

CLAY MINERALS AND THEIR INTERACTIONS WITH HEAVY METALS AND MICROBES OF SOILS IRRIGATED BY VARIOUS WATER RESOURCES AT ASSIUT, EGYPT

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ABSTRACT :

Soil samples were collected from four soil profiles at Assiut city, Egypt, that they have been irrigated by fresh and contaminated (sewage and agricultural drainage) Nile waters as well as artesian water for more than 40 years, in order to study the clay minerals and their interactions with heavy metals and microbes of these soils.

X-ray diffraction analysis of the clay fraction of the different irrigated soil samples revealed that smectites occurred as major clay minerals in all studied soil samples, and tended to increase with depth. Kaolinite was the second abundant clay mineral, followed by mica-smectite mixed layers, vermiculite, micavermiculite mixed layers, mica, sepiolite and palygorskite that are arranged in a decreasing order of abundance. The source of these clay minerals in the studied soils is largely due to the detrital materials from the Ethiopian Plateau that is mixed with the detrital materials from the sandstones and limestones plateaus surrounding the Nile river course.

Concerning the interaction between clay minerals and heavy metals, the significant relations between smectite and Fe or Cu, kaolinite and Pb, mica-smectite mixed layers and Mn, mica-vermiculite mixed layers and Zn, Cu or Ni, vermiculite and Cu, sepiolite and Fe or Pb, and palygorskite and Mn or Cu may be mainly attributed to the adsorption of these heavy metals on these clay minerals.

Regarding the interaction between clay minerals and microbes, the examination of the soil surface samples using the scanning electron microscope (SEM) pointed out that the bacillus and coccus bacteria were observed in all investigated samples, but they were found as colonies in the soils irrigated by sewage contaminated Nile water. Fungi were shown in all examined samples, but they were in smaller amounts than bacteria that were probably associated with smectite. Some of fungi types were characterized to specific soils, such as *Triscelophoius monosporous Ingold* in the soil irrigated by artesian water, and *Pilobolus* (Zygomycota) in the soil irrigated by sewage contaminated Nile water. However, actinomycetes appeared in the soil irrigated by fresh Nile water. These reflected the influence of irrigation water source, which led to the diversity of bacteria, fungi and actinomycetes in these different soil samples.

INTRODUCTION:

A knowledge of clay minerals is important to evaluate the magnitude of soil fertility, to provide a clear indication of the role played by weathering processes and sometimes to reduce soil and water pollution (Miller and Donahue, 1992). Clay minerals are among the major materials that interact with almost all soil contaminants (Prost and Yaron, 2001). Clays often represent a short-term sink of heavy metals in soils, because of their adsorptive properties. Contaminated soils can often contain more than one heavy metal species. It is possible that the behaviour of a particular metal species in a soil system will be affected by the presence of other metals (Srivastava *et al.*, 2004).

Along with microorganisms, clays provide some of the most catalytic surfaces in sedimentary environments, which are important to a variety of biogeochemical cycles (Kostka et al., 2002). Soils are a major habitat for microorganisms. The interaction of clay particles and microbial cells is dependent on the size and charge of exchangeable cations and on electrolyte concentration, just as for other negatively charged colloidal particles (Oades, 1989). Stotzky (1986) stated that surface interactions between clays and microbes in soils occur in situ, e.g. 1) lack of movement of large number of microbes from surface to underlying soil layers during floods and irrigation and 2) partial removal of microbes from waste waters in percolation beds. He added that, in soils with low levels of organic substrates, the clay minerals concentrate these substrates at the solid-liquid interface, where they reach levels high enough to support the growth of microbes which adhere to these surfaces in response to the nutrient enrichment.

The objectives of this work are to study the clay mineral compositions, and their interactions with some heavy metals and microbes in different soils irrigated by fresh and contaminated (sewage and agricultural drainage) Nile waters as well as artesian water.

