by Hainsworth (1994) provides the best overall fit to the observed $^{36}Cl/Cl$ distribution in this study. A process-oriented model by Phillips (2000) generally overestimates $^{36}Cl/Cl$ in most parts of the country and has several significant local departures from the empirical data.

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Résumé La distribution naturelle du rapport $^{36}Cl/Cl$ dans les eaux souterraines des États-Unis a été récemment présentée par Davis et al. (2003). Dans ce travail, les processus à grande échelle et les sources atmosphériques de ^{36}Cl et de chlorure responsables du contrôle de la distribution observée du rapport $^{36}Cl/Cl$ sont discutés. Le processus dominant qui affecte le rapport $^{36}Cl/Cl$ dans les eaux souterraines d'origine météorique à l'échelle continentale est l'apport atmosphérique de chlorure stable, qui provient pour l'essentiel de sources océaniques. La circulation atmosphérique transporte des chlorures marins vers l'intérieur des continents, où la distribution de chlorure est définie par la distance à la côte, la topographie et les régimes des vents. La seule exception majeure à ce schéma est observée dans le nord de l'Utah et le sud de l'Idaho où l'on suppose qu'il existe une source continentale de chlorure dans les bas-fonds salés de Bonneville. Au contraire de précédentes études (Knies et al. 1994; Phillips 2000), on trouve que le flux atmosphérique de ^{36}Cl vers le sol est approximativement constant sur l'ensemble des États-Unis, sans forte corrélation entre la retombée locale de ^{36}Cl et les précipitations annuelles. Cependant, la corrélation entre ces variables devient significative (R^2=0.15 à 0.55) lorsqu'on supprime les données du sud-est des États-Unis, dont on pense qu'elles présentent des concentrations en ^{36}Cl atmosphérique inférieures à la moyenne. Le flux total moyen de ^{36}Cl sur les États-Unis continentaux et le flux moyen global de ^{36}Cl sont respectivement évalués à 30.5 ± 7.0 et 19.6 ± 4.5 atomes.m^{-2}.s^{-1}. La distribution du rapport $^{36}Cl/Cl$ calculée par Bentley et al. (1986) sous-estime l'ordre de grandeur et la variabilité observés pour la distribution mesurée du rapport $^{36}Cl/Cl$ sur les États-Unis continentaux. Le modèle proposé par Hainsworth (1994) fournit le

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meilleur ajustement d'ensemble à la distribution du rapport ^{36}Cl/Cl observée dans cette étude. Un modèle orienté vers les processus proposé par Phillips (2000) surestime dans l'ensemble le rapport ^{36}Cl/Cl dans la plupart des régions du pays et présente plusieurs désaccords locaux avec les données empiriques.

Resumen Davis et al. (2003) han informado de la distribución natural de la proporción ^{36}Cl/Cl en las aguas subterráneas de la parte continental de los Estados Unidos de América [EUA]. En este artículo, se discute cuáles son los procesos a gran escala y las fuentes atmosféricas del ^{36}Cl y del cloruro que dan lugar a la distribución observada de ^{36}Cl/Cl.

El proceso dominante que afecta a la relación ^{36}Cl/Cl en las aguas subterráneas de origen meteórico a escala continental es el aporte de cloruro estable desde la atmósfera, que procede principalmente de los océanos. La circulación atmosférica transporta el cloruro marino hacia el interior, donde la distancia a la costa, topografía y corrientes del viento definen la distribución del cloruro. La única desviación principal de este esquema tiene lugar al norte de Utah y en el sur de Idaho, donde se deduce que existe una fuente continental de cloruro en los Rellanos Salados de Bonneville (Salt Flats).

En contraste con estudios previos (Knies et al. 1994; Phillips 2000), se ha descubierto que el flujo atmosférico de ^{36}Cl hacia la superficie terrestre es aproximadamente constante en todos los estados, sin deducirse una correlación fuerte entre el aporte de ^{36}Cl y la precipitación anual. Sin embargo, la correlación entre estas variables se ve mejorada de forma significativa, con coeficientes de regresión comprendidos entre 0,15 y 0,55, cuando se excluyen los datos recogidos en el sudeste de los EUA, que tienen concentraciones de ^{36}Cl atmosférico presuntamente inferiores a la media. El flujo medio total de ^{36}Cl calculado en la zona continental de los Estados Unidos vale 30,5±7,0 átomos por metro cuadrado y segundo, mientras que el flujo total global de ^{36}Cl es de 19,6±4,5 átomos por metro cuadrado y segundo.

La distribución de ^{36}Cl/Cl calculada por Bentley et al. (1986) infravalora la magnitud y variabilidad observada en los valores medidos a lo largo de los Estados Unidos. El modelo propuesto por Hainsworth (1994) proporciona el mejor ajuste conjunto a la distribución observada de ^{36}Cl/Cl en este estudio. El modelo orientado a procesos de Phillips (2000) sobreestima por lo general la distribución de ^{36}Cl/Cl en la mayoría del país y difiere significativamente de algunos valores locales empíricos.

Keywords Atmospheric deposition · Chlorine-36 · Groundwater · United States

Introduction

Quantitative studies of groundwater systems using environmental nuclides have become popular because of the realization that the distribution of such nuclides constitutes a historical record of flow and transport processes within aquifers. To interpret this record one must first understand the inputs of the isotopes to the hydrologic system under study (Cecil and Vogt 1997; Davis et al. 1998). Such a need explains the extensive research that has been directed at quantifying the sources and behavior of both stable nuclides, such as ^{2}H and ^{18}O, and radionuclides, such as ^{3}H, ^{14}C, and ^{36}Cl, in groundwater (Clark and Fritz 1997).

The use of ^{36}Cl (half-life, $t_{1/2}$=301 ka) in groundwater studies, however, has been hampered by a deficiency in our understanding of its spatial and temporal input function to the subsurface (Fabryka-Martin et al. 1987; Beasley et al. 1993; Davis et al. 1998). In particular, there are few studies of the spatial distribution of naturally occurring ^{36}Cl in groundwater beyond the aquifer scale (e.g., Bentley et al. 1986; Phillips 2000). Moysey (1999) and Davis et al. (2003) inferred the natural distribution of ^{36}Cl/Cl over the continental United States from analyses of groundwater samples. In this paper, the natural variability seen in the empirical distribution is explored and discussed with an emphasis on identifying the large-scale processes that are most important in shaping the observed distribution. Additionally, the measured distribution is compared to three models of the ^{36}Cl/Cl distribution (Bentley et al. 1986; Hainsworth 1994; Phillips 2000).

The empirical ^{36}Cl/Cl distribution presented in this paper is based on that of Moysey (1999) and is essentially the same as that given by Davis et al. (2003); both have been generated from the same database. These data, however, are treated differently in constructing the two ^{36}Cl/Cl models. Moysey (1999) interpreted clusters of data jointly to determine regional estimates of ^{36}Cl/Cl and assigned a weight to each of these values according to a qualitative measure of reliability. Davis et al. (2003) used only data from the samples that have the lowest chloride concentrations and that are believed to be free from anthropogenic influence. The consistency between the ^{36}Cl/Cl maps produced by each of these studies, using different data interpretation techniques, shows that the underlying data set is robust. The minor differences that exist between the two models do not change the conclusions in this paper.

