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The Rise and Fall of Chloride in Larderello Steam

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ABSTRACT

Significant amounts of Cl appeared in steam from upflow zones of Larderello in the early 1960s. The general pattern of Cl concentration includes: a rapid rise, a more or less constant plateau (coincident with a low, stabilized flow rate), and rapid decrease generally after 1980. The rise correlates with a general decrease in pressure throughout the field presumably due to the disappearance of liquid in the two-phase, vapor-dominated reservoir. The plateau may be related to the distance from centers of upflow and inhomogeneities in the temperature and chemistry of deep brine. The decrease is correlated with the start of liquid introduction from reinjection and drilling losses and particularly with the time of breakthrough of steam formed from injected water. This history suggests that when liquid was exhausted in some areas and pressure dropped, other areas suffered accelerated boiling and dried out as well, allowing high-Cl steam formed from boiling of deep brine to move upward to reach the wells. The introduction of liquid after 1980 reversed the process. A similar situation may be occurring now at The Geysers, California.

INTRODUCTION

Chloride in steam from Larderello was mentioned by Allegrini and Benvenuti (1970) as an important factor in corrosion of pipelines. At that time (1960s), Cl in steam in "considerable quantities" was "rather rare," with only 22 of 188 wells listed carrying >5 ppm and only 10 wells with >10 ppm. The Cl was considered to be carried as NH_4Cl (which is neutral in solution) despite evidence that some condensate drains carried high concentrations of Fe^{++} as well as Cl^-, indicating H^+ corrosion. They contributed the critical observation that Cl in superheated steam was noncorrosive but became corrosive when it contacted liquid water. D'Amore et al. (1977) showed that steam with more than 1 ppm Cl was limited to zones of intense exploitation (Fig. 1), where superheating of steam is most common. HCl was identified as the probable form of Cl in steam, but the formation of HCl was considered to be limited to reaction of $CaCl_2$ with silica. D'Amore and Truesdell (1979) considered Cl in steam to indicate boiling of a deep, high-Cl brine and pointed out that Cl in steam from three Larderello wells had appeared in low concentrations (0-5 ppm) when first analyzed in 1963 and increased until at least 1977, reaching 22 ppm in steam from one well.

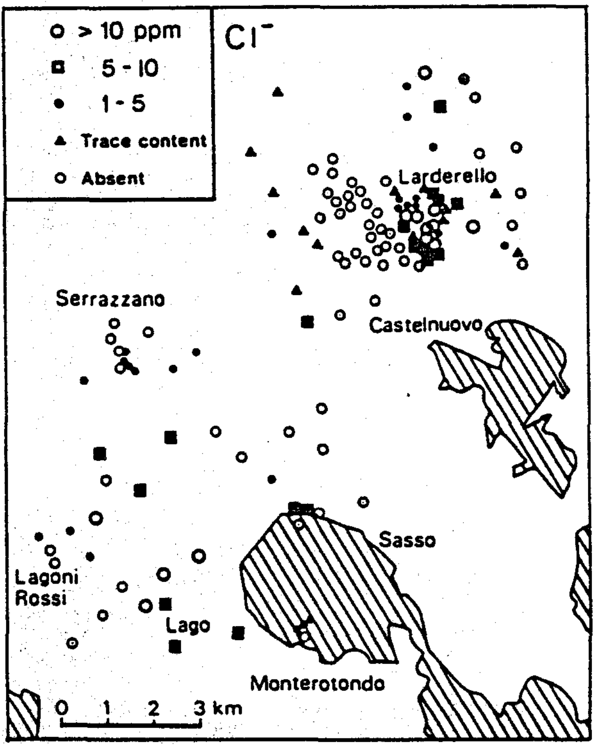

Figure 1. Chloride concentrations in Larderello steam from D'Amore et al. (1977).

