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Atmospheric Mg^{2+} wet deposition within the continental United States and implications for soil inorganic carbon sequestration

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ABSTRACT

Little is known about atmospheric magnesium ion (Mg^{2+}) wet deposition in relation to soil inorganic carbon sequestration. Understanding the conversion of carbon dioxide (CO_2) or organic carbon to a form having a long residence time within the soil (e.g., dolomite, magnesian calcite) will greatly benefit agriculture, industry, and society on a global scale. This preliminary study was conducted to analyze atmospheric Mg^{2+} wet deposition within the continental United States (U.S.) and to rank the twelve major soil orders in terms of average annual atmospheric Mg^{2+} wet deposition. The total average annual Mg^{2+} wet deposition for each soil order was estimated with geographic information systems (GIS) using the following data layers: (1) atmospheric Mg^{2+} wet deposition data layers covering the continental U.S. for a 10-yr period (1994–2003) and (2) a soil order data layer derived from a national soils database. A map of average annual Mg^{2+} wet deposition for 1994–2003 reveals that the highest deposition (0.75–1.41 kg ha^{-1}) occurred in Oregon, Washington, parts of California, and the coastal areas of East Coast states due to magnesium enrichment of atmospheric deposition from sea salt. The Midwestern region of the U.S. received about 0.25–0.75 kg ha^{-1} Mg^{2+} wet deposition annually, which was associated with loess derived soils, occurrence of dust storms and possibly fertilization. The soil orders receiving the highest average annual atmospheric Mg^{2+} wet deposition from 1994 to 2003 were: (1) Mollisols (3.7×10^7 kg), (2) Alfisols (3.6×10^7 kg) and (3) Ultisols (2.8×10^7 kg). In terms of potential soil carbon sequestration, the average annual atmospheric Mg^{2+} wet deposition was equivalent to formation of the following theoretical amounts of dolomite: (1) Mollisols (2.8×10^8 kg of $CaMg(CO_3)_2$), (2) Alfisols (2.7×10^8 kg of $CaMg(CO_3)_2$) and (3) Ultisols (2.1×10^8 kg of $CaMg(CO_3)_2$). The soil orders receiving the lowest average annual atmospheric Mg^{2+} wet deposition were: (1) Andisols (3.3×10^6 kg), (2) Histosols (3.4×10^6 kg) and (3) Vertisols (5.0×10^6 kg). The methods proposed here to estimate soil inorganic carbon sequestration potential from atmospheric wet deposition data can be useful for preliminary carbon accounting on a global scale.

1. Introduction

Soils are a large reservoir of both organic and inorganic carbon, and thus play an important role in the global carbon budget (Schlesinger, 2002). The largest fraction of soil inorganic carbon is present in the form of soil carbonates (Schlesinger, 2002), which account for up to one-third of the total carbon in soils (Ming, 2002). Calcite is the most common carbonate mineral found in soils, although the presence of dolomite, magnesian calcite, magnesite, nahcolite, trona, soda, aragonite and siderite have also been reported for certain soils (Sposito, 1989; Ming, 2002). Carbonates may be formed from the parent soil material (lithogenic) or newly formed as a result of soil processes (pedogenic) (Ming, 2002). Atmogenic soil dolomite precipitation sequesters atmospheric CO_2 and requires Mg^{2+} additions outside of the soil system, for example from precipitation, fertilizer addition, irrigation, etc. (Monger and Gallegos, 2000). Attempts to decrease the atmospheric CO_2 concentration require an understanding of soil inorganic carbon and its role in the sequestration of carbon through pedogenic carbonate mineral formation in soils.

Soils with pedogenic dolomite ($CaMg(CO_3)_2$) occur on a variety of substrate materials, but are most commonly associated with calcium and magnesium rich parent materials resulting in variable quantities of $CaMg(CO_3)_2$ within the soil profile (Capo et al., 2000; Whipkey et al., 2002). Soils developed on calcium

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and magnesium rich substrate typically contain both lithogenic and pedogenic carbonates. Dolomite dissolution or precipitation in soils is dependent on a number of environmental/geological factors, including the atmospheric concentration of CO_2, the concentrations and relative stoichiometric amounts of Mg^{2+} and Ca^{2+}, the alkalinity and pH of the soil solution, and temperature (West and McBride, 2005):

$$CaMg(CO_3)_2(s) + 2H_2O(l) + 2CO_2(g) \leftrightarrow Mg^{2+}(aq) + Ca^{2+}(aq) + 4HCO_3^-(aq) \quad (1)$$

