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# Effect of soil parameters on the corrosion of archaeological metal finds

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**Abstract**

Archaeologists have observed an increasing tendency for freshly excavated iron artefacts to deteriorate due to accelerated corrosion. Iron artefacts and associated soil samples from German archaeological sites have therefore been examined and analysed. The approximate aggressiveness of the soil toward buried iron objects has been estimated by means of an existing rating standard. In addition, correlation coefficients have been calculated to investigate the relationship between soil properties and the state of corrosion. The results showed that the most heavily corroded artefacts came from sandy and acidic soils as well as from urban soils. The investigation showed that there is the risk of complete destruction of iron artefacts in well-drained soils with low buffering capacities. Conservation of the archaeological heritage requires the collaboration of soil scientists and metallurgists to develop suitable methods for the protection of buried artefacts.


*Keywords:* archaeology; artefacts; corrosion; iron; soil acidity

## 1. Introduction

In recent years, archaeologists have observed an increasing level of deterioration in freshly excavated metal artefacts. There is little research in this field (Booth et al., 1962; Tronner et al., 1995; Scharff and Gerwin, 1996) and only a few studies have been able to confirm a correlation between soil properties and the corrosion of archaeological metal finds (Mattsson et al., 1996).

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Iron and bronze objects have been studied by Mattsson et al. (1996) and Scharff and Huesmann (1997). They compared archaeological finds excavated in the first half of this century with those excavated during the past few years from the same sites. Significantly, the Swedish investigations of bronze artefacts by Mattsson et al. (1996) confirmed a continual increase in the degree of deterioration since the beginning of the 20th century. This study also showed the influence of road salt on the corrosion process. The bronze objects (corrosion products around the objects) excavated near a road contained a higher chloride content than those from a longer distance. Scharff and Huesmann (1997) were able to confirm a spatial correlation between the corrosion products formed on archaeological iron objects and regional air pollution load. In particular, finds at archaeological sites in the southern part of the former German Democratic Republic contained high sulphate loads. A more recent study has tried to develop suitable ranking criteria for archaeological artefacts in terms of their metallographical condition and environmental impacts (Wagner et al., 1998).

Research on the corrosion of metals buried in soil is not very familiar to archaeologists or soil scientists. Some knowledge about the influence of soil properties on the corrosion of buried iron artifacts is available from material sciences. Often, leaking iron water pipes were the cause for such studies (e.g., Palmer, 1989; Gimelfarb, 1990). Only a few soil scientific investigations have been carried out in this field (e.g., Corcoran et al., 1977; Moore and Hallmark, 1987; Jarvis and Hedges, 1994). Several standard methods (e.g., DIN 50929, Part 3, 1985: corrosion of metals; probability of corrosion of metallic materials under external corrosion conditions; pipelines and structural component parts in soil and water) have been derived from results of these studies to estimate the aggressiveness of different soil types toward buried iron structures. The most important soil parameters are considered to be soil texture, soil acidity and the amount of soluble salts.

Nowadays, various methods have been developed to alleviate corrosion of buried iron structures. However, other environmental pollutants are considered to be responsible for some different and increasing corrosion problems of buried metal (iron) structures. These corrosion problems affect both modern fabricated structures as well as archaeological finds. Recent investigations suggest that possible reasons for an increase in damage may be:

- higher levels of sulphuric and nitric deposits from the atmosphere;
- soil acidification; and
- a higher salt content in agricultural soils due to intensive application of artificial fertilizers (Scharff and Gerwin, 1996).

Laboratory investigations have shown a correlation between the degree of corrosion of buried iron and artificial acid rain (Levlin, 1991). The influence of different salts has been investigated by Nürnberger (1989). A marked increase of the corrosion rate was observed after the application of chloride and sulphate.

In general, it was supposed that archaeological iron and bronze artefacts were little damaged while buried in soil. But in recent years, corrosion of archaeological metal finds does appear to be an increasing problem. For instance, most of the bronze artefacts excavated at the end of the last century in the abovementioned Swedish investigation were well-preserved. In contrast, most of the objects excavated in 1990–1995 at the same site were strongly corroded (Mattsson et al., 1996). Other archaeologists from European countries also report an increasing deterioration of iron objects (Fjaestad et al., 1998). As a result, the effort required to excavate, restore and store the objects is also growing.

Well-preserved iron artefacts are exceptional and may only have survived because the soil in which they have been buried has been non-aggressive or because some layers of corrosion products on their surfaces protected them (Wranglén, 1985; Cronyn, 1990). Such protective layers on the surfaces of metal objects may also save important scientific information about the object itself (decoration, etc.). Fig. 1 shows a schematic drawing of a typical cross-section of an iron artefact. Often, the former surface of an archaeological object ('original