Origin of ^{36}Cl and Stable Chloride in Nature

Most ^{36}Cl found in actively circulating meteoric groundwater, not affected by anthropogenic sources, originated in the atmosphere. The spallation of ^{40}Ar by cosmic rays (Jiang et al. 1990; Lehmann et al. 1993) is the primary natural atmospheric source of ^{36}Cl. Subsequent to production, ^{36}Cl atoms are deposited at the land surface and eventually carried to the subsurface by recharging groundwater. In some cases, additional ^{36}Cl may also be introduced through secondary production at or below the ground surface (Bentley et al. 1986; Phillips 2000).

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About 60–75% of atmospheric ^{36}Cl is produced in the stratosphere with the remainder produced in the troposphere (Lal and Peters 1967; Bentley et al. 1986). Meridional circulation of the atmosphere causes the majority of exchange between the stratosphere and troposphere to take place at mid-latitudes (Reiter 1975; Seinfeld and Pandis 1998). Once in the troposphere, ^{36}Cl reaches the ground surface relatively quickly as either wet (i.e., associated with precipitation) or dry fallout. Lal and Peters (1967) developed a model for the latitudinal dependence of deposition based on observations of fallout produced during thermonuclear weapons testing in the 1950s.

Precipitation and evaporation rates in the recharge area of an aquifer can vary greatly through time, so groundwater studies often consider ^{36}Cl/Cl rather than absolute ^{36}Cl concentrations. This ratio is insensitive to changes in the supply of recharge water while the absolute concentration is not. Understanding variability in ^{36}Cl/Cl, therefore, requires knowledge of the historical sources for stable chloride as well as those for ^{36}Cl.

The most important sources of chloride to the continental landmasses are sea-spray and salt-based aerosols derived from the oceans (Eriksson 1960; Semonin and Bowersox 1982; Li 1992; Simpson and Herczeg 1994). The deposition of chloride from air masses originating over the oceans decreases approximately exponentially with distance from the coast (Slinn et al. 1982; Simpson and Herczeg 1994). With the probable exception of extensive playas in northwestern Utah, continental sources of wind-blown chloride appear to be only of local importance (Wood and Sanford 1995).

Methods

This study uses ^{36}Cl/Cl measured in 183 groundwater samples collected from 38 locations across the United States. Sampling locations were selected to obtain water recharged between 50–10,000 years ago. The younger age limit was chosen to minimize the influence of anthropogenic ^{36}Cl recharged after 1952 (Bentley et al. 1986; Davis et al. 1998; Phillips 2000); the older age limit was chosen because prior to 10,000 years ago, the global production rate of ^{36}Cl in the atmosphere was significantly different than it is today (Plummer et al. 1997; Phillips 2000). Waters with low chloride concentrations were also targeted to minimize dilution of ^{36}Cl/Cl due to the addition of connate chloride or dissolution of halite and other minerals in the subsurface. In general, clusters of samples were collected within a given region so that both local and continental scale variability in ^{36}Cl/Cl could be investigated. A description of the project and site-selection objectives is given by Davis et al. (2003) and specific site descriptions are provided by Moysey (1999).

Samples for ^{36}Cl/Cl measurement were prepared at the University of Arizona and measured by accelerator mass spectrometry (AMS) at PRIME Lab, Purdue University. All analytical results, as well as detailed descriptions of analytical procedures, are given by Moysey (1999). Unless otherwise stated, all ^{36}Cl/Cl reported in this work have been multiplied by 10^{15} and, therefore, have the implied units of x10^{-15}atoms ^{36}Cl/atoms Cl.

Results

The regional estimate of ^{36}Cl/Cl for each of the 38 study areas is given in Table 1; each regional value represents the synthesis of between 1 and 12 samples. The ^{36}Cl/Cl reported for each region was assigned based on data collected during this study, existing geochemical and hydrologic models of the local flow system, and information from the literature. The use of multiple samples from an area to provide a context for the determination of the regional meteoric ^{36}Cl/Cl signal is critical because variability at the local aquifer scale, caused by processes such as the dissolution of minerals or secondary ^{36}Cl production in the subsurface, can be of the same order of magnitude as continental scale variability (Moysey 1999). The "confidence index" (CI), also reported in Table 1, is a qualitative indicator of the reliability of the regional ^{36}Cl/Cl estimate and was assigned based on the amount of data collected from the area, the current understanding of the aquifer system as inferred from the literature, how well the data from this study can be explained by models of the aquifer system, and whether the data show signs of being influenced by anthropogenic or subsurface sources of ^{36}Cl or chloride. A CI rank of 1 implies high confidence in the ^{36}Cl/Cl estimate, while a rank of 3 implies low confidence. Moysey (1999) gives details of how each regional ^{36}Cl/Cl estimate in Table 1 was determined.

Figure 1 shows the locations of the 38 regional-control points used in this study. Nine control points were considered unreliable (CI =3), and were not used for the analyses discussed in this paper. Thirteen previously published studies that obtained estimates of the meteoric ^{36}Cl/Cl signal (Table 2) are also used in this study. The distribution of ^{36}Cl/Cl derived from these measurements is given in Fig. 1. The isopleths of ^{36}Cl/Cl in this figure were drawn by hand because automated interpolation techniques are not justified given the degree of uncertainty related to the ^{36}Cl/Cl estimates and the relatively sparse distribution of data points compared to the small-scale spatial variability of the underlying process. The ^{36}Cl/Cl map presented here, therefore, is not meant to be a quantitative estimate of local ^{36}Cl/Cl values, but rather act as a tool for understanding the large-scale processes that control the ^{36}Cl/Cl distribution.

Note that the map shown in Fig. 1 is similar to that interpreted by Davis et al. (2003). The most significant discrepancy between the two ^{36}Cl/Cl maps is a shift in the location of the maximum observed ^{36}Cl/Cl approximately 400 km (~250 miles) to the north in Fig. 1 compared with the map of Davis et al. (2003). In general, the differences between the two maps do not impact the conclusions of this paper.

Table 1 ^{36}Cl/Cl from the present study (regional estimates)

