STRATEGIES TO IMPROVE AND PROTECT SOIL QUALITY FROM THE DISPOSAL OF OLIVE OIL MILL WASTES IN THE MEDITERRANEAN

LIFE - PROSODOL

LIFE07 - ENV/GR/000280

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RESULTS AND ACHIEVEMENTS OF A 4-YEAR DEMONSTRATION PROJECT

"This book was created by the scientific team of Soil Science Institute of Athens-ELGO DEMETER in cooperation with the scientific teams of the Center for Agricultural Experimentation and Assistance (CERSAA), Department of Soil and Water Conservation and Organic Resources Management of CEBAS-CSIC, the Technical University of Crete (TUC) and of Institute of Mediterranean Studies-FORTH

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SUMMARY

This book contains objectives and achievements of the LIFE07 ENV/GR/000280 project "Strategies to improve and protect soil quality from the disposal of Olive Oil Mill Wastes (OMW) in the Mediterranean region-PROSODOL".

The project's overall objectives were to develop and disseminate innovative, environment friendly, low cost technologies for the protection of soil and water from pollution caused by the disposal of olive oil mills' wastes, to design; implement and support a monitoring system for the assessment of soil and water quality affected directly or indirectly from mills' activities in relation to factors pressures and responses; to identify potential safest uses of mills' wastes in the agricultural sector and to establish an info-library/knowledge base system to assess environmental impacts from olive oil mills' wastes to Mediterranean region.

All foresaw activities were implemented at two demonstration-pilot areas; one in Greece and one in Italy. The Greek demonstration area is located in the municipality of Rethymnon (former Municipality of Nikiforos Fokas), in the north of Rethymnon prefecture, Crete. The selected pilot municipality is one of the many municipalities in Greece but also in Mediterranean facing the same problem of the uncontrolled disposal of untreated olive mills wastes. Five olive oil mills are in function in the selected pilot area for more than 10 years, whereas there are also two more mills that have stopped their activities before almost 10 years. Some of the active mills use evaporation lagoons while other dispose their wastes directly in rivers and streams.

During the past years no detailed study had been carried out in the region but also in the neighbouring municipalities to evaluate the quality of the soil and water resources and the way that mills' activities affect the surrounding environment. The second implementation site of the project was set up in Liguria Region, Italy where young olive trees were grown under controlled conditions and with the addition of specific amounts of OMW during experimentations aiming to the identification of the optimum conditions for OMW use at olive trees orchards.

As regards the potential threats for soil quality due to the uncontrolled disposal of OMW it was deduced that risk for soils in OMW disposal and neighboring areas is high since a number of soil parameters exceeded normal, high or toxic thresholds. It was also revealed that there are some soil properties that protect soils from degradation; clayey soils have very strong adsorption capacity and may remove big percentages of phenols and other contaminants after one application of OMW. However, this capacity is substantially reduced though after additional applications increasing thus contaminant concentration in infiltrating leachates and thus risk for deeper soil horizons overloading and groundwater contamination, as well. Consequently, higher risk is anticipated if disposal of OMW takes place on soils poor in clay and $CaCO_3$ and with low pH, on soils close to the sea or other water resources and if groundwater table is shallow.

The obtained results highlighted the need for establishing soil quality standards for some soil parameters in order to declare soils affected by induced human pollution like disposal of OMW. There is strong indication that the long-term application of OMW has the potential to induce soil or groundwater contamination. Therefore, long term use of OMW might require monitoring to assess any risk of environmental pollution.

In order to assess the risk for water bodies at areas close to OMW disposal ponds a carefully designed water monitoring system was implemented in the project area and water samples were collected every 2 months for all the four years of the project form surface streams, springs, water supply pipes, old wells, existing water abstraction wells and from piezometers that were installed at OMW disposal areas to collect soil pore water. Results revealed that the risk for groundwater is highly depended on the soil type, the presence of limestones and the depth of groundwater table. Moreover, the presence of clays in soils reduces substantially the toxic load during infiltration.

It is anticipated that any impacts in the OMW disposal areas will affect mainly recipients at local scale. In case though of more intense activities, larger affected areas and different soil qualities (e.g. sandy soil) risk for humans and ecosystems will be much higher. It is therefore proposed that due to scattering of olive oil production units in the Med region, simple and cost effective measures should be considered including neutralization and/or dilution of OMW prior to disposal in ponds or on agricultural soils as well as construction of impermeable evaporation ponds; in the latter case geo-membranes or alternatively clayey soils may be considered as a cheaper option.

Having performed thorough studies on soil quality and its dependence on OMW disposal in the framework of PROSODOL project and considering the specific climatic conditions of the Mediterranean countries, it is recommended that a monitoring tool fully suited to OMW disposal areas should include: (1) an optimized set of soil quality indicators; (2) threshold values for soil quality indicators; (3) a system that enhances decision making regarding the suitability of soil for OMW disposal/application (i.e. a land application system) to ensure safe disposal/use/application of OMW on soil in the Mediterranean region; (4) guidelines for periodical soil quality monitoring; (5) software application tools for soil monitoring that will facilitate adoption of the monitoring system by authorities and individuals; (6) guidelines for periodical water bodies monitoring; and (7) a code of good practices for soil management. The proposed soil monitoring system was fully described and explained and is presented in this book.

One of the main project's objectives was to develop and implement cost effective soil remedial actions that will remediate or, at least, protect soils from further degradation. It should be, however, highlighted that the development and implementation of soil remedial actions, appropriate and specific for OMW disposal areas, have been never implemented and demonstrated before and thus, the selection among available soil remedial methodologies was not existed as an option and, most significant, there was no possibility to compare the obtained results with results obtained from other already implemented and demonstrated methods. Therefore, all potentially applicable soil remedial methods were recorded and evaluated. It was, however, clear that a soil remediation and protection plan suitable for OMW disposal areas, should include methodologies for polyphenols reduction and retention or immobilization of inorganic constituents. Therefore, for the reduction of polyphenols concentration in soil, in situ-bioremediation was selected since it targets to the biodegradation of organic pollutants in soil by taking full advantages of the natural biodegradation process of organic molecules by soil microorganisms.

For the reduction of inorganic soil constituents, the use of natural zeolite, clinoptilolite, as soil amendment was considered the most suitable for this case because of the already well-known properties of natural zeolites to attract, retain and slowly release many inorganic cations, such as K^+, Na^+, Fe^{3+}, Cu^{2+} and others. Moreover, the method is of very low cost and very easy to be implemented, even by no qualified personnel.
The two methodologies applied at a pilot disposal area and the results obtained were very much satisfactory, indicating that these methods could efficiently be used for soil improvement and protection from the disposal of OMW.

Field pilot composting was implemented during the PROSODOL project using as raw materials solid OMW (i.e. sludge from inside the evaporation ponds), straw, caw manure, fresh and dry leaves, and different ratios of zeolite dust (0.00-0.80mm). After the evaluation of the results and the chemical analysis of the composts the most appropriate composition was selected and proposed.
PROSODOL focused also on the development and demonstration of low-cost OMW pretreatment techniques with the use of various reactive agents. These reagents were used to remove solids, add alkalinity, remove some of the toxic load and degrade organic contaminants so that

the main treatment that follows becomes easier or disposal to land as fertilizer is feasible. Various materials were used in lab scale in pretreatment experiments to investigate sorption of organic contaminants, increase pH, initiate precipitation of metals in stable forms and/or remove solids from OMW. Most of these materials are low cost, by-products of other processes and are abundant in Mediterranean countries. For the pretreatment of OMW the materials used include magnesite by-products, natural zeolite, limestone, two different types of soils, goat manure (GOM), zero valent iron (ZVI) and activated carbon (AC). GOM and ZVI show promising results in terms of phenol removal and pH increase.

The investigation of the potential impacts of OMWs used for olive trees irrigation/fertilization on soil quality and the potential contribution to yield increase were studied at a pilot orchard for 2 years. For this, field trials were performed in Albenga, Italy, during which controlled distribution of OMWW and husk took place in a pilot olive orchard. A pilot scale experimentation site of around 1.500 m2 for the controlled use of OMWW for tree land fertilization was set up and almost 200 two-years-old olive tree plants belonging to 3 different varieties (Taggiasca, Pignola, Leccino) were transplanted. OMWW and husk were distributed and the impacts on soil quality, on leachates quality and on yield were recorded and evaluated.
Finally, a fast and easy methodology was developed for the measurement of wastes' COD even in the mills itselves.

For the integration of PROSODOL activities, an extensive analysis of European Union legislative framework on the subject of olive oil waste management was performed, including the relevant regulations of waste, water and soil. The analysis integrated the relevant legislative framework of the partner countries, i.e. Italy, Spain and Greece as well as of Portugal and Cyprus, as well.
In specific, the study includes (a) an analysis of the olive oil industry and the relevant environmental issues; (b) waste management and the relevant EU and national legislation on waste, water and soil; (c) legislative recommendations for olive oil waste management, both statutory and volunteer; (d) legislative recommendations as well as technical specifications and proposed strategies to monitor, protect and improve soil quality at olive oil mills' disposal areas.
For the promotion of soil protective and remedial actions at OMW disposal areas, PROSODOL proposes a set of recommendations to be included in the national/European legislative frameworks. The recommendation are those derived after evaluation of the project's outcomes and mainly from the soil monitoring actions performed at olive mills waste disposal areas, and their fulfillment is considered necessary for soil quality protection. It is believed that their incorporation as Member States obligations in the legislative framework of the EC or/and of the Med Member States will ensure future effective monitoring of the legal and illegal disposal areas, which in turn will facilitate the sustainable management of these areas.

