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ORGANIC CARBON CYCLING IN LANDFILLS: MODEL FOR A CONTINUUM APPROACH

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SUMMARY: Organic carbon cycling in landfills can be addressed through a continuum model where the end-points are conventional anaerobic digestion of organic waste (short-term analogue) and geologic burial of organic material (long-term analogue). Major variables influencing status include moisture state, temperature, organic carbon loading, nutrient status, and isolation from the surrounding environment. Bioreactor landfills which are engineered for rapid decomposition approach (but cannot fully attain) the anaerobic digester end-point and incur higher unit costs because of their high degree of environmental isolation and control. At the other extreme, uncontrolled land disposal of organic waste materials is similar to geologic burial where organic carbon may be aerobically recycled to atmospheric CO₂, anaerobically converted to CH₄ and CO₂ during early diagenesis, or maintained as intermediate or recalcitrant forms into geologic time (>1000 years) for transformations via kerogen pathways. A family of improved landfill models are needed at several scales (molecular to landscape) which realistically address landfill processes and can be validated with field data.

1. INTRODUCTION: THE CONTINUUM CONCEPT

Often, landfills are viewed through a very narrow time perspective including only the period of construction and post-closure monitoring at highly controlled sites, where specific practices are regulated by government and industry standards. More realistically, landfills throughout the world encompass a wide range of designs with various degrees of control and biodegradation rates. Here we propose a broader view of landfilling within a logical continuum of shorter-term and longer-term processes as shown in Fig. 1.

Organic Carbon Processes

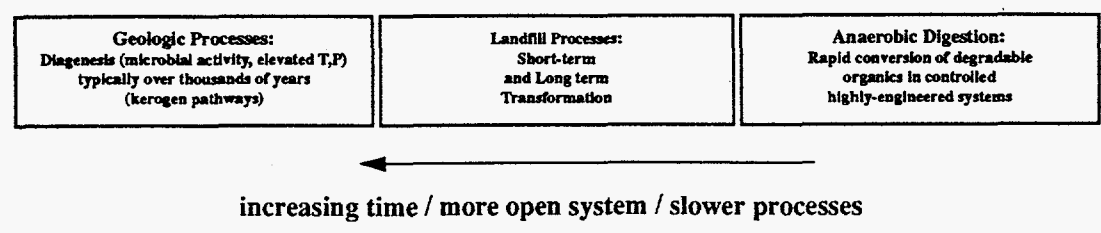


Figure 1. Placement of landfill processes within a logical continuum.

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The short-term endpoint is the engineered anaerobic digester; the long-term endpoint is geologic burial of organic matter under anaerobic conditions, where preservation and diagenetic transformations are promoted over long time frames. Distinctions between landfilled organic carbon and geologic burial tend to merge when one considers, at one extreme, documented cellulose preservation from the early Tertiary (Degens, 1967) and, at the other extreme, generation of crude oil from buried organic carbon in a few thousand years in an active tectonic environment (Peter *et al.*, 1991). Conservatively, it can be estimated that 30 million Tg of organic carbon are annually buried in landfills in developed countries which will not be converted to biogas end-products (CH₄ and CO₂) over decadal time frames (Bogner, 1992; Bogner and Spokas, 1995).

Our purpose in this paper is to develop a broader view of organic carbon cycling in landfills through examination of a continuum of long-term and short-term analogues. We will provide a common framework for comparison and draw on literature from both end-points to compare rates and controlling variables at several scales. We will also provide guidance for a family of improved landfill models which more realistically address scaling issues (both spatial and temporal) and can be validated directly with field data. Such models can better guide consideration of the ecological, economic, and human health aspects of landfilling as it is practised worldwide. Past landfill decomposition models have generally favoured the anaerobic digestion endpoint, the short-term analogue of landfilling. A number of models have been proposed, ranging from estimates based on simple stoichiometric yields to more complex first-order kinetic models. Most are based on organic carbon decomposition in a landfill which functions primarily as a batch anaerobic digester (*e.g.*, EMCON, 1980; Halvadakis *et al.*, 1983; Young, 1995; El Fadel *et al.*, 1996). There are many field situations where such models may not be appropriate and others where there is much reliance on default values since direct validation with field data is difficult, expensive, and may not be possible for some variables. We suggest that alternative models are needed at various scales which focus less on engineered closed-system solutions for the whole landfill and more on specific internal landfill processes in an open or semi-open setting. Such open-system models would be simpler, could be validated directly with focused field data, and address particular mass and energy fluxes at a given site without attempting to develop an all-inclusive model.

