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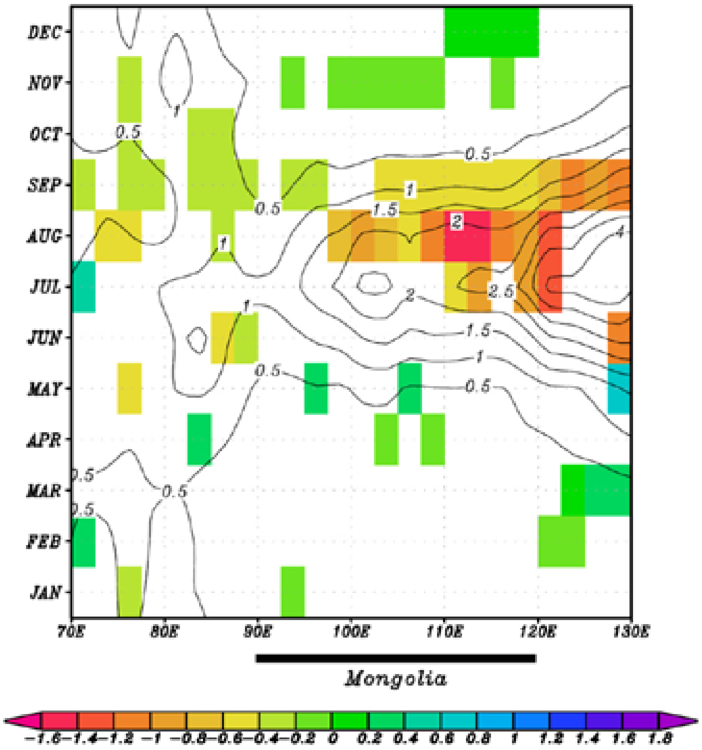

Fig.1: Climatology of monthly precipitation along 45-50N (solid lines) for Mongolian longitudes (70E to 130E) and trends during 1979 to 2008. Positive (negative) values indicate increasing (decreasing) trends.

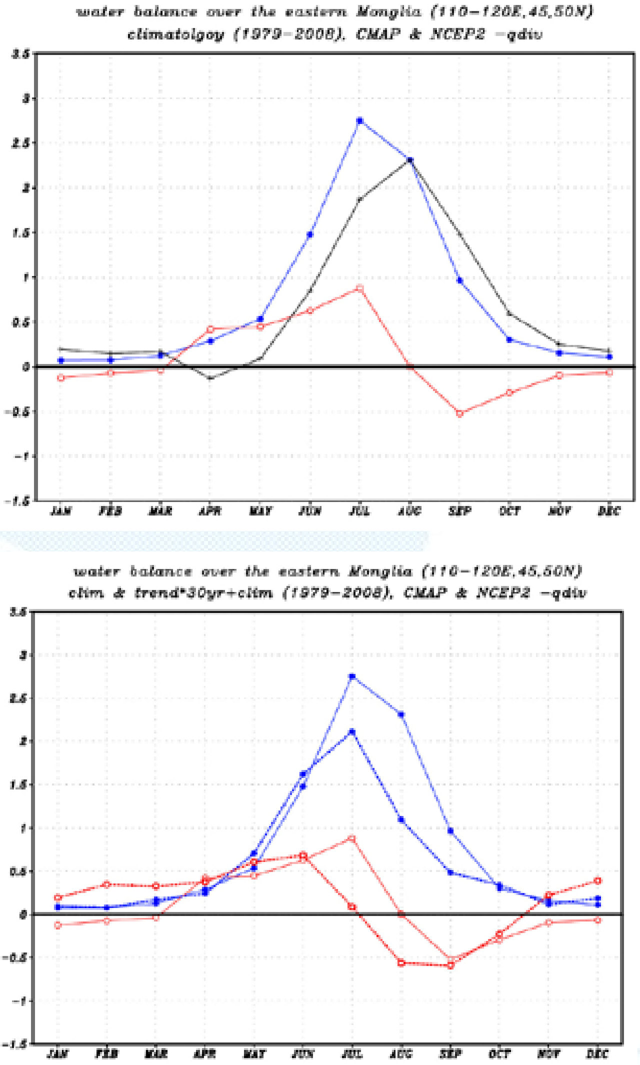

Fig 2: (left) monthly climatology (1979-2008) of atmospheric water balance over northeast Mongolia (110-120E, 45-50N) . precipitation (blue), moisture convergence (red) and evapo-transpiration (black). (right) same as the left figure but for the recent years (dashed) by adding the linear trend values for the 30 years.