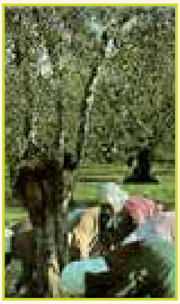
Moreover, PROSODOL proposes a set of technical standards which could be utilized either as Best Available Techniques for Soil Monitoring and Soil Quality Improvement or as Annexes in future Directives and legislative acts, which will assist national local/regional/governmental authorities to implement strategies to monitor, protect and improve soil quality at olive oil mills' waste disposal areas.

1. PROSODOL AIMS AND ACTIVITIES

1.1 OLIVE OIL PRODUCTION IN THE MEDITERRANEAN REGION

Over 750 million olive trees are cultivated worldwide, 95% of which are in the Mediterranean region. Most of global production comes from Southern Europe, North Africa and the Near East (Maps 1 and 2).

Of the European production, 93% comes from Spain, Italy and Greece. Spanish province of Jaén is well known for the biggest olive groves in the world.



Spain *is the country with the highest number of olive trees (more than 300 million), and is nowadays the world's leading olive and olive oil producer and exporter. Of the 2.1 million hectares (5.19 million acres) of olive groves, 92% are dedicated to olive oil production. The average annual production varies due to the cyclical nature of the harvest, but typically runs between 600,000 and 1,000,000 metric tons, only 20% of which is exported. About 80% of the crop is concentrated in Andalusia, (Jaén), the biggest olive growing area on the planet.*

In **Andalusia***, the most important olive oil producing areas are in the province of Jaén, where the main olive type is Picual, and other authorised varieties include Verdala, Real, and Manzanilla de Jaén, and in the province of Córdoba, where the authorised DO olive varieties include Picuda (a.k.a. Carrasqueña de Córdoba), Picual, Lechín, Chorrío, Pajarero, and Hojiblanco. DO certified Andaluz olive oils tend to be full bodied and tasty; class "A" oils have a maximum acidity of 0.4%, while class "B" oils have up to 1% acidity.*

Catalonia *also produces olive oil, which tends to be on the lighter side. The principal cultivation and production areas are Les Garrigues, in the province of Lleida, and Siurana, very nearby, in the province of Tarragona, where the Arbequina variety is the main olive grown, but where other DO authorised varieties include Real [Royal], Verdiel and Morrut olives.*

Italy *is the second European producer; two-thirds of the production is represented by extra-virgin oil with 37 DOP (Protected Origin Appellation) widespread on all the national territory. In Italy there are about 6.180 olive oil mills and the overall amount of processed olives in 2006/2007 was about 3.500.000 t with a production of about 600.000 t of oil. 90% of the entire oil production comes from Southern Italian Regions: Sicily, Calabria and Puglia. The introduction of new mills has increased the productivity and has decreased*

Map 1. *Olive cultivation-world production (source: FAO)*

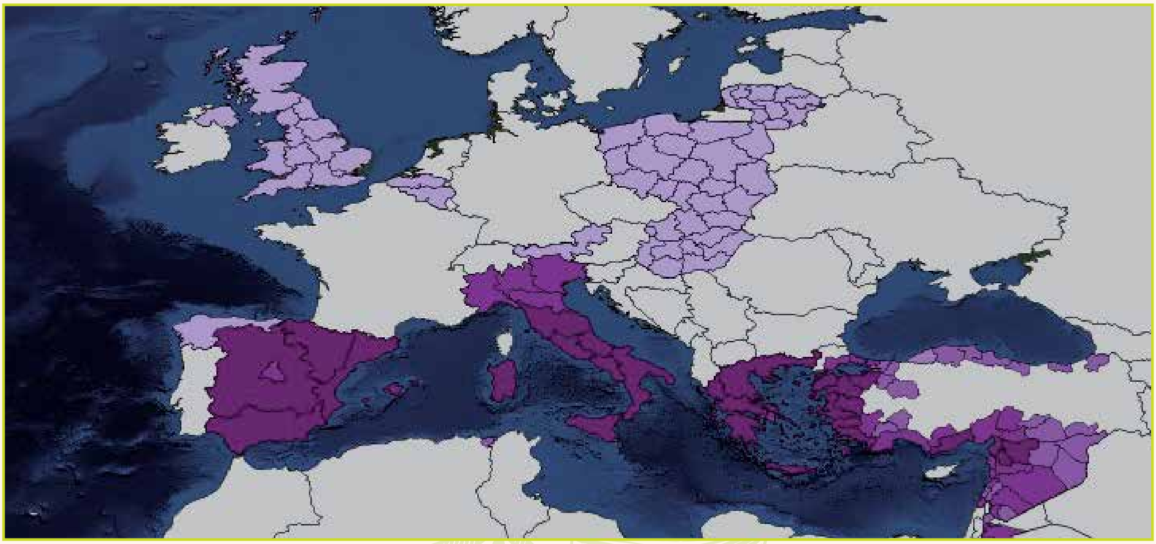
the need for manpower, heightening the problem related to the disposal of olive mills' wastes due to an increased production of wastes themselves. In Italy more than 2000 t/year of olive oil wastes are produced and half of them come from Puglia Region.

***Greece** devotes 60% of its cultivated land to olive growing. It is the world's top producer of black olives and has more varieties of olives than any other country. Greece holds third place in world olive production with more than 132 million trees, which produce approximately 350,000 tons of olive oil annually, of which 82% is extra-virgin . About half of the annual Greek olive oil production is exported, but only some 5% of this reflects the origin of the bottled product. Greece exports mainly to European Union (EU) countries, principally Italy, which receives about three-quarters of total exports. Olives are grown for oil in Greece, with **Peloponnese** being the source of 65% of Greek production, as well as in **Crete**, the **Aegean Islands** and **Ionian Islands**. The most prized Greek olive variety for oil production is the Koroneiki, originating from the area of Korone in Messenia, Peloponnese. This variety grows well on mountain slopes and produces very small fruit; the high ratio of skin to flesh giving the oil its coveted aromatic qualities. The variety is also suited to the production of agourelaio, oil from olives that are slightly unripe. When crushed in presses that are not capable of grinding the stone, this oil is entirely free of acidity and possesses top-tier organoleptic characteristics. Because not crushing the stones reduces oil yield, production of agourélaio is limited to "boutique" presses run by entrepreneurs and small cooperatives.*

*Among the many different olive varieties or cultivars in **Italy** are Frantoio, Leccino Pendolino, and Moraiolo; in **Spain** the most important varieties are the Picual, Alberquina, Hojiblanca, and Manzanilla de Jaén; **in Greece**, Koroneiki; in France, Picholine; in **California**, Mission; in **Portugal**, Galega; in **Croatia**, Oblica and Leccino. The oil from the varieties varies in flavour and stability (shelf life).*

***Australia** now produces some of the world's finest olive oils, primarily due to the remarkably good growing conditions, rich soils and lack of traditional pests and diseases. Many Australian producers only make premium oils, whilst a number of corporate growers operate groves of a million trees or more and produce oils for the general market. Australian olive oil is exported to Asia and Europe where the consistent high quality is respected.*

*The Republic of **South Africa** also produces extra virgin olive oil, with production increasing to meet demand.*



Map 2. *Olive cultivation-Mediterranean production (source: FAO)*

1.2 OLIVE MILL WASTES: ORGANIC AND INORGANIC CONSTITUENTS

Olive oil mills wastes constitutes an important pollution factor for the olive oil-producing regions but also a significant problem to be solved for the agricultural industry. The main reasons are:

1. *The large wastes amounts produced in relatively small time interval, which should, ideally, be processed or be disposed with safety for the environment before the beginning of the next productive period. Although the produced waste volumes depend on a lot of factors, as the variety of olive-crop, the stage of maturity, the storage before the processing, the time of segregation of olive oil from the crop, the available water for the processing and its cost, in general lines, for each 100 kilos of olive-crop they are produced 100-120 kilos of humid waste, while the medium daily production per mill varies between 15 and 20 tons.*
2. *The physico-chemical waste characteristics; some of them may cause significant problems to the recipients, where they are disposed (e.g. eutrophication, toxic phenomena to the aquatic fauna, phytotoxicity, aesthetic degradation).*
3. *The substantial high organic load, which is constituted by compounds/substances that can be easily decomposed (e.gg sugars, organic acids, amino-acids, proteins) and by substances that are decomposed with difficulty (e.g. fats, polyphenols). Wastes contain very high concentration of polyphenols, which may cause the appearance of bio-toxic phenomena in the recipients.*

Olive Mill Wastes are very rich in many organic and inorganic substances and elements; some of them may be toxic to the environment, while others are considered as necessary nutrients and for this reason their recycling under control conditions in the agricultural sector has been proposed as an alternative senario for their management (Chartzoulakis et al. 2010).