D'Amore and Pruess (1986) extended the observations of D'Amore and Truesdell (1979) on temporal changes in steam composition and correlated Cl in steam with the increase of the apparent reservoir vapor saturation ("y" values). Recently, Haizlip and Truesdell (1988) and Truesdell et al. (1988) reconsidered the origin of Cl in geothermal steam and concluded that it is carried as HCl volatilized from Cl brine under conditions of low pH, high Cl, and/or high temperature. Condensate at normal reservoir temperatures would completely scrub HCl from steam, so transport is limited to superheated steam with a dry pathway from the boiling brine. The occurrence of Cl in Larderello steam was briefly described by Truesdell et al. (1988) but will be discussed here at greater length, and the reasons for its rapid rise and (after 1980) fall will be considered in relation to reservoir processes.

OBSERVED CL IN LARDERELLO STEAM

Chloride in steam was not routinely analyzed at Larderello before 1963. Earlier, only steam from wells showing high corrosion was analyzed for Cl, as well as Na^+, SiO_2, and other nonvolatile salts, which indicate entry of liquid from the reservoir. The low values for Cl found in the first fieldwide analyses suggest that Cl was below detection (<1 ppm) in most wells before 1960. Of 45 wells for which we have detailed chemistry, Cl when first measured (in 1963 for most wells, up to 1982 for newer wells) was below detection in 14 wells, 1-2 ppm in 15 wells, 2-5 ppm in 7 wells, 5-15 ppm in 6 wells, and >15 ppm in 3 wells.

Thirteen wells located in different zones of Larderello were chosen for detailed study (Fig. 2). These wells have a range of Cl contents and temperature (Table 1) typical of wells considered to be in upflow zones on the basis of relatively high Cl and B and relatively low CO_2 and NH_3 in produced steam (D'Amore and Truesdell, 1979). Changes in Cl concentration with time for these wells are shown in Figures 3-5. The patterns shown are similar. Cl concentrations start low (below 5 ppm for 5 wells, below about 15 ppm for all but 2 wells) and increase rapidly, reaching a more or less broad plateau from 7-10 years after the first measurement until 1980 to 1983. After 1980 most wells showed a sharp decrease in Cl, which was temporary for some wells, and continued for others at least through 1985-1986, which is the limit of our data. Other data for these wells are shown in Table 2, including typical gas analyses and reservoir temperatures calculated from wellhead physical data and from gas geothermometers.

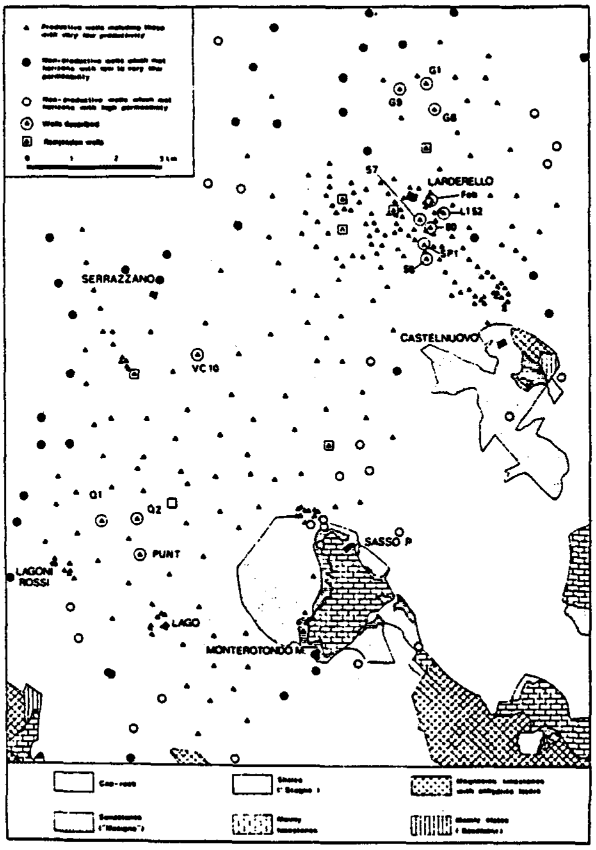

Figure 2. Locations of wells studied and reinjection wells.