# The Role of Moisture and Pollutants in Rock Weathering

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## Abstract

*All the geomaterials exposed as natural outcrops at the Earth's surface are subjected to the destructional physical, chemical, and biological weathering. The moisture absorption and pollutant uptake are the most important factors triggering the rock weathering. Moisture in rocks is of greatest importance, both as a dissolution and disruptive agent and as a vehicle of transport for pollutant salts. Air pollutants, such as* $SO_2$ *and* $NO_x$*, induce physical and chemical weathering. Chemical weathering is resulted from the reaction of mineral constituents of the rock with water, acid rain, and oxygen from air, and it takes place at the sub-microscopic level on the exposed rock surfaces containing pores, fracture surfaces and grain boundaries where chemical reactions including dissolution and precipitation can occur. Physical weathering is caused by the differential stress which have resulted from salt crystallization, swelling of clays, freeze-thaw process and wet-dry cycle. As results of these processes, the rock undergoes a progressive fragmentation along anisotropic surfaces, such as intra- and inter-crystalline micro-cracks, cleavage planes, twin lamellae, joints, etc. Acids generated from microorganisms also can corrode and attack the minerals in rocks. Rock damage happens in the zone of the maximum moisture content which is necessarily the zone of the salt precipitation. Weathering forms of the surface of rock are variable depending on the depth of maximum moisture content of the rock. Distribution of salt in rock is controlled by the drying speed. It is important and necessary to protect the environment from air pollution so that we can not only live in the clean atmospheric environment, but also mitigate the rate of natural weathering of rocks and land degradation.*

## 1. Introduction

Lands on the earth are degraded by the slowly occurring weathering and erosion as well as the abruptly occurring natural disasters, such as flood, earthquake, and desertification. Natural weathering of rocks progresses near or at the ground surface, influenced by various weathering agents: atmosphere, rainwater, ground moisture, stream water, lake water and sea water. For the land to be eroded, rocks must disintegrate into particles fine enough to be moved by water or wind. In order to reduce the disasters due to land degradation, we have to fully understand the nature of rock weathering and its mechanisms.

Weathering is the thermodynamic readjustment of rocks to the environmental conditions of the Earth’s surface where meteoric water and atmospheric gases prevail. Minerals are oxidized by the presence of oxygen, and hydrated or dissolved by the presence of moisture by chemical weathering, and decayed by physical weathering. The chemical weathering on the other hand results in the precipitation of new weathering minerals that are in equilibrium with the new environmental conditions, whereas the physical environmental changes lead to the generation of strain to rocks, resulting in the destruction of rocks.

The atmosphere of the twentieth and this centuries are creating special environmental problems to exposed rock surfaces because of its contamination with pollutants. Industrial pollutants are becoming the most damaging factors of rock weathering. The chemical weathering process is aided by mechanical disruption by pollutant salts which leads to the rapid increment of the mineral surfaces, prevailing accessibility to oxygen and moisture and so resulting in accelerated weathering. Weathering increase strongly with time. Different rock types show different weathering rates. Isotropic siliceous rocks such as granite will be more resistant than anisotropic rocks such as sandstone and limestone (Loefvendahl et al., 2000). The aim of the present paper is to review on the importance of the moisture and pollutants both of which are the most damaging factors in the rock weathering.

## 2. Principal Factors Triggering the Rock Weathering

The principal factors triggering the rock weathering are moisture absorption and pollutant uptake. The important sources of moisture reacting with rocks are from rain, stream water, lake water, sea water, ground water and atmospheric water. Water absorbed onto rocks are either liquid water or water vapor. Even if rain falls for a short span of time, the surface of permeable rocks transmit moisture easily and allow it to penetrate deeper into the rock.

The moisture absorption into rocks takes place in warm humid air when moisture comes into contact with cooler rocks. Moisture is adsorbed readily to capillary walls in an oriented manner by electrostatic attraction in capillaries 0.1 μm or smaller. The positively charged $H^+$ ions of the water molecules are attracted to the negatively charged pore walls. The water molecules adhere tightly to the capillary walls with two or possibly three layers on each side of the capillary wall (Winkler, 1994). Such ordered water is believed to be unfreezable to near -40°C (Dunn and Hudec, 1966). Pore diameter ranges of 20-6 μm (0,02-0.006 mm) should enable a maximum vertical capillary rise of 3-10 m; a diameter range of 6-2 um (0.006-0.002 mm) permits 10-30 m rise (Kieslinger, 1957), Water travels twice as far in a horizontal direction as in a vertical direction. Moisture in rocks are of the greatest importance, both as a vehicle of transport for salts and as a disruptive agent.