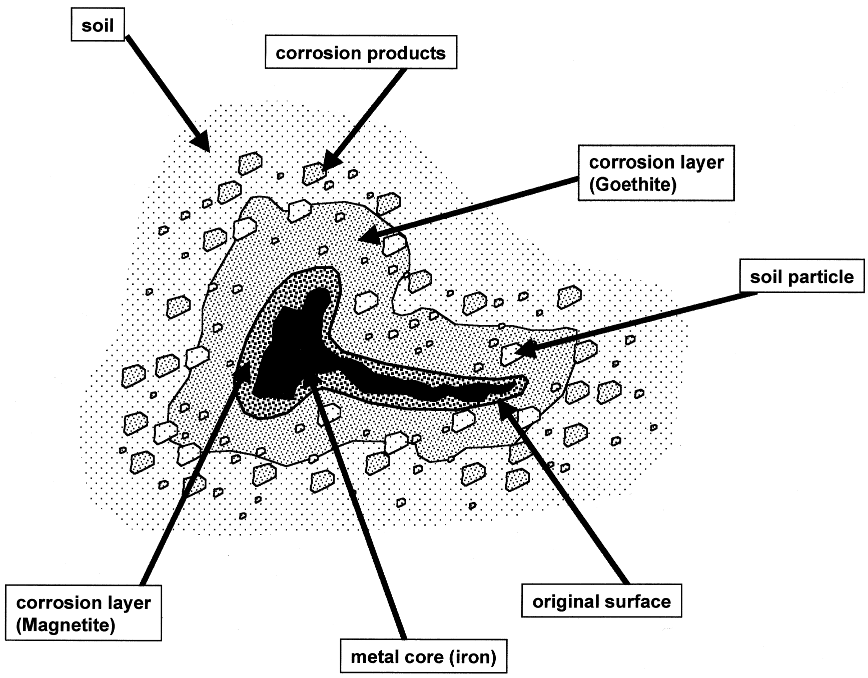


Fig. 1. Schematic cross-section of an archaeological iron artefact (after Cronyn, 1990; Knight, 1990).

surface') has been preserved under a sequence of corrosion layers consisting mainly of goethite (Knight, 1990). This original surface is critical for archaeologists trying to reconstruct the former function of an object. Finds with carbonate or phosphate layers around the metal core are especially well-preserved (Booth et al., 1962; Wranglén, 1985).

This paper presents some selected results of an interdisciplinary research project investigating the relationship between the condition of archaeological iron objects and soil properties. Interactions and relations between soil properties and soil pollution, on one hand, and condition of iron archaeological objects, on the other hand, have been analysed. One objective of the work was to evaluate the practicality of existing standards for estimating potential soil corrosiveness. Another aim of the investigation was the detection of interaction between iron objects and the surrounding soil.

## 2. Materials and methods

### 2.1. Sampling

Iron objects and soil samples have been taken from five important archaeological sites in Germany (see Fig. 2). Principal archaeological and pedological

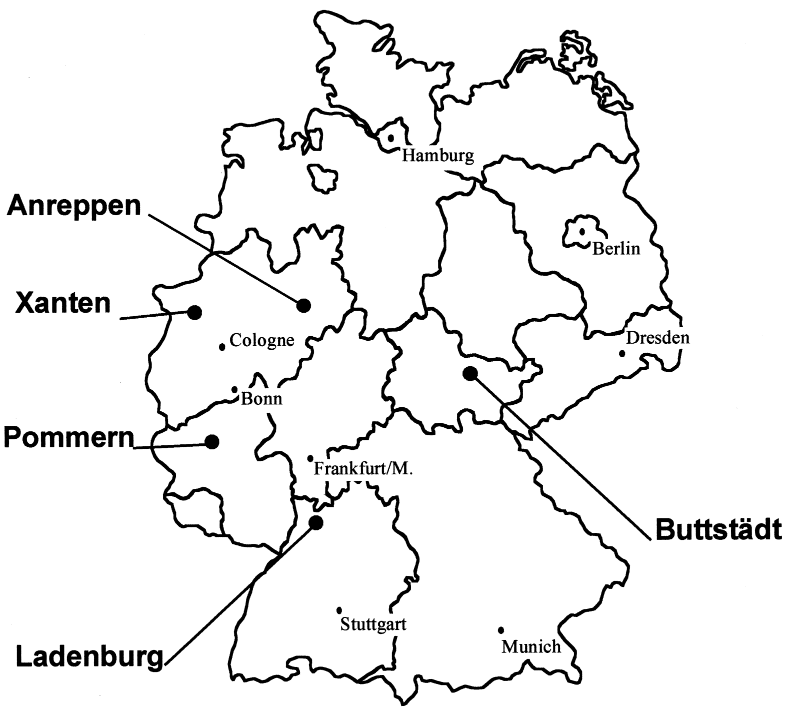


Fig. 2. Location of sampled archaeological sites.

features are given in Table 1. All sites are situated in rural environments except one which is located in the city centre of Ladenburg (Baden-Württemberg, near Heidelberg).

The iron objects were recovered by the Landesdenkmalamt Baden-Württemberg (LDA: the Baden-Württemberg Department of Historical Sites). The finds have been taken from selected archaeological profiles and from different depths. Metallurgical investigations at the University of Freiberg (Saxonia) have confirmed that the iron objects are comparable.

Each iron object was carefully assigned to a corresponding soil layer. These soil layers (archaeological deposits) were identified as being relatively homogeneous. Iron objects buried close to bones or mortar particles were excluded from the analysis. Fig. 3 shows the sampling method schematically. This approach to sampling has been adopted for a large number of archaeological profiles (between four and eight profiles at each site; see Table 2). At each site, the following soil samples were taken.

