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Yungay area, the nature of the oxidant remains unexplained. Photochemical reactions initiated by sunlight continually produce oxidants in the lower atmosphere and surface. However, in most soils, biological production of reduced organic material completely dominates the net redox state of soils. When biological production is less than the photochemical production of oxidation, then the soil will become oxidizing. The transition from biologically dominated soils to photochemically dominated soils appears to be abrupt. Whichever process dominates will shift the redox state in one direction or another. In the Atacama, there is a gradual decline in biological activity as conditions became drier, yet near the extreme arid region there is an abrupt transition to very low bacterial levels and low organic content.

It is unlikely that the oxidizing conditions are due to high ultraviolet flux, because the site is only 1 km above sea level. Instead, the dry conditions in the Atacama must inhibit biological production of reductants and possibly enhance the survival of photochemically produced oxidants. Our results suggest that in the extreme arid core of the Atacama, we have crossed the dry limit of microbial survival in extreme environments. The net result is that photochemical processes dominate. Thus, in the Atacama Desert, we find almost no microorganisms and low levels of organic material, and the organic material present appears to have been oxidized. The LR experiments confirm the presence of as-yet-unidentified oxidants in the Atacama soil. In many respects, these soils are similar to the Mars soils investigated by the Viking Biology Experiment and may provide a valuable testing ground for instruments and experiments designed for future Mars missions.

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A Reservoir of Nitrate Beneath Desert Soils

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A large reservoir of bioavailable nitrogen (up to ~10^4 kilograms of nitrogen per hectare, as nitrate) has been previously overlooked in studies of global nitrogen distribution. The reservoir has been accumulating in subsoil zones of arid regions throughout the Holocene. Consideration of the subsoil reservoir raises estimates of vadose-zone nitrogen inventories by 14 to 71% for warm deserts and arid shrublands worldwide and by 3 to 16% globally. Subsoil nitrate accumulation indicates long-term leaching from desert soils, impelling further evaluation of nutrient dynamics in xeric ecosystems. Evidence that subsoil accumulations are readily mobilized raises concern about groundwater contamination after land-use or climate change.

Increased deposition of bioavailable nitrogen (N) at the land surface has adversely affected water quality, biodiversity, and ecosystem functioning around the world (*1–6*). Understanding such impacts requires quantification of N sources, reservoirs, and cycling rates (*1, 5, 7, 8*). Desert soils, which cover approximately one-fourth of the conterminous United States and one-third of the land surface worldwide, are reportedly low in total N (*9, 10*). Studies of N cycling in terrestrial ecosystems have traditionally examined only the biologically active soil zone, defined operationally as extending to ~1 m in depth (*9, 11*). Within this zone, N turnover is rapid (*6*), and N concentrations decrease with depth (*7, 10, 11*). Natural sources of N in desert ecosystems include nitrate (NO_3^-) and ammonium (NH_4^+) in precipitation, eolian deposition of nitrate salts, and biological assimilation of atmospheric N_2 by N-fixing organisms (*5, 7, 8, 10*). Mechanisms of N removal include plant uptake, volatilization to ammonium and other gases, wind erosion, and denitrification (*6, 7, 12*). Nitrogen loss from the soil zone by leaching is generally assumed to be negligible in desert ecosystems (*5, 10, 12*). Our findings challenge this assumption, demonstrating that substantial quantities of N, as NO_3^-, have leached and accumulated beneath the soil zone over millennial time frames.

Soil-water N generally follows a nutrient-type profile, with concentrations that decrease sharply with depth because of biological uptake and cycling (*11*). In contrast, soil-water chloride (Cl^-) follows a conservative solute-type profile, with concentrations that increase with depth because of progressive evaporation and water extraction by plant roots. In desert settings, Cl^- typically exhibits an exaggerated conservative solute-type profile resulting from the accumulation of thousands of years of atmospheric Cl^- deposition (*13*). A recently developed model (*14*) (supporting online material) quantitatively explains these Cl^- profiles by considering geothermally driven water vapor transport toward the atmosphere, together with the hydraulic sink created in the soil by the roots of desert plants. Physical and biological processes selectively remove water, concentrating Cl^- (Fig. 1A).