Pollutant uptake of rocks is important for both the physical and chemical and weathering of rocks. The Earth's atmosphere has a basic composition of 78% nitrogen, 21% oxygen and 1% $CO_2$ by volume, There are also traces of argon plus a number of impurities (pollutants), such as $SO_2$ and $NO_x$, and organic oxidants. These are introduced into the atmosphere from automotive and industrial sources. These eventually settle out on the ground and on the rocks surfaces as aerosols (dry fallout). Aerosol with sizes ranging from molecules to rain drops usually settle with rain and react with the rock substance in aqueous solution (Winkler, 1994), resulting in the formation of deleterious soluble salts. Important soluble salts that easily decay the rocks are sulfates, nitrates, and chloride of Ca, Na, Mg which are mainly derived from dissolution of rocks by acidic rain water.

Groundwater is an ample source of salts, mostly as chlorides, sulfates, and nitrates of Na, Mg, Ca, P and occasionally rare earth elements (Winkler, 1994). Rising ground moisture is the vehicle of salt transport upward to the surface of rocks. A considerable amounts of salts are supplied from the ocean to continents by sea spray. NaCl, the most common salt in the oceans, may be carried as fine spray up to about 300 miles inland, settling on the ground. Sodium rarely exceeds 0.7% of the total rock substance. Pollutant salts derived from ground moisture, stone weathering or polluted air travel readily in rocks both by diffusion and capillarity. Salts may interact variously with humidity and moisture of materials and the air.

## 3. Role of Moisture in Rock Weathering

### 3.1. Silicate mineral-water reaction

Moisture plays the main role in chemical weathering. The main process of chemical weathering is dissolution that is the dissociation of rock materials in a solvent, such as water. The aluminum silicate and carbonate rocks are primarily dissolved in water acidulated by dissolved $CO_2$, $SO_2$ and $NO_x$. $SO_2$ is combustion by-product of coal, oil and natural gas. Burning coal (power generation) contributes most to the formation of these acidic pollutants (approx. 55%), combustion of oil and the smelting of sulfate ores with approx. 20% and 13%, respectively. A highly corrosive sulfuric acid ($H_2SO_4$) is formed by dissolution of $SO_2$ in water. The rain water is naturally acidic (pH 5.7) because of the presence of carbon dioxide in the atmosphere. It becomes more acidic (pH 4.0-6.0) because of natural emission of sulphur dioxide and other species, or likewise by sulfur or nitrogen oxides ordinarily supplied by fumes, bacterial action, or other sources.

The weathering of silicate minerals is chiefly a hydrolysis reaction, but it also includes dissolution, hydration, oxidation, and carbonation. Fig. 1 shows the variation of pH and ion release from orthoclase- (Fig. 1a) and biotite-water (Fig. 1b) interactions at initial pH 5.7. pH of solution increased very rapidly in a few minutes in the beginning and then slowly toward the final pH. It suggests that these minerals are unstable in alkaline solution, thus leading to dissolution (Lee and Kim, 1994). Over the whole range of pH, $Si^{4+}$ is more soluble than $Al^{3+}$ or $Fe^{3+}$ (Lee, 1996).

Orthoclase has silanol, aluminol and potassium ions in solution. The initial rapid uprise of pH may be due to the rapid uptake of protons by the formation of aluminol and silanol plus the exchange of cations with protons and the gradual down of pH is due to the increase of proton and decrease of OH in the reaction. Biotite shows maximum pH 9.5-10.5 and final pH 8-9 when the initial pH is 5.7. It is characteristic of biotite that pH decreases slowly from maximum pH compared with orthoclase. This may be ascribed to the negative structural charge of biotite which accommodate more protons in relatively high pH solution and the presence of brucite-like sheets which have high isoelectric point (Lee and Kim, 1994).

Swelling (expansion) of clay is due to the expansion of basal spacings by hydration of exchangeable cations in the interlayer of its crystal structure. Water molecules are electrically attracted to this ions, so they penetrate into interlayer and surround the alkali ions, resulting in expansion (Scherrer and Gonzalez, 2005). The basal spacings varies depending on the vapor pressure. Smectite with layer charge 0.2-0.9 has basal spacing 10 Å for anhydrous state, 15 Å for humid state and 17 Å for wet state.