A comparison of anaerobic digestion (AD) and geologic settings is given in Table 1. AD is widely used for commercial treatment of many types of biodegradable organics -- traditional feed stocks are sewage sludges and wastes such as manure, food processing wastes, and industrial wastewaters containing high BOD (biological oxygen demand) such as paper mill effluent (Welandar, 1989). Different biomass "energy crops" have also been used, *e.g.* grass (Jarlsvik, 1995). AD has been developed for both unsorted MSW or various fractions, either alone or in combination with other wastes. Gaseous end-products of anaerobic digestion are methane and carbon dioxide ("biogas"). The anaerobic digestion process involves several groups of microorganisms - hydrolytic and fermentative groups, acidogens and acetogens, and methanogens - which have evolved complex and interacting functional strategies through geologic time for energetic gain and, hence, survival. Conversion of some fraction of the organic carbon placed in a commercial digester occurs within periods of approximately 2-30 days, depending on numerous variables, particularly substrate chemistry, digester design (*e.g.*, low solids/high solids, stirred tank/plug flow), temperature, and operating strategies (one-phase, two-phase). At present about a dozen types of AD systems are marketed, the most common type being the low solids, continuously-stirred, one-step processes. The overall digestion process in a reactor vessel is highly controlled and may include frequent monitoring of operational variables known to be diagnostic of system performance for multiple interacting microbial groups. Conversion rates are optimised according to pre-set engineering goals. Existing landfill models based on anaerobic digestion concepts may be run with variable temperature, pressure, and other environmental variables to mimic field settings (*e.g.*, Young, 1995 and previous work).

Table 1. Comparison of Anaerobic Digester and Geologic Endpoints*.