Interactions among the various environmental/geological factors are more apparent by examining the individual component reactions leading to eq. (1) (Sposito, 1989; Stumm and Morgan, 1996):

$$CaMg(CO_3)_2(s) \leftrightarrow Mg^{2+}(aq) + Ca^{2+}(aq) + 2CO_3^{2-}(aq) \quad (2a)$$

$$2CO_3^{2-}(aq) + 2H^+(aq) \leftrightarrow 2HCO_3^-(aq) \quad (2b)$$

$$2H_2CO_3^*(aq) \leftrightarrow 2H^+(aq) + 2HCO_3^-(aq) \quad (2c)$$

$$2CO_2(g) + 2H_2O(l) \leftrightarrow 2H_2CO_3^*(aq) \quad (2d)$$

For example, the reactions in eqs. (2a)–(2d) indicate that the dissolution of dolomite is favored with increasing atmospheric CO_2 concentration or decreasing soil solution pH and/or alkalinity, whereas dolomite precipitation is favored with decreasing atmospheric CO_2 concentrations or increasing pH and/or alkalinity.

The reactions above are crucial in understanding how inorganic carbon can be sequestered in soils through pedogenic dolomite formation. In the case of well-drained soils (i.e. rapid downward water flux), 'new and recycled' magnesium originates from weathering, rain, dust, decomposing plant and root material, soil organic complexes, irrigation and fertilization. Organic matter decomposition in the A horizon below the soil surface and root respiration produce a high $CO_2(g)$ partial pressure, which leads to enhanced CO_2 partitioning into the soil solution and additional HCO_3^- production (Chadwick and Graham, 2000). Mg^{2+} and HCO_3^- are leached with soil water to a depth of decreased $CO_2(g)$ partial pressure and below the zone of major biological activity where it precipitates as dolomite (Chadwick and Graham, 2000). Modern dolomite precipitation is often associated with dissimilatory sulfate-reducing bacteria that remove sulfate, produce alkalinity, and presumably drive dolomite formation, and methanogenic bacteria have also been reported to lead to the formation of dolomite in dilute natural waters (Roberts et al., 2004).

Most pedogenic dolomite in soil forms under the same environmental conditions as the soil itself and therefore its formation is controlled by the six soil-forming factors: climate, parent material, biota, topography, time and land-use (Jenny, 1941; Hoosbeek et al., 1997). Climatic factors such as precipitation and air-borne dust can be significant sources of Mg^{2+}. A recent study of Mg^{2+} cycling in a Mexican tropical dry forest ecosystem (659 mm yr^{-1} in annual rainfall) reported that mean inputs from the atmosphere were 0.80 kg ha^{-1} for Mg^{2+} (Campo et al., 2000). Magnesium wet deposition can be estimated for the area of study, although some of the deposited Mg^{2+} would have been re-distributed by wind and water runoff and/or taken up by plants. Pedogenic carbonates can form on any type of parent material, but the chemical composition of the parent material can favor and accelerate pedogenic carbonate formation (Alonso-Zarza, 2003). For example, formation of pedogenic carbonate has been reported to be faster in parent material enriched in Mg^{2+} (Wright, 1990) and having a smaller grain size (Gile et al., 1966).

Recent studies suggest the presence of a large terrestrial carbon sink, which is linked to climatic variation, nutrient deposition, afforestation, agricultural abandonment and others (Cao et al., 2005). However, little is known about atmospheric nutrient deposition in relation to soil carbon sequestration. While it has been known that pedogenic carbonates form in soils, the role of atmospheric Mg^{2+} deposition in relation to soil inorganic carbon sequestration potential is not well understood. Maps of potential soil inorganic sequestration can increase our understanding of terrestrial ecosystem inorganic carbon exchange and the way it can be manipulated to decrease carbon dioxide emissions (Blasing et al., 2005).

The objectives of this study were to: (1) analyze atmospheric Mg^{2+} wet deposition within the continental U.S. and (2) rank the twelve soil orders in terms of average annual atmospheric Mg^{2+} wet deposition.

2. Data acquisition

Data were acquired from national datasets available from U.S. government agencies. The sources of the data used in this study have been summarized in Table 1.

