Mineralization of Monomeric Components of Biodegradable Plastics in Preconditioned and Enriched Sandy Loam Soil Under Laboratory Conditions

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Abstract In the last 20 years, a new generation of materials was developed: the biodegradable plastics. They reduce the accumulation of plastic in the environment and the cost of waste management because they can be fed in composting plants or, if used in agriculture (mulch films), they are applied to the soil and left there. Ten monomers were chosen among the most used in the synthesis of biodegradable polymers (1,2-ethanediol, 1,4-butanediol, 1,6-hexanediol, adipic acid, azelaic acid, sebacic acid, terephthalic acid, glucose, lactic acid, and succinic acid) and tested according to ASTM 5988-96 (a standard test method for determining aerobic biodegradation in soil of plastic materials measuring the carbon dioxide evolution). Two sandy loam soils, collected in two different sites in Italy, were used to evaluate the mineralization rate of the monomers. Four tests (two replicates each) were carried out for 27–39 days. Experimental data show no relevant differences in the respirations of the two soils and in the carbon dioxide productions of the tested monomers. The final mineralization percent was 42–45% for glucose, succinic, and lactic acid and 50–56% for the other monomers.

Keywords Biodegradable plastic · Biopolymer · Monomer · Mineralization

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1 Introduction

Plastic is a general common name given to synthetic, organic, and high molecular weight polymers suitable for the manufacture of industrial products. It refers to their malleability or plasticity during manufacture that allows them to be cast, pressed, or extruded into an enormous variety of shapes and objects (films, fibers, plates, tubes, bottles, boxes, and much more) which found wide applications in every aspect of life and industries. Plastic objects are easy to produce, have high performances, and generally cannot be attacked by microorganisms and so they are not biodegradable. A problem in their use is the accumulation in the environment. To overcome this problem, the efforts of academic and industrial worlds have joined to produce a new generation of plastic materials: the biodegradable plastics, often made of polyesters.

Besides reducing the risk of accumulation of plastic materials in the environment, the production and use of biodegradable polymers involves considerable reduction in the cost of waste management (Tokiwa et al. 2009). Furthermore, biodegradable plastics can be recycled to useful metabolites (monomers or oligomers) by microorganisms.

In the last 20 years, the increasing development of biodegradable plastics has promoted initiatives to develop formal standards and laboratory test methods to assess the ultimate environmental behavior of plastics (Chiellini et al. 2007). Several international and national organizations have issued standard test methods to simulate the fate of biodegradable plastics in different environments. In the 1990s, most of the work was focused on biodegradation under composting conditions (ASTM D6400 2004; ASTM D6868 2003; UNI EN 13432 2002). The reason for this preference was linked to the concurrent trend in solid waste management policy, which aimed at reducing the use of landfill to as least as possible while promoting recycling. Consequently, criteria and standard test methods were needed in order to verify the compatibility of plastics with composting (Degli Innocenti 2005). In the so-called controlled composting tests, the material is mixed in mature compost, incubated at 58°C, and the CO_2 evolution is monitored. The material must disintegrate sufficiently during the composting process, and at least 90% of its organic fraction must be converted into CO_2 within 6 months. The test material must not have negative effects on the composting process, the compost quality must not be modified, and no toxic effects should occur (UNI EN 13432 2002). With the increasing use of biodegradable plastic mulch films in agriculture, the study of the biodegradation in soil has received intense interest, but the standardization of test methods is difficult (Mueller 2003). Soil is a complex matrix: Its natural properties can be very different in different sites and cannot be controlled in nature. In soil, the biological activity and the temperatures are lower than in composting conditions, so biodegradation in soil is slower than in compost. However, the agricultural soil is the medium for the production of food for humans and cattle, so standard test methods and specific criteria to verify the biodegradability and the absence of eco-toxic effects in soil are required (Degli Innocenti 2005). In 1996, a standard method was published for determining

aerobic biodegradation in soil of plastic materials by measuring the amount of carbon dioxide evolved in a closed respirometer (ASTM 1996). In 2003, ISO published a standard method for determining the biodegradation rate of plastic material in soil by measuring the oxygen demand or the amount of carbon dioxide evolved in a ventilated respirometer (ISO 17556 2003).