It is a well known fact that all porous systems exhibit both hygric (in the range between 0-95% RH) and hydric (immersion into water) dilatation (expansion) and contraction as a result of moisture changes (Snethlage and Wendler, 1997). Rock damage due to repeated and/or rapid wetting and drying is attributed to differential strain from expansion of wet region of the rock. For a temperature range of 30°C, granite expands 0.15% by heating-cooling, while water expands 1.5% uncompressed. The expansion of granite by moisture ranged from 0.0004 to 0.009%, with an average of 0.0039%; for quartz sandstone it was 0.01-0.044% (Kessler and Hockman (1950).

### 3.2. Examples of chemical weathering

If the aluminum silicate rock is reacted with acidic water (rain, dew, moisture) in nature, firstly $H^+$ ions will displace the alkali and alkali earth cations, forming a thin film of possibly “acid feldspar” (Garrels and Howard, 1959; Keller et al., 1963), and alkaline fluid (pH 7.5-9.8). $H_4SiO_4$ produced from dissolution of aluminum silicate minerals will form colloidal or amorphous $SiO_2$ or crystalline minute quartz. $Al_2O_3$ in fluid will form bauxite minerals when silica is intensively leached out. It forms kaolinite or halloysite when leaching of silica is not severe. In the presence of $K^+$ and $Mg^{2+}$, silica and alumina will form illite or smectite. $Fe^{2+}$ resulted from dissolution of iron-bearing mineral such as biotite will combine with $H_2O$ and $O_2$ to form iron oxide (hematite or hydroxide (goethite). $Mn^{2+}$ resulted from dissolution of manganese-bearing mineral will react with $H_2O$ and oxygen to form manganese oxide (pyrolusite) or hydroxide (manganite). The iron and manganese oxides occur frequently as surface crusts of weathered rocks. It is significant to understand that numerous recent studies suggest that organic acids and organisms may play role in chemical weathering (Huang and Keller, 1972). Excretion of organic acids by microorganisms and lower plants such as lichen, algae and fungi contribute to the chemical decomposition of minerals (Valsami-Jones and McEldowney, 2000).

Extremely vulnerable rock to acid rain (commonly 5 to 5.5, although it may range from below 4) is limestone which is essentially composed of calcium carbonate. Carbonate minerals such as magnesite ($MgCO_3$) and siderite ($FeCO_3$) are also attacked by the acid rain. If the calcium carbonate is attacked by $H_2SO_4$, gypsum ($CaSO_4 . 2H_2O$) is formed.

Presence of large amounts of gypsum is considered to be an indicator of acid rain attack.

If the rain water is in contact with solid $CaCO_3$, it quickly rises to about 8 to 8.2, as observed in pools of water standing on bare limestone. In the typical range of pH 7 to 9 of waters draining limestone, the dominant anion associated with dissolved $Ca^{2+}$and $Mg^{2+}$ is $HCO_3^-$. Thus, $CaCO_3$ is dissolved predominantly as $Ca^{2+}$, with $HCO_3^-$, and less as $Ca^{2+}$ with $CO_3^-$ (Keller, 1978).

Dissolution of minerals along the fracture surface, grain boundaries and rock surfaces results in the generation of new pore structure having higher porosity compared to the original rock. But the precipitation of new minerals along or at the newly formed pores result in the formation of new textures of different configuration depending on the degree of weathering.

Saprolite is a typical product of moderate degree of chemical weathering of rocks. The large or small fissures in the original rock mass are often filled with clay minerals admixed with iron hydroxide minerals. Manganese and iron hydroxide crusts are occasionally found on the surfaces of weathered rocks. Megascopic boxwork pattern is the result of migration of dissolved mineral matter, calcite, silica and iron to the surface, often along a system of narrow spaced intersecting joints and fissures. Microscopic boxwork fabric of halloysite (Fig. 2) (Jeong and Kim, 1993) in the kaolin deposits of Hadong-Sancheong, Korea, is the result of precipitation of halloysite from dissolved anorthite matter along more or less widened cleavages and fissures and then complete dissolution of remaining anorthite grains. Fig. 2 shows that exactly what are taking place during weathering process. It is important to remember that the original minerals of anorthosite has been completely dissolved by chemical weathering, but the secondary minerals and fabric have been newly constructed by chemical weathering.