Group ID	Group name	Lat. °N	Long. °W	^{36}Cl/Cl	^{36}Cl/Cl Error[a]	Confidence index (CI)[b]	Precipitation[c] (cm/year)	Wet chloride deposition[d] (kg ha^{-1} year^{-1})	Wet ^{36}Cl deposition[e] (atoms m^{-2} s^{-1})	Total ^{36}Cl deposition[e] (atoms m^{-2} s^{-1})
1	W Georgia	32.5	83.7	156	8	1	159	3.1	26.0	35.0
2	E Georgia	33.2	82.5	97	5	1	170	4.0	20.9	28.1
3	N Florida	30.4	84.3	389	17	3	–	6.5	136.2	182.8
4	Florida	27.5	81.5	45	2	2	160	10.0	24.2	32.5
5	Florida	29.1	81.8	183	252	3	–	13.0	128.1	172.0
6	Arkansas	35.3	90.6	139	98	2	144	14.0	104.8	140.7
7	Mississippi	31.2	89.4	50	3	2	138	6.8	18.3	24.6
8	Oklahoma	35.6	97.4	277	78	2	84	1.7	25.4	34.0
9	North Dakota	46.0	97.8	900	100	1	45	0.4	20.4	27.3
10	South Dakota	44.4	103.5	1215	191	1	35	0.4	23.6	31.6
11	Nebraska	41.1	100.8	493	19	2	37	0.2	5.3	7.1
12	West Virginia	39.6	78.2	386	20	2	130	2.0	41.6	55.8
13	New York	43.1	73.8	9	4	3	–	1.5	0.7	1.0
14	St.. David (AZ)	31.9	110.2	364	16	2	30	0.9	17.6	23.7
15	S Utah	37.2	113.6	403	32	2	25	0.5	10.9	14.6
16	N Utah	41.9	111.6	699	961	3	–	15.0	564.8	758.1
17	Idaho	44.1	111.3	352	309	2	30	0.7	13.3	17.8
18	E Oregon	44.4	121.2	499	202	2	100	3.0	80.6	108.2
19	W Oregon	45.0	123.0	219	164	2	200	6.0	70.8	95.0
20	Washington	45.7	121.5	47	52	3	–	16.0	40.5	54.4
21	W Colorado	39.2	104.8	952	142	2	40	0.5	23.1	31.0
22	Kansas	38.0	97.5	500	200	2	60	1.4	37.7	50.6
23	E Colorado	39.2	102.9	2123	624	3	–	0.4	45.7	61.4
24	Tahoe (CA)	39.1	120.0	952	517	2	15	0.2	7.7	10.3
25	Fresno (CA)	36.7	119.6	219	7	1	84	1.6	18.9	25.3
26	White Mountains (AZ)	33.8	109.1	1842	93	3	–	0.3	29.8	40.0
27	N Wyoming	43.8	104.2	930	233	1	35	0.4	18.0	24.2
28	S Wyoming	41.2	104.8	1210	60	2	24	0.4	24.1	32.4
29	Indiana	41.0	85.2	275	75	1	84	1.5	22.2	29.8
30	Ohio	40.1	83.8	252	147	3	–	1.5	19.7	26.4
31	Kentucky	37.5	83.1	218	75	2	130	1.5	17.4	23.3
32	E Alabama	32.0	85.6	110	35	1	140	3.5	20.7	27.8
33	W Alabama	32.7	87.6	129	16	1	120	3.7	25.7	34.5
34	Montana	48.3	114.3	1,087	163	1	67	0.3	17.6	23.6
35	Iowa	42.4	92.8	492	108	1	70	0.8	21.2	28.5
36	Wisconsin	43.1	89.5	368	215	3	–	0.9	17.8	23.9
37	Minnesota	45.1	93.2	657	159	1	75	0.6	19.5	26.1
38	Arizona	32.2	110.9	373	17	1	40	1.0	20.1	27.0

[a] The reported error is the largest of the analytical error, group standard deviation, or model error

[b] CI is a subjective measure of confidence in the regional estimate of ^{36}Cl/Cl; CI=1 is the highest confidence, CI=3 is the lowest

[c] Precipitation values are estimated from the NADP (2000) total precipitation map for 1994

[d] Wet chloride deposition is estimated from the mean of NADP (2000) measurements between 1979–1997

[e] Calculated values. Total deposition assumes that wet deposition accounts for 74% of total fallout

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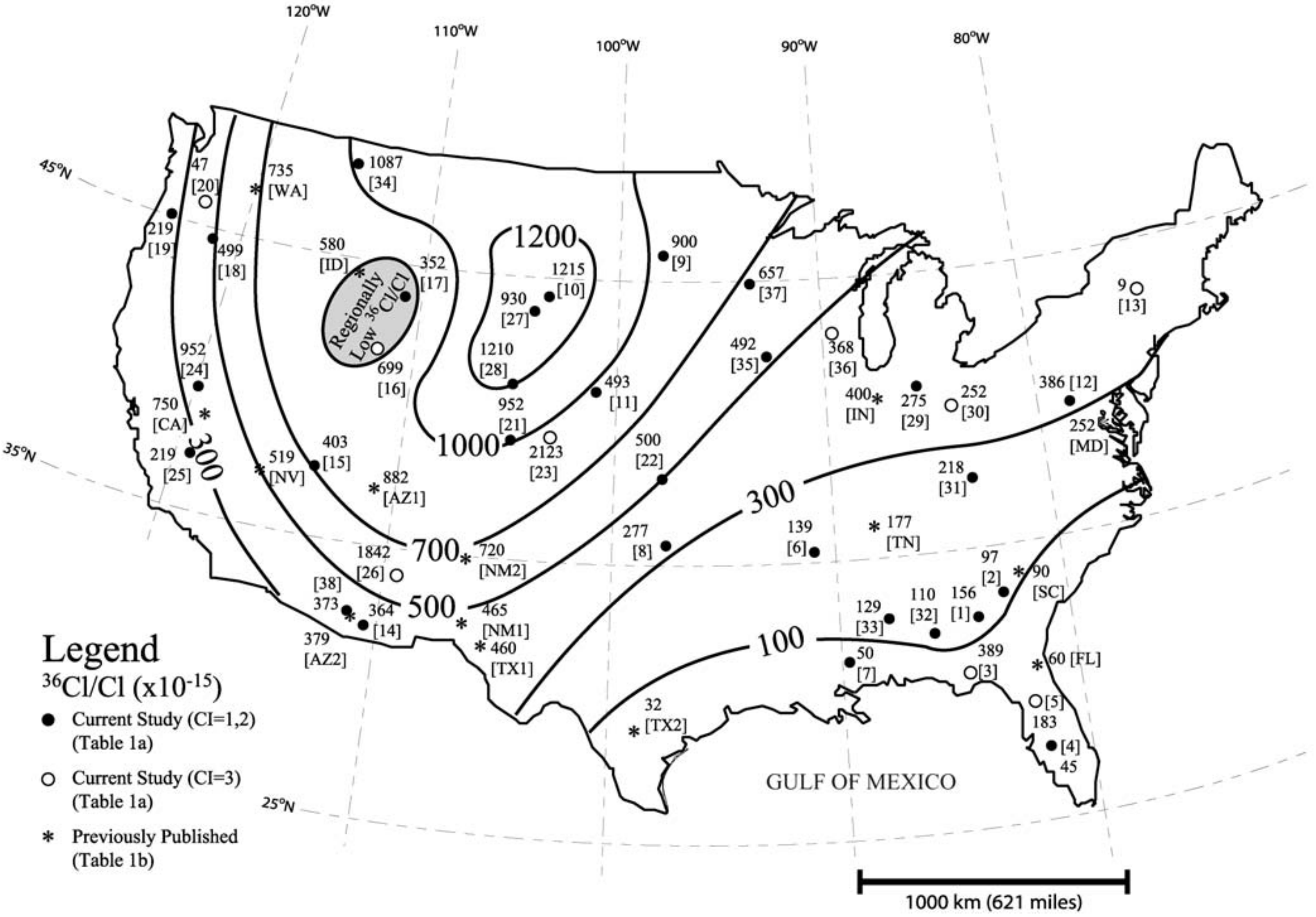

Fig. 1 Empirical ^{36}Cl/Cl distribution in the United States inferred from groundwater measurements. ^{36}Cl/Cl are shown with control point ID from Table 1 in parenthesis

The ^{36}Cl/Cl ranges from a minimum of less than 100 near the oceans to a maximum of about 1,200 in the interior of the continent over the central High Plains region of Wyoming (Fig. 1). The ^{36}Cl/Cl isopleths are approximately parallel to the west coast, where they form a steep inland gradient. In contrast, ^{36}Cl ratios in the eastern United States slowly increase with distance from the Gulf of Mexico and northward along the eastern seaboard, showing only secondary influence from the Atlantic coastline. The specific arrangement of ^{36}Cl/Cl isopleths along the northeastern coastline is uncertain due to a lack of data in this area. A region of low ^{36}Cl/Cl is also apparent in northwestern USA (northern Utah and southern Idaho).