| Anaerobic Digestion: | Geologic: |
|--|---|
| <i>Starting Material and Alteration Pathways:</i> | |
| <p>In AD biopolymers are hydrolysed, broken down to monomers, and degraded to C1-C2 compounds, CO₂ and H₂, which are utilised in biological CH₄ production. AD is used both in combination with pre- and post-composting. Usually a one step process is employed, in some cases a treatment yielding organic acids and alcohols is used as a primary step. MSW is dominated by the cellulose fraction, estimated to hold 63 % of the methane potential of Swedish MSW (Chen 1995), and about 73 % for US MSW (Barlaz <i>et al.</i> 1990). MSW also contains proteins and fats, COD/TSS ratios of about 1.3 have been observed in the biodegradable MSW fraction, whereas cellulose would give a ratio close to 1.2 (Lagerkvist 1987).</p> | <p>Terrestrial organic material (mainly lignocellulosics). Predominately preserved under conditions of anaerobic burial with early diagenetic generation of biogenic methane. Also early aerobic bacterial and fungal decomposition of lignocellulosics. Competition of methanogenesis with sulfate reduction in environments of elevated sulfate (marine, near-marine). If organic carbon in altered or unaltered forms survives early diagenesis, mainly evolves to Type III kerogen [H/C atomic ratio usually <1.0 and O/C ratio 0.2 to 0.3; containing condensed polyaromatics and oxygenated functional groups, with minor aliphatic chains]. End products include peat, coal, or thermogenic natural gas. (Tissot and Welte, 1976, and references cited therein)</p> |
| <i>Occurrence</i> | |
| <p>MSW is generated where there are people. Johnson (1984) estimated daily generation rates from above 0.4 kg d⁻¹c⁻¹ for low income countries and up to 1.8 for high income countries. The difference between high and low average income countries often lies more in the waste composition than in the specific amounts generated. Two waste fractions which may vary much are the biowaste and the ash/dust fractions. Carbon content of MSW varies in the range of about 35-45 %, e.g. 38.3 % of TS was reported by Savage <i>et al.</i> (manuscript) for a Californian waste. C-content [% of TS] can be roughly estimated as the VS·1.8⁻¹ (Halmø 1984). In recent years, about a hundred AD plants for MSW/MSW fractions have been built, most in Europe. Few plants have a capacity above 50 Gg y⁻¹, the total commercial AD plant capacity in 1996 was estimated to 1.8 Tg per y⁻¹ (Lusk <i>et al.</i> 1996).</p> | <p>Disseminated in sediments or concentrated in peats, lignites, and coals (vitrinite). Current terrestrial biomass contains about 560 Pg (10¹⁵g) organic carbon with mean residence time (live biomass) of about 9 years; soils contain about 1500 Pg organic carbon; estimates of annual accumulation of organic carbon in soils is only about 0.4 Pg y⁻¹ due to efficiency of aerobic decomposers (Schlesinger, 1995). In sedimentary rocks, siltstones and sandstones typically contain <1% organic carbon; carbonaceous shales and "petroleum source rocks" up to 10% organic carbon; and bituminous coals >70% "fixed carbon" (all weight %) (Bostick, 1979). Peats may contain recognisable cellulose (Tissot and Welte, 1976). Recognisable plant material may be present in coals as old as the Paleozoic; such plant materials can be classified petrographically (Alpern, 1970).</p> |
| <i>Temperatures:</i> | |
| <p>Typical operation temperatures for AD are either around 35 °C or around 55°C . The former is referred to as "mesophilic" and the latter "thermophilic".</p> | <p>For methanogenesis (early diagenesis only), temperatures range from less than 10 °C in arctic environments (Svensson, 1984; Kotsyurbenko <i>et al.</i>, 1993) to >40 °C in low latitudes. Later diagenetic temperatures to several hundred degrees (Celsius).</p> |

Table 1. Continued.

Pressure:

Ambient atmospheric to slightly elevated. The partial pressure of various gases, such as H₂, may influence process pathways.

Ambient atmospheric pressure to slightly elevated during early diagenesis. Later diagenesis may occur at pressures of several hundred bars or more with burial to depths of several thousand meters.

Methane production and organic matter conversion rates:

Methane yield of AD-plants using MSW or the organic fraction of MSW varies much. *E.g.* a range of 2.3-3.5 kmol per Mg (fresh weight) of waste treated has been reported for a Swiss plant using the biowaste fraction, the range for a French plant using mixed household was 2.7-3.1 (de Jong *et al.* 1994). CH₄/CO₂ depends on substrate, but is also changed by the process and especially if it promotes CO₂ absorption and stripping. This is mainly linked to the water usage; surplus wastewater is generated in a range from 0 up to about 0.5 m³ per tonne of treated waste. Solids retention times varies between 2-30 days and sometimes more. Usually a higher number of treatment steps leads to shorter retention times as does also increased mixing. Observed first order rate constants vary much, examples of k-values for organic MSW presented by (Owens & Chynoweth 1993) was about 0.075 d⁻¹.