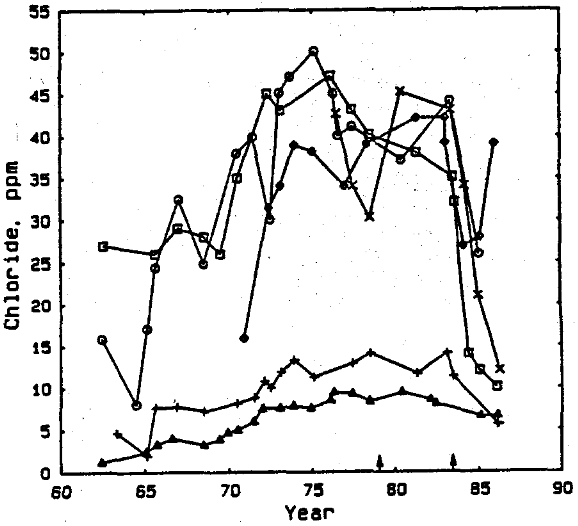

Figure 3. Chloride concentrations in steam from central Larderello wells (L56, pluses; L57, squares; L80, circles; L152, crosses; Fab, triangles; SP1, diamonds). The earlier arrow indicates the start of injection. The later arrow indicates the breakthrough of steam vaporized from this water.

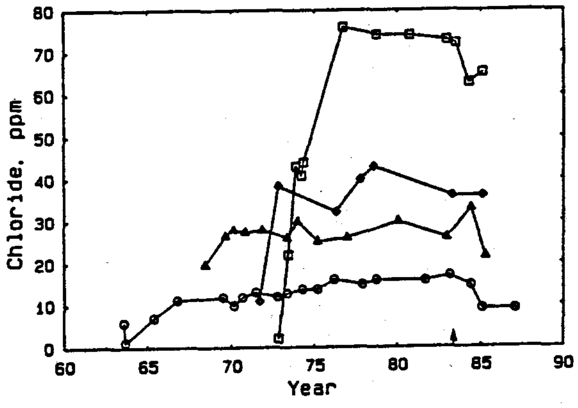

Figure 4. Chloride concentrations in steam from western Larderello wells (VC10, circles; Q1, triangles; Q2, squares, Punt, diamonds). The arrow indicates the start of injection.

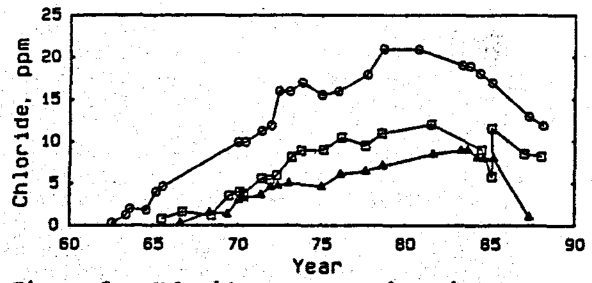

Figure 5. Chloride concentrations in steam from northern Larderello wells (G1, circles; G6, squares; G9, triangles).

Data in D'Amore and Truesdell (1979) and D'Amore and Pruess (1986) suggest that, for wells in the central zone of Larderello drilled before 1950, the appearance of Cl in steam (extrapolated to zero ppm) correlates with the stabilization of steam flow at 10-20% of initial values. For these earlier wells the time of chloride appearance seems to be about 1957.

INDICATIONS OF RESERVOIR DRYING AT LARDERELLO

The accepted model for vapor-dominated geothermal systems (after White et al., 1971) consists of a liquid-saturated condensate zone, a vapor-dominated two-fluid-phase zone, and a deep boiling brine. In the natural state, steam produced from the boiling brine flows upward in the vapor-dominated zone along large fractures and conduits, and condensate flows downward on surfaces and discontinuous small fractures to replenish the deep brine. Although deep temperatures (>350°C) have been measured at Larderello (Cappetti et al., 1985), the deep brine has never been satisfactorily sampled; however it must exist to provide upflowing steam to maintain system temperatures and heat flow.

In the natural state the vapor-dominated reservoir at Larderello was drier than that at The Geysers (1 wt % of total fluid at Larderello compared with 5% at The Geysers, Nathenson and Muffler, 1975 using methods from Nathenson, 1975) and extensive production has dried it further. The production of super-heated steam from wells in all parts of Larderello indicates complete drying of the upper parts of the reservoir near well bottoms but does not prove that the entire vapor-dominated reservoir above the brine was dry.

