^{36}Cl in the vadose zone: Matching rainfall to recharge.

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^{36}Cl has become a popular tool for hydrogeologists to use when evaluating flow rates, ages and origins of groundwaters, in particular in systems where flow paths are long and/or flow rates are slow. The ratio $^{36}Cl/Cl$ and total chloride are measured in water samples, and a series of waters can be assessed in light of the different processes acting upon the stable and radioactive isotopes. Thus, figure 1 illustrates trends expected to occur on a ^{36}Cl vs. Cl^- plot for different processes.

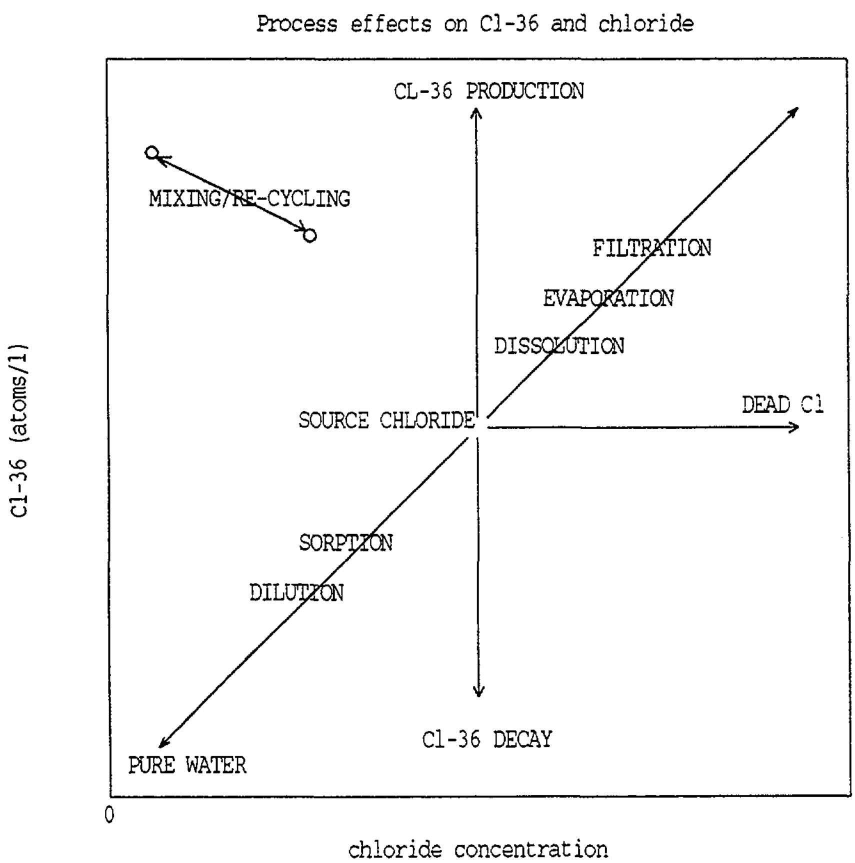

Figure 1: Processes affecting ^{36}Cl and chloride levels in groundwaters.

Use of this figure relies on knowledge of the input values for ^{36}Cl and Cl^- to the system being studied, or the analysis of a large number of samples from a single aquifer, or system, to extrapolate to potential end member water bodies. We are attempting to marry theoretical calculations for the input functions with observed values for recharge waters in aquifers around Australia.

Following from the work of Keywood et al. (1997, and in press) we can now compare the theoretical fallout of ^{36}Cl to that observed in rainfall, and then to that seen in shallow, recharge groundwaters, to evaluate the processes involved in the incorporation of chloride to groundwater systems.

The seminal work by Lal and Peters (1967) still gives the best theoretical estimate of ^{36}Cl fallout, though we should scale their values by a factor of 1.3 in light of the empirical study across western Australia by Keywood et al. (in press). Keywood et al (1997) have also synthesised a comprehensive analysis of the chloride fallout across the western half of Australia, permitting an accurate assessment of the expected ^{36}Cl/Cl ratio to be measured in rainfall. Correspondence with observed values is good, though local climatic effects are important, and local variability in the chloride fallout outlines the difficulty in providing inputs based on theoretical estimates alone.

For Australia, the presence of playa lakes across much of the interior of the continent poses additional constraints. Measurement of brines and salt crusts from playas through central Australia give ^{36}Cl/Cl ratios in the relatively restricted range: 30-60x10^{-15}. This is the upper limit to expected steady-state concentrations from neutron-capture on ^{35}Cl via neutrons derived from fission of trans-uranic elements in the subsurface (Bentley et al, 1986). Alternatively, it may represent surface re-working of chloride with a mean age of 500,000a - 1Ma, depending on rainfall concentrations. The effect of this re-working is evident in recharge waters of the Amadeus and Ngalia Basins, central Northern Territory, where observed values for the ^{36}Cl/Cl ratio in recharge waters are 40% lower than expected. This can be explained by less than 10% addition of lake salt to rainfall prior to recharge into the aquifers. The critical nature of this mixing, which is dominated by the high chloride content of the brines and surface salts, means that recharge can only be acertained through measurement of shallow bore-waters close to basin margins.

Along the eastern margin of the Australian interior, however, recharge to the Great Artesian Basin is via a distinct outcropping of sandstones along the Great Dividing Range. Shallow bores just inland of this recharge area give ^{36}Cl/Cl ratios commensurate with values estimated from theory and observed at comparable distance from the western coast of Austalia. Recharge to the shallow, unconfined aquifers of the Murray Basin in southern Australia also exhibit ratios that relate well with expected values. In areas where discharge of groundwaters predominates, in the lower reaches of the basin, however, the recharge signal is diluted by older emerging salts.

In northern Australia we have observed extreme levels of ^{36}Cl in rainfall that appear to be linked to the tropical monsoon. As yet we cannot fully explain the disrepancies between the observed and estimated values, even if we include scavaging of ^{36}Cl from the stratosphere, and correct for the increased rainfall during this period (Keywood, et al., in press).

In summary, we now have sufficient coverage of Australia to start speculating on the discepancies between the observed ^{36}Cl/Cl ratios in rainfall and the ratios measured in shallow, supposedly recharge, waters in groundwater systems. The basic premise of mixing at the surface and within the vadose (unsaturated) zone of the chloride in precipitation with remobilised salts from the local environment can now be given some empirical basis.

Salts measured from salt lakes around Australia reveal a restricted range of $^{36}Cl/Cl$ suggestive of a dominance of equilibrium concentrations of ^{36}Cl due to neutron capture on ^{35}Cl. Our rainfall measurements indicate values closer to those expected on theoretical grounds, though there are some complications, particularly where rainfall has a marked seasonality and high intensity. These values are almost always higher than the observed values in shallow groundwaters considered to be the recharge waters for major deep groundwater aquifers. Shallow groundwater aquifers seem to more reasonably reflect theoretical and rainfall values, though again, weather patterns are important.

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