- Composite samples representative of each soil layer were taken near the iron objects to study the interrelation between soil properties and corrosion. These samples were considered to be unaffected by the corrosion process but represent the environment in which the finds were buried.
- Soil samples were also taken from the immediate vicinity of archaeological iron artefacts. These samples may have been affected by the corroding artefacts and have been used to study possible changes of soil properties caused by corrosion.

The samples were taken when the archaeological artefacts were recovered. Soil particles adhering to the freshly excavated iron objects were removed from their metal surfaces. The weight of these samples was about 10–50 g and sample volumes were calculated using an average bulk density of 1.4 g/cm$^3$.

Table 1
Archaeological and pedological features

| Location | Archaeological features | Today's land use | Soil type | Substrate |
|---|---|---|---|---|
| Anreppen | Roman military camp | agriculture | Podzols | sand |
| Buttstädt | Medieval settlement | agriculture | Chernozems | loess |
| Ladenburg | Roman/Medieval/recent settlement | town centre | Anthrosols (urban soil) | anthropogenic material (rubbish, ash, etc.) |
| Pommern | Celtic/Roman sanctuary | agriculture | Gleysols | loess |
| Xanten | Roman military camp | agriculture/forestry | Cambisols | sand |

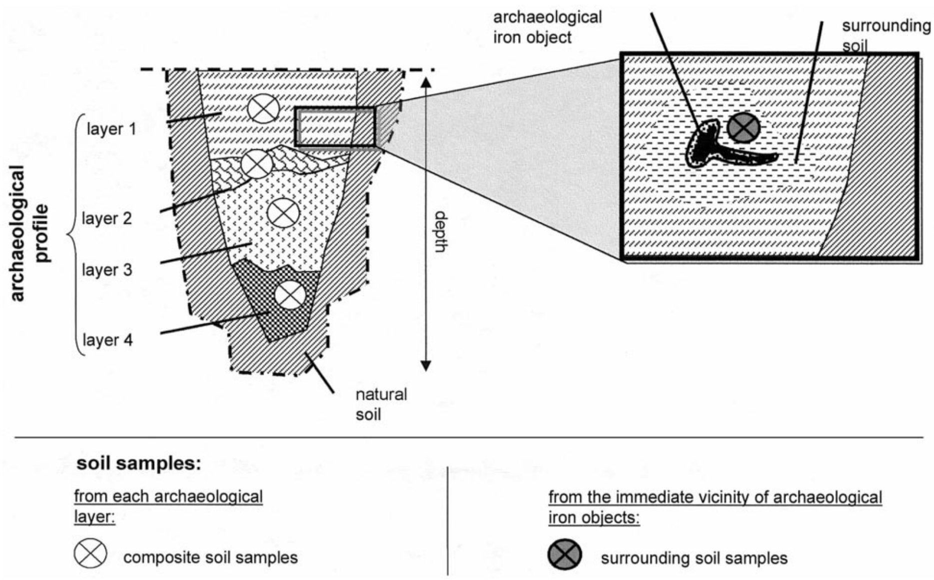


Fig. 3. Location of soil samples (schematic).

These volumes were used to judge approximately how thick a zone around the object the samples represented.

## 2.2. Corrosion of archaeological artefacts

The degree to which the archaeological iron artefacts have been corroded was established by visual analysis of X-radiographs. The amount of metal core remaining as compared with the original size of each object indicated the loss as a result of corrosion.

Fig. 4 shows, on the left, a model X-radiograph of an iron artefact. The contours of the metal core and the former original surface (deduced from the X-radiograph) are drawn on the right. A total of more than 300 iron artefacts have been analysed in this way.

## 2.3. Soil analysis

The following soil parameters (with a brief description of their determination), which are commonly thought to be relevant to soil corrosion processes, were

Table 2
Number of archaeological profiles at each site

| Anreppen | Buttstädt | Ladenburg | Pommern | Xanten |
|---|---|---|---|---|
| 4 | 6 | 4 | 8 | 6 |

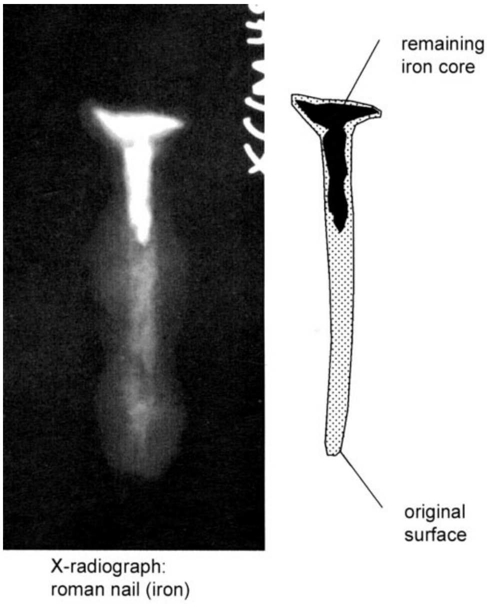


Fig. 4. X-radiograph of an archaeological iron nail (left) and an explanatory diagram showing the metal core and the original surface (right).

measured in the collected samples. For more detailed descriptions of the methods, see Rowell (1993) and Schlichting et al. (1995) for the following.