By far the most toxic OMW constituent is the polyphenolic compounds. Phenols are very soluble in water, oils, carbon disulfide and numerous organic solvents, at high concentrations are toxic and mutagenic substance and may be absorbed through the skin. Phenols are, for the most part, biodegradable. Populations residing near phenol spills, waste disposal sites, or landfill sites may be at risk for higher exposure to phenol than other populations. If phenol is present at a waste site near homes that have wells as a source of water, it is possible that the well water could be contaminated. If phenol is spilled at a waste site, it is possible for a person, such as a child playing in dirt containing phenol, to have skin contact or to swallow soil or water contaminated with phenol. Skin contact with phenol or swallowing products containing phenol may lead to increased exposure. This type of exposure is expected to occur infrequently and generally occurs over a short time period (Ahmaruzzaman, 2008).

Both the stone and pulp of olives are rich in phenolic compounds. Polyphenols, once released or formed during processing of olives, are distributed between the water and oil phases. Another part of the phenols is trapped in the olive cake. The distribution of the released amount of the phenols between water and oil is dependent on their solubilities in these two phases. The olive phenols are amphiphilic in nature and are more soluble in the water than in the oil phase. Due to their low partition coefficients (Kp), only a fraction of the phenols enters the oil phase. In general, the concentration of the phenols in the olive oil ranges

from 50 to 100 mg/g of oil depending on the olive variety. This amount corresponds to 1–2% of the total phenolic content of the olive fruit, while the rest is lost in OMWW (53%) and the olive cake (45%) depending on the extraction system (Rodis P.S. et al., 2002).

Phenolic compounds are present in olive mill wastewaters at concentrations in the range from 0.5 to 24 g/l, and are strictly dependent on the processing system used for olive oil production (Borja-Padilla R. et al., 1990a,b). Phenolic compounds generically include a great many organic substances that have the common characteristic of possessing an aromatic ring with one or more substitute hydroxyl group and a functional chain.

As regards the inorganic constituents, OMW have high potassium concentration and notable levels of nitrogen, phosphorus, calcium, magnesium, and iron. The highest potassium concentrations were observed in wastewaters (OMWW), while the sludges showed higher levels of the other nutrients, especially iron (Kavvadias et al., 2010, 2011).

In the case of wastewaters (OMWW) obtained by pressure, K was the predominant metal (17.10 g/l) followed in decreasing order by Mg (2.72 g/l), Ca (2.24 g/l), Na (0.40 g/l), Fe (0.129 g/l), Zn (0.063 g/l), Mn (0.0147 g/l), and Cu (0.0086 g/l). Lower concentration levels of cations were detected in OMWW samples obtained by centrifuge due to the dilution of the water during the centrifugal processing of the olive oil. With regard to the anions, the prevailing anion proved to be Cl- followed by the biacid phosphate H2PO4, which was in this form as a consequence of the acid waste pH. In OMWW samples obtained by pressure, the anions F- and SO42- presented very similar concentrations, whereas in the OMWW samples obtained by centrifuge the concentrations of the same anions were slightly different. With respect to the other anions, NO3- ions were present at very low concentrations in both kinds of wastewater. Most of the metal cations found to be bound to the organic polymeric fraction (Niaounakis and Halvadakis, 2006).

The wastes contain also high concentrations of Cu, which in the reductive environment of OMWW copper is present in monovalent form as crystals of copper oxide (cuprite Cu2O). Cuprite is insoluble in water and has a red color. The red color, which may be observed on the surface of OMW lagoons, may be caused by cuprite. In addition to the fact that copper is used against fungus attacks, copper is also toxic towards algae and other lower vegetation, but in concentrations of one-tenth of what has been found in OMWW. Fertilizer being used on trees is the real cause of copper presence in OMWW and its content is enriched in the bottom sludge of a lagoon. If this sludge is used as fertilizer it may poison the soil. The cause is claimed to be the contents of polyphenols in OMWW. But, polyphenols are produced in nature and is a natural conserving agent with a temporary toxicity, whereas the toxic action of copper is permanent The essential properties of OMWW depend on the process and the quantity of the added water. (Niaounakis and Halvadakis, 2006).

The very high pollutants load of the OMW characterizes them as hazardous wastes if they disposed in the environment untreated; one ton of processed olives produced a polluting load equivalent to that of 50–100 inhabitants; the average BOD5 concentration of undiluted OMWW is 120–150 kg/m3 while the dilution of OMWW with processing waters does not affect substantially the polluting load.

1.3 PROSODOL OBJECTIVES AND ACTIVITIES

The project's overall objectives were to develop and disseminate innovative, environment friendly, low cost technologies for the protection of soil and water from pollution caused by the disposal of olive oil mills' wastes, to design; implement and support a monitoring system for the assessment of soil and water quality affected directly or indirectly from mills' activities in relation to factors pressures and responses; to identify potential safest uses of mills' wastes in the agricultural sector and to establish an info-library/knowledge base system to assess environmental impacts from olive oil mills' wastes to Mediterranean region.

In this framework, the project was divided into 20 actions focused on:

1. *the establishment of an info-library with a lot of information regarding Olive Oil Production and OMW management in the Mediterranean. The info-library is available from the web site of the project (http://www.prosodol.gr) in four languages (English, Greek, Spanish and Italian). The library provides information regarding:*
   - Olive oil mills' activities, places of activities, volume and type of wastes, disposal type, soil pollution extent, existing studies of regional pollution, actions have been taken to reduce environmental impacts.
   - Methods of soil chemical analyses.
   - Soil monitoring systems, strategies developed and applied.
   - Soil remedial/protective methods that have been applied or can be potentially applied, results, benefits, required equipment and cost.
   - Composting technology, equipment and cost, economical data.
   - National environmental legislative framework.
   - Methods of water chemical analyses
   - Water monitoring systems, strategies developed and applied, chemical analyses methods
   - Water remedial/protective methods have been applied or can be potentially applied, results, benefits, required equipment and cost.
   - OMW management strategies in relation with environmental impacts.
   - Waste treatment technologies which have been applied or can be potentially applied, case/pilot studies have been funded, economical data of the application, required equipment and operational cost, benefits, social acceptance and extent of adoption.
   - Methods of olive oil wastes chemical analyses.
   - Methods for solid wastes treatment applied so far, or investigated and applied in pilot scale, benefits, composting technology, equipment and cost, economical data.
   - Bioremediation.
2. *the design and implementation of a monitoring system to assess soil and water quality at areas where the many years disposal of untreated olive oil wastes characterizes them as highly risky sites, aiming to develop methodologies and procedures, capable to identify soil/water quality parameters and extent of pollution over time and to provide national authorities with effective tools, useful for the control of such areas.*
3. *the identification of a set of soil parameters, suitable to be used as soil quality indicators to assess soil quality at OMW disposal areas.*
4. *the evaluation of the potential effects of wastes application on tree-land fertilization and the development of a useful guide for the safe use of mills' wastes in the agricultural sector.*
5. *the development of rapid methods of wastes analyses which can be used in field and in mills.*
6. *the development and implementation of soil remedial and protective technologies. Among the several protective/remedial methods for polluted or degraded soils, bioremediation and natural zeolite addition in soil were selected, conformed to the specific environmental conditions and implemented in a pilot field.*
7. *the development and implementation of low cost methodologies for OMW pre-treatment at field scale with the use of low-cost reactive agents, such as metallic iron, manure and zeolites.*
8. *the identification of optimum conditions for OMW composting with the use of clinoptilolite (natural zeolite) as additive.*

9. the development of an integrated scenario, financially and technically evaluated, suitable for OMW disposal areas, which includes soil protection/remediation, recycling of the wastes by composting and the cost effective pre-treatment of wastes in order the toxic substances to be reduced.
10. The development of an integrated approach of actions, measures and means suitable for the Mediterranean region.
11. the dissemination of project's achievements so that the knowledge gained to be actively communicated to those stakeholders that may best make use of it and apply the lessons learnt from the project.

1.4 PROSODOL BENEFICIARIES

Five Mediterranean Institution were the Beneficiaries of the project and responsible for its successful fulfillment. These were:

	The Soil Science Institute of Athens (SSIA) ***Hellenic Agricultural Organization DEMETER.*** *Team Leader and coordinator of the project: Dr. Maria Doula.*
	The Department of Mineral Resources Engineering ***Technical University of Crete (TUC).*** *Team Leader : Prof. Konstantinos Komnitsas.*
FORTH	***The Institute of Mediterranean Studies*** ***Foundation of Research and Technology (IMS-FORTH).*** *Team Leader: Dr. Apostolos Sarris.*
CSIC	***The Center of Soil Science and Applied Biology*** ***Spanish National Research Council (CEBAS-CSIC).*** *Team Leader : Dr. Jose Luis Moreno Ortego.*
Camera di Commercio Savona	***The Regional Center of Experimentation*** ***and Technical Assistance, Italy (CERSAA).*** *Team Leader : Dr. Federico Tinivella.*

1.5 PILOT AREAS OF PROSODOL PROJECT

The PROSODOL project included two demonstration-pilot areas; one in Greece and one in Italy.