Methane production rates of 250-350 μmol m⁻² h⁻¹ accounting for about 20% of organic matter decomposition were observed in Lake Washington (USA) (Kuivila *et al.*, 1988). High methane production rates of 10 mol C m⁻² y⁻¹ were measured in Cape Lookout Bight (summer, North Carolina, USA) (Crill and Martens, 1986). A methane production rate of 0.6 mol C m⁻² y⁻¹ was measured in Skan Bay, Alaska, USA (Alperin *et al.*, 1992). Some first-order rate constants (1/y) for Cape Lookout Bight sediments (anoxic; 3-5% total organic carbon): acetate 300; free amino acids 7500; total hydrolyzable amino acids 1.4; total organic carbon 0.7 to 11; fatty acids 0.76; carbohydrates 0; lignin 0 (Crill and Martens, 1986; Burdige and Martens, 1988, 1990; Martens and Klump, 1987; Haddad and Martens, 1987; Haddad, 1989; see references for methods and scale of measurements)

Recalcitrance of organic carbon conversion

Depending on substrate and process the mass of the solid residues varies from about 20 to 70 % (by weight) of the treated solid wastes. In many cases, the 35-45 % range applies. As no external electron acceptor is utilised, the mean oxidation number of carbon will be similar in feed and products. A difference may develop between different products, if for example biomass generation is significant, the sludge may become more reduced (lower mean oxidation number of carbon) than a substrate dominated by cellulose. Different fractions of the solid wastes will be degraded, depending on retention time and process. *E.g.* it has been noted that about 15 % of the TS of biowaste from MSW can be washed out/mobilised rapidly (Assarsson *et al.* 1994) while the further degradation is slower.

Recalcitrance of organic carbon during early diagenesis is generally related to structural characteristics (polymerisation, branching, bonds which are not readily hydrolysed; heterocyclic, polycyclic compounds); also related to protection by biogenic mineral matrices; adsorption by sediment particles, incorporation by refractory macromolecules (humic substances), or complexation by metals (Henrichs, 1993 and numerous references cited therein). For example, in Buzzards Bay (Massachusetts, USA) sediment cores representing 200 years deposition, there was decomposition of only 40% of the total organic carbon, total hydrolysable amino acids, and fatty acids; there was no appreciable decomposition of the long-chain normal alkanes (Henrichs, 1993).

* AD endpoint limited to MSW digestion. Geologic endpoint limited to diagenesis of terrestrial organic matter (mainly plant material which evolves to "type III kerogen" as discussed below (Tissot and Welte, 1976). "Early diagenesis" generally limited to burial depths of 1 m or less over time frames of 1000 years or less (Henrichs, 1993); after that, generally only minor structural changes in organic matter with time until organic matter is subjected to elevated temperatures through kerogen pathways.

Geologic burial of organic carbon lies at the other end of the continuum shown in Figure 1. The processes encompassing the physical and chemical alteration of buried sediments are collectively termed "diagenesis." Preservation of organic carbon is promoted under conditions of anaerobic rather than aerobic burial. The global cycle of organic carbon can be reasonably divided into two sub-cycles: a short-term cycle where organic carbon is rapidly recycled back to atmospheric carbon dioxide; and a long-term cycle where some portion of the organic matter enters geologic storage for further transformations through kerogen pathways under elevated temperature and pressure conditions. The cross-over point between the two cycles, termed "early diagenesis" in the geochemical literature, is characterised by microbial decomposition processes at temperatures and pressures slightly above ambient. Such early diagenetic conditions are also characteristic of the landfill setting. (Tissot and Welte, 1976; additional discussion with reference to landfills in Bogner and Spokas, 1995). If we confine our discussion to terrestrial organic matter dominated by terrestrial plant materials with high percentages of lignocellulosic materials, we find similarities to municipal solid waste. Some general characteristics of this pathway are summarised in Table 1. Kerogen is indistinctly defined as the altered organic matter fraction which cannot be dissolved in organic solvents; it consists of condensed cyclic nuclei linked by heteroatomic bonds or aliphatic chains. Kerogen pathways refer to the chemical changes undergone by buried organic matter through diagenetic survival to become pre-cursors of fossil fuels at elevated temperatures and pressures. Elevated temperatures occur in environments of deep burial (hundreds to thousands of meters) or shallow burial where geothermal gradients are high. One type of kerogen (type III) is dominated by terrestrial plant materials (high percentages of lignocellulosics) and is the most relevant to issues relating to buried organic material in landfills. Chemical changes along the type III pathway mainly yield pre-cursors of vitrinite (coal) and thermogenic natural gas. A number of analytical tools (*e.g.*, vitrinite reflectance, infrared spectroscopy, and many others) combined with geochemical modelling are used to evaluate diagenesis.