- Particle size analysis by sedimentation (Köhn method) and sieving.
- Carbonate content by the gas volumetric method (Scheibler method).
- Soil pH values determined in 0.01 M $CaCl_2$ suspensions (1:2.5) with a glass electrode.
- Loss on ignition by placing dry samples in a furnace at 450°C.
- Electrical conductivity of the soil solution, as a measure of the total salt content of the soil, in a 1:5 extract using a standard conductivity cell.
- Water-soluble sulphate and chloride as measured by an ion chromatograph (Metrohm 610) with a 1:2.5 extract.
- Water-soluble phosphate by the colorimetric Murphy–Riley method.
- Dithionite-soluble iron oxides ($Fe_d$) have been determined (AAS Philips Unicam SP 9) in order to study the enrichment of iron oxides in the surroundings of corroding iron artefacts (i.e., levels in the surrounding soil samples minus the levels in the corresponding composite soil samples from the particular layer).

The enrichment was related to the approximate volume of the soil samples. As mentioned above, the volume of the sample taken from the material immediately surrounding the object can be used indirectly to judge the distance of the sample from the object surface.

### 2.4. Evaluation of soil aggressiveness

An approximate evaluation of the aggressiveness of the surrounding soil has been made following DIN 50929, Part 3 (1985). The DIN standard considers the following soil parameters: clay content, organic matter, electrical conductivity, pH value, buffering capacity, sulphate and chloride content. The sum of the values for the individual parameters provides a measure of the aggressiveness of the soil.

### 2.5. Statistical methods

Rank correlation coefficients after Spearman have been calculated for the relation between the soil properties and the degree of corrosion of the archaeological iron finds. The degree of corrosion of 219 iron finds from rural sites and properties of the associated soil layers (particle size, carbonate, loss on ignition, sulphate, phosphate and pH value) have been analysed. Coefficients have not been calculated for the only urban site considered here.

## 3. Results and analysis

### 3.1. Soil properties

Particle size distribution measurements showed large differences between the sites (Fig. 5). At Anreppen and Xanten, the archaeological deposits had low silt and clay contents. The average sand content was over 70%. These soils were well-drained and well-aerated. The soil at the Buttstädt site had a loamy texture

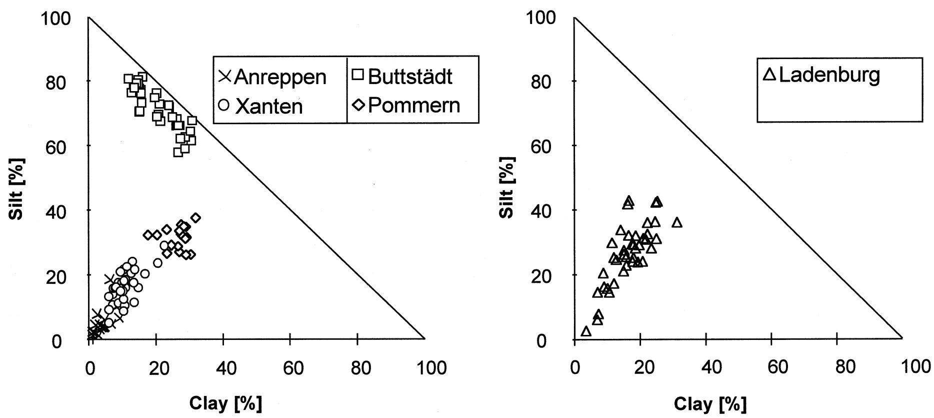


Fig. 5. Particle size distribution.

with a high silt content derived from the loess substrate. This soil is well-aerated and drained as a result of numerous lumbricid burrows and has a well-aggregated soil structure. In contrast, the archaeological deposits at the Pommern site, with a clay content of about 30%, are poorly drained. The profiles showed hydromorphic properties. The texture of the urban soil at the Ladenburg site was a sandy loam. It contained large amounts of rubble from former buildings and these coarse soil components result in comparatively good drainage and aeration.

Fig. 6 shows box plots of the carbonate content, the pH values, the electrical conductivity and the loss on ignition. The results are summarised below:

- Soil pH values range from neutral or weak alkaline (Buttstädt, Pommern and Ladenburg) to strongly acidic (pH values $< 5.0$) in Anreppen and in some parts of the Xanten site.
- The soil samples from Anreppen, Pommern and Xanten commonly did not contain any carbonates. On the other hand, the soils from Buttstädt and from Ladenburg are calcareous.
- The lowest salt levels of all the sites analysed (low electrical conductivity) were found in the soils from Anreppen, Pommern and Xanten. The highest EC values were measured in the Ladenburg soil.
- Loss on ignition date showed that the Buttstädt soil is rich in organic matter.