The Greek demonstration area is located in the municipality of Rethymnon (former Municipality of Nikiforos Fokas), in the north of Rethymnon prefecture, Crete (Maps 3, 4 and 5).

The selected pilot municipality is one of the many municipalities in Greece but also in Mediterranean facing the same problem of the uncontrolled disposal of untreated olive mills wastes. Five olive oil mills are in function in the selected pilot area for more than 10 years, whereas there are also two more mills that have stopped their activities before almost 10 years. Some of the active mills use evaporation lagoons while other dispose their wastes directly in rivers and streams. During the past years no detailed study had been carried out in the region but also in the neighbouring municipalities to evaluate the quality of the soil and water resources and the way that mills' activities affect the surrounding environment.

Map 3. The island of Crete, where the Greek demonstration area is located.

Map 4. The Municipality of Rethymnon.

Map 5. Overall view of the pilot area in Crete.
Three active and two inactive disposal ponds are located near the different marks.

Map 6. Liguria Region in Italy.

Photo 1. Pilot olive tree orchard in Liguria

2. PROSODOL RESULTS

2.1 SOIL RISK ASSESSMENT

Soils that accept wastes, apart from progressive degradation, may cause serious problems to the surrounding environment (humans, animals, plants, water systems, etc), and thus, soil quality should be necessarily monitored. The changes caused to soil properties, due to waste disposal, are strongly depended on the specific soil type, on waste type as well as, on the climatic conditions of the area.

For Mediterranean countries disposal of Olive Mill Wastes (OMW) is considered a major environmental problem.

The olive oil extraction industry represents an important activity in the Mediterranean area. In general, for each ton of olive oil production about 1,600kg organic pollutants (dry basis) are produced while the polluting load related to the disposal of 1 m^3 of OMW is equivalent to 100-200 m^3 of urban wastes (corresponding to the ones produced by 100,000 people).

The uncontrolled disposal of OMW on soil may cause strong phytotoxic and antimicrobial effects, may increase soil hydrophobicity, decrease water retention and infiltration rate (Abu-Zreig and Al-Widyan, 2002), may also affect acidity, salinity, N immobilization, microbial activity, nutrient leaching, lipids concentration, organic acids and naturally occurred phenols (Sierra at al., 2007).

In general, soils that accept OMW are rich in organic matter, inorganic nutrients as well as, in polyphenols. Although organic matter and nutrients could be beneficial for soil fertility and necessary for plant growth, potential serious soil degradation should always be considered due to very high concentrations of inorganic elements and polyphenols, sometimes near or above thresholds. Moreover, the addition of the insufficiently stable organic matter of wastes, although it leads to the general increase in soil organic matter, may induce a number of negative effects on soil properties and plant growth, such as increase in mineralization rate of native organic carbon, induction of anaerobic conditions and release of phytotoxic substances that may have negative effects on plant growth (Kavvadias et al., 2010).

The experience gained through the PROSODOL project indicates that in the three major olive oil productive countries in Europe (i.e. Spain, Italy and Greece), different OMW management systems have been developed, adopted and are implemented. In Italy, the law No 574 of 1996 rules the management of OMW as well as the Ministerial Degree of 6th July 2005 with the majority of the mill owners already conformed to them. In Spain the wastes of the olive oil industry are mainly used for the production of heat and energy, or they are composted; however large amounts are still deposited in evaporation ponds. In Greece, due to the lack of specific legislative framework and effective monitoring by the responsible governmental, regional and

local authorities, OMW are mainly disposed in evaporation ponds, on soil and in water systems. Thus, as also proved during the implementation of PROSODOL project, uncontrolled disposal of OMW increases substantially the risk of soil degradation. In specific, the results indicated that :

1. *For soils rich in CaCO3 content and initial pH>7.5 the long term application of OOMW did not markedly affect soil pH because most of the acidity present in OMW is neutralized by the soil carbonate. For soils with low CaCO3 (<40%) content the reduction in soil pH should be a major concern if long-term application of OMW takes place on soils. Moreover, the pH values of the leachates that are produced after wastes disposal are by almost 1 pH unit lower than the pH of the leachates produced after only water addition. For soil rich in sand (i.e. Loamy Sandy and Sandy) the decrease in leachates' pH is substantial.*

2. *The electrical conductivity (EC) of soils that accept wastes disposal is higher than EC of soils unaffected by OMW, but still below the EC threshold value for salinity (4mS/cm). Wastes cause also a significant increase in EC of the leachates's produced by all soil types after disposal. Rainfall or soil washing could decrease the EC of the leachates, although they still remain high. Increase in soil EC is mainly due to ionic species, namely potassium, chloride, sulfate, ammonium and nitrates, present either in OMW or generated through waste mineralization and transformation. Therefore the increase in soil salinity can be a major concern if long-term application of OMW takes place on soils uncontrolled or at high rates.*

3. *The carbonate content of the upper soil layers in evaporation ponds and at surface disposal points may be reduced since carbonates buffer OMW's acidity by generating soluble calcium bicarbonate that moves to lower horizons and precipitates again as calcium carbonate. Thus, there is a considerable risk for soils poor in CaCO3 for which the decrease in carbonate content could be significant with a subsequent decrease in soil pH. For soils rich in CaCO3, the risk is medium, however, the continuous uncotrolled OMW disposal may lead to non-recoverable soil degradation.*

4. *The soil organic matter shows to be a significant component of the soil profile characteristics of pond soils up to a depth of 100 cm. For the sites of direct or indirect disposal (but no evaporation pond soils), high amounts of organic matter are restricted to upper soil layers (0-50 cm). Since organic matter is closely related to nutrient availability, it is postulated that contradictory observations may be drawn either due to the increase in the concentration of some less soluble nutrients, making them more available, or to make them less available and hence less toxic. It is anticipated that organic matter will not increase noticeably with depth and distance from pond, since transfer of large organic molecules through soil layers is difficult, unless specific circumstances or the geomorphology of the disposal area enhnance the transfer to deeper soil horizons.*

5. *The total polyphenols content of soils that accept OMW disposal is very high, and remains high even many years after the last disposal. The content of OMW in polyphenols is extremely high, however, it was observed that all soil types (from fine to coarse textured soils) retained a noticeable percentage of the added phenols and leached only a small part of the initially added amount. This is a very interesting result, which could be positive for the environment, since large amounts of polyphenols are kept from soil and are not leached to reach groundwater systems. However, an issue to be identified is the fate of the retained polyphenols. In other words, if the retained polyphenols are not biodegraded from soil fauna and remain as toxic compounds in the soil environment, then protective measures should be taken. It is known, and was confirmed during PROSODOL, that the polyphenols content of soils at OMW disposal areas are very high and remain high even if no disposal took place for years. However, soil texture affects the leached amounts of polyphenols. As it was observed, the capability to retain polyphenols is reduced following the order from fine textured soils to coarse textured soils. The fact that soils rich in clay retain polyphenols has positive and negative consequences. Thus, the* *high retention results in lower risk of leaching and subsequently in contaminating ground-waters, however high phenols concentrations in soil may result in phytotoxic incidents as recorded during the pilot actions of the PROSODOL project. On the other hand, the risk of phytotoxicity appearance is limited to soils with lower content in clay and higher in sand, however, the risk of groundwater contamination is high. Therefore, as long as the waste remains on soil, surface soluble phenolic substances can be released and leached down to deeper horizons, which, in turn, can enhance the risk for groundwater contamination. The risk is higher for soils poor in clay content.*

6. *A buildup of nitrogen shall be anticipated due to elevated total N concentrations in the evaporation pond soils. For sites that accept surface disposal it is very likely to measure N concentrations higher than 3mg/kg, mainly found in the top 0-50 cm. Moreover a considerable percentage of inorganic N (NH4+ and NO3-) found to be varied in very high levels in pond soils (>10 mg/kg and >30 mg/kg, respectively), mainly found at surface soil layers (0-75 cm). Therefore, the high N in evaporation pond soils may be a significant source of potential pollution of surface soil and waters. Moreover, the N data collected from inactive disposal sites shows that mineralization of organic N in soils where disposal of OMW have been ceased 8 years ago, is retarded.*

7. *In the case of exchangeable K, it was found that in the most of the cases, evaporation pond soils have values from >2 up to 26cmol/kg which were found throughout the soil profile. For soils that accept surface disposal K concentration ranged from >2 up to 17cmol/kg which were concentrated up to 125cm soil depth. It should be also highlighted that, even 10 years after the last disposal inactive evaporation pond soils have values >2 up to 9.0cmol/kg, which were found at all soil depths indicating significant increase in the potential potassium reserve in soil. The capability to retain potassium is reduced following the order from fine textured soils to coarse textured soils. The fact that the soils with high clay content retain potassium has positive and negative consequences. Thus, the high retention results in lower risk of leaching and subsequently in contaminating groundwaters, however the high concentrations of these constituents in soil may result in phytotoxic incidents as recorded during the pilot actions of the PROSODOL project. On the other hand, the risk of phytotoxicity appearance is limited to soils with lower content in clay and higher in sand, however, the risk of groundwater contamination is high. Concluding, the long-term application of OMW cause the build-up of soil potassium and may deteriorate physical properties of the receiving soils and increase the risk for leaching of K to ground waters.*