The pattern observed for the spatial distribution of ^{36}Cl/Cl is similar to that measured by the National Atmospheric Deposition Program (NADP 2000) for stable-chloride deposition (Fig. 2), though the magnitudes are inversely related. A notable exception is along the eastern seaboard where the chloride contours tend to more closely parallel the coastline. The strong correlation between ^{36}Cl/Cl and chloride deposition is shown quantitatively in Fig. 3 for data obtained in this project as well as 13 other studies conducted around the United States (Table 1). Based on this high level of correlation, the deposition of stable chloride is inferred to be the primary control on spatial variability of ^{36}Cl/Cl.

Most of the chloride that is deposited on the continent originates as particulates released to the atmosphere from the oceans (Eriksson 1960; Semonin and Bowersox 1982; Li 1992; Simpson and Herczeg 1994). Once chloride is suspended in the atmosphere, it can be carried inland by winds. Atmospheric chloride availability decreases exponentially with distance from the ocean, resulting in higher chloride deposition rates and lower ^{36}Cl/Cl near the coast compared with the interior of the continent. Synoptic scale weather patterns and topographic barriers also play an important role in controlling the migration of chloride particulates over the continent (Semonin and Bowersox 1982). The western mountain ranges act as an effective barrier to the inland movement of air masses carrying chloride. This results in the sharp gradient in ^{36}Cl/Cl along the west coast, as shown in Fig. 1. In the eastern USA, northward-trending storms are generally limited to coastal areas while air masses moving north from the Gulf of Mexico are the primary source of moisture for the

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Table 2 $^{36}Cl/Cl$ from previously published measurements

ID	State	$^{36}Cl/Cl$	Precipitation[a] (cm/year)	Wet chloride deposition[b] (kg ha^{-1} year^{-1})	Wet ^{36}Cl deposition[c] (atoms m^{-2} s^{-1})	Total ^{36}Cl deposition[d] (atoms m^{-2} s^{-1})	Reference
AZ1	Arizona	882	45	0.3	14.3	19.1	Lopes and Hoffmann (1997)
AZ2	Arizona	379	40	1.0	20.4	27.4	Bentley et al. (1986)
CA	California	750[d]	110	1.0	40.4	54.2	Phillips et al. (1995)
FL	Florida	60	140	6.0	19.4	26.0	Phelps (1997, personal communication)
ID	Idaho	580	25	0.8	25.0	33.5	Cecil et al. (1992)
IN	Indiana	400	94	1.1	23.7	31.8	Vogt et al. (1994)
MD	Maryland	252	126	4.0	54.3	72.9	Purdy et al. (1987)
NM1	New Mexico	465	41	0.5	12.5	16.8	Phillips et al. (1988)
NM2	New Mexico	720	30	0.4	15.5	20.8	Phillips et al. (1988)
NV	Nevada	519	40	0.8	22.4	30.0	Norris et al. (1987)
SC	South Carolina	90	155	4.0	19.4	26.0	Beasley et al. (1992)
TN	Tennessee	177	140	2.2	21.0	28.2	Vourvopoulous et al. (1990)
TX1	Texas	460	41	0.7	17.3	23.3	Scanlon (1990)
TX2	Texas	64	47	5.0	17.2	23.1	Bentley et al. (1986)
WA	Washington	735	50	0.5	19.8	26.6	Prych (1992, personal communication)

[a] Precipitation values are estimated from the NADP (2000) total precipitation map for 1994
[b] Wet chloride deposition is estimated from the mean of NADP (2000) measurements between 1979–1997
[c] Calculated values. Total deposition assumes that wet deposition accounts for 74% of total fallout.
[d] Originally estimated from a chloride budget.

interior of the continent (Barry and Chorley 1992). These air masses, which also carry chloride from the Gulf of Mexico are, therefore, the predominant influence on chloride deposition in the southeastern interior of the USA, resulting in the broad Gulf-dominated isopleths shown in Fig. 1.

The low $^{36}Cl/Cl$ observed near the border of Utah and Idaho is the only clear exception to the oceanic forcing described above. This deviation from the continental pattern is likely caused by a continental source of chloride, predominantly the Bonneville Salt Flats in northern Utah. Local winds can pick up and transport particles from the salt flats resulting in abnormally high chloride deposition in this area. This effect is apparent in chloride deposition measurements that are locally two to six times higher than those measured in nearby regions (NADP 2000).

The historical depositional flux of ^{36}Cl may be estimated by taking the product of the $^{36}Cl/Cl$ ratios, measured in groundwater, and modern chloride-deposition rates. This approach relies on two assumptions: (1) after their initial deposition, ^{36}Cl and chloride behave conservatively with no subsurface sources, and (2) modern chloride deposition is representative of the average chloride deposition over the last 10,000 years. From the data in Tables 1 and 2, excluding Arkansas and eastern Oregon (items 6 and 18 in Table 1), the mean wet ^{36}Cl flux over the United States is 22.7±1.8 atoms m^{-2} s^{-1}. The deposition rates from eastern Oregon and Arkansas were not included in the calculation of the mean flux as these values were found to be statistical outliers using Chauvenet's Criterion (Taylor 1982).

The chloride-deposition rates measured by the NADP (Fig. 2) reflect only the wet component of modern chloride fallout and, therefore, the calculated ^{36}Cl flux represents a "wet-only" estimate. Few reliable estimates of chloride deposition currently exist for the dry component of fallout over the continent. Using the wet ^{36}Cl flux calculated above and the slope of the regression line from Fig. 3 (16.9±1.2 atoms m^{-2} s^{-1}), it is possible to estimate that 26±11% of the total mean ^{36}Cl fallout for the continental United States occurs as dry deposition. This is in good agreement with Eriksson (1960) and Bentley et al. (1986) who suggested that dry fallout should be approximately 23% of total deposition. Taking the relative fraction of dry deposition to be 26±11%, the total mean flux of ^{36}Cl over the United States is 30.5±7.0 atoms m^{-2} s^{-1}. The resulting ^{36}Cl fluxes calculated in this way (Tables 1 and 2) agree with the model used by Bentley et al. (1986) as they show little dependence on latitude (Fig. 4) and closely match the deposition rate used by these authors. If the latitudinal dependence proposed by Lal and Peters (1967; Fig. 4) is assumed to hold, then the estimated global "wet-only" fallout of ^{36}Cl is 14.6±1.1 atoms m^{-2} s^{-1} and the total global fallout, i.e., combined wet and dry deposition, is 19.6±4.5 atoms m^{-2} s^{-1}.