The drying of the reservoir and the production of superheated steam are clearly related to the decline of reservoir pressure. Measurements in newly exploited zones as well as thermodynamic arguments (James, 1968; Truesdell and White, 1973) indicate that natural state Larderello pressures and temperatures at the top of the vapor-dominated reservoir were near 32-35 ata and 235-240°C. These conditions are consistent with coexistence of vapor and liquid. Pressures in intensely exploited zones of Larderello dropped rapidly (Ferrara et al., 1970; Celati et al., 1976). Early shallow wells in central Larderello showed lower pressures (30 ata) probably resulting from natural discharge. By 1943 pressures in the top of the reservoir in the central Larderello and Castelnuovo zones had declined to 10 ata, and by 1952 the entire Larderello and Castelnuovo zones showed similar low pressures. In 1975 pressures in the reservoir top were near 5 ata.

Although pressures dropped rapidly in response to production, temperatures in the vapor-dominated zone changed very little. The wells studied by D'Amore and Truesdell (1979, 1980) showed nearly constant wellhead and reservoir temperatures after the end of production from vaporized near-well liquid. Declining pressures with near-constant temperature result in drying of the reservoir and superheating of steam. Indications of drying from pressure decline and steam superheating are, however, limited to the upper part of the vapor-dominated zone where most wells have their main steam entries. Processes occurring deeper in the vapor-dominated zone, particularly near the deep brine, would not be revealed by these observations.

Calculations by Haizlip and Truesdell (1988) show that the production and transport of HCl in geothermal systems require the boiling of high-Cl (or low-pH) brine at temperatures above 300°C and the existence of dry pathways through zones of lower temperature. The only part of the system where Cl-containing steam could be generated is the deep brine because sufficiently high temperatures and Cl contents of liquids are found only there. The rapid increase of Cl in Larderello steam is unlikely to have resulted from changes in the deep brine such as a sudden increase in brine salinity, acidity, or temperature. The drying of the vapor-dominated zone is the most likely factor. Cl-bearing steam must have come from the >300°C boiling brine and must have traveled by an entirely dry path to the well because any lower-temperature, lower-Cl liquid would have scrubbed almost all Cl from the steam.

IMPLICATIONS FOR RESERVOIR BEHAVIOR

The nearly simultaneous appearance of high-Cl steam from wells within intensely exploited zones (e.g., central Larderello, Fig. 3) or in nearby wells (e.g., Gabbro, Fig. 4) indicates drying of these parts of the reservoir within a short time period. This probably resulted from pressure equalization and lateral transfer of steam. With continued production, pressure in the reservoir will be maintained as long as both liquid and vapor coexist. As soon as drying is complete, the pressure drops and steam from still-wet zones, where pressures are higher, flows to the dry zone. The lateral flow accelerates the drying of wetter parts and they eventually dry. As soon as steam conduits to the boiling brine are dry, pressures drop and steam in these conduits becomes superheated. The pressure drop increases boiling of deep brine and the now dry conduits no longer scrub Cl from steam boiled off the brine. Cl contained in deep steam is now able to reach the wells and does so at about the same time throughout the connected volume.

The appearance of Cl in widely separated areas at the same time might result instead from similar exploitation histories with drying of these areas occuring coincidentally. However, exploitation histories at Larderello are not the same. Intense modern exploitation of central Larderello began about 1950 with many wells drilled earlier. In 1951 total flow reached 2500 t/h (Sestini, 1970). This was two times the flow from earlier producing wells. Some of our study wells from the central Larderello area started production early (Fab, 1941; L57, 1949; L56, 1950; and L80, 1951) and some later (SP1, 1963 and L152, 1976). Other zones were developed later. Gabbro was produced after 1960 (G1, 1962; G6, 1964; and G9, 1966). The western area was producing less than 600 t/h before 1957 and only reached 1000 t/h in 1962. Our examples also started production relatively late (VC10, 1963; Q1, 1967; Q2, 1972; and Punt, 1971).