Figure 2 summarises the products of some major microbial decomposition reactions affecting organic carbon cycling in digesters, in landfills, and during early diagenesis. For simplicity, numerous interacting nutrient (N,P,K), sulfur, and other cycles are not shown. The figure is divided into anaerobic and aerobic sides with feed-backs pertaining to the several settings in which carbon dioxide is produced and consumed. The complexities of carbon dioxide cycling can be seen from 1) production through direct oxidation of organic carbon (including root zone respiration), anaerobic decomposition reactions, and methane oxidation; and 2) consumption during methane production via a carbon dioxide reduction pathway with hydrogen. The anaerobic side is well-known in the microbial ecology literature as a depiction of substrate flow through methanogenic ecosystems (Zehnder *et al.*, 1982), where acetate and carbon dioxide are the major precursors to methane.

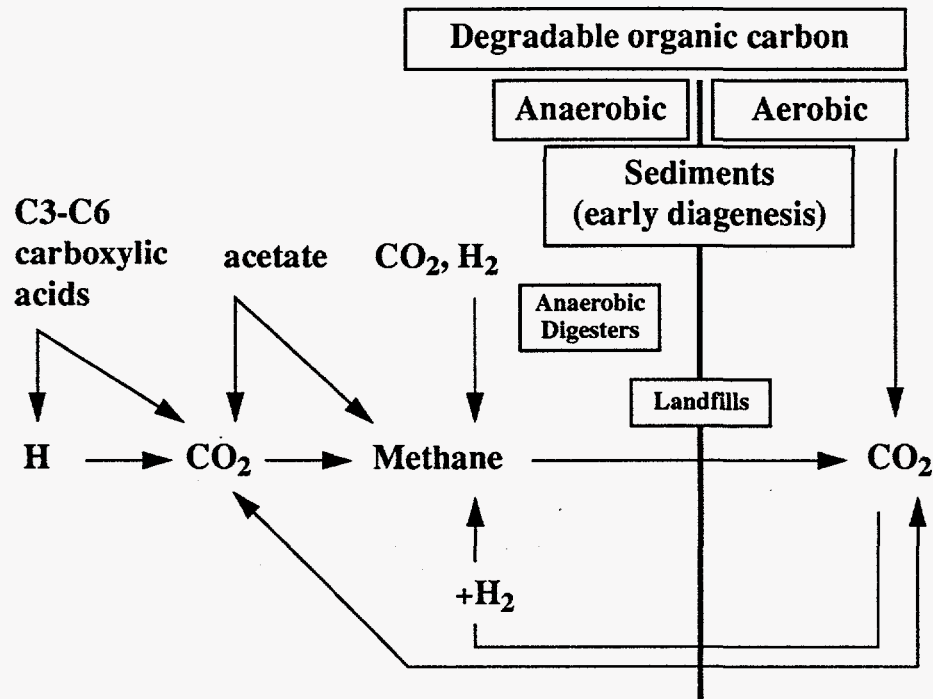


Figure 2. Some major microbial pathways for organic carbon cycling in landfills, anaerobic digesters, and buried sediments (early diagenesis).