2.1. Annual trends in atmospheric Mg^{2+} wet deposition (1994–2003)

Annual average Mg^{2+} wet deposition for a maximum of 253 stations in the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) was downloaded from the NADP website (http://nadp.sws.uiuc.edu/) for the period of January 1994 through December 2003 (NADP, 2005). Mg^{2+} wet deposition data were available as annual summaries at each station in kg ha^{-1}. These data were summarized by calculating an average Mg^{2+} kg ha^{-1} using all available stations for each year. The summarized data were then statistically analyzed to determine if temporal depositional trends over the time period existed.

2.2. Atmospheric Mg^{2+} wet deposition (1994–2003)

Annual isopleth maps for atmospheric Mg^{2+} wet deposition were downloaded from the NADP website (http://nadp.sws.uiuc.edu/isopleths/) for the period of January 1994 through December

Table 1. Data sources and descriptions.

Data layer	Description	Source
Mg^{2+} wet deposition (kg)	Isopleth maps in ArcGIS grid format – 2500 m resolution	http://nadp.sws.uiuc.edu/isopleths/
Loess	Gridded 0.1 × 0.1° from maps from the U.S. Geologic Survey	Lineback et al., 1983; Miller et al., 1988; Hollbrook, et al., 1990; Gray et al., 1991; Hallberg et al., 1991; Denne et al., 1993; Whitfield et al., 1993; Swinehart et al., 1994
Precipitation (cm)	Isopleth maps in ArcGISGrid format – 2500 m resolution	http://nadp.sws.uiuc.edu/isopleths/
Soil Order	Derived from STATSGO	USDA/NRCS

2003 (NADP, 2005). The files were available in ArcGIS Grid format and were created using spatial interpolation of annual Mg^{2+} wet deposition at sites of the NADP/NTN. Precipitation samples were collected weekly at each NADP/NTN site using standardized equipment and protocols and analyzed at NADP's Central Analytical Laboratory (NADP, 2005). Precipitation-weighted annual mean Mg^{2+} concentrations were calculated at each NADP/NTN site. The Mg^{2+} wet deposition at each site was calculated by multiplying the precipitation-weighted mean Mg^{2+} concentrations by the total annual precipitation at the site. These data were spatially interpolated using the Inverse Distance Weighting (IDW) algorithm, and only sites meeting NADP's data completeness criteria (http://nadp.sws.uiuc.edu/documentation/completeness.asp) were used in the spatial interpolation.

2.3. Precipitation (1994–2003)

Annual precipitation maps were downloaded from the NADP website (http://nadp.sws.uiuc.edu/isopleths/) for the period from January 1994 to December 2003 (NADP, 2005). These files were also available in ArcGIS Grid format and were created using spatial interpolation of annual precipitation at sites of the NADP/NTN. Precipitation samples were collected weekly at each NADP/NTN site using standardized equipment and protocols and analyzed at NADP's Central Analytical Laboratory (NADP, 2005).

2.4. Soils order map

Soil order information for the country-level analysis was derived from the State Soil Geographic (STATSGO) Data Base (NRCS, 1995). This national soils database has been created at a scale of 1:250 000 and is composed of map units averaging between 2800 and 24 200 hectares in size with a minimum mapping unit of 628 hectares. This level of mapping is designed to be used for broad management uses covering state, regional and multi-state areas (NRCS, 1995) and is an appropriate resolution of data for the present study. Soil component tables, which provide information on the soil types within each mapping unit, were queried to determine the dominant soil order for each mapping unit. This query was used to create a vector map of dominant soil orders for the Continental United States.

2.5. Loess distribution

A generalized loess thickness map of the conterminous U.S. was gridded 0.1 × 0.1° from U.S. Geological Survey (USGS) maps (Lineback et al., 1983; Miller et al., 1988; Hollbrook et al., 1990; Gray et al., 1991; Hallberg et al., 1991; Denne et al., 1993; Whitfield et al., 1993; Swinehart et al., 1994) by Kohfeld and Harrison (2001). This data set contains boundaries for loess deposits downwind of source regions. Loess thicknesses are greatest near their original dust sources (e.g., along rivers, at the southern edge of the Late Wisconsin ice sheet, directly downwind of the Rocky Mountains), and rapidly decrease to the east of these source regions (Kohfeld and Harrison, 2001).