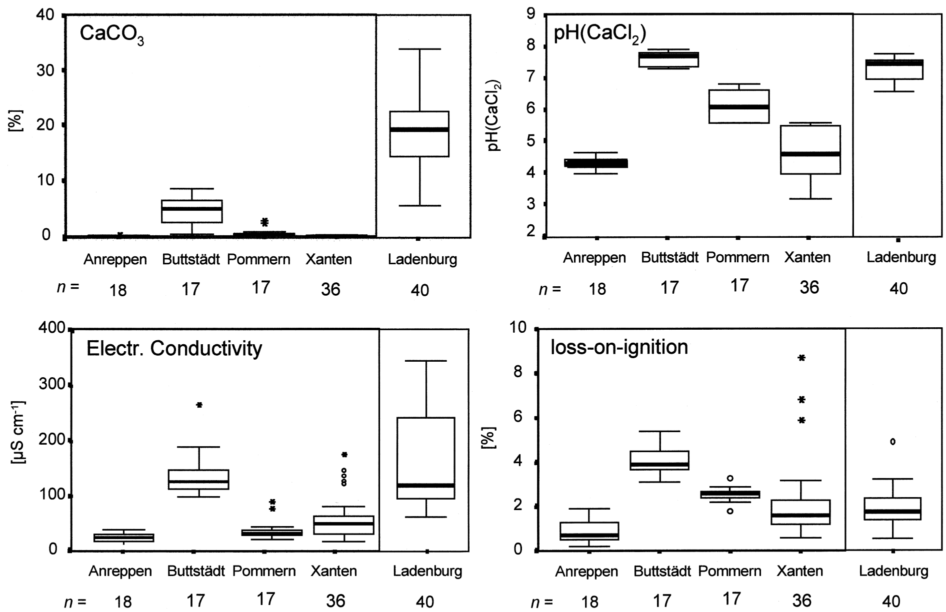


Fig. 6. Soil properties of the archaeological sites studied: carbonates, loss on ignition, electrical conductivity, pH values.

The average chloride and sulphate content and the phosphate concentrations are given in Fig. 7. Chlorides are leached easily by percolating water under humid climatic conditions so the concentrations are generally low except at the Ladenburg site. The highest sulphate levels among the rural soils studied were found at Buttstädt. Particularly high phosphate levels were measured in the urban soils at Ladenburg. Large amounts of phosphates were also extracted from the Anreppen soils and some samples from the Xanten site.

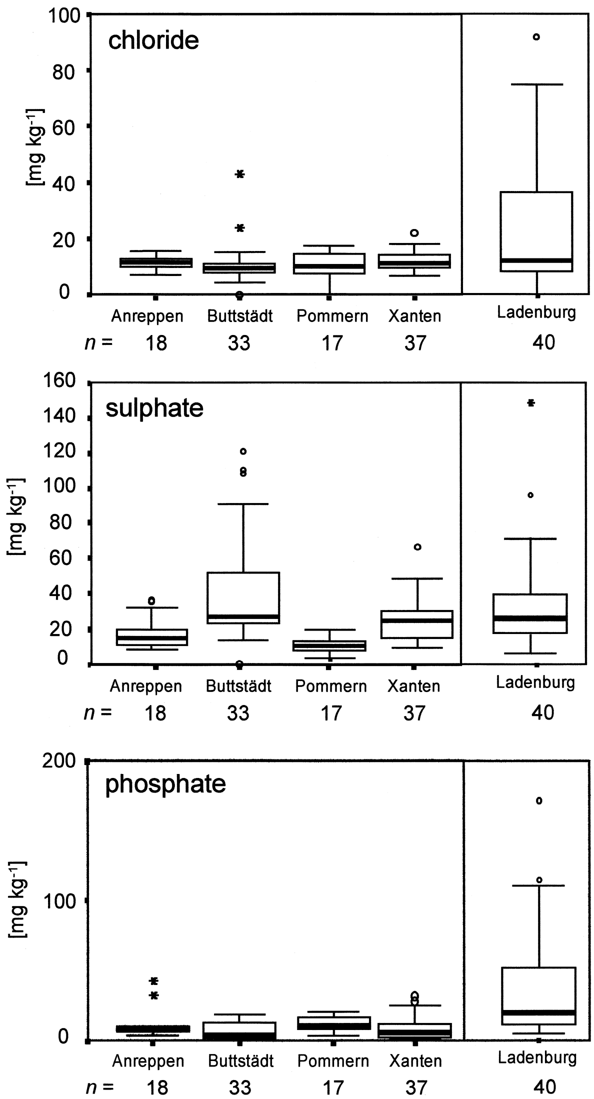


Fig. 7. Soil properties of the archaeological sites studied: water-soluble chloride, sulphate and phosphate.