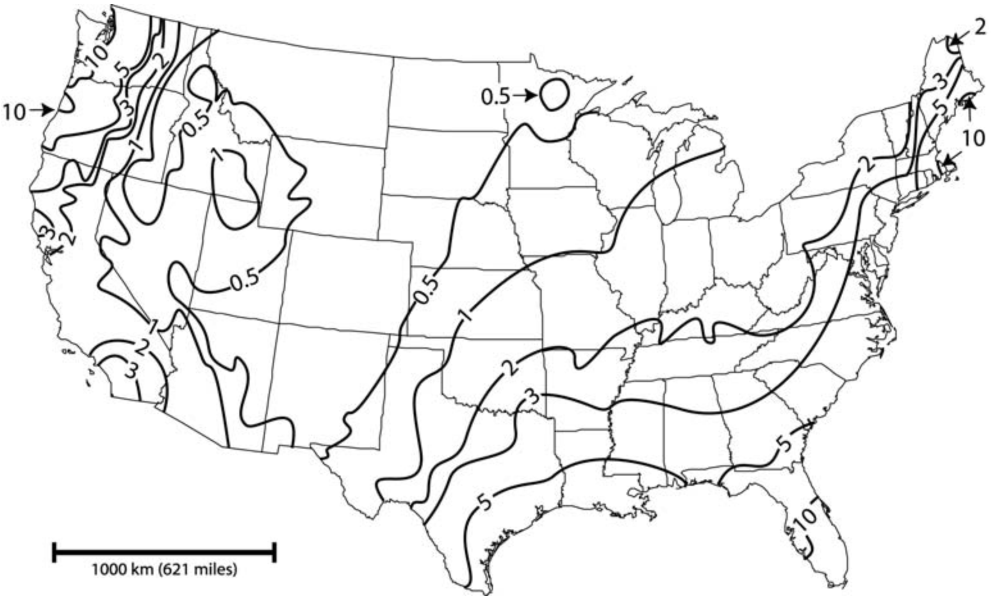

Fig. 2 Isopleth map of average annual wet fallout of stable chloride (kg/ha) in the United States for the period 1979–1997 (data from NADP 2000)

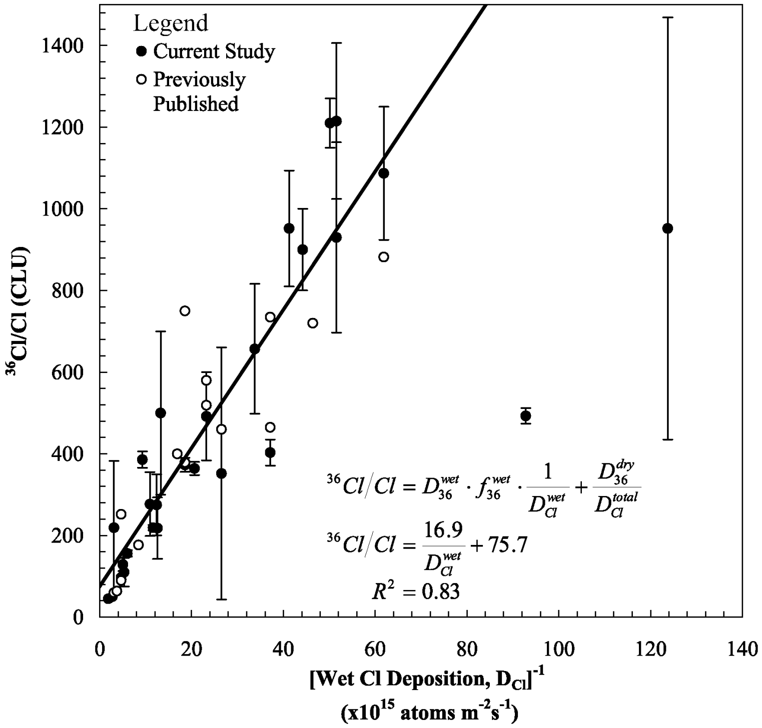

Fig. 3 Correlation between ^{36}Cl/Cl and chloride deposition. The single most important factor controlling ^{36}Cl/Cl is stable-chloride deposition. *Closed circles* are from this study (Table 1); *open circles* represent data from 13 previously published studies (Table 2). The samples from Nebraska and Lake Tahoe (items 11 and 24 from Table 1) appear as outliers in the figure and were not included in the regression. Chloride deposition was inferred from measurements by the NADP (2000)

Discussion

Previous attempts at modeling ^{36}Cl/Cl variability over the United States have been made by Bentley et al. (1986), Hainsworth (1994), and Phillips (2000). The approach taken in constructing these models has been to estimate the fallout of ^{36}Cl and stable chloride as a function of geographic location and then combine these to produce a map of the resulting ^{36}Cl/Cl. In all three cases, the investigators have included measured stable chloride deposition over the United States as part of their calculations; Bentley et al. (1986) used chloride fallout from Eriksson (1960), and Hainsworth (1994) and Phillips (2000) used data from the NADP (2000).

Bentley et al. (1986) and Phillips (2000) used the latitude dependence of the ^{36}Cl depositional flux (Fig. 4) proposed by Lal and Peters (1967) in their models (Fig. 5 a, c). Each author, however, used a different estimate of the global ^{36}Cl production rate (16 atoms m^{-2} s^{-1} for Bentley et al. 1986; 30 atoms m^{-2} s^{-1} for Phillips 2000). Phillips (2000) also attempted to account for variability of ^{36}Cl deposition by allowing local deviations from the

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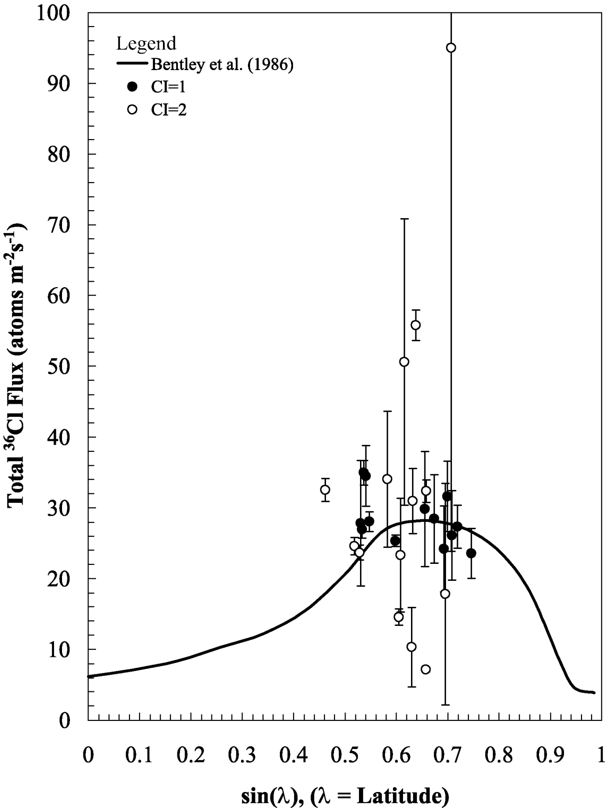

Fig. 4 Plot of ^{36}Cl vs. latitude. No correlation can be observed between the ^{36}Cl flux and latitude given the range of this study. The ^{36}Cl flux was calculated as the product of ^{36}Cl/Cl and stable-chloride fallout (NADP 2000) assuming that dry deposition accounts for 26% of total fallout. The *solid line* is the model from Bentley et al. (1986) after correction of dry deposition from 23% to 26%; *closed circles* are for qualitatively more reliable data (i.e., confidence index (CI) =1), *open circles* are for qualitatively less reliable data (i.e., CI =2)

mean flux proportional to the difference between local precipitation and the average precipitation rate for a given latitude. Hainsworth (1994; Fig. 5b) took a considerably different approach; she attributed 70% of the geographic variability of fallout to the transfer of ^{36}Cl between the stratosphere and troposphere. The stratospheric component of the ^{36}Cl flux was calibrated using the measured fallout of ^{90}Sr produced by thermonuclear weapons tests. The tropospheric component of ^{36}Cl fallout at a given location was assumed to be proportional to the ratio of the mean local rainfall to the mean zonal-precipitation rate. Hainsworth (1994) used the global ^{36}Cl flux calculated by Blinov (1988) of 19 atoms m^{-2} s^{-1}.