Surprisingly, soil-water concentration profiles of NO_3^- N in five arid-to-semiarid sites in the western United States (Fig. 2) (*15*) follow the conservative solute-accumulation profiles of Cl^- (Fig. 3) rather than the expected progressive nutrient depletion profiles. Maximum NO_3^- N

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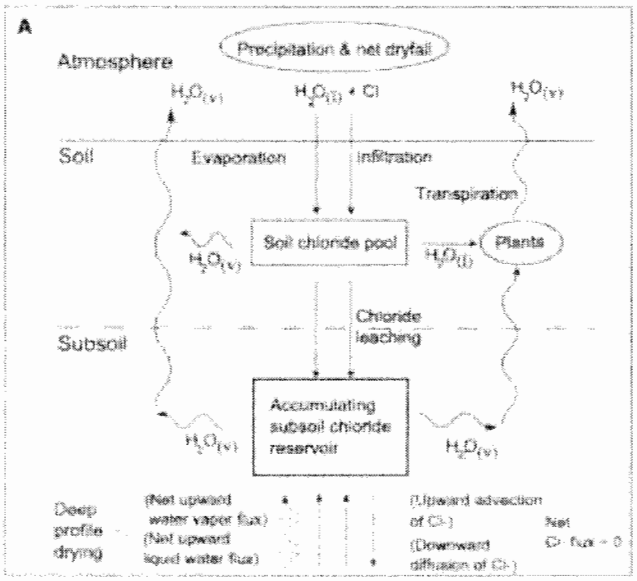

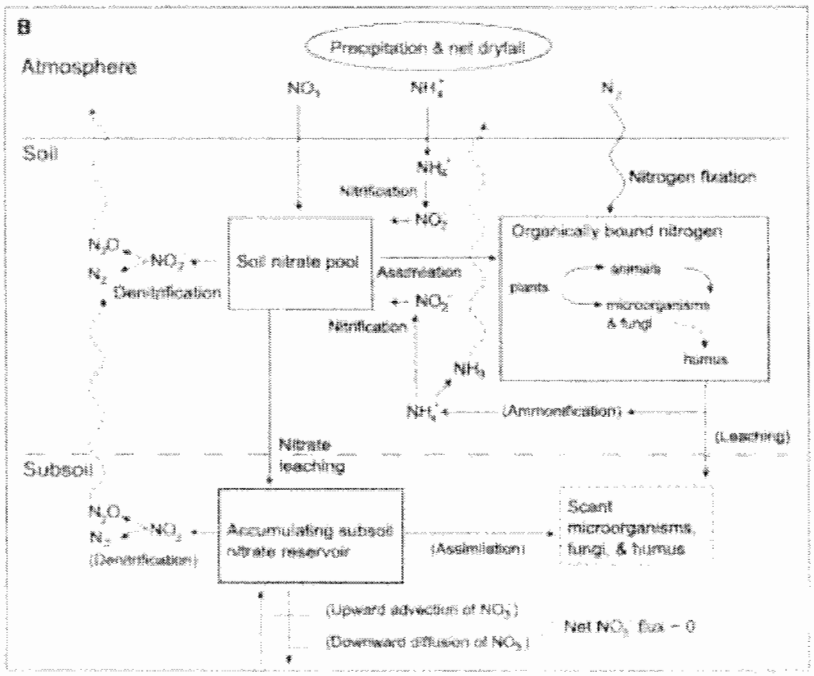

Fig. 1. Main transformations and transport pathways for water, Cl^-, and NO_3^- in desert systems. Straight lines denote liquid (l) pathways; wavy lines denote vapor (v) pathways; dotted lines denote minor pathways. (**A**) Water and Cl^- pathways. Cl^- arrives at the land surface in dust and precipitation, accumulating near the land surface (soil Cl^- pool) during normal conditions of limited rainfall, separated by prolonged droughts. Conservative Cl^- anions are completely excluded from soil vapor and preferentially excluded by cell membranes from plant transpiration. Soil Cl^- leaches to the subsoil reservoir during infrequent major wetting events. Water returns to the soil zone and atmosphere as vapor, leaving nonvolatile Cl^- behind. Small net fluxes of water and Cl^- beneath the upper subsoil are directed upward, reflecting deep-profile drying under current climatic conditions. (**B**) NO_3^- pathways. NO_3^- acts like Cl^- with respect to leaching and exclusion from soil vapor. Unlike Cl, however, NO_3^- is preferentially taken up by plants and is reactive. Assimilation, nitrification, denitrification, and ammonification are all biologically mediated. Subsoils, beneath the root zone, are virtually devoid of organic matter and active organisms, where leaching and evaporative concentration are the main processes affecting NO_3^-. Soil-pool flushing after extended dry periods, when limited bioavailable carbon reserves are exhausted, leaches accumulated NO_3^- to the subsoil below the reach of plants.