The above-mentioned standard methods have some limits. In particular, as observed by Briassoulis and Dejean (2010), they do not impose prerequisites for soil, and their results depend on the soil used for the test. Therefore, their application for a general description of the biodegradation process cannot always be accepted. The problem should be solved by developing international guidelines taking into account the transferability of results to different soils under real conditions and the soil quality at the end of the test.

Plastic films obtained from biodegradable polymers can replace the traditional ones commonly used in agriculture for soil mulching. During their use in field, biodegradable films must ensure the same performances of the traditional ones, but at the end of their life they can be directly left in soil or disposed in composting plants as normal organic waste (Kapanen et al. 2008). Different authors (Kapanen et al. 2008; Kijchavengkul et al. 2008a; Briassoulis 2007) proposed field studies on biodegradable polymer available on the market (Mater-Bi or Ecoflex) in order to verify their applicability in mulching. Generally in these tests, the physical, thermal, and mechanical properties of the applied films, their resistance to UV exposure, or their disintegrability was taken into account. The biodegradability that occurs when films are mixed to the soil was measured as mineralization percent under laboratory composting conditions (Kijchavengkul et al. 2008b) or evaluated indirectly as weight loss of buried samples of the films under investigation in the soil under real field conditions (Briassoulis 2007).

The biodegradation of polymers is normally referred to as an attack by microorganisms on non-water soluble polymer-based materials (plastics; Mueller 2003). It is a complex process in which the carbon of the polymer is converted into carbon dioxide (mineralization) and biomass. Extracellular enzymes (i.e., enzymes released by microbial cells into the surrounding environment) are secreted by the organisms performing the first step of the degrada-

tion. In case of polyesters, enzymes are expected to catalyze the hydrolysis of the ester bonds and cut the solid, hydrophobic polymer into oligomers and monomers, which are released in the environment. Due to their low molecular weight and water solubility, these can pass through the cell membrane and be then metabolized by microorganisms (Tokiwa and Suzuki 1974, 1977; Herzog et al. 2006; Mueller 2006). The simple depolymerization can actually lead to the buildup of environmental concentrations of oligomers and monomers and to their transfer from one environmental component to another (Degli Innocenti 2005).

The aim of this work was to evaluate the mineralization in soil of different monomers which are generally used in the production of biodegradable plastics. Two sandy loam soils were used in order to test ten monomers according to ASTM 5988-(96), the above-mentioned standard method for determining aerobic biodegradation in soil of plastic materials.

2 Materials and Methods

2.1 The Tested Substances

Ten monomers, used in the synthesis of potentially biodegradable polymers, were tested by respirometric test in soil: 1,2-ethanediol, 1,4-butanediol, 1,6-hexanediol, adipic acid, azelaic acid, sebacic acid, terephthalic acid, glucose, lactic acid, and succinic acid.

1,2-Ethanediol ($C_2H_6O_2$) It is an organic compound widely used as automotive antifreeze and as raw material in plastic production. It is an odorless, colorless, and sweet-testing liquid produced from the reaction between ethylene oxide and water. The reaction can be catalyzed by either acids or bases, or can occur, at neutral pH, at high temperatures (Rebsdat and Mayer 2002). The ethanediol is an important monomer used in the synthesis of polyester fibers and resins: Polyethylene terephthalate used to make plastic bottles for soft drinks (for example) is prepared from ethylene glycol. It is one of the constituents of Sky Green (made of adipic acid, succinic acid, butanediol, and ethylene glycol), a biodegradable polymer produced by SK Chemicals (Korea; Lee et al. 2002).