All three models qualitatively reproduce the main features of the ^{36}Cl/Cl distribution observed in this study. Low ratios occur near the coast and high ratios occur in the interior of the continent (Fig. 5). The reproduction of this large-scale variability is tied to the dominating effect of stable-chloride fallout. All of the models, nevertheless, deviate somewhat from the empirical values listed in Table 1 and shown in Fig. 1. The significance of some of these differences, such as the behavior of the ^{36}Cl/Cl isopleths along the northeast coastline, cannot be assessed due to insufficient data. However, each of the Bentley et al. (1986), Hainsworth (1994), and Phillips (2000) models do exhibit several features that are significantly different from the empirical distribution in Fig. 1.

The Bentley et al. (1986) model generally underestimates the empirical ^{36}Cl/Cl. This model (Fig. 5a) reaches a maximum ^{36}Cl/Cl of only 640 and predicts a region where the distribution is approximately constant in the interior of the United States. These low ^{36}Cl/Cl ratios are primarily related to overestimation of stable-chloride fallout. The chloride-deposition rates used by Bentley et al. (1986) were taken from Eriksson (1960). Comparison with modern data collected by the NADP (2000) indicates that the Eriksson chloride deposition values overestimate the flux in the interior of the United States, leading to the low ^{36}Cl/Cl predicted by Bentley et al. (1986).

The model by Hainsworth (1994) in Fig. 5b captures similar patterns of variability as seen in the empirical results (Fig. 1). The maximum ^{36}Cl/Cl predicted by her model (1,600), however, is somewhat higher than that observed in our study (~1,200). The significance of this discrepancy is difficult to evaluate given the lack of ^{36}Cl/Cl measurements from eastern Montana and North Dakota. The model of Hainsworth (1994) also tends to underestimate ^{36}Cl/Cl in the western and southwestern parts of the country. The cause of this underestimation is uncertain, though it could be related to annual fluctuations in the stable-chloride measurements used in the model, differences between the spatial distribution of ^{36}Cl fallout and that of ^{90}Sr, or over correction of the ^{36}Cl flux to account for precipitation.

The ^{36}Cl/Cl map constructed by Phillips (2000), shown in Fig. 5c, is considerably different from the pattern shown in the empirical data (Fig. 1) in four ways. First, the irregular tight curves in the isopleths in California and Nevada reflect large regional variations in precipitation that control the ^{36}Cl/Cl values calculated for the Phillips (2000) model. This type of small-scale variability cannot be captured by the empirical data due to the sparsity of sample locations. Second, the Phillips (2000) model shows only small variations in ^{36}Cl/Cl over a large region encompassing northern Colorado, western Nebraska and South Dakota, and all of Wyoming. The empirical data (Fig. 1) show large variations in the same region. An explanation of the contrast is not evident. Third, the measured distribution (Fig. 1) shows that a zone of low ^{36}Cl/Cl exists in northern Utah and southeastern Idaho. This probably reflects contributions of "dead" chloride, i.e., ^{36}Cl/Cl≅0, from the Bonneville Salt Flats, Utah, and other playas in the region. This zone is not evident in the Phillips (2000) model. Fourth, values of ^{36}Cl/Cl in the Phillips (2000) model (Fig. 5c) generally tend to overestimate those of the empirical result (Fig. 1), especially in the southeastern USA (e.g., Mississippi, Alabama, Georgia, and Florida) and the Midwest (e.g., Minnesota, Iowa, and Indiana) where the predicted ^{36}Cl/Cl are roughly twice the observed values. This is a large-scale trend that illustrates a significant difference between the Phillips

Fig. 5 Isopleth maps of ^{36}Cl/Cl based on previously published models of ^{36}Cl/Cl ($x10^{-15}$) in the United States: **a** Bentley et al. (1986); **b** Hainsworth (1994); **c** Phillips (2000)

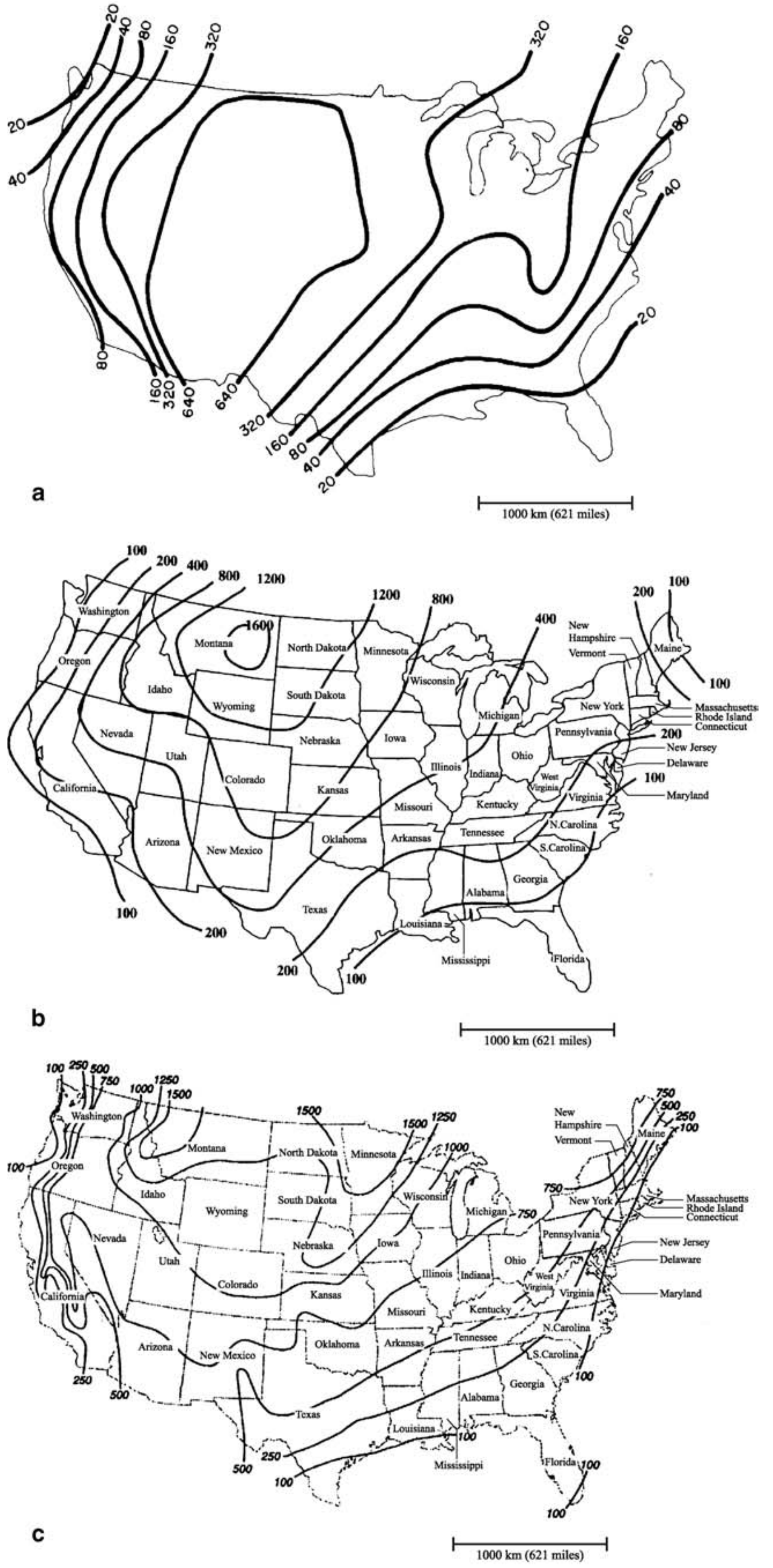

Table 3 Previously published estimates of the global ^{36}Cl flux

Global production/ fallout rate (atoms m^{-2} s^{-1})	Estimation method	Hemisphere of measurement	Reference
11	Calculated	–	Lal and Peters (1967)
19	Calculated	–	Masarik and Beer (1999)
19	Calculated	–	Blinov (1988)
17–26[a]	Calculated	–	Oeschger et al. (1969)
14–24[a]	Calculated	–	Huggle et al. (1996)
24	Measured	Northern	Andrews et al. (1994)
28	Measured	Northern	Hainsworth et al. (1994)
30	Measured	Both	Phillips (2000)
15	Measured	Southern	Keywood et al. (1998)
10–14	Measured	Southern	Nishiizumi et al. (1979) Finkel et al. (1980) Nishiizumi et al. (1983)