8. *Exchangeable Mg data indicated that the long term surface disposal of OMW may potentially endanger soil quality due to excess Mg accumulation. The most of the data collected during the PROSODOL have values >2.2 up to 11cmol/kg while, it reaches up to 2.51cmol/kg in pond surface soils. Regarding Mg leaching after wastes disposal, this does not seem to be affected by soil texture, except for the case of sandy soils. However, it is anticipated that the leaching will be higher for soils naturally rich in Mg.*

9. *For exchangeable Ca, it was observed that soils rich in CaCO3 undergo considerable reduction in exchangeable Ca content because of the OMW acidity which cause dissolution of CaCO3. For soils low in CaCO3, no significant differences were found between pond and control samples. As for Mg leaching, the leaching of Ca is higher for soils naturally rich in Ca. The leaching of Ca does not seem to be affected by soil texture, except maybe for the case of sandy soils. Wastes contain Ca but in low concentration in relation to the concentrations of the other wastes' constituents (e.g. K), however it seems that the disposal of wastes on soil cause the release of soil Ca due to dissolution of solid CaCO3 and the subsequent neutralization of the wastes in soil.*

10. *Regarding the availability of phosphorous, results indicate a high potential mobility for P and a potential threat for soil and surface water contamination in evaporation pond sites and in*

the direct disposal sites.The most of the measured available P concentrations had values from >60 to 669mg/kg in active pond soils, from >60 to 591mg/kg in direct disposal site and from >60 to 365mg/kg in inactive pond sites. Levels of water soluble phosphates were also found in high levels. Phosphates concentration is significantly affected by the addition of wastes, which could be also present in high concentration in the leachates produced after OMW disposal.
The environmental concern of P accumulation in soil regarding the long period required to reduce P to levels normally allowed for agronomic production was confirmed during PROSODOL, as well.

***11.** As it was observed there is an environmental risk for soluble boron due to OMW disposal in evaporation ponds, since it was found that available B in pond soils had, in many cases, concentrations from >3.0 up to 4.8mg/kg, found in soil layers 0-50 cm.*

***12.** A high percentage of available iron (DTPA extractable) data in pond soils and in the direct disposal site reached very high levels (up to 360 mg/kg) across the soil profile suggesting potential risk for Fe contamination due to disposal of raw OMW.The long period of cease of OMW disposal in ponds (almost 10 years), is not an adequate period to reduce the available Fe concentration in soils to normal levels.*

***13.** Results of available copper (DTPA extractable) indicated that potential toxic Cu concentrations can be found in both active (up to 21 mg/kg) and inactive (up to 18 mg/kg) pond soils, however very high Cu levels were not measured (only 5% of the studies areas).*

***14.** Long term disposal of OMW in evaporation ponds increases availability of zinc to high levels (up to 31mg/kg) in surface soil (0- 50cm) but not to toxic levels.*

***15.** For available manganese (DTPA extractable), it was detected that there were cases for which increased Mn concentrations were measured. The increase was slight or substantial. However, there were also cases for which the high natural Mn content was decreased after OMW disposal, mainly due to waste's acidity, which causes the dissolution of naturally occurring metals.*

***16.** Regarding heavy metals and by integrating the collected data, there is an elevated risk due to the OMW acidity, which may cause the dissolution of naturally occurring metals.*
The risk becomes higher due to the overloading of soils with Fe, Cu, Zn and Mn added to soils as OMW constituents. As far as Ni, Cr and Mo are concerned; if their background concentrations are high then there is a substantial risk of dilution and thus polluting deeper horizons and groundwater. The risk is considered higher for soils with low carbonate content, low pH and poor in clay.
An additional risk for Ni, Cr and Mo pollution should be anticipated in case of old mills equipment, since these recalcitrant metals could be produced from the corrosion of inferior quality steel equipment, which is used for olive processing.

- *It is therefore deduced that risk for soils in OMW disposal and neighboring areas is high since a number of parameters exceed normal, high or toxic thresholds. If the entire study area is considered, risk is assessed as average.*
- *Clayey soils have very strong adsorption capacity and may remove big percentages of phenols and other contaminants after one application of OMW; this capacity is substantially reduced though after additional applications increasing thus contaminant concentration in infiltrating leachates and thus risk for deeper soil horizons overloading and groundwater contamination, as well. Even higher risk is anticipated if disposal of OMW takes place on soils poor in clay and CaCO3 and with low pH, on soils close to the sea or other water resources and if groundwater table is shallow.*
- *A high risk for soil and ground water exists due to potential dissolution of Ni and Cr and transport to deeper soil horizons. Regardless if Ni and Cr come from mills' poor quality steel equipment or their presence is due to the high background levels of the area, the risk is considered high when*

metals are in contact with the acid OMW.

• Absence of vegetation on soils that accept the disposal of OMW directly, is a sign of phytotoxicity for seed germination and plant growth not only due to the presence of polyphenols, but also of other organic and inorganic constituents which may remain phytotoxic even after complete removal/degradation of polyphenols.

• An actual risk for humans exists in cases when exposure exceeds the Maximum Permissible Risk for humans (MPRhuman) value. MPRhuman is defined as the dose of a contaminant, based on body weight for oral intake or air volume for inhalation intake, which forms a risk of one additional case of lethal tumor in 10,000 lifelong exposed individuals. For example, the Maximum Permissible Risk for intake of Cu is 140 μg/kg/d. If a mean human weight of 70 kg is considered, the maximum allowable daily intake for Cu is 9800 μg. Considering the mean Cu concentration in the disposal sites, it is deduced that a human should consume almost 3.8 kg of contaminated soil per day to be at risk, which is impossible. Thus, it is anticipated that the risk for humans is low.

• Potential soil and plant toxicity due to polyphenols and metals may affect health of grazing animals due to ingestion of soil (or water drinking). It is therefore proposed that no grazing animals are allowed in areas around evaporation ponds before a complete risk assessment is carried out and the calculated risk is low.

• Humans may be also affected by OMW polyphenols and metals through consumption of products derived from grazing animals. Food, particularly animal-derived products (e.g. meat, milk, cheese), represents the most important source of human exposure to several recalcitrant pollutants. Thus, determination of animal tissue concentration is important for assessing potential risk to animal as well as human health (Rhind et al., 2005).

***In conclusion**, the obtained results highlight the need for establishing soil quality standards for some soil parameters in order to declare soils affected by induced human pollution like disposal of OMW. There is strong indication that the long-term application of OMW has the potential to induce soil or groundwater contamination. Therefore, long term use of OMW might require monitoring to assess any risk of environmental pollution.*

2.2 WATER RISK ASSESSMENT

A carefully designed monitoring system was implemented in the project area to assess the quality of all existing water bodies.

Water samples were collected every 2 months for all the four years of the project form surface streams, springs, water supply pipes, old wells, existing water abstraction wells and from piezometers that were installed at OMW disposal areas to collect soil pore water.

The parameters measured in situ included pH, electrical conductivity and liquid dissolved oxygen. Other parameters measured in the laboratory include COD, phenols, tannic acid, total hardness, NO_3^-, SO_4^{2-}, PO_4^{3-}, NH_3-N and elements such as Ni, Mn, Cl, K, Fe, Cu, Cd and Zn.

2.2.1 WATER SAMPLES COLLECTED FROM SURFACE STREAMS, SPRINGS, WATER SUPPLY PIPES AND OLD WELLS

Samples collected were analyzed to identify potential contamination sources, concentration of the most important contaminants, transport mechanisms and fate of contaminants in aquatic media.

Table 1 shows the quality of all water samples, as well as drinking water standards for the parameters measured.

Chemical analyses revealed that water samples collected between May 2009 and January 2010 as well as between October and November 2010 from water bodies close to OMW disposal areas, were characterized by high phenol concentrations (2.5-5 mg/L). Phenol concentrations for the samples collected from the above sampling points between March - July 2010 and January - March 2011 were lower varying between 0-1.5 mg/L. European Council Directive 98/83/EC proposes an indicative acceptable value of 5 μg/L phenols. Moreover, in some water samples the concentration of Ni and Mn reached up to 0.1 and 1.2 mg/L, respectively, exceeding drinking water standards (<0.07 and <0.05 mg/L, respectively as shown in Table 1). Concentration of other parameters such as NO_3^-, SO_4^{2-}, PO_4^{3-}, Cl, NH_3-N, K, Fe, Cu and Zn were lower than drinking water standards.