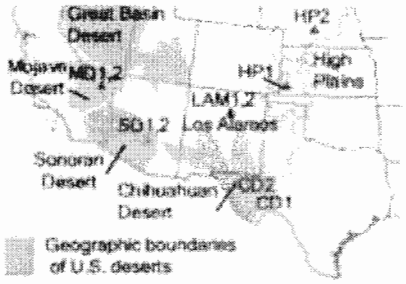

Fig. 2. Map showing locations of vadose-zone pore-water concentration profiles used in this analysis.

concentrations in the subsoil below these nutrient-limited vegetation communities (*10*) can exceed 2000 mg liter^{-1}, surpassing N concentrations applied in hydroponic agriculture by a factor of 10. Clearly, not all NO_3^- N is consumed in the soil zone. We infer that NO_3^-, like Cl^-, leaches from the soil pool to the subsoil reservoir, just beyond the reach of roots, during occasional deep-wetting events. Once there, NO_3^- concentrates as water moves upward as vapor along energy gradients and ultimately returns to the atmosphere via plants (Fig. 1B). The sustained absence of downward water movement below the subsoil reservoir has enabled NO_3^- to accumulate for thousands of years (*13*, *14*). Desert subsoils are persistently low in organic matter, low in microbial populations, low in water content, aerobic, and neutral to basic in pH (*16*), all of which promote NO_3^- stability and inhibit denitrification (*17*).

Integration of the NO_3^- N profiles from 1 m to the maximum depth sampled yields subsoil NO_3^- N inventories that vary from 30 to 13,600 kg of N ha^{-1} (table S2). The NO_3^- N inventories show high intra- and interregional variability relative to Cl^- inventories. This is not surprising, as N gains and losses within the soil zone are controlled by unevenly distributed plant and microbial activity, in addition to hydraulic controls (Fig. 1B). Despite the large variability, general trends are apparent. For example, the pinyon-juniper woodland in semiarid northern New Mexico (Los Alamos) has the lowest NO_3^- N inventory, suggesting a lower limit for environmental conditions under which subsoil NO_3^- N accumulates in appreciable quantities. A nearby ponderosa pine woodland that receives moderately more rainfall shows little to no subsoil NO_3^- N accumulation (*18*).

One key factor contributing to contrasting NO_3^- behavior in arid and humid soils is the establishment of a persistent hydraulic sink at the base of the soil zone in deserts. Cl^- mass balance calculations provide an estimate of the time scales over which conditions required for solute accumulation have been maintained (*13*, *14*) (supporting online text). Estimated accumulation times for the desert sites range from 10,000 to