[a] Minimum and maximum production rates reflect changes in activity due to sunspot cycle

(2000) model and the empirical results of the current study. Parts of the West and Southwest (e.g., regions of Arizona, New Mexico, Nevada, Utah, Colorado, Wyoming, and South Dakota) are an exception to this trend as the predicted and observed ^{36}Cl/Cl are of similar magnitude in these areas.

The higher ^{36}Cl ratios predicted by Phillips (2000) could be related to the relatively high estimate of the mean global ^{36}Cl flux used in his model (30 atoms m^{-2} s^{-1}) compared with that found in this study (~20 atoms m^{-2} s^{-1}). The precipitation correction to the ^{36}Cl flux used in the Phillips model would have reduced the effective ^{36}Cl deposition rate in the Southwest, which could explain why the model and empirical results are in better agreement in this region. Several estimates of the global mean ^{36}Cl flux are given in Table 3. The theoretically calculated ^{36}Cl production rates are in good agreement with the flux calculated in this study, particularly the calculation of 19 atoms m^{-2} s^{-1} by Masarik and Beer (1999). Estimates of the ^{36}Cl flux derived from measurements are highly variable (Table 3).

The correction of the ^{36}Cl flux for local variations, suggested by Phillips (2000), relates local deviations from mean latitudinal precipitation to local deviations in mean latitudinal ^{36}Cl deposition:

$$D_{36}^{m} = \bar{D}_{36}(\lambda) + S_D(\lambda)(P_{local} - \bar{P}(\lambda)) \tag{1}$$

where D_{36}^{m} is the local or measured ^{36}Cl flux, $\bar{D}_{36}(\lambda)$ is the mean ^{36}Cl flux for a given latitude band λ, P_{local} is the local mean annual precipitation rate, and $\bar{P}(\lambda)$ is the mean precipitation rate for a given latitude band. The parameter $S_D(\lambda)$ describes the relationship between ^{36}Cl deposition and precipitation rate for a given latitude band, i.e., is the slope of a regression line on a plot of the ^{36}Cl flux vs. precipitation. This correction assumes a linear dependence between ^{36}Cl deposition and precipitation. In the current study, a strong linear correlation is not observed between the ^{36}Cl deposition rate and precipitation (Fig. 6). The reason for this lack of correlation is unclear; however, it may be related to spatial variability of depositional processes.

The net flux of a constituent from the atmosphere to the land surface at a given location can be expressed as the sum of wet and dry deposition rates, which are a function of concentration (Davidson 1989):

$$F_T = F_{wet} + F_{dry} = C_P P + v C_a \approx (KP + v^*)\bar{C}_a \tag{2}$$

where F_T, F_{wet}, and F_{dry} are the net, wet and dry depositional fluxes of the constituent to the surface. The concentration of the constituent in precipitation is given by C_P, C_a is the concentration in air at the ground surface, $\bar{C}_a$ is the mean air-phase concentration over the entire atmospheric column, K is a vertically averaged partitioning coefficient which describes scavenging between the air and precipitation phase, P is the local precipitation rate, and v is a dry deposition velocity, which describes processes other than precipitation that carry the constituent to the ground surface. The last variable in Eq. (2) is v^*, which is a dry deposition velocity that is scaled by the ratio between surface-air concentration and mean-air concentration (i.e., $v^* = v \cdot (C_a/\bar{C}_a)$.

From Eq. (2), it is inferred that for the linear relationship expressed in Eq. (1) to hold, the parameters K, v^*, and $\bar{C}_a$ must be approximately constant within a given latitude band or vary in a complementary manner. It is likely that there are many situations in nature where these conditions will not hold. For example, the atmospheric parameters which control deposition, i.e., K and v^*, are spatially variable because site-specific conditions, such as topography, have an important effect on local atmospheric conditions, such as turbulence. Furthermore, and perhaps more importantly, if the atmospheric concentration of ^{36}Cl (i.e.,$\bar{C}_a$) varies spatially within a latitude band, then the net ^{36}Cl flux cannot depend linearly on precipitation. Spatial variability of the atmospheric ^{36}Cl concentration can be expected if there exists spatial heterogeneity in the mean location of stratosphere–troposphere exchange within a given latitude belt.

At a given location, the deposition parameters (K and v^*) and atmospheric ^{36}Cl concentration may be approximately constant through time. According to Eq. (2), this would result in a linear relationship between precipitation and the ^{36}Cl flux for a particular site. The high degree of linear

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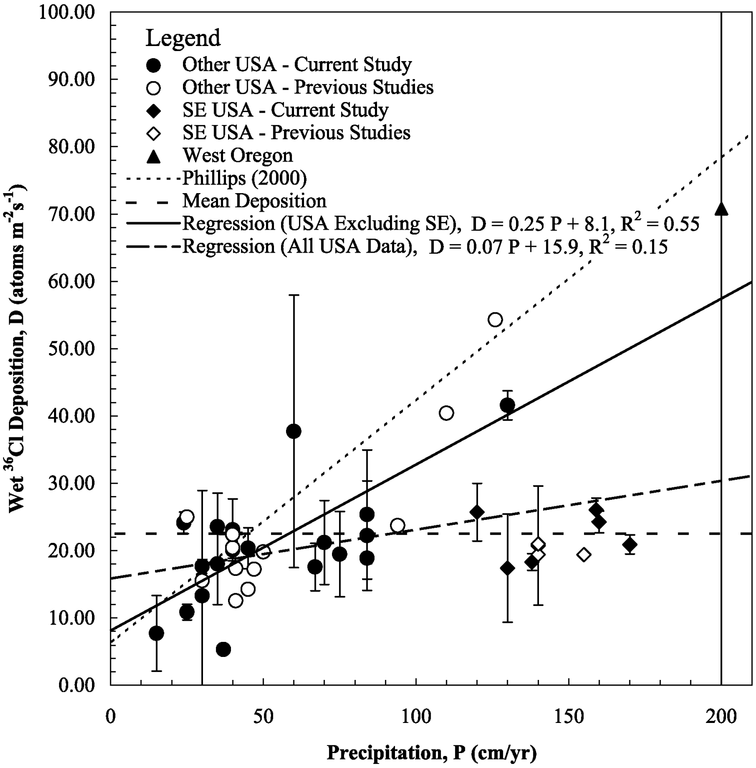