MATERIALS AND METHODS:

Twelve samples were collected from four localities representing Nile alluvial soils at Assiut city that have been irrigated with various water resources for more than 40 years, (Fig. 1). The soils at location 1 are irrigated by fresh Nile water, whereas those of location 2 are irrigated by artesian water. The third and fourth locations have soils that are irrigated by Nile waters contaminated with incompletely treated sewage water and agricultural drainage water, respectively. Water samples were taken from each irrigation water source used. The physical and chemical analyses of the studied soils (Table 1) and collected water samples (Table 2) were carried out according to Page (1982) and Klute (1986). The heavy metals (Fe, Mn, Cu, Zn, Ni and Pb) were extracted from the soil samples (Table3) using the diethylene triamine pentaacetic acid (DTPA) method (Lindsay and Norvell, 1978) and determined using a model 1999 Perkin-Elmer atomic absorption.

The clay fraction ($<2\mu$) of each soil sample was separated after removing the soluble salts, organic matter and iron oxides for investigation by XRD. Three treatments i.e., oriented air dried, glycolated and heated at 550°C were performed on each sample. Running between 2 and 40 of 2 θ was executed using the X-ray Diffractometer. The clay minerals of the studied samples were estimated following the method of Schultz (1964).

The surface samples of the studied soils were selected to demonstrate interaction effects between clay minerals and microbes using Scanning Electron Microscope (SEM) according to Cervini-Silva and Dong (2004) and Bennett (2004). Clod samples and clay fractions of the studied soil surface samples were prepared by mounting onto aluminum stubs and then coated

with gold for the best resolution using a model JEOL-JSM-5400 LV SEM.



Fig. (1): Location of the studied soil profiles

Table (3)	: DTPA extrac	table-heavy m	etals (ppm) of	the studied so	il samples

Profile No.	Depth(Cm)	Fe	Mn	Cu	Zn	Ni	Pb
	0-15	13.71	9.68	3.08	1.65	0.43	0.39
1	15-45	14.89	7.56	1.79	0.52	0.26	0.28
1	45-150	10.21	5.22	0.54	0.31	0.22	0.36
	Average	12.94	7.49	1.80	0.83	0.30	0.34
	0 - 25	15.25	18.97	2.44	1.16	0.64	0.43
2	25-65	15.62	6.44	1.75	0.5	0.35	0.39
2	65-150	15.65	16.5	2.69	0.91	0.6	0.29
	Average	15.51	13.97	2.29	0.86	0.53	0.37
	0-10	8.17	13.73	3.63	1.63	0.48	0.49
2	10 30	12.41	24.9	2.55	1.62	0.5	0.24
3	30-150	14.24	4.84	1.06	0.52	0.28	0.36
	Average	11.61	14.49	2.41	1.26	0.42	0.36
4	0-25	7.67	5.58	2.49	0.58	0.49	0.06
	25-45	11.4	22.83	2.61	0.63	0.32	0.16
	45-150	4.24	28.18	3.24	0.53	0.48	0.25
	Average	7.77	18.86	2.78	0.58	0.43	0.16

RESULTS AND DISCUSSIONS:

1- Clay mineral composition:

X-ray diffraction (XRD) patterns of the clay fraction of the studied soils are shown in Figures 2, 3, 4 and 5. Scanning electron microscope (SEM) images of some clay minerals are observed in Figures 6 and 7. Semi-quantitative measurements of the identified minerals and their relative abundance in the studied soils with depth are given in Tables 4 and 5. Clay minerals distribution of the studied soils with depth is illustrated in Figure (8).

Smectites occur as major clay minerals in all soil samples with percentages vary from 36.0 to 58.7% with an average of 47.2%. They are mainly much abundant in these samples. In most studied soils, smectites tend to increase with depth (Fig. 8). This trend suggests that the deepest layer of these soils is largely made up of preserved Nile sediments that are rich in smectites. However, the surface layers have been mixed with the aeolian materials of the surrounding plateau (El-Attar and Jackson, 1973).

Kaolinite represents the second abundant clay mineral in the studied soils with amounts range from 7.9 to 15.7% and an average of 10.3% (Table 4). Its relative abundance is little in all investigated samples, except the second layer of profile 4 which is moderate (Table 5). There is no general trend for kaolinite distribution throughout the studied soils.

Interstratified minerals in the studied soils are represented by little amounts of regular mica-smectite mixed layers and trace to little amounts of irregular mica-vermiculite mixed layers. Mica-smectite mixed layers vary between 5.6 and 9% with an average of 7.4%, whereas mica-vermiculite mixed ones have amounts ranging from 4.2 to 7.3% and an average of 5.5%. No clear trend for the distribution of these minerals with depth in the profiles (Table 4).