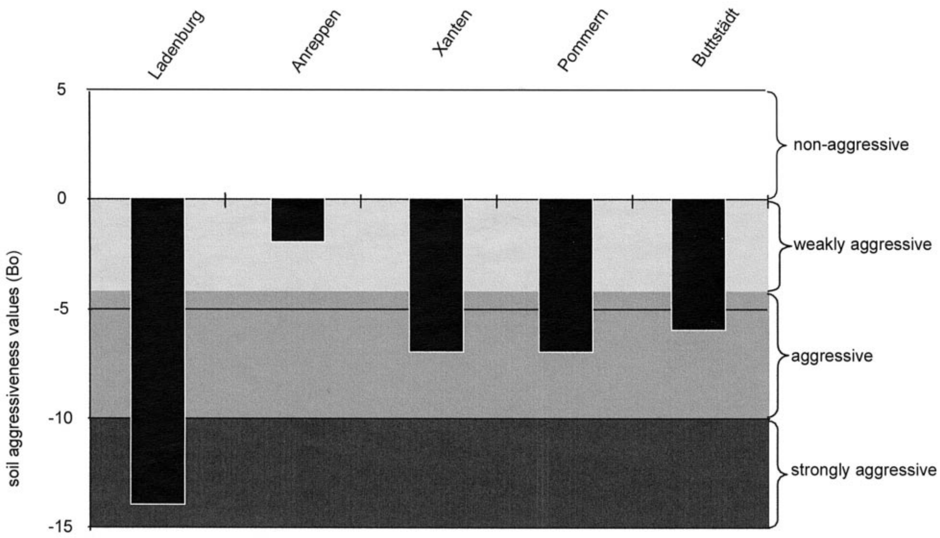


Fig. 8. Evaluation of soil aggressiveness after DIN 50929 (Bo: sum of the single rating values).

Based on the soil properties, an attempt has been made to estimate potential soil aggressiveness using the DIN standard (DIN 50929, Part 3, 1985). Fig. 8 shows the assessment average for each site. The larger the negative rating, the more aggressive the soil is towards buried iron objects. The urban soil layers at Ladenburg were the most aggressive. In contrast, the archaeological deposits at the rural sites appear to be less corrosive.

### *3.2. Degree of corrosion and soil aggressiveness*

Fig. 9 shows the average degree of corrosion of finds from the various sites. It is clear that there are significant differences between the various sites. The average loss of metal due to corrosion is over 50%, the only exception being artefacts from Buttstädt, which were all in better condition. Objects from Ladenburg and Anreppen were extremely corroded (more than 80% metal loss).

The correlation between the degree of corrosion and the estimated degree of soil aggressiveness appears to be very low (Fig. 10). Therefore, it is assumed that the evaluation method in use is not completely suitable for estimating aggressiveness toward archaeological artefacts. The DIN standard has been developed for structures like pipelines and does not appear to allow for the special case of archaeological objects which have been buried for several centuries.

One possible reason for this poor assessment may be the negative classification of the clay content. Moore and Hallmark (1987) also reported similar

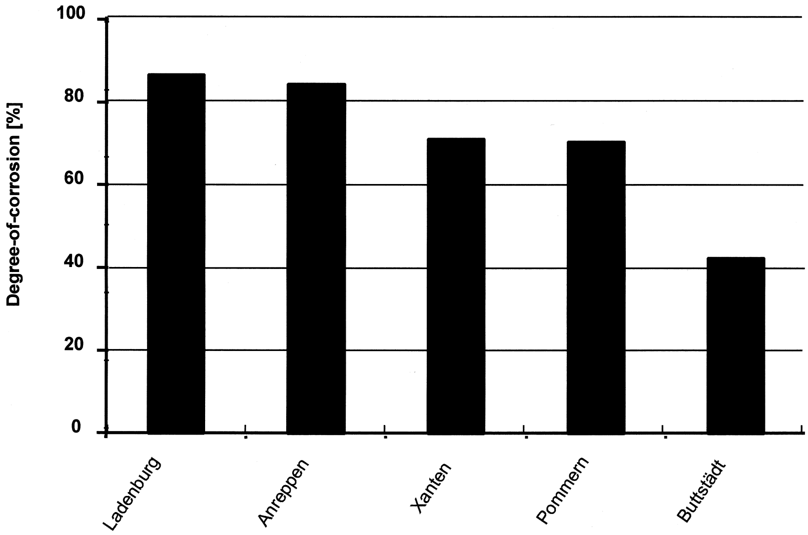


Fig. 9. Average degree of corrosion of archaeological iron objects.

problems of a too cautious negative rating of loamy soils. In reality, corrosion of buried iron often decreases in loamy soils because of oxygen deficiency. The occurrence of an aggressive microbiologically induced type of corrosion under anaerobic conditions requires the presence of certain species of microorganisms and of sulphate (Wranglén, 1985). The sulphate content of all the investigated