Parameter	Water samples	Drinking water standards
pH	6.5-8.9	6.5-8.5 (FPTC, 2008)
Electrical conductivity (EC), μS/cm	92-904	~2500 (20 oC) (98/83/EC)
Liquid dissolved oxygen (LDO), mg/L	3-10	No health-based guideline value (WHO, 2008)
COD, mg/L	0	No health-based guideline value (WHO, 2008)
Tannic acid, mg/L	0-0.6	No health-based guideline value (FPS, 1999)
Phenols, mg/L	0-5	No health-based guideline value (suggested safe levels <5*10-4) (CMD Y2/2600/2001)
Hardnesstot, mg/L CaCO3	130-420	No health-based guideline value (suggested safe levels around 100-200) (WHO, 2008)
Mn, mg/L	0-1.2	<0.05 (98/83/EC; FPS, 1999; FPTC, 2008)
NO_3^-, mg/L	0-18	<45 (FPS, 1999; FPTC, 2008)
SO_4^{2-}, mg/L	5-62	≤500 (FPS, 1999; FPTC, 2008; WHO, 2008)
PO_4^{3-}, mg/L	0-1.4	No health-based guideline value (suggested safe levels <5) (WHO, 2008)
Cl, mg/L	5-40	<250 (98/83/EC; FPS, 1999)
K, mg/L	0.1-9.3	<12 (98/83/EC)
Fe, mg/L	<0.1	<0.2 (98/83/EC)
Cu, mg/L	<0.1	<2 (98/83/EC)
Ni, mg/L	<0.1	<0.07 (WHO, 2008)
NH_3-N, mg/L	0	<3 (WHO, 2008)
Zn, mg/L	0	≤5 (FPTC, 2008)

Table 1: Characterization of waters collected bi-monthly and drinking water standards

2.2.2 WATER SAMPLES COLLECTED FROM DRAWING WELLS

Additional representative water samples were collected from existing water drawing wells located at the study area.
These as well as other water samples collected from streams, springs, water supply pipes and old wells were analyzed for various parameters of interest.
However, no contamination was seen (pH ~8, electrical conductivity ~505 μS/cm, LDO ~8 mg/L, phenols not detected, tannic acid<0.1 mg/L, COD=0 mg/L, total hardness ~200 mg/L $CaCO_3$, Cl<21 mg/L, NO_3^-<16 mg/L, SO_4^{2-}<13 mg/L, Cu<0.08 mg/L, NH_3-N=0 mg/L, Zn=0 mg/L, Fe=0 mg/L).
The only contaminant that exceeded the drinking water standards was Mn (0.2-07 mg/L); contamination though cannot be attributed to OMW since Mn is one of the most common elements in earth's crust and is characterized by high mobility.

2.2.3 WATER SAMPLES COLLECTED FROM PIEZOMETERS

In order to assess the fate of organic and inorganic (mostly recalcitrant metals) contaminants present in OMW and soils and the subsequent risk for groundwater contamination, piezometers were installed in 4 drillholes in the wider affected area of the pilot area; one control drillhole was drilled about 5 km away from the evaporation ponds.
Drilling and installation of piezometers took place in the first week of November 2010, to allow monitoring of pore water quality for a period of almost 26 months until the end of the project (31/12/2012).
Monitoring of pore water quality took place normally every two months.
Results showed that pore water pH ranges between 6.9 and 8. Electrical conductivity values ranging between 538-1689 μS/cm are lower than the acceptable value of ~2500 μS/cm for drinking water as shown in Table 1.
The highest phenol and COD concentrations (3.04 and 692 mg/L, respectively) were seen for piezometer which was located between two disposal ponds. Mn and Zn concentration ranged between 0.01-0.9 and 0.03-0.3 mg/L, respectively while Ni and Cu were below detection limits.

Experimental results and chemical analysis carried out indicate that:
- ***the risk for groundwater is low due to the soil type, the presence of limestone in low depth and the depth of groundwater table which normally exceeds 50 m.*** *Therefore phenols, as well as other recalcitrant contaminants, migration from surface to deeper soil horizons and especially groundwater is quite improbable.*
The presence of clays in soils reduces substantially the toxic load during infiltration.
- ***the risk for humans is also low; higher risk is anticipated if humans drink water from public wells where high concentration of phenols has been determined in specific periods (average concentration though is rather low).***

It is anticipated that any impact foreseen in the area under study will affect mainly recipients at local scale. In case though of more intense activities, larger affected areas and different soil qualities (e.g. sandy soil) risk for humans and ecosystems will be much higher.
It is therefore proposed that due to scattering of olive oil production units in the Med region, simple and cost effective measures should be considered including neutralization and/or dilution of OMW prior to disposal in ponds or on agricultural soils as well as construction of impermeable evaporation ponds; in the latter case geo-membranes or alternatively clayey soils may be considered as a cheaper option.

2.3 SOIL MONITORING SYSTEM

Having performed thorough studies on soil quality and its dependence on OMW disposal in the framework of PROSODOL project and considering the specific climatic conditions of the Mediterranean countries, it is recommended that a monitoring tool fully suited to OMW disposal areas should include:

- *An optimized set of soil quality indicators*
- *Threshold values for soil quality indicators*
- *A system that enhances decision making regarding the suitability of soil for OMW disposal/application (i.e. a land application system) to ensure safe disposal/use/application of OMW on soil in the Mediterranean region*
- *Guidelines for periodical soil quality monitoring*
- *Software application tools for soil monitoring that will facilitate adoption of the monitoring system by authorities and individuals*
- *Guidelines for periodical water bodies monitoring*
- *A code of good practices for soil management.*

2.3.1 SET OF SOIL INDICATORS FOR OMW DISPOSAL AREAS

The continuous monitoring of the pilot area revealed that not all of the measured parameters are affected by the disposal of OMW.

In particular, some of the measured parameters remained almost unchanged or the changes recorded were not significant relative to the control soil samples used for comparison (e.g. exchangeable Ca), other were subjects to changes but their values were depended also on different seasons and thus were inappropriate to be used as indicators (e.g. Cl^{-}*,* NH_4^{+}*,* SO_4^{2-}*,* PO_4^{3-}*,* NO_3^{-}*, microbial activity). Other parameters were significantly changed due to wastes disposal but this change lasted for short time after ceasing of wastes disposal although the area was still very much degraded (e.g. N, B).*

Finally, there were parameters that exhibit significant changes strongly depended on OMW disposal (e.g. organic matter, exchangeable K, available Fe).

From the evaluation of the obtained results it was clear that, since soil degradation at OMW disposal areas remains significant also for inactive-abandoned disposal areas, the indicators to be established should cover these two potential cases namely, active disposal areas and inactive disposal areas.

Thus, after statistical evaluation of the collected data and considering that an indicator should be characterized by four features, i.e. relevance; understandability; reliability; and accessibility of data, the following soil parameters were proposed as indicators for monitoring soil quality in areas of OMW disposal:

- *Electrical Conductivity*
- *Organic Matter*
- *Total Nitrogen*
- *Total Polyphenols,*
- *Available Phosphorous*
- *Exchangeable Potassium*
- *Available Iron, and*
- *Soil pH (mainly for acidic soil types)*

All these soil parameters are characterized by the four basic features:

- *Relevance: All indicators are related to the disposal of OMW and as it was observed during the soil sampling campaigns and the analyses performed for many soil samples (affected and control) and during different seasons, their values depend only on disposal activity.*
- *Understandability: All indicators are soil parameters that have been used for many years to characterize soil systems and thus are very much understandable, even by people who are not experts.*
- *Reliability: The proposed indicators are reliable as proved by many soil analyses, by periodically sampling from the same sites and by data evaluation.*
- *Accessibility of data: Indicators provide timely information and as it was proved by the monitoring of the disposal areas. One waste application was enough to increase these parameters to values much higher than control samples. It is also significant to be mentioned that, during soil sampling campaigns there were sites that were recognized as disposal sites after having analyzed these parameters.*

The Ni, Cr and Mo issue

Regardless if Ni, Cr and Mo come from poor quality steel or their presence is due to the high background levels of the area, attention should always be paid to the fact that if they are in contact with the acidic OMW there is always risk for metals dissolution and transport to deeper soil horizons and potentially ground water. Contamination of soils with recalcitrant heavy metals is an issue that needs to be seriously considered in OMW disposal sites.

Soil colour

Apart from the above soil parameters, one more that should not be ignored is soil colour. When soil degradation takes place, both texture and colour change, and this change is often one of the first obvious indicators of soil degradation.
Colour changes were observed for soils that accept surface disposal of OMW (Photo 2), while such degraded systems do not seem to recover after many years of last disposal (Photo 3).

Munsell soil-colour charts give a full description and code for soil colors.

It is, thus, necessary to standardize the moisture level of the soil for the color determination and to record soil colour both in dry and wet/moist representative soil samples. Moreover, for soil-degradation assessment it is necessary to compare colors between un-degraded and degraded conditions.
The monitoring of soil quality indicators within a defined ecological zone requires (Arshad and Martin, 2002):

- *Direction of change-positive or negative increase or decrease, etc*
- *Magnitude of percent change over the baseline values*
- *Rate of change-duration: months, years*
- *Extent of change-percentage of the area being monitored i.e. what percentage of the area has changed with respect to the selected indicator during a specified period*

Monitoring of soil indicators needs to set up sampling strategies allowing assessment of changes in soil quality.