1,4-Butanediol ($C_4H_{10}O_2$) It is a colorless and viscous liquid derived from butane by placement of alcohol groups at the end of the chain. In its industrial synthesis, 1,4-butanediol is produced with the method of reacting acetylene with formaldehyde (Reppe process; Küksal et al. 2002), or, with an alternative bio-based process, from corn-derived glucose. Glucose is fermented to succinic acid which is then purified and reduced catalytically to 1,4-butanediol (Cooper and Vigon 2001). It is a constituent of biodegradable plastics such as Ecoflex (polybutilene adipate and/or polybutylene therefthalate) produced by BASF (Steinbuchel and Doi 2002) and of the previously mentioned Sky Green.

1,6-Hexanediol ($C_6H_{14}O_2$) It is a white solid organic di-alcohol, with two primary terminally located hydroxyl groups. It is a valuable intermediate product for chemical industry, and it finds applications in a variety of polymeric systems. Its configuration results in a rapid and simultaneous reaction in the formation of numerous di-substituted products. 1,6-Hexanediol is used in the production of polyesters, coatings, adhesives, and polymeric plasticizers. In these end-use areas, it contributes significantly to many high-performance characteristics such as hydrolytic stability, high flexibility, good adhesion, and surface hardness (BASF intermediates description available in internet). It is used in the synthesis of biodegradable high molecular weight aliphatic–aromatic copolyesters (Li et al. 2009).

Adipic Acid ($C_6H_{10}O_4$) It is a white crystalline powder not very soluble in water because of its long aliphatic chain. It is a synthetic molecule normally prepared from cyclohexane by two oxidation steps, which can also be produced starting from natural raw materials (Asahi Kasei Kogyo 1991). Adipic acid is largely used as a monomer for the production of nylon, but also as plasticizer and lubricant component. It is a constituent of biodegradable plastics such as Ecoflex and Sky Green.

Azelaic Acid ($C_9H_{16}O_4$) It is a saturated dicarboxylic acid naturally present in wheat, rye, and barley or produced by *Malassezia furfur* (Ashbee and Evans 2002), a yeast that lives on normal skin. For its antibacterial properties, azelaic acid is used for the treatment of skin irritations such as acne (Liu et al. 2006). It is also used in the production of plasticizers, polyamides, and alkyd resins.

Sebacic Acid ($C_{10}H_{18}O_4$) In its pure state, sebacic acid ($C_{10}H_{18}O_4$) is a white flake or powdered crystal natural substance. For industrial purposes, it is derived from castor oil and is typically used for the production of candles as well as of plasticizers, lubricants, and cosmetics, besides and bio-based plastics.

Terephthalic Acid (C_8H_6O) It is an aromatic colorless commodity chemical used mainly as a precursor to the non-biodegradable polyester such as polyethylene terephthalate (used to make bottles or clothes) or polybutylene terephthalate. Its aromatic component provides excellent material properties, so terephthalic acid is frequently used to improve material properties in polyesters such as Ecoflex (Mueller et al. 2001).

Glucose ($C_6H_{12}O_6$) It is the most widespread monomer in nature; it is a simple sugar produced by photosynthesis and used as energy source by respiration. Cellulose, the most common polymer in nature, derives from the dehydration of glucose, and polymers from cellulose are used as biodegradable plastics (e.g., cellophane).

Lactic Acid ($C_3H_6O_3$) It is an α-hydroxy acid involved in biochemical processes. It can be produced both by chemical synthesis and by fermentation of carbohydrates by *Lactobacillus* (Sotergard and Stolt 2002). It is used as a monomer for producing polylactic acid, which has application as a biodegradable plastic.

Succinic Acid ($C_4H_{16}O_4$) It is a solid colorless and odorless dicarboxylic acid which plays an important metabolic role in the citric acid cycle by which organisms draw energy. It is a constituent of polybutylene succinate or polybutylene succinate-*co*-butylene adipate copolymers, commercially known as Bionolle, produced by Showa Highpolymers (Japan; Tserki et al. 2006). The main properties of the tested substances are reported in Table 1.