2. ORGANIC CARBON CYCLING IN LANDFILLS

As indicated in Fig 2, the conversion of organic carbon in landfills takes place both under aerobic and anaerobic conditions. However, most of the mobilisation of carbon will usually occur under anaerobic conditions, since the mass flow of oxygen into the landfill is much too limited to support a complete aerobic degradation. The anaerobic degradation of organic biopolymers in landfills lead to similar degrees of degradation as AD or early diagenetic processes, but the rates of conversion are different. Often first order models are used to model the conversion of organic materials in landfills. The basic assumption of such models is that the degradation is limited by substrate availability, *i. e.* a high initial gas generation is followed by an exponential decrease over time. However empirical results indicate differently, Augenstein (1996) analysing gas abstraction from 17 landfills, where the abstraction was maximised, found that zero order and first order models all had a similar predictive ability. Results from the Brogborough test cells in the UK indicate linear increases of specific gas generation over the first five years (Knox 1997). Data from test cells at Helsingborg indicate almost constant gas generation over the first years (Lagerkvist 1987). There are several reasons why such results are fairly reasonable. To begin with, waste data from full scale landfills are rarely of high precision and assuming a high precision of gas abstraction data, the abstraction will not equal gas generation. Emission measurements at Swedish test cells indicate that the carbon loss over the landfill surface may reach as high as up to about 30 % of the total carbon mobilised even at specific methane abstraction levels as high as 3-5 Nm³/tonnes of fresh waste and year. (Lagerkvist & Maurice 1996). With regard to the degradation processes, a number of factors may limit substrate utilisation, *e.g.* the availability and distribution of water and methanogenic microorganisms. Different barriers like

polyethylene bags will contribute to heterogeneity in the landfills. Methanogenesis may also be inhibited in landfill environments over extended periods through the build-up of high concentrations of organic acids (Lagerkvist 1994). Table 2 gives a sampling of data on carbon conversion in landfill environments. The data all originate from test cell studies.

Table 2. Data on conversion of organic material in landfill test cells. If not otherwise indicated, the data are from Glaub *et al.* (1986) for Mountain View, (ETSU 1996) for Brogburrough and Lagerkvist (in preparation) for the Co-ordinated Swedish test cell programme.

| | Mountain View | Brogburrough | Co-ord. Swedish |
|--|--------------------------|--------------|------------------------|
| Number of cells | 6 | 6 | 12 |
| Time [y] ^a | 5 | 7 | 2.3-4 |
| Waste/cell [Gg of TS] | 5.3-6.5 | 9.5-11 | 2-8 |
| TS at start [%] | 54-72 | 66-69 | 53-72 |
| Gas Yields [kmol Mg ⁻¹ y ⁻¹] ^b | 0.44-1.2 | 0.33-0.54 | 0.27-0.79 ^e |
| Av. CH ₄ content [vol-%] | 55-57 | 54-57 | 35-50 |
| TS [%] | 31-67 ^c | | 43-82 ^f |
| VS [% of TS] | 32-51 ^{c,d} | | 27-88 ^f |
| Temperature [°C] | 45-60 | 25-40 | 15-35 |
| BMP [kmol Mg ⁻¹ of TS] | 0.98-5.4 ^{c,d} | | 1.5-7.5 ^g |
| Cellulose [% of TS] | 16.3-32.8 ^{c,d} | | 8-40 ^g |
| Cellulose/Lignin [-] | 1.2-2.4 ^{c,d} | | |

^a Number of years studied ^b refers to initial wet waste mass ^c after five years ^d (Knox 1997) ^e Normalised to 50 % CH₄ ^g sampled after 2-4 years ^f sampled after 2-3 years.

3. A NEW PERSPECTIVE ON MODELLING OF LANDFILL CARBON CYCLING IN LANDFILLS

A major conclusion to be drawn from Tables 1 and 2 is that, while previous modelling of landfill processes has drawn heavily on the anaerobic digestion literature, it is also appropriate to develop process models similar to those used in more open geologic settings. The anaerobic digestion "ideal" becomes increasingly more relevant for optimised "biofill" landfills, but a well-mixed digester concept may be totally inappropriate for many conventional controlled landfills with non-shredded or baled refuse, high compaction rates, discrete cell construction, or "semi-aerobic" operating practices. The digester concept would also be inappropriate for uncontrolled landfills in many places where high rates of aerobic decomposition occur.