Photo 2. *Changes in soil colour due to OMW surface disposal in an active disposal area.*

Photo 3. *Changes in soil colour due to OMW surface disposal in an inactive, for more than 10 years, disposal area.*

2.3.2 THRESHOLD VALUES FOR SOIL QUALITY INDICATORS

In general, changes in soil quality can be assessed by measuring appropriate indicators and comparing them with desired values (critical limits or threshold level) at different time intervals, for a specific use in a selected area-system.
A critical limit or threshold level is the desirable range of values for a selected soil indicator that must be maintained for normal functioning of the soil ecosystem health. Within this critical range, the soil performs its specific functions in natural ecosystems. (Arshad and Martin, 2002).
Thus, when a set of indicators is proposed, this list should be accompanied by thresholds level for each one of the indicators in order to assist evaluation of collected data and of the chemical analyses results.
The thresholds could be identified based on EU directives, on national laws, but also on the international literature.
The peculiarity of the indicators proposed for the case of OMW disposal is that they mainly correspond to soil properties associated with fertility and not to pollutants in the classical sense, such as heavy metals and therefore are not included in national laws or EU directives.
Nevertheless, international literature can provide general limits as these properties have been extensively studied for many years.
Given the complexities of setting limits and the uniqueness of each targeted area/region, it may be more efficient to develop guidelines that can help in setting up limits under certain land and environment conditions.
Thus, although a general definition of indicators thresholds could be performed after searching in international literature and national or EU legislative frameworks, it should be highlighted that the definition of indicators thresholds would be more effective and representative of each target area if they would be determined after evaluation of data collected from the areas of interest and by taking into account local characteristics and values of the indicators of representative control samples.
Especially for polyphenols, for which the assessment of their concentration in soil is considered difficult and with high degree of uncertainty due to the lack of generally accepted threshold, it is recommended to use local and site specific thresholds as guidelines/normal values (Zhou, 1996; Swartjes, 1999; Sierra et al., 2001; Mekki et al., 2007; Di Serio et al., 2008; Kavvadias et al., 2010).
A GIS based land information inventory of the area should be then designed and developed to store all collected data for further evaluation by local authorities, scientists, a.o.
General threshold values and the respective literature are included in Annex I for each one of the proposed quality indicators.

2.3.3 APPLICATION OF OLIVE MILLS WASTES ON SOILS - A LAND APPLICATION SYSTEM

If land distribution is planned (e.g. disposal, irrigation) the organic load and the toxic substances (e.g. polyphenols) of treated or untreated wastes should not be the only issues of concern. Specific care should be taken also for inorganic constituents and especially for K, Cl^-, NO_3^-, SO_4^{2-}, P, Mg, Fe, Zn and others, since the very high amounts disposed on soil change its quality properties drastically, while the concentrations of the inorganic soil constituents (especially K, P, Fe, Cu, SO_4^{-2}) and the electrical conductivity remain high even many years after the last disposal (Kavvadias et al., 2010).
Therefore, for the safe disposal or use of OMW, soil and land data have to be considered in combination with bioclimatic conditions and management practices in order to develop a system for assessing land suitability. A Geographical Information System is necessary to define the application of OMW to agricultural or other type of lands because of the importance of spatial accuracy in the application. Also, it is necessary to include information on land, soil and OMW properties, processes and composition;

climate variability; land use and management; and possible environmental risks.

The land suitability system for spatially manipulating soil and land data that is proposed has been designed and developed in Soil Science Institute of Athens (SSIA) for other Greek areas in the past and it was adapted to the peculiarities of OMW disposal. It follows the basic concepts and ideas of Theocharopoulos et al., (1998) and is also based on the philosophy of the UN Food and Agriculture Organization's 1976 Soils Bulletin, "A framework for land evaluation" (FAO, 1976) as far as its structure, nomenclature and definitions are concerned. The criteria are based on published guides of other Scientific Organizations (MAFF, Dept. of Environment, 1989; Soil Science Society of America, 1986) for assessing the suitability of soil for different kinds of use and experience of soil properties and seasonal variability of soil processes gained in the field.

The system requires a complete initial soil survey at region or larger level, which should include systematic soil sampling and then mapping of the results. An example is presented in Annex 2 for Viotia prefecture, Greece for the application of sewage sludge performed at prefecture level from SSIA.

Each mapping unit was categorized according to the following: polygon type (mapped, not mapped, lake, sea), drainage class, (assessed from profile morphology), texture (classes and for 3 depths i.e. 0-25 cm, 25-75 cm and 75-150 cm), gravel (classes), slope (classes), erosion (classes), calcium carbonate (classes), soil order, suborder and great group, irrigability (availability of water for irrigation), variability class and limitations, rainfall, and geology of the parent material.

Also infiltration rate and the presence and depth of the impermeable layer were recorded in some mapping units. In each mapping unit, analytical data from profile samples or auger sampling for each horizon were also stored.

The proposed system present the following specifications:

a) is adapted to Greek and generally speaking to Mediterranean bioclimatic conditions,

b) is general and can be used throughout Mediterranean where OMW is produced,

c) incorporates soil behavior and functions,

d) incorporates all or most of the principles of other countries

e) considers the properties and pollution charges of the OMW

f) considers soil physical and chemical properties

g) it is based on velocity of water movement, soil map interpretation and on the combination of limiting factors and downwards water movement.

The system allocates soil map units to Suitability Orders (S for suitable and N for unsuitable) and Suitability Classes according to the degree of their limitations (S1 for slight, S2 for moderate and S3 for severe limitations; N1 for currently not suitable and N2 for permanently not suitable for waste application.

Especially, the application/disposal of OMW directly on soil has to consider location, geology, physiography, geomorphology, hydrogeology, land use, soil structure, texture, water permeability, coefficient of hydraulic conductivity (saturated or unsaturated), porosity, presence and depth of impermeable soil layers. Moreover, it is necessary to include the soil quality indicators, which for the case of OMW disposal are pH, electrical conductivity, organic matter, total nitrogen, polyphenols, exchangeable potassium, available phosphorous and available iron. Total salts content, Sodium Adsorption Rate (SAR) and toxicity indicators are also recommended to be included. Toxicity could be assessed by using the standard methods for the determination (a) of nitrogen mineralization and nitrification in soils and the influence of chemicals on these processes (ISO 14238); (b) of the effects on earthworms (ISO 11268-1); (c) of the chronic toxicity in higher plants (ISO 22030); and (c) of soil biomass or soil respiration (ISO 14240). The selection among these standard methods should be based on several factors, such as current soil quality, present and future use of the area, amounts of produced waste and treatment level, and others.

OMW should also be analyzed in terms of macronutrient content, pH, BOD, COD, electrical conductivity, polyphenols, solids content, Ni, Cr, and Mo. The criteria of the system, adapted to the case of OMW disposal are presented in Table 2.

The proposed system has built up by using five steps (Fig. 1).

Step 1: Suitable or unsuitable soils for OMW application

Firstly, soils with the potential to receive or soils that should be excluded from OMW application are identified based on permanent physical and/or chemical characteristics (Table 2).

Step 2: Estimation of the maximum permitted OMW amount

The soils that are suitable for OMW application are further studied in order to define the maximum permitted amount (or the maximum amount they can afford) of OMW based on the physicochemical properties of targeted soils and of OMW and considering legally applied thresholds for these properties.

Provided that no Regulation or Directive exist in the EC, the rate of annual OMW application estimation is suggested to be performed taking into account the maximum permitted levels of potentially toxic elements as defined by the European Commission (EC Council Directive 86/278) for sewage sludge application and the thresholds as derived from the literature, especially for the non-toxic macronutrients (P, K, N) and for the available forms of metals. As regards heavy metals, the EC Council Directive defines thresholds for Cd, Cu, Ni, Pb, Zn and Hg. For the case of OMW potential risk may arise from Ni, Cr and Mo (from the steel parts of mills' equipment), which may be present in OMW (liquid and solid). However, high concentration values for these three metals (and also of the other heavy metals) are not expected in OMW and thus, heavy metals amounts that exceed the thresholds defined in the Directive are unlikely to be detected.

High concentrations of the other heavy metals that are not mentioned in the Directive are also unlike to be measured in OMW. Yet, the limit values for concentrations of heavy metals in soil as defined in the Directive should be considered prior the estimation of the maximum permitted OMW amount.