Fig. 6 Plot of ^{36}Cl deposition rate vs. precipitation. A strong correlation between ^{36}Cl fallout and annual precipitation, as proposed by Phillips (2000), is not observed for data distributed throughout the United States (R^2=0.15). The correlation is significantly improved when data from the southeastern USA, presumably an area with low atmospheric ^{36}Cl concentrations, are not included in the regression (R^2=0.55). Previously published values refer to those studies listed in Table 2. The Phillips (2000) curve has been adjusted assuming that wet deposition accounts for 74% of total deposition. Western Oregon was not included in the regressions due to the large error associated with this datum point

correlation observed in the data of Knies et al. (1994) may be the result of such an effect. The opposite was shown by Hainsworth et al. (1994) who found little correlation between precipitation and ^{36}Cl fallout at a site in Maryland, USA.; they concluded that seasonal differences in the tropospheric concentration of ^{36}Cl were responsible for the poor correlation. Recently, Sterling (2000) has suggested that variability in the atmospheric concentration of chloride leads to the need for spatially and temporally dependent deposition parameters [i.e., $S_D(\lambda)$ in Eq. (1)] to accurately model the deposition of chloride. It is likely that the same is true for modeling the deposition of ^{36}Cl.

Phillips (personal communication, 2002) has speculated that the southeastern United States may have anomalously low atmospheric ^{36}Cl concentrations. This is due to the persistent inland flow of air masses from lower latitudes of the Atlantic, where ^{36}Cl availability is diminished (Fig. 4). Equation (2) indicates that, for a given precipitation rate, lower atmospheric ^{36}Cl concentrations will lead to lower ^{36}Cl deposition rates. Since the precipitation rate in the southeastern USA is higher than the continental average, a low atmospheric ^{36}Cl concentration could result in a ^{36}Cl flux for this region that is equal to the mean flux over the United States, as observed in Fig. 6. By excluding the data from the southeastern part of the country (i.e., Florida, South Carolina, Tennessee, Georgia, Alabama, Kentucky, and Mississippi), the coefficient of determination for a linear regression is significantly higher (R^2=0.55) than when this region is retained in the data set (R^2=0.15). This improvement in the correlation between ^{36}Cl deposition and precipitation is a good indicator that factors, such as atmospheric ^{36}Cl concentrations, must be considered in addition to precipitation when modeling ^{36}Cl deposition.

Conclusions

The use of ^{36}Cl as a hydrologic tracer requires that the input of this nuclide to groundwater systems be known. As a result, an intensive groundwater sampling program was undertaken by Davis et al. (2003) to estimate ^{36}Cl/Cl in the United States. Significant variability in ^{36}Cl/Cl exists at the aquifer scale due to processes such as dissolution of chloride in the subsurface and mixing of young water, which contains an anthropogenic component of ^{36}Cl, with old water having natural levels of ^{36}Cl. By attempting to remove the effects of these processes, the natural distribution of ^{36}Cl/Cl in meteoric groundwater across the continental United States was inferred by Moysey (1999) and Davis et al. (2003).

Processes that are responsible for the continental scale distribution of ^{36}Cl and chloride fallout from the atmosphere ultimately control the meteoric ^{36}Cl/Cl signal in groundwater. The single most important factor controlling the ^{36}Cl/Cl distribution is the spatial dependence of stable-chloride fallout, which can be shown to statistically account for almost all of the variability observed in ^{36}Cl/Cl across the United States. The lowest ^{36}Cl/Cl ratios (<100) are observed in coastal areas, which coincide with the location of high chloride deposition. The influence of the oceans steadily decreases inland and a maximum ^{36}Cl/Cl of around 1,200 is reached over the central High Plains in eastern Wyoming. The overall pattern is modified somewhat by topography and dominant wind patterns, resulting in high ^{36}Cl/Cl gradients along the west coast and low gradients extending from the Gulf of Mexico in the east. An important continental source of chloride, associated with the Bonneville Salt Flats, is apparent in northern Utah where a large depression in ^{36}Cl/Cl is observed.

The general pattern of variability observed in the empirical distribution of ^{36}Cl/Cl was captured in the models presented by Bentley et al. (1986), Hainsworth (1994), and Phillips (2000). This is due to the dependence of ^{36}Cl/Cl on stable-chloride deposition, which was incorporated explicitly into these models through the direct use of empirical data. The best general agreement with the empirical distribution found in this study was provided by the Hainsworth (1994) model, which captured most of the observed spatial variability. The model of Bentley et al. (1986) underestimates the observed ^{36}Cl/Cl throughout the country. The model proposed by Phillips (2000) generally overestimates the observed ^{36}Cl/Cl in the eastern part of the United States, but matches the measured values in the Southwest.

The mean flux of ^{36}Cl over the United States was calculated to be 30.5±7.0 atoms m^{-2} s^{-1} (assuming that dry deposition is on average 26% of the total) and was found to have no dependence on latitude, given the range of the investigation. This average continental fallout corresponds to a mean global ^{36}Cl flux of 19.6±4.5 atoms m^{-2} s^{-1}, which was calculated using the latitude dependence for ^{36}Cl fallout proposed by Lal and Peters (1967). This estimate is in good agreement with the flux of 19 atoms m^{-2} s^{-1} calculated by Masarik and Beer (1999) from theoretical considerations, but is low compared to the flux of 30 atoms m^{-2} s^{-1} estimated by Phillips (2000) from measurements of ^{36}Cl deposition obtained by various studies from around the globe.

The ^{36}Cl deposition rates in dry areas, such as the Southwest, were generally found to be lower than the mean flux for the continental USA. However, the strong linear correlation between ^{36}Cl deposition and precipitation found by Knies et al. (1994) and Phillips (2000) was not observed for the data in this study, which are representative of a large range of the climactic conditions throughout the continental United States. The lack of this linear relationship is likely due to spatial variability of depositional processes and, in particular, atmospheric ^{36}Cl concentrations over the continent. This hypothesis is supported by a significant improvement in the coefficient of determination, R^2, from 0.15 with all data considered, to 0.55 with data from the southeastern USA excluded. Since air masses in the South typically originate at low latitudes and travel north through the Gulf of Mexico, the atmospheric ^{36}Cl concentrations in this region are probably low compared to those found at similar latitudes in the rest of the United States. The diminished chloride availability in the atmosphere would lead to lower than expected ^{36}Cl deposition rates. This example emphasizes the fact that a combination of processes, at global and local scales, contributes to the removal of ^{36}Cl from the stratosphere and subsequent distribution of this nuclide at the land surface. Accurate modeling of the ^{36}Cl/Cl distribution will require a quantitative understanding of these processes.

The map of ^{36}Cl/Cl presented in this study is not intended to provide a rigorous estimate of the initial ^{36}Cl/Cl for the United States. In previous publications (Moysey 1999; Davis et al. 2003), the high degree of local variability that exists in ^{36}Cl/Cl was discussed in detail. The results presented here should be taken as a tool to improve our understanding of ^{36}Cl and chloride deposition and variability. For individual aquifer studies, the initial value of ^{36}Cl/Cl should be estimated locally from the available data whenever possible. The map provided here can be used, however, as a general reference as long as care is taken to note the uncertainty underlying Fig. 1. The empirical ^{36}Cl/Cl distribution presented in this study will be most useful as a reference for comparison against future models of ^{36}Cl/Cl variability, for identifying significant local variations from the continental pattern of ^{36}Cl/Cl, and for better understanding the processes of stratosphere–troposphere exchange and atmospheric deposition.

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