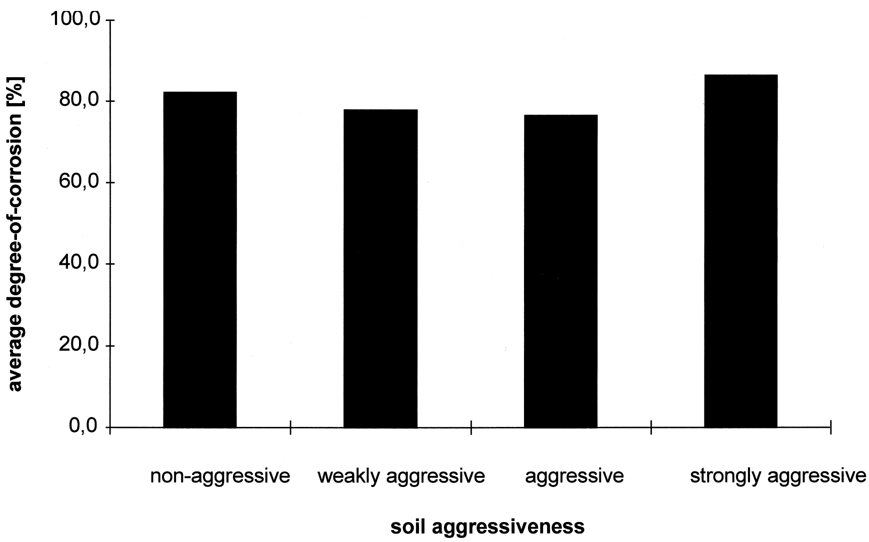


Fig. 10. Correlation between the average degree of corrosion and estimated soil aggressiveness.

soils was comparatively low and, apart from the Pommern site, the soils were well-aerated. Therefore, microbiological corrosion seems unlikely.

As a general point, the estimation of the aggressiveness of soils toward archaeological iron will be difficult because the former environment in which the artefact was buried cannot be reconstructed. In addition, there is much uncertainty about influences during burial.

### 3.3. Soil properties and degree of corrosion

Rural and urban sites clearly differ with respect to their soil properties. There is reason to suppose that these differences are responsible for the variability of the corrosion of ancient metals. To analyse relationships between soil properties and the amounts of corrosive decay of archaeological iron finds, correlation coefficients (Spearman) have been calculated for the rural sites because urban sites are affected by the deposition of garbage, rubble from old buildings, ash and other artificial matter over a period of several centuries, and the soils can differ strongly from natural soils (Burghardt, 1994).

In general, the correlation coefficients are quite low, but the large number of objects ($n = 219$) is responsible for the apparent statistical significance. Considering only the rural sites (Anreppen, Buttstädt, Pommern and Xanten) as presented in Fig. 11, the negative correlation coefficient between the clay

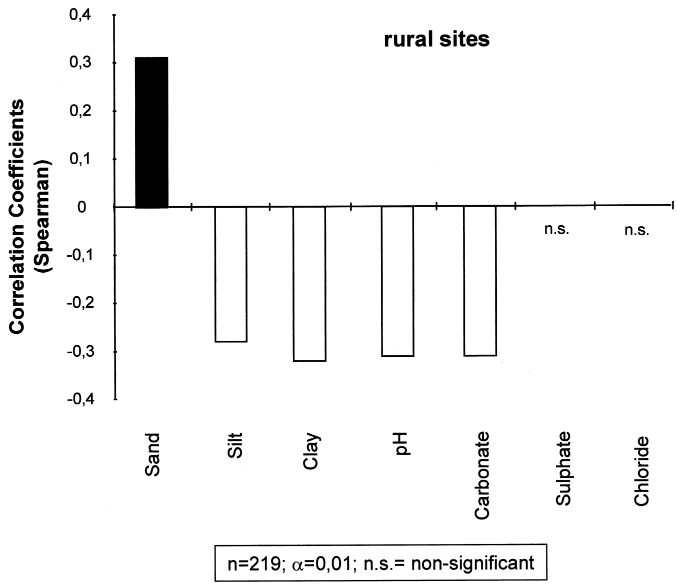


Fig. 11. Correlation coefficients between corrosion levels of archaeological iron objects and soil parameters.

content and the degree of corrosion suggests an apparent positive influence of fine-textured soils on the preservation of buried iron objects. This is, as mentioned above, in contrast to the negative rating of loamy or clayey soils by the DIN standard. A possible explanation may be the poor aeration of clayey soils. In this case, the corrosion process can be slower due to oxygen deficiency. Well-preserved iron finds recovered from the poorly drained Pommern site have been excavated from deeper archaeological layers. Finds from the upper horizons (from beneath the plough sole) were usually in a worse state of corrosion. On the other hand, the leaching of corrosive substances, such as acid precipitation or salts from fertilizers, is much faster in a well-drained soil. So many of the iron finds from the two sandy sites (Anreppen and Xanten) show serious deterioration even though they have been excavated, in part, from deeper archaeological layers.

It is clear from Fig. 11 that the extent of corrosion is greater in acidic soils than in neutral soils or those containing carbonates. This accords with the general knowledge that the corrosion of buried iron structures is much stronger in acidic soils (Camitz and Vinka, 1989). Soil acidification has been exacerbated by acidic precipitation in recent decades and is probably partly responsible for increasing soil aggressiveness (Levlin, 1991).

High soil carbonate levels correlate with less deterioration in metal artefacts and finds in these soils are in better condition as a result of protective layers being formed around their metal cores. Carbonates, phosphates (e.g., from bones) and some organic compounds in archaeological middens form low-solubility corrosion products around the surface of metal objects (Wranglén, 1985; Cronyn, 1990). Analysis of the Buttstädt artefacts by the University of Freiberg/Saxony, Institute of Metallurgy, demonstrated that such carbonate layers existed. Negative correlation between corrosion and loss-on-ignition data suggested that specific organic substances may also be protective (Farrer et al., 1953; Wranglén, 1985).