Vermiculite is detected in little abundant with values vary from 5 to 8.4%, with an

average of 6.7%, and without a specific pattern of distribution throughout the soils.

Micas are also present in the examined samples, but in trace to little amounts. Their amounts range between 3.0 to 7.4%, with an average value of 4.9%. It is obvious that amounts of micas increase from surface to subsurface soil layers of the first three profiles (Table 4). This may be attributed to the similarity in the sedimentation regime. Most mica minerals in soils are mainly primary as they were inherited from soil parent materials. They present in the clay of young and less weathered soils (Fanning *et al.*, 1989). Presence of micas indicates that these soils are pedologically young.

Sepiolite is found in trace amounts in most investigated samples with exception of the surface soil layers of profiles 2 and 3 which have little amounts (7.2 and 7.3%, respectively). Moreover, palygorskite is present in trace amounts in all examined samples with values up to 4.1%. These minerals do not show any specific trend in the studied soils with depth. Occurrence of Sepiolite and palygorskite in all examined samples indicated that they were inherited from calcareous sedimentary rocks of the surrounding plateaus during the formation of these soils.

Associated minerals that are represented by quartz, feldspars and calcite are detected in the investigated samples with averages reach 4.6%, 4.6% and 1.9%, respectively. Presence of feldspars emphasizes that these soils are young in the pedological viewpoint.





Fig. (4):X-Ray Diffractograms of the clay fractions of the soil irrigated by sewage contaminated Nile water.

Fig. (5):X-Ray Diffractograms of the clay fractions of the soil irrigated by agricultural drainage contaminated Nile water.



Fig. 6. Scanning Electron Microscope photomicrograph showing smectite (Sm) and Kaolinite (Ka) of the soil irrigated by fresh Nile water.



Fig. 7. Scanning Electron Microscope photomicrograph showing smectite (Sm) and Kaolinite (Ka) of the soil irrigated by agricultural drainage contaminated Nile water.



Fig. 8. Distribution of the identified clay minerals throughout the studied profiles.

In general, the clay minerals are formed from weathering of different rock types and under specific climatic conditions. Smectites are inherited from weathering of basic volcanic rocks. Pedogenic smectites may be precipitated form solution (neoformed smectite) or by transformation of other silicate minerals (degraded smectite). They are most commonly formed in soils of tropical wet- and- dry to warm-temperate climates (De Visser, 1991). Potassium-bearing micas alter to expansible 2:1 minerals (smectite and vermiculite) by replacement of K in the interlayer with hydrated cations through a simple transformation by both layer and edge weathering (Fanning et al., 1989). The occurrence of mixed layers in soils can most often be ascribed to incomplete rock weathering (De Visser, 1991). Formation of interstratified minerals might be due to hydrothermal transformation, weathering involving partial removal of interlayer K of micas. Reversibly uptake of K and formation of brucite or gibbsite interlayers in vermiculite and smectite form interstratified layers (Sawhney, 1989). Kaolinite is derived from nearly all types of igneous, metamorphic and sedimentary rocks if rainfall is frequent and water flow and hydrolysis are sufficiently strong under tropical to subtropical climate (De Visser, 1991). Kaolinite is formed in the lithologic environment from feldspars and, to a lesser extent, micas in sandstones (Rossel, 1982). Vermiculite is originated from weathering and low temperature hydrothermal alteration of basic igneous rocks. It is most common in humid temperate and mountainous regions (De Visser, 1991). Vermiculite is most often reported to be transformed from muscovite, biotite, or chlorite (Douglas, 1989). Most mica minerals in soils are mainly primary as they are inherited from sedimentary and metamorphic rocks. Dominance of micas indicate an essentially

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physical weathering, found in cool or hot, dry regions and in mountainous regions (De Visser, 1991). Sepiolite and palygorskite minerals are believed to be formed mainly in calcareous crusts of semi-arid to arid regions (De Visser, 1991).

From the above-mentioned discussion, it is concluded that the identified clay minerals in the studied soils are detrital and were originated from the Ethiopian Plateau and latter on they were mixed with the detrital materials derived from the sandstones and limestones plateaus which surrounding the Nile River course during the transportation and precipitation of these Nile sediments. This conclusion is in full agreement with that reported by Said (1981).