Given this uneven history of exploitation it seems unlikely that coincidence produced the similarity of chloride appearance and increase between, for example, the Gabbro wells developed in the mid-1960s and Fab, L56 and L80 developed from 1940-1950. The distance from central Larderello to Gabbro is 3-4 km, so lateral flow is acceptable. Lateral flow to Larderello was in fact indicated in the study by Brigham and Neri (1979) of the Gabbro zone. The distance from central Larderello to Q1 and Q2 is 9-10 km and lateral flow is less likely. The similarity of the chloride curves of these wells to those for Larderello wells is less marked. However, these wells do not show evidence of a long period of production of low-Cl steam, indicating that somehow there was less available liquid in the vapor-dominated reservoir, perhaps through vaporization and lateral flow to other zones.

Gradual increase of Cl over a period of 7-10 years suggests that low-Cl steam generated from boiling of condensate mixes with an increasing fraction of deep, high-Cl steam. The mixing occurs after low-Cl steam has become superheated by passage through dry zones. Conduits from the brine to the zone of mixing and from there to the surface are dry but liquid water still is present further from the wells (and possibly in nearby matrix blocks) to provide low-Cl steam. Apparently the proportion of deep steam increases until almost all steam was from deep boiling brine and the Cl contents of produced steam stabilizes.

The plateau in Cl contents, which differs in concentration in different wells and different zones, may reflect the equilibrium concentrations of HCl boiled from brines of different temperature, pH, or Cl content. The calculations of Haizlip and Truesdell (1988) show that 100 ppm HCl could originate from boiling at 350°C of brine with 20,000 ppm Cl (like seawater) and pH near 5 or with 100,000 ppm Cl and pH near 6. pH is most likely buffered by feldspar-mica reactions to values near 6 so the more concentrated brines are indicated. At 300°C only 10 ppm Cl would be generated from these brines, suggesting higher temperatures in agreement with deep drill-hole results.

Variations of Cl concentrations with time (e.g., Figs. 3-5) suggest that drying of much of central Larderello and parts of western Larderello occurred in 1955-1960 and that drying of parts of the northern zone occurred in 1960-1965. Wells starting production after 1970 (SP1 in Fig. 3) show moderately high initial Cl and rapid increase suggesting a small amount of initial low-Cl steam from near wellbore liquid that was quickly exhausted. Within these zones there is apparently rapid lateral steam flow and high lateral permeability. More detailed examination of steam Cl could indicate details of connectivity between wells within zones and in interzone areas.

THE EFFECT OF WATER INJECTION AND DRILLING WATER LOSS

Drying of large parts of the Larderello reservoir has resulted in low steam flow and electricity production with large amounts of heat left in the reservoir. This additional heat may be exploited through injection of additional water to increase pressure and steam flow (Cappetti et al., 1982).

Water was introduced into the Larderello reservoir both as injected condensate from cooling towers and also from lost circulation during drilling of new wells. Relating changes in Cl to these events is complicated because reinjection and new drilling were part of a program for the rejuvenation of the Larderello field and both started at about the same time (Cappetti et al., 1982; Bertrami et al., 1985). The wells chosen for study are away from areas of natural recharge and water introduced by this means is not a consideration. We have shown on Figures 3 and 4 approximate times of the start of injection of cooling tower condensate and, for the central area, the time when breakthrough of steam generated from injected condensate was detected isotopically (D'Amore et al., 1987).

Although exact timing of liquid introduction is not available, the general correspondence of sudden decrease in steam Cl contents with liquid injection and, for the central zone, with breakthrough of steam from injected liquid are in reasonable agreement. The data suggest that introduction of liquid produced the Cl decrease through the action of newly generated low-Cl steam and liquid itself.

Mixing of new low-Cl steam may explain the data for some wells in which the decrease was small or temporary, perhaps resulting from a short period of water introduction (as from nearby drilling).

Wells with large, permanent (at least to 1986) decreases in Cl are apparently affected not only by the introduction of low-Cl steam but also of liquid that scrubbed Cl from deep steam. The mechanism of liquid introduction is suggested by the simulations of Calore et al. (1986), which indicated that injected liquid moves mostly vertically downward unless it encounters lower permeability that causes lateral spreading. At Larderello this lateral spreading of liquid would be expected to occur at the bottom of the two-phase vapor-dominated reservoir where increased permeability from mineral solution by downward-flowing condensate gives way to decreased permeability from mineral precipitation by boiling brine. (Pruess et al. (1987) also suggest decreased permeability at this location.) If the contrast in permeability is sufficiently great, liquid could be carried to some distance from its source. Direct introduction of liquid would scrub Cl from deep steam and eliminate superheating at least locally. After the steam left the liquid it might again become superheated, but it would no longer have a supply of HCl. The behavior of Cl in produced steam thus depends on the quantity and location of introduced liquid and also on details of lateral and vertical permeability within the reservoir. Quantitative study of the Cl data along with details of injection and drilling and of other reservoir observations (interference for example) could provide information about reservoir structure at the bottom of the vapor-dominated zone and the top of the brine zone.