2.2 Soil Samples

Two different sandy loam soils were sampled in two different locations and used to test the monomers. The texture of the first one (collected in Albenga, Italy) is made of about 70% sand, 24% silt, and 6% clay and its pH (in water) is 7.5 to 8. In the second one

(collected in Arborio, Italy) ,sand is about 55%, silt 43%, clay 2%, and pH (in water) 5 to 6.5. The soil samples were freshly collected, sieved (<2 mm) and used within a few days for biodegradation tests.

2.3 Biodegradation Tests

Biodegradation tests were carried out according to ASTM 5988-96 (ASTM 1996). To increase organic matter, soil was enriched with compost with a ratio of 1 g compost to 25 g soil which corresponds to a typical application of compost in agricultural land (ASTM 5988-96). Mineral salts dissolved in water were added to soil and compost to obtain the correct ratio of nutrients and the ideal moisture, around 50% of the water holding capacity of each soil. Mineral salts addition was adjusted to provide 0.2 g KH_2PO_4, 0.1 g $MgSO_4$, 0.4 g $NaNO_3$, 0.4 g NH_4Cl, and 0.2 g urea kg^{-1} of soil.

Soil–compost–salt mixtures (500 g) were incubated at room temperature (21±2°C) in the dark, in hermetically closed jars (3 l), with the test substances. Blank jars, with no test substance, were also prepared. Each jar contained a beaker filled with 0.5 M KOH (40 ml), which was regularly titrated with 0.25 or 0.5 M HCl in order to measure the CO_2 production within the jar. The measurement was carried out every 3 days in the first 2 weeks, during which biodegradation was expected to be faster, and weekly thereafter. When the beakers were taken away from the jars for titration, the jars remained open from 15 to 30 min, so that the air was refreshed before replacing fresh potassium hydroxide.

Tests lasted 27 to 39 days, according to the cumulative CO_2 evolution. Only for terephthalic acid the test duration was extended to 140 days because no plateau phase was reached after 40 days. Moisture was not adjusted during the biodegradation tests. In hermetically closed jars, water evaporating from the soil saturates the headspace in a very short time, and consequently, any further water loss is negligible; therefore, the soil moisture can be considered as constant during the test period.

Five monomers (adipic acid, succinic acid, sebacic acid, 1,4-butanediol, and glucose) were tested both with soil from Albenga and with soil from Arborio. 1, 2-Ethanediol, lactic acid, 1,6-hexanediol, azelaic acid, and terephthalic acid were tested only with soil from Arborio. On the whole, four tests (two replicates each) were carried out for each monomer and for the blanks.

Table 1 Main properties of the tested monomers

Monomer	Molecular formula	Molar mass (g mol^{-1})	Carbon fraction (−)	Physical state at lab conditions
1,2-Ethanediol	$C_2H_6O_2$	62	0.39	Liquid
1,4-Butanediol	$C_4H_{10}O_2$	90	0.53	Liquid
1,6-Hexanediol	$C_6H_{14}O_2$	118	0.61	Liquid
Adipic acid	$C_6H_{10}O_4$	146	0.49	Solid
Azelaic acid	$C_9H_{16}O_4$	188	0.57	Solid
Sebacic acid	$C_{10}H_{18}O_4$	202	0.59	Solid
Terephthalic acid	$C_8H_6O_4$	166	0.58	Solid
Glucose	$C_6H_{12}O_6$	180	0.40	Solid
Lactic acid	$C_3H_6O_3$	90	0.40	Liquid
Succinic acid	$C_4H_6O_4$	118	0.41	Solid

2.4 Chemicals and Analytical Methods

All chemicals were of analytical grade purity. Adipic acid, azelaic acid, 1,6-hexanediol, sebacic, and succinic acid were supplied by Gamma Chimica S.p.a. 1,4-Butanediol, 1,2-ethanediol, lactic acid, glucose, and terephthalic acid were provided by Sigma-Aldrich. Moisture content and pH of the soil–compost–salt mixture were measured according to ISO 11465 (ISO 1994a) and ISO 10390 (ISO 2005), respectively. Titration was carried out according to Standard Methods (APHA 1998).