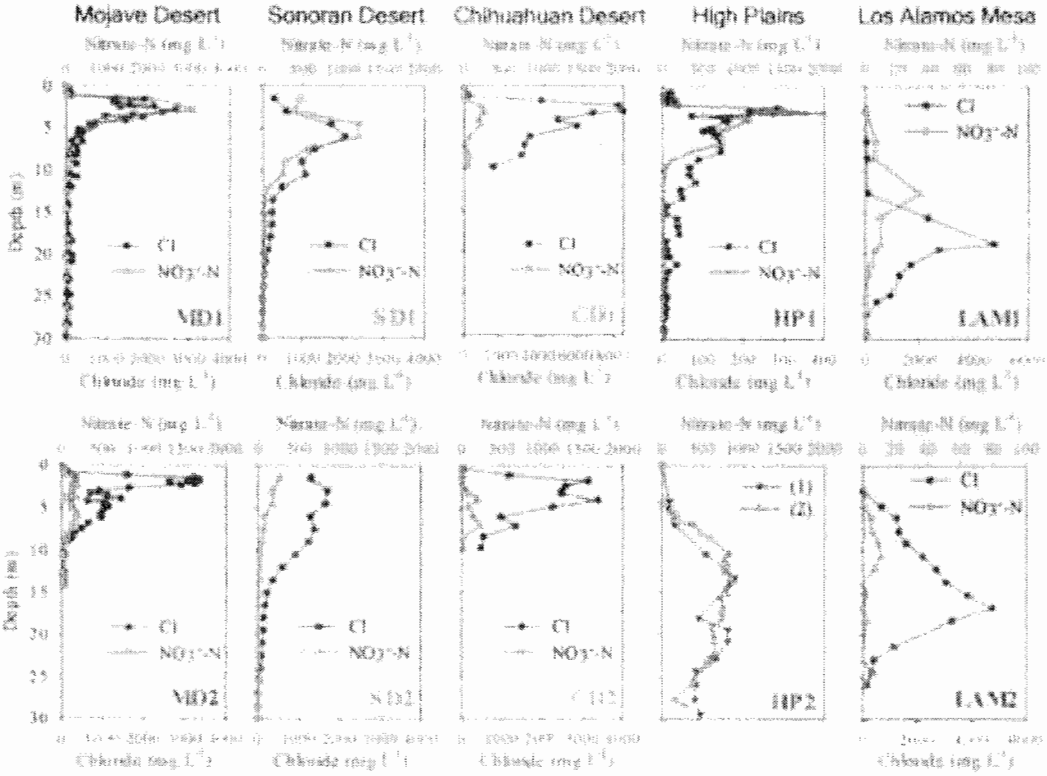

Fig. 3. Vadose-zone nitrate N and chloride pore-water concentration profiles from locations indicated in Fig. 1. (No chloride data are available for HP2.) Note the change of the Cl (**bottom**) scale for CD1, HP1, LAM1, and LAM 2 and the change of the NO_3^- N (**upper**) scale for MD1, LAM1, and LAM2. Data sources are as follows: MD1 (*14*), MD2 (*29*), SD1 and SD2 (*30*), CD1 and CD2 (*31*), HP1 (*32*), HP2 (*27*), and LAM1 and LAM2 (*33*).

16,000 years (table S2). These accumulation times are consistent with the hypothesis that the onset of and Holocene climatic conditions and succession to xeric vegetation (*19*) triggered subsoil solute retention (*13, 14, 17*). Although the presence of biochemical pathways makes NO_3^- transport much more complex than Cl^- transport (Fig. 1), the strong similarity of the two profiles at many sites (Fig. 3) suggests that subsoil input histories, transport behavior, and accumulation times are similar. In the sampled environments, inferred subsoil NO_3^- N retention times exceed the 3000-year soil organic N pool maximum retention time (*9*) by as much as a factor of 5.

Comparisons of subsoil inventories to soil inventories for the sampled regions (*7, 16, 12*) as well as for arid-to-semiarid soil inventories worldwide (*9*) indicate that subsoil N (as NO_3^-) inventories are similar in magnitude to total soil N inventories (Fig. 4 and table S2). Based on these comparisons, subsoil NO_3^- N likely accounts for a preponderance of total vadose-zone N (ground surface to water table) in nonriparian arid environments. The ratios of subsoil NO_3^- N to total vadose zone N are 44 to >92% for the Mojave Desert, 41 to 81% for the Sonoran Desert, and 41 to 62% for the High Plains region. Subsoil NO_3^- N accounts for >4 to 20% of the total vadose zone N in the Chihuahuan Desert and <4% in dry forests similar to the Los Alamos sites. Assuming that comparable inventories (1 to 5 kg ha^{-1}) exist in the 1.5×10^9 ha of Earth's warm deserts and arid shrublands, subsoil NO_3^- N inventories contain approximately 3 to 15 Pg of bioavailable N. This compares to global total estimates of 21 Pg in desert soils and 95 Pg in all soils (*9*). Consideration of subsoil NO_3^- N thus raises estimated global inventories of vadose-zone N by 14 to 71% for desert regions and 5 to 16% overall. Reducing uncertainty in these estimates will require substantial sampling efforts, given the variability among measured profiles. Nevertheless, the large amounts of subsoil NO_3^- N indicated by all of the sites warrants consideration in assessments of global as well as arid-land N distributions.