## 4. Role of Pollutants in Rock Weathering

### 4.1. Pollutants and Physical Weathering

The disruptive force of rocks in the weathering environment is primarily caused by the differential stress that has generated from 1) the expansion by wetting, 2) the swelling of clays, 3) the pressure from crystallization of ice and pollutant salts, and 4) heating-cooling process. Only a small strain is required to produce stresses exceeding the tensile strength of rocks, so differential swelling resulting from superficial wetting or drying of a rock surface can cause deterioration even for a rock whose swelling is on the order of 0.1% (Scherrer and Gonzalez, 2005). Although some clay exhibits enormous expansions, the greatest stress is exerted at small strains; the pressure required to suppress the expansion of clay typically drops exponentially as the volume is increased (Macey, 1942; McEwan and Wilson, 1980).

Pollutant salts have been known to damage porous materials through the production of physical stress resulting from the crystallization of salts in pores. Salt crystallization occurs by evaporation of solution between the states of saturation and supersaturation or by chemical reaction. Pressure developed by salt crystallization exerts high pressure of varying degree within small capillaries. Calcium dissolved from carbonate rocks tends to combine with $SO_2$ in pollutant contaminated air to form gypsum. White efflorescence of soluble sulfates or chloride on the surface of rocks are the crystallization products from salt solution.

The rocks having water in their pores are likely to susceptible to damage by pressure from crystallization of ice. Salts have been known to damage porous materials through the production of physical stress resulting from its crystallization in pores. The ranking of pressures developed by crystallization of salts: halite (NaCl) > nitratine ($NaNO_3$) > niter ($KNO_3$) > thenardite ($Na_2SO_4$) > gypsum ($CaSO_4 \cdot 2H_2O$) > trona ($Na_3H(CO_3)_2 \cdot 2H_2O$) > aphthitalite ($K_3Na(SO_4)_2$) > hexahydrite ($MgSO_4 \cdot 6H2O$) > epsomite ($MgSO_4 \cdot 7H_2O$) > mirabilite ($Na_2SO_4 \cdot 10H_2O$) (Alves et al., 2000). Important soluble salts that are found in rocks are gypsum, mirabilite, epsomite and halite. Halite can exert a maximum pressure of 605 atm at $25^{o}$C. This pressure is probably the largest occurring in nature under confining capillary conditions; the stress is strong enough to disrupt most rocks.

In general, salts of low solubility tend to crystallize at an early stage near the ground surface, like most sulfates; salts of high solubility rise high and often remain in solution, forming wet zones (Winkler, 1994). Less soluble salts, namely gypsum, will tend to crystallize inside the rocks (Alves and Sequeira Braga, 2000). Disruption of rocks may also take place by reason of the considerable contrasts in thermal expansion of entrapped pollutant salts in the pores of rocks.

Although hydration from thenardite ($Na_2SO_4$) to mirabilite ($Na_2SO_4 \cdot 10H_2O$) has been known to result in a large volume expansion (about 314%) (Winkler, 1994), recent studies using environmental electron microscope demonstrated that the transition from thenardite to mirabilite, although it looks like a hydration process, occurs by dissolution and precipitation, ruling out hydration pressure as a credible cause for the extensive damage caused by sodium sulfate (Rodriguez-Navarro and Doehne, 1999; Tsui et al., 2003).

Microorganisms and lower plants such as lichen contribute to the destruction of rocks through biophysical processes such as stresses produced by growing, wetting and drying (Lee, 2000).

### 4.2. Examples of physical weathering

Large cracks are often found on the weathered rock outcrops. If the rocks are under control of differential stress during weathering, they are likely to disrupt along the discontinuity planes, such as joint, cleavage, intra- and inter-crystalline micro-cracks, and twin lamellae. The rapid relief of stress results in the generation of micro-cracking in rocks. The joints parallel to the exposed surface of granite mass are derived from the relief of the overlying loading by weathering and erosion. Especially the large or small superficial shallow cracks in rocks indicate separation of the surface layer from the underlying rock. If the surface layer is detached, it is scale or flake. Such phenomena suggest that the surface zone has been subjected to repeated wetting-drying, repeated heating-cooling or salt damage, resulting in the generation of differential stress relative to the interior rock. The 1-3 mm thick scales in granites may be due to the salt crystallization or expansion-contraction cycle by ground moisture entrapped in the pores of the granite. Wetting- drying as well as freezing-thawing cause displacement of grains relative to each other and contribute to loosening of the grain structure (Snethlage and Wendler, 1997). Surface and subsurface rocks are mainly weathered and degraded by wetting-drying and diurnal heating-cooling cycles.

The weathering forms resulted from the differential stresses which exerted to the rock during past or on-going weathering process are: fissure, scale, flake, crumbling, sanding, break out, etc.