A CONCEPTUAL MODEL

The reservoir processes suggested by the changes in Cl of Larderello steam are illustrated by Figure 6. This cartoon is telescoped in time and space showing on the left side the evolution of an expanding dried and decompressed zone of the reservoir just after it has contacted the deep brine. Steam from the not yet dried two-phase (vapor and liquid) zone is flowing from the left into the dry zone. The source of this saturated, low-Cl steam is vaporization of condensate induced by exploitation pressure decline. The lack of Cl in this steam results from its source in boiling of low-Cl liquid at normal reservoir temperature and from passage through the still wet reservoir, which would scrub any Cl in the steam. This saturated, low-Cl steam becomes superheated by isothermal decompression as it flows through the dried zone.

A flow of high-Cl steam is shown entering the dry zone from below where it originates from boilng of saline, high-Cl brine. Temperatures are not shown but must include a relatively steep gradient between the part of the brine that is initially above 350°C and the 250°C vapor-dominated reservoir. The high-Cl steam mixes in the dried zone with low-Cl steam (from lateral flow) and flows to the well. The path from the boiling brine to the well bottom is entirely dry and low-Cl steam that was once saturated has become superheated before mixing. Thus, no scrubbing of chloride occurs and the well produces more or less diluted high-Cl steam. Before the dried zone encountered the deep brine, Cl in steam from the brine would have been scrubbed by condensate in the reservoir and would have contributed steam but not Cl to the production.

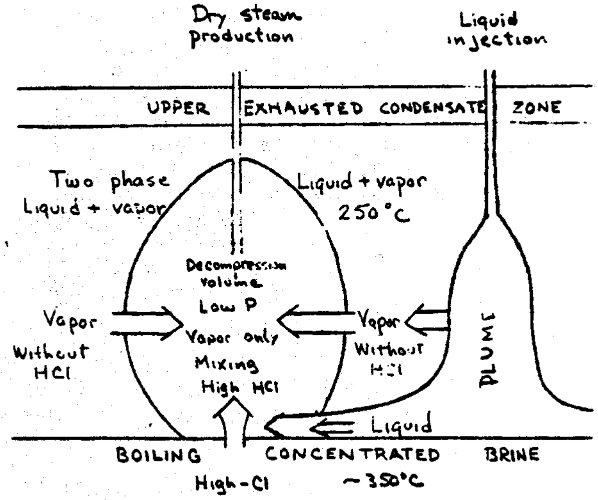

Figure 6. Cartoon of reservoir processes influencing chloride concentrations in steam.

On the right side of the cartoon is shown a plume of liquid (and saturated steam) from an injection well or circulation lost during drilling. Saturated steam from vaporization of liquid contributes to the mixture of high- and low-Cl steam in the dried zone feeding the production well. More importantly, liquid water from the plume is spreading laterally along the high permeability at the base of the vapor-dominated reservoir above the low-permeability original brine interface (at the center of the cartoon). This liquid water has just encountered the dried zone and is about to overtake the high-Cl steam rising from the boiling brine. When this happens steam from the high-Cl brine will become saturated and Cl will be scrubbed into the liquid. As the steam continues to flow upward through the dried zone it will become super-heated again but will not increase in Cl contents. If the flow of liquid diminishes, these processes will reverse themselves. All of these processes occur below the bottoms of most wells at depths sufficiently great that none of the physical characteristics of the produced steam would indicate their occurrence.