3. Data analyses

Summarized data of annual Mg^{2+} wet deposition for January 1994 through December 2003 showed no distinct increasing or decreasing trend in depositional values (Fig. 1). Statistical

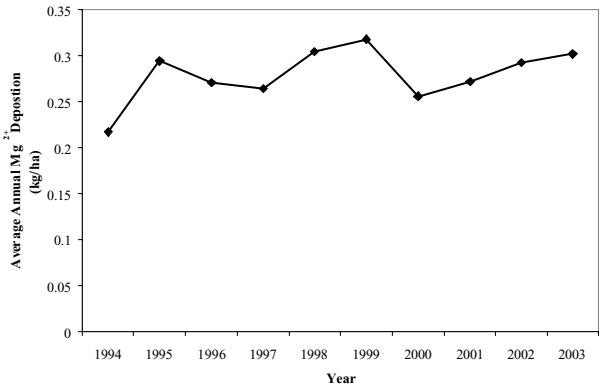

Fig. 1. Mean Mg^{2+} wet deposition temporal trends (1994–2003).

techniques including ANOVA and regression analysis conducted on summarized Mg^{2+} wet deposition data also established that no discernable trends existed.

All datasets were added to a new map project using Environmental Systems Research Institutes ArcGIS v. 9.0 software (ESRI, 2004). A model using ArcGIS Modelbuilder was used to automate the geoprocessing steps used to develop the final GIS layers. A mean Mg^{2+} data layer was created by combining yearly deposition layers using a cell statistics function within ArcGIS. An attribute table for the mean Mg^{2+} data layer, that contained the derived values for each area, was then created using a zonal statistics function.

Spatial data for the period from January 1994 to December 2003 (10 yr) were averaged using the 'Cell Statistics' tool located in Spatial Analyst and using a 'mean' statistical overlay to create a final map layer of average Mg^{2+} wet deposition in kg ha^{-1} (Fig. 2a). This process utilized the average of each cell of the data layer to derive a final data layer. The averaged Mg^{2+} data layer was multiplied by the cell size (625 ha) to derive Mg^{2+} data layer that contained only kilogram. The Cell Statistics tool was also used to create a final map layer of average precipitation for the same time period (Fig. 2b).

Zonal statistics were calculated using the final average Mg^{2+} wet deposition data layer in combination with the generalized soil order data layer to estimate the total Mg^{2+} wet deposition per soil order. Areas of soil orders were calculated within the GIS software as summarized below:

$$\text{Total average annual } Mg^{2+} \text{ wet deposition for each soil order (kg)} = \text{Area of soil order (ha)} \times \text{Average annual } Mg^{2+} \text{ wet deposition (kg ha}^{-1}\text{)} \quad (3)$$

4. Results and discussion

Soil orders having the closest proximity to oceans appear to have the highest Mg^{2+} wet deposition. For example, the map of average annual Mg^{2+} wet deposition for the years 1994–2003 (Fig. 2a) shows that the highest deposition (0.75–1.41 kg ha^{-1}) occurred in Oregon, Washington, parts of California, and the coastal areas of East Coast states. This finding is consistent with the expected enrichment of rainwater by magnesium due to sea salt (Berner and Berner, 1996). The map produced in Fig. 2a of average annual Mg^{2+} wet deposition for 1994–2003 is helpful in identifying those soils having the highest and lowest Mg^{2+} wet depositional loadings and subsequent potential to sequester CO_2 in the form of dolomite or other related magnesium carbonates. It is also an important component in estimating carbon fluxes at the regional and global scales. The NADP/NTN has measured the concentrations of the major chemical species, including Mg^{2+}, in precipitation since 1978 to characterize and evaluate the 'chemical climate' and its changes over time.

Magnesium is an alkaline earth metal and the sixth and ninth most abundant element in crustal rocks and soils, respectively (Sposito, 1989). Furthermore, Mg^{2+} concentrations have increased by 25% from 1985 to 2002 at 38 NADP sites in the southwest U.S., while its concentrations have decreased at 126 sites elsewhere (Lehmann et al., 2005). These findings demonstrate the dynamic nature of magnesium wet deposition and therefore the dynamic nature of potential carbon sequestration through pedogenic dolomite formation in soils. Such temporal variations and trends need to be accounted for in future carbon budgets.