3 Results

In a first set of experiments, five monomers were tested with the two different soils: Albenga soil and Arborio soil. The respiration curves (cumulative CO_2 production (milligrams) measured in the jars) of the blanks and of the tested monomers (adipic acid, sebacic acid, succinic acid, 1,4-butanediol, and glucose) are shown in Figs. 1 and 2, respectively. Black symbols refer to tests in Albenga soil and white symbols to tests in Arborio soil.

Experimental data show that CO_2 produced by the respiration in the blank jars is quite the same for the two soils. The same situation can be observed for the tested monomers. The most important difference of the two soils is their natural pH. It is known that, as temperature and soil moisture do, pH affects the final biodegradation in soil of polymeric materials (Shin and Eun 1999), but the present results show no

relevant difference in the respirations of the two soils and in the CO_2 production measured for the tested monomers. So, the final extent of mineralization was calculated as average from the experimental data obtained with both soils.

Figure 3 reports the mineralization curves for all ten monomers. Each point represents the experimental value obtained in each test, and the broken line represents their best fit. Table 2 reports the average mineralization percent at the end of the tests for each monomer. The reproducibility of the results is good, as shown by the low standard deviations always below 10%.

The smallest variations in the distribution of data were observed for the three di-alcohols (1,2-ethanediol, 1,4-butanediol, and 1,6 hexanediol) and for lactic acid. Their physic state, liquid at room temperature, and their
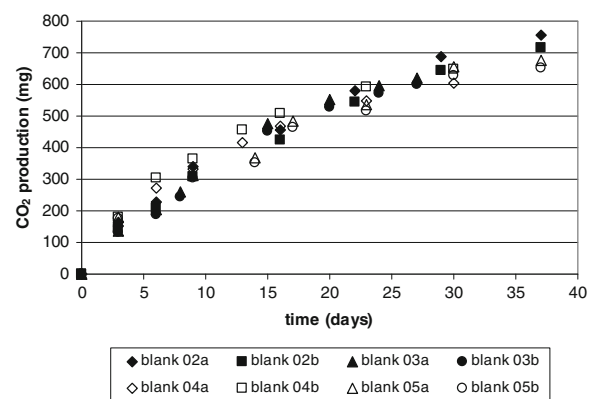