## 5. Behaviors of Moisture and Pollutant Salts in the Changing Environment

Moisture and pollutant salts, if both are present together, impact together in rocks. Behaviors of moisture and salts in rocks are determined depending on its saturation degree in the pore and atmospheric environment. It, therefore, is important to know the mechanical contribution of moisture and pollutant salts to the generation of stress leading to disruption of porous rocks.

Freezing of water causes expansion of its volume. Thus, in the rocks with full moisture saturation, the freezing causes an expansion of grain structure of rocks that is not reversible after thawing, due to the volume expansion of ice against liquid water. In the rocks with partial moisture saturation, however, the freezing causes a contraction of the pore structure (Snethlage and Wendler, 1997).

The expanding moisture in pore may develop as much as 7500 psi when heated from near freezing to 60°C. Observed spalling and surface flaking should be merely ascribed to the expansion action of pure water (Snethlage and Wendler, 1997). Interstitial water upon freezing expands about one tenth and exerts a powerful disruptive force up to about 2,100 atm at -22°C on rock (Winkler, 1973).

The most serious damage to rocks is normally associated with high salt concentration. The salt-contaminated rock contracts during wetting and expands during drying (Snethlage and Wendler, 1997). During drying, salts migrate to the rest of pore liquid and precipite there. Formation of gypsum enhances the decay of rocks and causes the detachment of the scales as a result of its crystallization pressure (Snethlage and Wendler, 1997). They show that expansion and contraction under the influence of moisture and dissolved ionic species results in the displacement of grains to each other. Swelling results by the interlayer hydration of expanding clay minerals and by hydration of common rock-forming minerals, such as iron oxides and anhydrite, and by other minerals taking on water of crystallization. Dissolved salts brought in by rain, rising from soil, or generated within the rock by weathering may, during the crystallization as the water evaporate, destroy the rock through granular disintegration or by spalling of surface shells (Keller, 1978).

Formation of clay minerals during weathering process contributes to the crystallization damage by creating small pores that are partially susceptible to high crystallization pressure of water and salts. Clay-containing rocks are inherently susceptible to damage for crystallization of ice and salts (McGreavy and Smith, 1984; Scherrer, 1999), because of the presence of a fraction of very small pores.

## 6. Moisture Distribution and the Weathering Form

Pollutant salts are usually associated with water due to its hygroscopic nature. Rock damage happens in the zone of the maximum moisture content which is necessarily the zone of the salt precipitation. If this zone is located on the surface, the damage type “sanding off” is observed in wet corner where the drying is low. If it is very close to the surface (e.g. 1 or 2 mm), a thin flake will form. If it is deeper in the rock, a scale of 1 or 2 cm will form (Snethlage and Wendler, 1997). Sun and drying of rocks also influence the distribution of salts. Where drying is rapid, salts are accumulated at or near the wetting front and ultimately resulting in contour scaling (Smith and McGreavy, 1988). Such hardened surface form a hardness profile (Snethlage, 1984). Where drying is slower, salt solution can migrate back toward surface where evaporation and crystallization is thought to favor granular disintegration (Smith et al., 1988).

The interactions of rock materials and weathering factors produce an intriguing array of weathering forms, such as cavern (tafoni), honeycomb, blistering, flaking (<5 mm), scaling (>5 mm), granular disintegration, cracking, fluting, efflorescence, case hardening, iron staining and pedestrial rocks as well as rock debris (Smith, 1994; Mottershead, 2000; Doehne, 2002). Where drying is rapid, salts are accumulated at or near the wetting front. In this case contour scaling is expected. Where drying is slow, salt solution migrate back to the surface. In this case evaporation and crystallization results in granular disintegration.

## 7. Conclusion

1) The principal factors triggering the rock weathering are moisture absorption and air pollutant (SO$_x$ and NO$_{x)}$ uptake. Moisture in rock is of greatest importance, both as a dissolution and disruptive agent and as a vehicle of transport for pollutant salts. Weathering is accelerated by acid rain due to air pollutants.

2) Rock damage happens in the zone of the maximum moisture content which is necessarily the zone of the salt precipitation. Weathering forms depend on the depth of maximum moisture content of the rock. Rock damage by pollutants is due to the differential stress from crystallization of pollutant salts from dissolved solution.

3) In order to protect the land degradation, we need both the deep understanding of the mechanisms of weathering and the pertinent endeavor to preserve our natural environment from air pollution which promotes the physical and chemical weathering of land and rocks.

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