The soil orders receiving the highest average annual atmospheric Mg^{2+} wet deposition from 1994 to 2003 were: (1) Mollisols (3.7×10^7 kg), (2) Alfisols (3.6×10^7 kg) and (3) Ultisols (2.8×10^7 kg) (Table 2). Mollisols and Alfisols are typical for loess dominant regions of the U.S. (Fig. 2a), and much of the Mg^{2+} present in the rainwater over these regions is likely

Table 2. Average annual atmospheric Mg^{2+} wet deposition by soil order within the continental U.S. for the 10-yr period 1994–2003.

Soil orders	Total area of soil order (ha)	Average annual Mg^{2+} wet deposition (kg)
Alfisols	1.3×10^8 (2)[a]	3.6×10^7 (2)
Andisols	5.9×10^6 (10)	3.3×10^6 (10)
Aridisols	7.8×10^7 (5)	8.7×10^6 (7)
Entisols	9.2×10^7 (3)	1.7×10^7 (4)
Gelisols	NS[b]	NS
Histosols	6.8×10^6 (9)	3.4×10^6 (9)
Inceptisols	6.0×10^7 (6)	1.5×10^7 (5)
Mollisols	1.8×10^8 (1)	3.7×10^7 (1)
Oxisols	NS	NS
Spodosols	2.6×10^7 (7)	9.1×10^6 (6)
Vertisols	1.5×10^7 (8)	5.0×10^6 (8)
Ultisols	9.1×10^7 (4)	2.8×10^7 (3)

[a]Numbers in parentheses indicate the relative ranking among soil orders for each column.
[b]NS, not significant.