FURTHER IMPLICATIONS OF THE MODEL

The general drying of the Larderello reservoir indicated by the appearance of Cl in produced steam has further implications for the exploitation of the field and explains some observations that were poorly understood. A negative implication is that for much of the field (where high stabilized Cl concentrations occurred) the supply of stored liquid in the condensate and vapor-dominated zones of the reservoir has been exhausted. The evaporation of this stored liquid provided the early high flows of low-temperature steam described by D'Amore and Truesdell (1979). A more positive implication is that the stabilized flow of wells with high Cl comes from the boiling of a deep brine. Boiling of deep brine might be expected to be a more stable source than evaporation of liquid in either the relatively thin condensate zone or the originally rather dry vapor-dominated zone. Using this model the long-term increase in average steam temperature at Larderello is simply the result of progressive drying of low-temperature sources of steam and possibly by their replacement by higher temperature steam from boiling of deep hot brine. This last requires the heating of conduits from the brine.

Finally, the model has implications for the future of The Geysers. Exploitation at The Geysers started in the 1960s compared with 1930s at Larderello. Cl in steam at Larderello appeared in 1960, signalling general drying of the upper parts of the reservoir. Cl in steam at The Geysers has appeared in the mid-1980s (Haizlip and Truesdell, 1988) and most probably also signals widespread drying of the vapor-dominated reservoir at The Geysers. This has the same implications for steam production as at Larderello except that The Geysers vapor-dominated zone is thicker and deep steam would have further to travel to the wells, possibly resulting in lower pressures.

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Table 1. Physical and Chemical Data for Typical Wells in Upflow Areas of Larderello

Well	Max. Steam Temp., (°C)	Depth (m)	Period	Average Steam (ppm)		
				Cl	H_3BO_3	NH_3
VC10	260	>1600	1976-84	15±2	335±29	130±10
Q1	250	>1000	1970-84	30±4	430±30	116±18
Q2	255	>1000	1977-84	72±5	487±44	90±8
PUNT	250	>1000	1973-85	37±4	352±36	103±16
LA80	240	~600	1970-83	39±8	344±26	125±7
LA57	248	~500	1970-83	37±7	330±32	118±17
SP1	250	>1000	1973-86	36±5	340±10	109±11
LA56	237	~600	1973-83	12±1	257±25	141±17
FAB	240	~600	1976-83	8±1	199±20	240±10
G1	242	>1000	1977-84	19±1	276±37	168±12
G6	250	~800	1973-84	10±1	206±16	209±27
G9	237	~980	1978-85	9±1	211±32	190±7
LA152	245	>1000	1976-84	40±6	362±37	100±10

Table 2. Typical Steam Chemistry for Wells from Table 1

Well	Date	Gas/Steam (molal ratio)	CO_2	H_2S	H_2	CH_4	N_2	$t_{(Res)}$ °C	$t_{(H2S)}$ °C	$t_{(H2)}$ °C
			(volume, percent)							
VC10	1981	12.4	91.29	3.15	3.43	1.52	0.65	260	254	262
Q1	1981	12.1	90.07	3.49	3.52	1.95	0.97	265	259	265
Q2	1981	15.3	90.11	3.68	3.69	1.81	0.71	275	266	271
PUNT	1978	12.0	90.91	3.19	3.70	1.66	0.54	265	254	266
LA80	1981	32.1	93.70	2.04	1.65	1.55	1.06	260	255	254
LA57	1981	32.9	93.72	1.87	1.67	1.63	1.10	265	254	254
SP1	1975	29.7	93.24	2.08	1.83	1.85	1.01	265	255	256
LA56	1978	29.8	94.31	1.73	1.58	1.37	1.01	255	250	250
FAB	1981	22.5	93.57	1.91	1.89	1.29	1.40	250	248	253
G1	1981	38.6	95.44	1.64	1.56	0.72	0.64	260	252	254
G6	1981	44.2	95.76	1.22	1.25	0.98	0.78	250	243	250
G9	1981	48.2	96.17	1.06	1.23	1.25	0.93	245	240	250
LA152	1981	26.5	93.40	1.80	2.00	1.60	1.10	260	249	257

$T_{(Res)}$ is measured from ENEL data (±10°C); $t_{(H2S)}$ and $T_{(S)}$ are values computed from semiempirical equations 12 and 11 of D'Amore and Truesdell (1980, 6th Stanford Res. Eng. Workshop).