Chlorides and sulphates in soil usually promote the corrosion of metal objects (Booth et al., 1967; Foley, 1970) but in this study, no significant correlation could be found between chloride and sulphate content and corrosion at the rural sites probably because the concentration of these anions was low.

The Ladenburg urban soil is particularly rich in phosphates and carbonates. The accumulation of phosphates in cultural layers of former settlements due to the deposition of household deposits (e.g., bones) is well-known (Sjöberg, 1976). However, neither high phosphate and carbonate level nor the neutral or weakly alkaline condition of the soil at Ladenburg has prevented heavy corrosion of iron objects during burial there. In fact, objects from Ladenburg show greater deterioration than those from rural sites. ‘Artificial’ or anthropic substrata in urban soils are commonly strongly aggressive (Nürnberger, 1995) because they usually contain large amounts of salts. Chloride, especially, is known to be a very corrosive anion (Foley, 1970; Cronyn, 1990).

## 3.4. Iron concentration in the surroundings of iron objects

Iron oxides and other iron compounds may enrich the surrounding soil as a consequence of the corrosive decay of iron objects nearby. The amounts of $Fe_d$ were measured in the soil immediately surrounding iron artefacts as well as in the composite soil samples from the specific archaeological layers (Fig. 3). Differences between the surrounding and composite soil samples may indicate iron enrichment in the surrounding soil. Results for Anreppen and Buttstädt are presented in Fig. 12 as examples of possible relationships between corroding iron objects and their immediate vicinity. It is likely that the impact of the corrosion process on the surrounding soil decreases with increasing distance from the corroding metal surface. Therefore, the calculated sample volumes in Fig. 12 may indicate the approximate distance from the objects' surfaces from where the samples were taken.

The enrichment of iron oxides in the surroundings of the artefacts at Anreppen is commonly low. Apparently, it does not depend on the distance from the object's surface. In contrast, a logarithmic decrease of the iron enrichment with increasing distance from the metal surface was found at Buttstädt. A marked enrichment of iron oxides occurs only close to the metal surfaces.

The sandy and acidic soil at Anreppen was not able to immobilise the iron oxides released by corrosion. The high permeability of these archaeological layers is responsible for the fast transport of corrosive substances to the buried

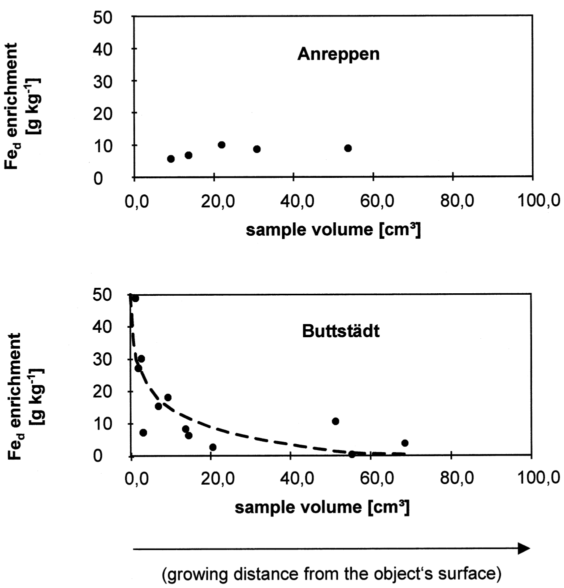


Fig. 12. Enrichment of $Fe_d$ in the soil surrounding corroded archaeological iron objects: sample volumes are calculated as a relative unit of measurement for the distance from the object's surface.

iron objects as well as for the removal of corrosion products away from the objects. Such conditions do not promote the formation of protective corrosion layers on the surface of buried iron. As a result, iron oxides constantly diffuse into the surrounding soil and the corrosion process is uninhibited. The typical appearance of such artefacts is a bulky brown mass of iron oxides (Cronyn, 1990). On the other hand, iron oxides are quite immobile in the calcareous soil at Buttstädt. Protective layers (oxides and other compounds) may be formed on the surface of buried objects and the corrosion process slows down with time.

## 4. Conclusions

The study identified the possible influence of certain soil properties on the corrosion of ancient iron objects. For instance, increasing soil acidity accelerates corrosion and destroys important information about the original surface features of an object. Probably, the high salt (chloride) content in the urban soil was responsible for the poor state of iron finds. Therefore, excessive salt loads need to be avoided. Preventative conservation of buried artefacts in situ almost certainly necessitates the reduction or avoidance of harmful environmental impacts on soil. Furthermore, preservation of our archaeological heritage requires a good knowledge of the causes of soil corrosivity and possible steps to reduce it. Soil research and metallurgical analysis is necessary to review the conditions at archaeological sites and to develop suitable protection methods (Wagner et al., 1997). A suitable method for estimating soil aggressiveness towards archaeological iron objects needs to be developed and evaluated.

Material scientists are also concerned in understanding the results of both soil science and metallurgical research into the impact of soil properties on corrosion of archaeological metal. Such results may be helpful, e.g., as ‘‘natural’’ analogues for metal containers used for the storage of radioactive materials (Miller and Chapman, 1995).

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