2-Interaction between clay minerals and heavy metals:

Generally, clays and clay minerals are used for a large variety of environmental applications such as water purification, waste treatment, mineral barriers for waste deposits, and slurry walls for the encapsulation of contaminated areas. This is due to their large specific surfaces and the resulting ability to adsorb cations (Bradl, 2002).

The average concentrations of the measured heavy metals in the different investigated soils are arranged in a decreasing order as follows: Fe > Mn > Cu > Zn > Pb > Ni in soil irrigated by fresh Nile water, Fe > Mn > Cu > Zn > Ni > Pbin the soil irrigated by artesian water, and Mn >Fe > Cu > Zn > Ni > Pb in both soils irrigated by sewage and agricultural drainage contaminated Nile waters (Table 3).

Correlation coefficients between clay minerals and DTPA-extractable heavy metals in all studied samples and in each individual irrigated soil are mathematically calculated (Tables 6 to 10). Generally, there are low correlation values between clay minerals and extractable heavy metals in all studied samples, which in most cases are insignificantly positive. The only exception appears in the case of micasmectite mixed layers with Mn (r=0.65*), where a slightly significant positive correlation (Table 6).

Data in table (7) clearly show that there are highly significant positive correlation between mica-vermiculite mixed layers and Zn or Ni (r=0.99**) as well as between sepiolite and Fe (r=0.99**) in soil irrigated by the fresh Nile water. Slightly significant positive correlations are between palygorskite and Mn or Cu (r=0.98* or 0.97*) as well as between micavermiculite mixed layers and Cu (r=0.95*). However, the correlation coefficients are low, and in most cases are positive.

Coefficients given in Table (8) demonstrate that there is a highly significant positive correlation only between mica-vermiculite mixed layers and Cu ($r=0.99^{**}$) in the soil irrigated by artesian water. However, three values of slightly significant positive correlations are found between kaolinite and Pb ($r=0.97^{*}$), vermiculite and Cu ($r=0.98^{*}$), as well as sepiolite and Pb ($r=0.97^{*}$). Insignificant correlations in the rest cases are observed.

The soil irrigated with sewage contaminated Nile water has a highly significant positive correlation between smectite and Fe (r=0.99**), and a slightly positive correlation between micavermiculite mixed layers and Fe, as well as between mica-smectite mixed layers and Mn. Although, there are insignificant correlations in most cases, they show a negative trend between some clay minerals and certain heavy metals (Table 9).

Low correlation values are recorded between some clay minerals and extractable heavy metals, which, in most cases they are positive in the soil irrigated by agricultural drainage contaminated Nile water (Table 10). Only a slightly significant positive correlation (r=0.97*) is obtained for smectite and Cu.

From the previous discussion, it is concluded that the significant relations between smectite and Fe or Cu, kaolinite and Pb, micasmectite mixed layers and Mn, mica-vermiculite mixed layers and each of Zn, Cu and Ni, vermiculite and Cu, sepiolite and each of Fe and Pb, and palygorskite and each of Mn and Cu may be mainly attributed to the adsorption of these heavy metals on these clay minerals of the studied soils. However, insignificant positive correlations between clay minerals and extractable heavy metals, in most cases, may be due to low amounts of these elements are exchanged on these clay minerals of the examined soils. On the other hand, in few cases, a negative trend between some clay minerals and some heavy metals may be reflecting that these heavy metals are not mainly exchanged on these clay minerals. Similar results, especially those concerned with smectite and kaolinite, are coincided with those obtained by Schulthess and Huang (1990) and Srivastava et al. (2004).

Clay Mineral	Fe	Mn	Cu	Zn	Ni	Pb
Smectite	0.27	0.05	-0.35	-0.35	0.04	-0.14
Kaolinite	-0.48	0.44	0.44	-0.15	0.07	-0.34
Mixed mica-Smectite	0.12	0.65*	0.38	0.15	0.33	-0.30
Vermiculite	-0.10	0.41	0.28	0.08	0.49	-0.24
Mixed Mica-Vermiculite	-0.04	0.15	0.05	0.05	-0.32	-0.15
Mica	0.01	-0.31	-0.18	0.24	-0.11	-0.04
Sepiolite	-0.11	0.19	0.43	0.33	0.47	0.47
Palygorskite	0.01	-0.41	-0.16	0.06	-0.18	-0.03