Fig. 1 Respiration curves measured for soil Albenga (in *black*) and Arborio (*empty symbols*) in the blank jars
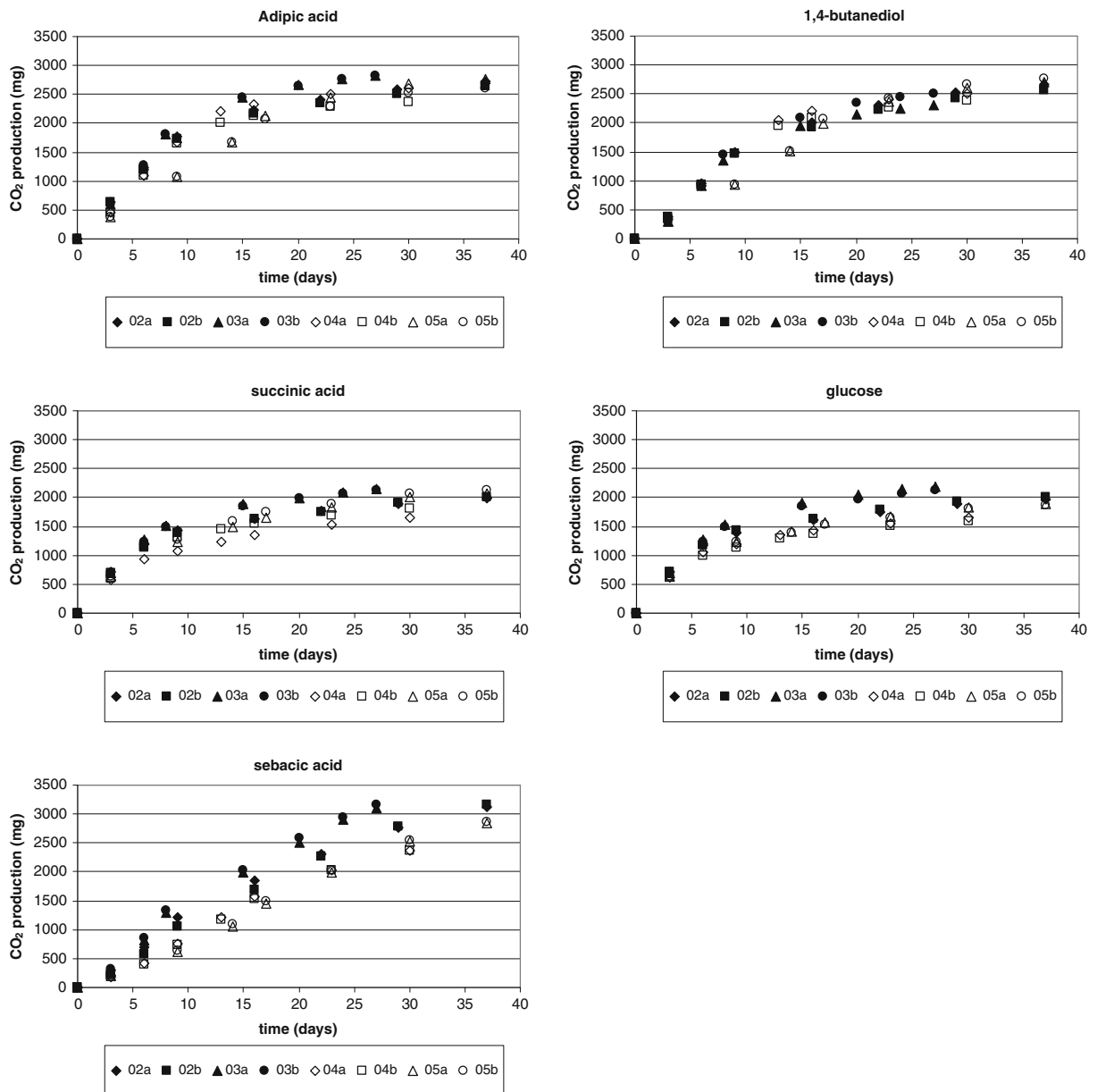
Fig. 2 Respiration curves of adipic acid, 1,4-butanediol, succinic acid, glucose, and sebacic acid obtained during incubation in Albenga soil (in *black*) and in Arborio soil (*empty symbols*)

high solubility in water favor their homogeneous distribution in the liquid phase of soil and their use by microorganisms. This could explain the high reproducibility of the results in the different tests.

Generally, a plateau phase was reached within about 3 weeks at more than 50% mineralization. A lag phase of about 3 to 4 days was observed for sebacic acid, 1,6-hexanediol, azelaic, and terephthalic acid whose final mineralization was about the 53%, 55%,

54%, and 56%, respectively (as previously mentioned, the final mineralization of terephthalic acid was estimated after 140 days incubation; after 40 days mineralization was below 40 %). For adipic acid, the final mineralization was comparable (55 %), but no lag phase was observed.