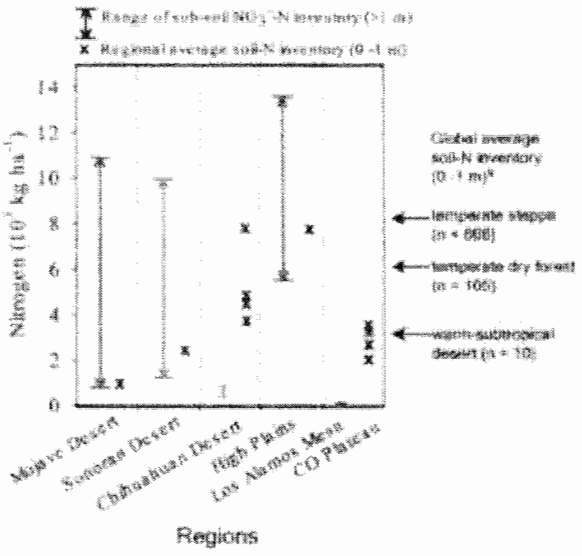

Fig. 4. Comparison of subsoil NO_3^- N inventories (table S2) with average regional and selected global soil N inventories. Multiple soil N regional averages correspond to average measurements collected under different species. Sources for soil N data are as follows: Mojave and Sonoran Deserts (*10*), Chihuahuan Desert (*34*), High Plains (*35*), and CO Plateau (*7, 36*). Los Alamos data are compared with CO Plateau data based on similar vegetation, proximity, and lack of local soil N data.

The indicated leaching of soil NO_3^- N to the subsoil reservoir affects long-term N cycling calculations that derive fluxes from residuals. For example, estimates of N lost to the atmosphere by volatilization and denitrification will

be inflated by the amount of NO_3^- N leached to the subsoil; the latter is substantial at some of the sites investigated. Averaged over Cl^--based accumulation times, long-term NO_3^- N soil losses via leaching to the subsoil reservoir range from 3×10^{-3} to 6.8×10^0 kg of N ha^{-1} $year^{-1}$ (table S2). For comparison, mean annual inorganic N in wet deposition (NO_3^- N plus NH_4^+-N) ranges from 0.8 to 4 kg of N ha^{-1} $year^{-1}$ in the western half of the United States (*20*). Our data do not permit precise generalization of NO_3^- N soil leaching to subsoil reservoirs. Even so, NO_3^- N soil leaching clearly constitutes an appreciable fraction of atmospheric N deposition over large areas.

Leaching of N from arid soil zones is unexpected, given the N-limited nature of desert ecosystems and the high nutrient utilization efficiency of xeric plants (*10*, *21*, *22*), and cannot be readily explained. The presence of large quantities of NO_3^- N sequestered below a depth of 1 m demonstrates that not all of the available NO_3^- N is consumed in the soil zone or returned to the atmosphere. Ecologic implications follow, given the strong linkages between nutrient cycling and plant community dynamics. Recent studies show that desert plants do not necessarily take up water and nutrients simultaneously (*23*). In addition, some species may rely solely on available N at the soil surface (*24*). Such behaviors may help explain the apparent paradox of NO_3^- N leaching from soils populated by N-limited vegetation.

Subsoil NO_3^- reservoirs also have implications for groundwater quality, as their mobilization may adversely affect public water supplies. Drinking water exceeding the maximum contaminant level established by the U.S. Environmental Protection Agency of 10 mg of NO_3^- N $liter^{-1}$ is associated with methaemoglobinaemia, miscarriages, and non-Hodgkin's lymphoma (*3*, *25*). Investigations in the 1970s reported large amounts of subsoil NO_3^- in southern California (*26*) and central Nebraska (*27*) that could not be attributed to agriculture or other human activities. Similarly, investigations of high NO_3^- levels in Las Vegas Valley groundwater near irrigated fields ruled out fertilizer, livestock, and septic systems as sources of pollution (*28*). Recent studies indicate that subsoil NO_3^- reservoirs are readily mobilized to groundwater when desert land is converted to irrigation (*29*) (fig. S3). Dam construction or changes in climate and vegetation could likewise mobilize subsoil nitrate reservoirs, with local to regional effects.

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African Droughts and Dust Transport to the Caribbean: Climate Change Implications

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Great quantities of African dust are carried over large areas of the Atlantic and to the Caribbean during much of the year. Measurements made from 1965 to 1998 in Barbados trade winds show large interannual changes that are highly anticorrelated with rainfall in the Soudano-Sahel, a region that has suffered varying degrees of drought since 1970. Regression estimates based on long-term rainfall data suggest that dust concentrations were sharply lower during much of the 20th century before 1970, when rainfall was more normal. Because of the great sensitivity of dust emissions to climate, future changes in climate could result in large changes in emissions from African and other arid regions that, in turn, could lead to impacts on climate over large areas.

Aerosols, including mineral dust, can affect climate directly by scattering and absorbing solar radiation and indirectly by modifying cloud physical and radiative properties and precipitation processes (*1*). Over large areas of the Earth, the atmospheric aerosol composition is dominated by mineral dust. Dust storms and dust plumes are the most prominent, persistent, and widespread aerosol features visible in satellite images (*2*). Dense dust hazes often cover huge areas of the Atlantic, Pacific, and Indian oceans down-