Table (6): Correlation coefficients between clay minerals and heavy metals in the studied soil samples

* Significant at 5% level

Table (7): Correlation coefficients between clay minerals and heavy metals in the soil irrigated by fresh Nile water

Clay Mineral	Fe	Mn	Cu	Zn	Ni	Pb
Smectite	-0.92	-0.94	-0.93	-0.74	-0.76	0.10
Kaolinite	-0.16	0.55	0.58	0.83	0.82	0.94
Mixed mica-Smectite	0.79	0.18	0.14	-0.23	-0.19	-0.92
Vermiculite	0.02	0.69	0.72	0.92	0.91	0.87
Mixed Mica-Vermiculite	0.47	0.94	0.95*	0.99**	0.99**	0.55
Mica	0.55	-0.16	-0.2	-0.54	-0.51	-0.99**
Sepiolite	0.99**	0.76	0.73	0.44	0.47	-0.46
Palygorskite	0.86	0.98*	0.97*	0.82	0.84	0.03

Table (8): Correlation coefficients between clay minerals and heavy metals in the soil irrigated by artesian water

Clay mineral	Fe	Mn	Cu	Zn	Ni	Pb
Smectite	0.58	0.30	0.68	0.11	0.35	-0.97*
Kaolinite	-0.90	0.19	-0.25	0.38	0.14	0.97*
Mixed mica-Smectite	0.43	0.46	0.80	0.28	0.52	-0.91
Vermiculite	0.02	0.79	0.98*	0.65	0.83	-0.66
Mixed Mica-Vermiculite	-0.23	0.92	0.99**	0.82	0.94	-0.45
Mica	0.09	-0.85	-0.99**	-0.73	-0.88	0.57
Sepiolite	-0.89	0.18	-0.27	0.36	0.12	0.97*
Palygorskite	-0.98*	0.44	0.01	0.61	0.39	0.87

 Table (9): Correlation coefficients between clay minerals and heavy metals in the soil irrigated by sewage contaminated Nile water

Clay mineral	Fe	Mn	Cu	Zn	Ni	Pb
Smectite	0.99**	-0.07	-0.89	-0.62	-0.55	-0.80
Kaolinite	-0.92	-0.16	0.76	0.42	0.34	0.92
Mixed mica-Smectite	-0.06	0.98*	0.37	0.72	0.78	-0.68
Vermiculite	0.77	0.44	-0.53	-0.14	-0.05	-0.99**
Mixed Mica-Vermiculite	0.95*	-0.53	-0.99**	-0.92	-0.88	-0.43
Mica	0.20	0.91	0.12	0.52	0.59	-0.84
Sepiolite	-0.92	-0.16	0.76	0.43	0.34	0.92
Palygorskite	0.86	-0.69	-0.98*	-0.98*	-0.95*	-0.24

* Significant at 5% level, ** Significant at 1% level

Clay mineral	Fe	Mn	Cu	Zn	Ni	Pb		
Smectite	0.58	0.63	0.97*	-0.90	0.52	0.81		
Kaolinite	0.74	0.50	-0.14	0.73	-0.98*	0.26		
Mixed mica-Smectite	0.56	0.70	0.10	0.54	-0.91	0.49		
Vermiculite	-0.97*	0.43	0.90	-0.98*	0.71	0.65		
Mixed Mica-Vermiculite	0.25	0.90	0.43	0.23	-0.72	0.75		
Mica	-0.21	-0.92	-0.47	-0.18	0.69	-0.78		
Sepiolite	-0.99**	0.28	0.81	-0.99**	0.81	0.52		
Palygorskite	-0.29	-0.88	-0.39	-0.27	0.75	-0.72		

Table (10): Correlation coefficients between clay minerals and heavy metals in the soil irrigated by agricultural drainage contaminated Nile water

* Significant at 5% level, ** Significant at 1% level

3-Interaction between clay minerals and microbes:

Based on the examination of the soil surface samples using the scanning electron microscope (SEM), bacillus and coccus bacteria are observed in all investigated samples, particularly in the soil irrigated by sewage contaminated Nile water which are found as colonies (Figures 9, 10 and 11). Fungi are shown in all examined samples but in small amounts than bacteria; most of fungi are similar in all studied samples but some types are characterized to specific soils, such 88 Triscelophoius monosporous Ingold which is manifested in the soil irrigated by artesian water, and Pilobolus (Zygomycota) that is shown in the soil irrigated by sewage contaminated Nile water (Figures 12 and 13). However, actinomycetes appear in the soil irrigated by fresh Nile water (Figure 14). These results reflect the influence of the type of irrigation water, on the diversity of bacteria, fungi and actinomycetes in the studied soils. Bacteria are more abundant than fungi in all studied soil samples; also smectites are abundant. The interaction between this type of clay minerals and microbes could be interpreted that in montmorillonite containing soils. populations of bacteria are able to be developed in a greater extent than fungi due to 1) the buffering capacity of this clay because of its high CEC and the clay was able to maintain a suitable pH for continued bacterial development by the exchange of metabolically produced H^+ for basic cations on the exchange complex of the clay (Stotzky, 1966a and b, 1974; Stotzky and Rem, 1966), and 2) the ability of the clay to adsorb heavy metals that are more toxic at the same concentration to bacteria than to fungi (Babich and Stotzky, 1980 and 1983). Bacteria grow about twice as fast in the soils containing montmorillonite (either naturally or amended with mined clay), whereas fungi grow about twice as fast in that soils not containing this clay; the addition of mined kaolinite had little effect on growth (Stotzky, 1974). Bacteria growth on the smectite is dependent upon its available surface area. Iron (Fe⁺³) bound in clay minerals should be considered an important electron acceptor supporting the growth of bacteria in soils or sedimentary environments (Kostka et al., 2002).

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تم جمع عينات تربة من أربعة قطاعات تمثل ترب مروية بمياه النيل الطبيعية والملوثة (بمياه صرف صحى وأخرى بصرف زراعى) ومياه إرتوازية بمدينة أسيوط لأكثر من 40 سنة.

لقد أوضحت نتائج التحليل باستخدام جهاز حيود الأشعة السينية للجزء الطينى أن السمكتيت هو معدن الطين الرئيسى فى كل عينات الدراسة ووجد أنه يزيد بالعمق. والكاولينيت يمثل المعدن الطينى الثانى يليه معادن الطين المختلطة من السمكتيت والميكا ، والفيرمكيوليت ، والمختلطة من الميكا والفيرمكيوليت ، والميكا ، والسبيوليت والباليجروسكيت. ووجود هذه المعادن بهذا التنوع فى الترب تحت الدراسة يعكس الأصل الفتاتى من الهضبة الأثيوبية والمختلط مع رواسب الهضاب الرملية والجيرية المحيطة بمجرى نهر النيل.

بالنسبة للتأثير المتبادل بين معادن الطين والعناصر الثقيلة، فقد وجدت علاقات معنوية بين بعض المعادن وبعض العناصر الثقيلة مثل السمكتيت مع الحديد أو النحاس، الكاولينيت مع الرصاص ، معادن الطين المختلطة من السمكتيت والميكا مع المنجنيز ، المعادن المختلطة من الميكا والفيرمكيوليت مع الزنك أو النحاس أو النيكل ، الفيرمكيوليت مع النحاس ، السبيوليت مع الحديد أو الرصاص ، والباليجروسكيت مع المنجنيز أو النحاس. وربما تعزى هذه العلاقات نتيجة إدمصاص هذه العناصر على أسطح المعادن المذكورة.

أما بالنسبة لتواجد معادن الطين والميكرويات في التربة فقد أظهر الفحص باستخدام الميكروسكوب الإلكترونى الماسح أن البكتيريا العصوية والكروية شائعة فى كل العنات وخاصة فى التربة المروية بمياه النيل الملوث بالصرف الصحى حيث كانت فى صورة مستعمرات. كما أوضح الفحص وجود الفطريات فى كل العينات ولكن بكميات أقل من البكتريا التى ربما تكون مصاحبة للسمكتيت. وهناك أنواع معينة من الفطريات ظهرت فقط فى التربة المروية بمياه النيل الطبيعية وأخرى فى التربة المروية بالمياه الأرتوازية، كما وجدت الأكتينوميسيتات فقط فى التربة المروية بمياه النيل الطبيعية. وكل ذلك يعكس أثر نوع مياه الرى على نوع الكائنات الحية الموجودة فى التربة.