The faster mineralization was observed for glucose, lactic, and succinic acid for which the plateau phase was reached in about 10 days, but the final mineralization
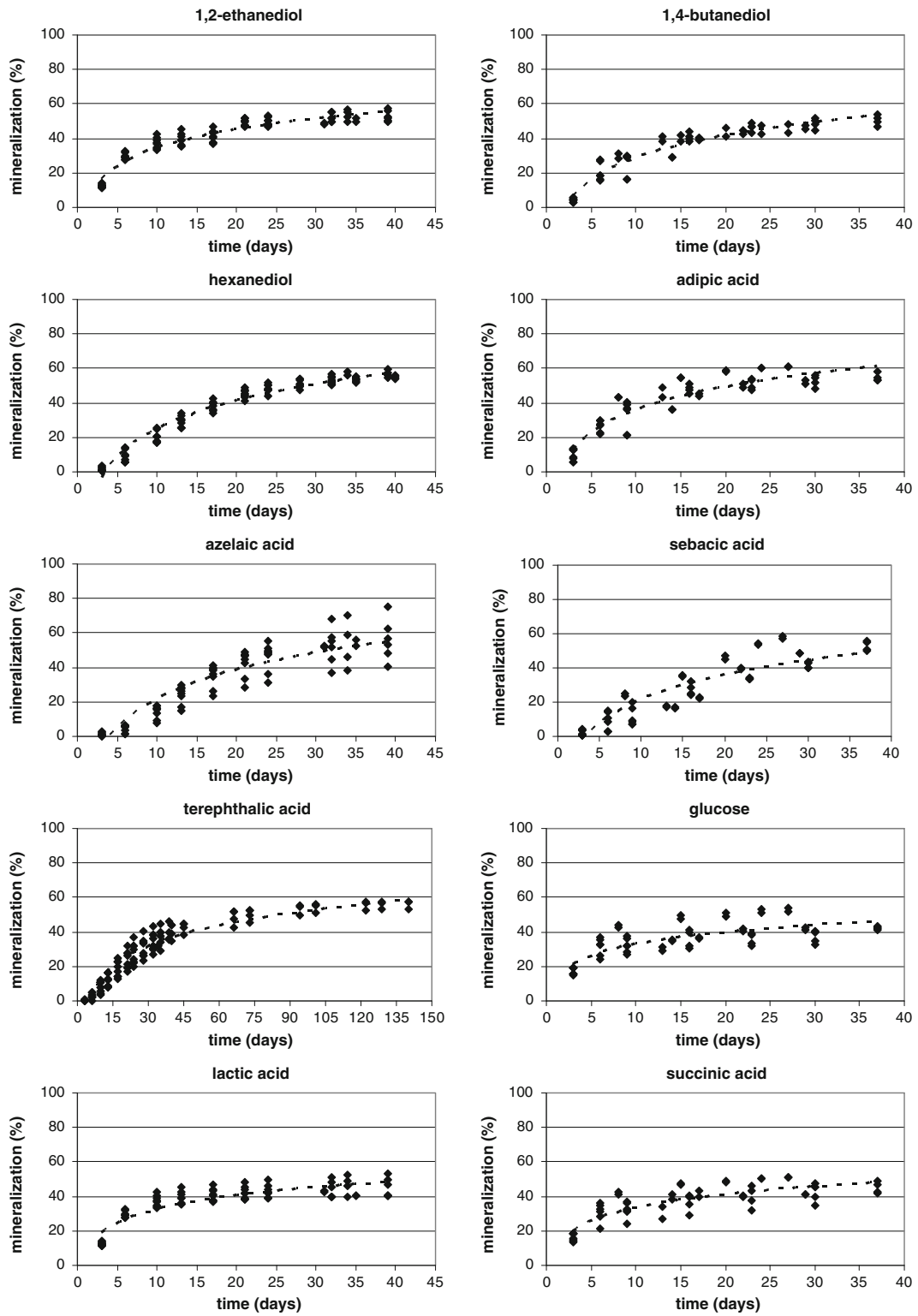
Fig. 3 Mineralization curves of the tested substances

Table 2 Mineralization percentages and standard deviations at the end of mineralization tests

Monomer	Mineralization (%)
1,2-Ethanediol	53.05±3.15
1,4-Butanediol	50.47±3.15
1,6-Hexanediol	55.87±1.88
Adipic acid	54.78±2.26
Azelaic acid	53.86±8.35
Sebacic acid	52.71±2.90
Terephthalic acid	55.94±2.45
Glucose	42.22±0.95
Lactic acid	45.26±5.52
Succinic acid	45.17±3.39

was lower than for other tested monomers (45% for lactic and succinic acid and 42% for glucose). For sebacic and azelaic acid, the plateau phase seemed to be reached by the end of the tests, and the mineralization was about 53% and 54%. It is possible that for such monomers, longer lasting tests could show better defined plateau and greater mineralization percents, as it was the case for terephthalic acid.