The more open-ended approach used in modelling geologic and soils systems typically proceeds from a general diagenetic equation in which the flux of a given component is referenced to a critical horizon, permitting the development of open-system equations which describe the various diffusive and advective fluxes plus diagenetic reactions in various liquid, solid, and gaseous systems (Berner, 1983). Although the resulting models could be extremely complex, in practice there is an emphasis on critical controlling variables and diagnostic indicators of specific processes using focused field data. This approach is attractive for landfill settings for several reasons. First, it presents a logical open system framework in which to consider liquid, solid, and gaseous

fluxes and reactions. Within this framework, there is a great deal of flexibility with respect to the complexity of processes included. Secondly, general diagenetic equations can be adapted to any interface of interest in a landfill setting (1D) or expanded to a whole-landfill model (3D). Third, such equations are similar in form to widely-used contaminant transport equations which consider advection/dispersion and biodegradation of specific contaminants in open geologic systems, thus encouraging combined models for specific components of interest (chlorinated compounds, for example) within larger landfill process models.

There are also landfill situations where the anaerobic digestion model may be appropriate. For example a biofill of finely ground, homogenous substrates operated under high hydraulic load would resemble some commercial "dry" batch reactors, and may be the closest a landfill will get to the "digester endpoint". However it still has to be demonstrated that such systems can operate consistently over a degradation cycle, *i. e.* with an even distribution of water through the solid wastes.

A prime consideration for any model is scale. A family of landfill models is needed at various scales to more realistically address landfill processes. We suggest that existing landfill models have focused on equations more appropriate for a closed system at microbial scales but have been applied to whole-landfill processes in an open system. Some guidance for landfill systems can be found in recent literature discussing proposed scaling for spatial domains in ecological models. For example, Holden and Firestone (1997) criticised current biodegradation models, which are typically based on interactions between defined domains (boxes) of bacterial physiology, mass transfer, and environmental variables (pH, temperature, nutrients, redox). Such separate domains have encouraged consideration of numerous processes at whatever scale is chosen and tended to suppress improved selectivity using ecological considerations of scale. Instead, they proposed a nested hierarchy of models at various scales within a microbial ecology framework, where scale dictates both the selection and magnitude of various controlling processes, as shown in Figure 3. This figure could be used to guide development of a family of improved landfill models at various scales that may be more useful in field settings than either complex all-encompassing models or simplistic one-equation models. For example, if modelling degradation of a specific chlorinated hydrocarbon at the microbial level, the kinetics of specific enzymatic reactions may largely define the model framework. Conversely, when modelling organic carbon cycling in the context of overall landfill processes at the landscape or ecosystem level, it may be more appropriate to consider bulk mass transfer (through use of diffusional/advective transport terms and empirical field relationships) without resorting to microbial kinetic approaches. There have been several studies where such an approach has proven useful at the ecosystem level (for example, see references in Table 1 for studies at Cape Lookout Bight, North Carolina).