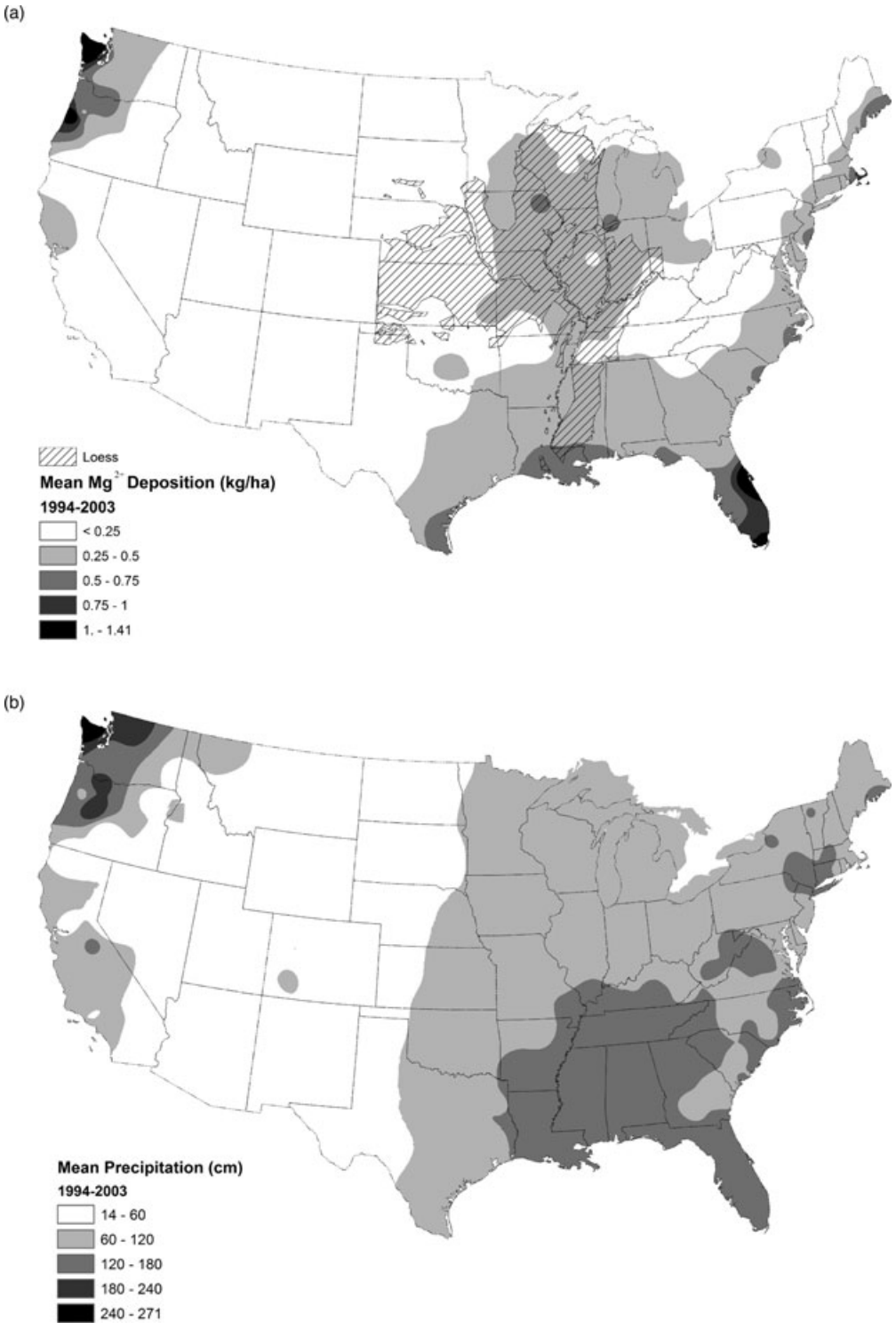

Fig. 2. (a) Mean Mg^{2+} wet deposition (1994–2003) and (b) mean precipitation (1994–2003) (based on data from http://nadpsws.uiuc.edu/isopleths/).

derived from soil dust (Berner and Berner, 1996). In addition, Mollisols and Alfisols are the soil orders typically located in the 'bread-basket' region of the U.S. and thus may receive fertilizers containing Mg^{2+} (West and McBride, 2005). Ultisols receive relatively high amounts of wet Mg^{2+} deposition as a result of the magnesium enrichment of atmospheric deposition from the seawater. The soil orders receiving the lowest average annual atmospheric Mg^{2+} wet deposition from 1994 to 2003 were:

(1) Andisols (3.3×10^6 kg), (2) Histosols (3.4×10^6 kg) and (3) Vertisols (5.0×10^6 kg) (Table 2). Our findings are similar to the study of Armbruster et al. (2002), who observed that throughfall fluxes of Mg^{2+} in 71 forest ecosystems were related to distances to the sea and the amount of particulate matter in the rainwater.

As shown in Table 2, Mollisols receive most of the total Mg^{2+} wet deposition in the continental U.S. In terms of potential soil carbon sequestration, the average annual atmospheric Mg^{2+} wet deposition can form the following theoretical amounts of dolomite: (1) Mollisols (2.8×10^8 kg of $CaMg(CO_3)_2$ equivalent), (2) Alfisols (2.7×10^8 kg of $CaMg(CO_3)_2$ equivalent) and (3) Ultisols (2.1×10^8 kg of $CaMg(CO_3)_2$ equivalent). The soil orders with the lowest soil carbon sequestration potential based on Mg^{2+} wet deposition are: (1) Andisols (2.5×10^7 kg of $CaMg(CO_3)_2$ equivalent), (2) Histosols (2.6×10^7 kg of $CaMg(CO_3)_2$ equivalent) and (3) Vertisols (3.8×10^7 kg of $CaMg(CO_3)_2$ equivalent). Of course, not all of the Mg^{2+} wet deposition on soils will lead to pedogenic dolomite formation, because some fraction of the Mg^{2+} will be utilized by plants while other fractions may remain in the upper soil or be flushed completely out of the soil (West and McBride, 2005).

Traditionally, Aridisols have been identified as soils with major carbonate accumulations. However, pedogenic carbonates in Aridisols often precipitate in the top 1 m of the soil profile, makeing them easy to collect and study. Pedogenic carbonates tend to be readily-soluble in soil solutions, and their depth of formation via precipitation can be a function of rainfall (Royer, 1999). Because the maximum pedogenic carbonate accumulations in Mollisols often occur below 1-m depth, they therefore have not received much research attention in past studies (Mikhailova and Post, 2006).

5. Conclusions

Soil receives continuous Mg^{2+} wet deposition, some of which is used for pedogenic dolomite formation and therefore carbon sequestration. Different soil orders receive various amounts of Mg^{2+} wet deposition and thus will have different potentials to sequester carbon in dolomite or other magnesium carbonate minerals, which are more stable forms than organic matter. Atmospheric wet deposition of Mg^{2+} is dependent on both geomorphological and atmospheric processes, and important sources of Mg^{2+} in rainwater include sea salt and soil dust. It is important to account for potential soil carbon sequestration resulting from atmospheric Mg^{2+} wet deposition in global carbon budgets.

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