4 Discussion and Conclusion

In this kind of research, an important point is the influence of soil and its characteristics on the final results of mineralization. Two types of sandy loam soils with different natural pH (alkaline the first, sub-acid the second) were used in this study. The results obtained in this work reveal that the two soils sustained similar mineralization curves of tested monomers.

The level of mineralization reached by the different substances is not complete. Mineralization percent was 42–45% for glucose, succinic, and lactic acid and 50–56% for the other monomers. This is expected considering microbial anabolism. The carbon fraction which is not released as CO_2 is incorporated in the cell biomass. A simulation carried out by AQUASIM code estimated that cellular biosynthesis could account for over 50% for lactic acid, succinic acid, and glucose (Siotto et al. 2011).

The availability of data on the biodegradation of monomers in soil is limited, as the attention is generally focused on the biodegradation of polymers.

However, the possibility of microorganisms to use the monomers derived from the biodegradation of polymers and the way they use them can have important implications on the one hand in stimulating the overall microbial activity and on the other hand in determining oxygen consumption in soil. Some research studies about microorganisms that use the tested monomers are available in literature. Terephthalic acid, for example, is used by three bacterial strains isolated from soils contaminated with oil and plastic waste (Vamsee-Krishna et al. 2006). Orchard and Goodfellow (1980) reported strains of *Nocardia* which can grow on adipic and sebacic acid as sole carbon source. Moreover, 1,4-butanediol, 1,2-ethanediol, and adipic acid are used by *Pseudomonas* strains isolated from soil (Stieglitz and Weimer 1985).

The comparison of the present experimental data to literature data is quite difficult because, where data are available and experimental protocols are similar, specific aspects such as some soil properties or the time duration of the tests can affect the results. Kim et al. (2001) and Sharabi and Bartha (1993) measured mineralization in soil of some of the monomers studied in present research and obtained, for some compounds, higher mineralization percent values. However, Kim et al. (2001) used a mixture of forest/agricultural soils and perlite as substrate for incubation and carried out the tests for 32 days. This mixture was used as the core in a multilayer system where the bottom and the top layers were made of perlite. The mineralization percent obtained by Kim et al. (2001) was comparable to that observed in the present study (27–45 days) for adipic acid and 1,4-butanediol and higher for succinic acid (+19%) and terephthalic acid (+10%). Sharabi and Bartha (1993) used a freshly collected sandy loam, buffered with $CaCO_3$ 5 days before the beginning of the tests, and measured the CO_2 evolution over 22 days. Soil moisture was adjusted to 60% of field capacity and nutrients were added by a 1% solution of $(NH_4)_2PO_4$. In that case, the mineralization percent was higher for both adipic acid (+40%) and glucose (+36%).

Standard respirometric methods are important tools for evaluating the mineralization of the tested compounds in soil, but the evaluation of their results must take carefully into account the specific experimental conditions. So, the reported results can be held as representative of the mineralization process in preconditioned and enriched sandy loams having the described properties, under laboratory conditions. This should be

considered and taken into account when the biodegradation of new generation materials and of their possible by-products (the monomers) is studied. As observed by Briassoulis and Dejean (2010) in a recent critical review, the presently available standard test methods have been developed for studying the biodegradation under laboratory controlled conditions, and their results could be not extended to all soil environments. Moreover, they cannot fully describe the biodegradation process which includes also biomass incorporation. This component can be of particular interest in the case of monomers and should be measured along the biodegradation tests by specific protocols to provide the needed data for a complete carbon balance.

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