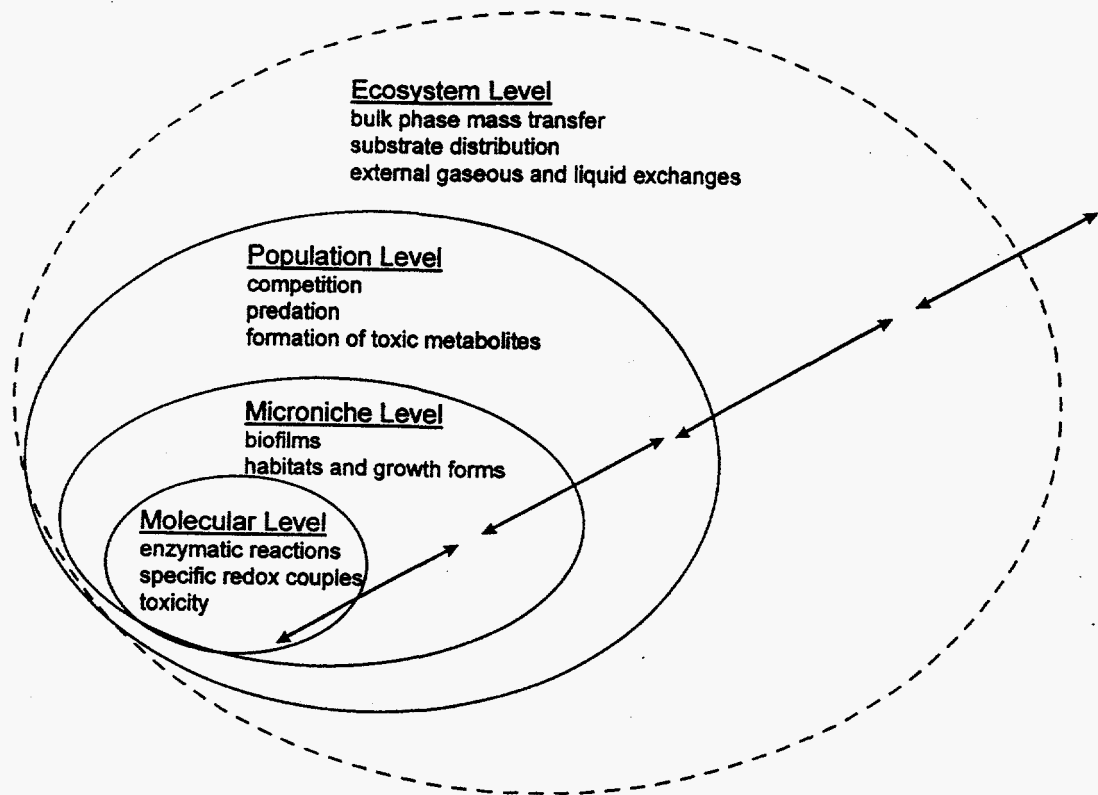


Figure 3. Suggested domains based on microbial ecology that could be used to develop a family of improved landfill models at various scales. (Modified from Holden and Firestone, 1997).

4. CONCLUSIONS: NEW APPROACHES FOR IMPROVED MODELS

A family of more realistic landfill models are needed to address internal landfill processes in the context of various landfill designs and operating strategies. For many less controlled landfill settings, geologic models for early diagenesis of organic carbon may be more appropriate than digester models. Important issues for landfilling over extended time frames include: (1) ecological considerations of long-term storage and mobilisation of organic carbon, often with considerable anthropogenic pre-processing and post-depositional alteration; (2) human health aspects of extended habitation near land burial of waste under various degrees of control; and (3) economic issues of achieving a specified degree of control at a reasonable cost. In particular, models are needed which include:

- o More rigorous consideration of scale
- o More realistic simulation of field data (some empirical terms allowed) for improved simulation of natural variability (*e.g.*, several orders of magnitude for methane generation, oxidation, and net emission processes; Bogner *et al.* 1997).
- o More use of selective site-specific data to validate models at various scales.

Such models could provide improved guidance for risk assessment, ecological issues, and economic issues related to landfilling practices. For example, with respect to risk assessment, the scale determines the population affected; more realistic models can better address short-term vs. long-term affects; and selective site-specific data can provide a better database to address the

health of landfill workers, scavengers, and plant and animal populations. Ecological issues related to modelling have been discussed above; however, modelling at various levels (microbial to landscape) must be coordinated with a particular scale of event and to site-specific data. The scale of economic modelling is usually a local scale, since landfilling in many countries is guided by waste disposal decisions made at the local level. Concurrently, economic models would benefit from community-specific data.

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