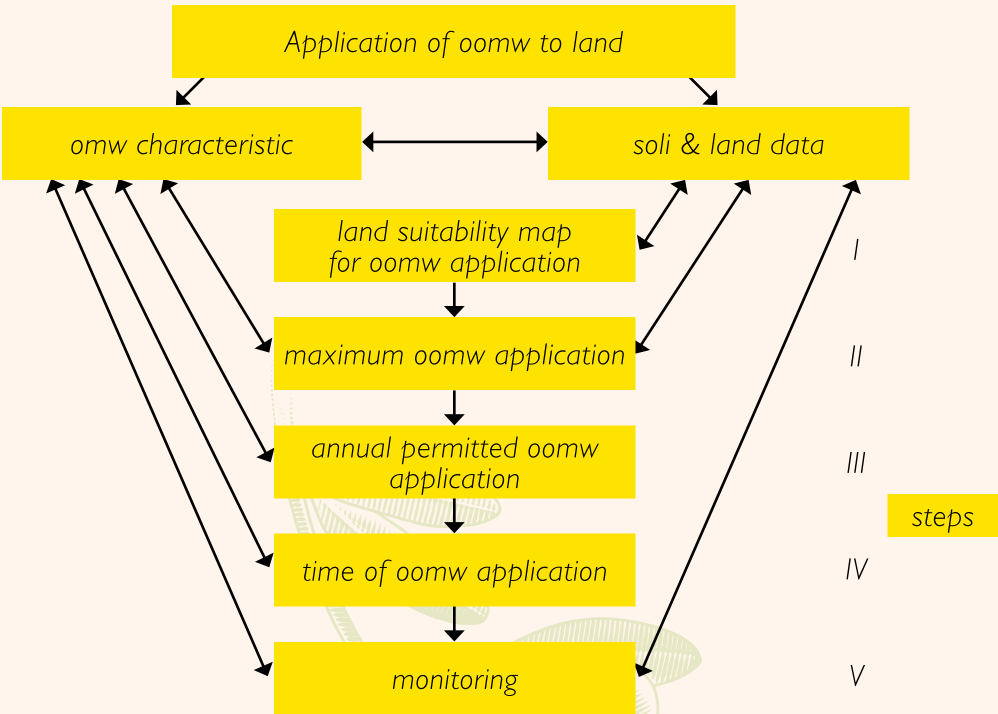


***Figure 1**. The proposed system for land application of OMW*

Property	S1	S2	S3	N1	N2
Flooding	never	seldom	often	always	As N1
Depth to bedrock,cm	>300	>180	100-180	<100	<50
Depth to impermeable layer, cm	>200	>180	100-180	<100	<50
Coverage with water	never	never	seldom	often	always
Groundwater level, cm	>300	>180	100-180	<100	<50
Infiltration rate, cm/h 30-150cm	2.0-6.5	0.5-6.5	0.5<,>6.5	0.5<,>6.5	As N1
Slope, %	<3	3-8	8-12	>12	>15
Stones, % (>7.5 cm)	<20	<35	>35		
Texture	All except CL, SC, SiCL, SiC, C, LS, S or with gravels	All except SiC, C, S or with gravels	All except C or with gravels	Clay (vertisols), very Sandy	As N1
Structure	granular angular-blocky	blocky or prismatic	massive platy compacted	vertic	As N1
SAR	<12	<12	>12		
pH	7.3-8.4	6.6-7.3	5.6-6.5	<5.6	As N1
EC, mmhos/cm	<4	<8	<16	16-40	>40
CEC, meq/100g	>16	8-16	<8	<8	
Salt, %	<0.09	0.09-0.16	0.16-0.26	>0.41	

Table 2: *Criteria for land suitability for OMW sludge application.*

Step 3: Estimation of annual permitted application of OMW

The annual rate and timing of OMW application could be determined by taking into account the maximum permitted levels of potentially toxic metals as defined by the European Community (EC Council Directive 86/278) for sewage sludge application and the thresholds as derived from the literature, especially for the non-toxic macronutrients (P, K, N) and for the available forms of metals. The latter, although general could be very helpful. However, it is strongly recommended to estimate the annual permitted application after evaluation of the specific local environmental conditions and soil quality. Since the most of the OMW constituents are non-toxic and are considered as important nutrients (N, P, K, organic matter, Fe, etc), the application of OMW could be beneficial for soil quality and may improve fertility. Thus, and as regards the nutrients' content, the OMW could be considered as nutritional material (like fertilizers) and thus annual dose estimation should follow the general rules of soil fertilization. However, due to the very high load of OMW in these constituents, the disposal on soil should follow restrictions and rules *and the annual application should be estimated by considering:*

- *The concentration of the specific elements/substances in soil*
- *The concentration of the specific elements/substances in OMW*
- *The specific climatic, geomorphologic and environmental conditions of the area that may affect the behavior of these elements/substance in soil (leaching, adsorption, decomposition, etc)*
- *The maximum permitted amount of each of these elements/substances that can be disposed on soil without changing its quality*

The distribution of OOMW may increase soil content in polyphenols substantially and thus, this is a very significant limiting factor for the agronomic use of OMW. All studies performed so far have reported noticeable increase in the content of phenolic compounds in soils immediately or after some months from waste application (Kavvadias et al., 2011). However, a healthy soil is capable to reduce the concentration of phenols through natural biodegradation processes (Mechri et al. 2008; Nikolaidis et al. 2008). Several studies demonstrate that toxicity in OMW-amended soil tends to disappear just few months after application.

However, if soil distribution is planned, some characteristics of polyphenols behavior in soil should be taken into consideration. Thus, one should consider that phenols do not move rapidly across the soil profile (Chartzoulakis et al. (2010), their leaching was shown to be negligible in soils rich in carbonates and clay materials while they can be adsorbed by soil organo-mineral components and thus can be detected in high concentrations even at depth of 125 cm (Sierra et al. 2007).

Low values of OMW's pH should always be an issue of concern as well as the impact of low acidity on many physicochemical and biological soil properties.

Step 4: Time of OMW application for different crops

In case of OM wastewater use for irrigation, the time of application has to be defined considering the annual rainfall rate, intensity and distribution throughout the year and the temperature, in relation to water balance, soil properties and processes, microbial activity and OMW decomposition. The background philosophy is to apply OMW at periods where rainfall induced leaching of the soil water is not expected.

Step 5: Soil Monitoring

The next step is monitoring the impact of OMW application on soil, on water bodies and the environment under the specific bioclimatic conditions of the Mediterranean areas through a systematically planned sampling scheme combined with different eco-bio-toxicological test. The most efficient way to produce a single-factor soil map or a land evaluation map for OMW disposal/application is to write a macro routine or command file. This specifies the selection criteria that are required to produce the land suitability map. The mapping unit and/or sample points are then interrogated to determine if they meet those required conditions. The fundamental process in the whole system, in order to evaluate for OMW application, is the comparison or matching of land use requirements (Table 2) with the attributes of the land-mapping units.

Using the steps illustrated in Fig. 1 and the land information system described above, land suitability maps for OMW application could be produced.

2.3.4 GUIDELINES FOR PERIODICAL SOIL QUALITY MONITORING

An initial geo referenced grid or free (based on the main soil types of each target area) soil sampling should take place at depth increments in order to define the current situation in representative, benchmark soils of the area.

Emphasis should be given to identify control soils i.e. soils that have never accepted OMW or other wastes in the area as well as soils in which OMW have been applied intensively. It is recommended that for the initial characterization of the area, the collected soil samples should be analyzed for all soil physicochemical properties.

After the initial characterization of the area, soil samples should be collected annually from hot spots, which would have been identified during the initial characterization of the area, and be analyzed only for the proposed soil quality indicators.

For the annual monitoring of the area, a geo-referenced soil sampling scheme should be planned and implemented by local authorities or through them by disposal areas owners, while the collected data is recommended to be stored and evaluated through GIS Land Information System. This would facilitate data management by local authorities.

2.3.5 SOFTWARE APPLICATION TOOLS FOR SOIL MONITORING

A software monitoring application tool for the sustainable management of OMW disposal areas was developed by IMS in the frame of PROSODOL project, which includes

(1) a sophisticated version suitable to be used by local authorities and,

(2) a more simple version suitable for individuals (mills' or disposal areas' owners).

Through this tool, farmers and local/regional authorities will have the opportunity to screen disposal or irrigated areas rapidly, identify potential risky conditions and proceed to detailed monitoring, if necessary, implement resources monitoring at field and municipal scale allowing, thus, continuous monitoring of the areas.

The first version of the application tool uses interpolation surfaces that indicate the distribution of the different chemical parameters in the area of interest, so the user can rapidly obtain an idea of the possible diffusion of the chemical parameters and the degree of risk in the vicinity of the waste disposal areas. This, potentially, allows also the establishment of an Operational Centre, which could be located, for instance, in cooperation with the Environmental Protection Office of the Local Government (District), in the premises of a Municipality, and can undertake the continuous monitoring of areas under risk and the scientific and consulting supporting of the owners. Such an operational center will enhance local authorities/ other stakeholders to screen disposal or OMW application areas rapidly, identify potential risky conditions and proceed to detailed monitoring, if necessary, implement resources monitoring at field and municipal scale allowing, thus, continuous monitoring of the cultivated areas.

The design of the particular software package needs to monitor a number of private fields that are spread around and make queries based on various spatial and chemical attributes.

The second simpler version of the tool is addressed mainly to individuals, such as mills' owners, disposal areas owners and farmers who use OMW for irrigation. It is user friendly and requires no specific knowledge and skills in order to be correctly applied under real conditions. The application provides interested individuals with the potential to monitor soil quality of their property periodically, identify on time potential risks and take the appropriate measures.

Interpolation surfaces-The sophisticated tool

Among the goals of the project was the development of an application soil-monitoring tool, which will facilitate the control of targeted areas as well as, others with similar activities, by monitoring several basic chemical parameters that will reflect the wastes' disposal activity of the areas. Having taken a number of data around the vicinity of the OMW disposal areas in the pilot municipality of the project, it was possible to create surfaces that indicate the distribution of the different chemical parameters in the area, so that to obtain an idea of the possible diffusion of the chemical parameters and the degree of risk in the vicinity of the waste disposal areas.

A number of interpolation algorithms were tested to verify the most appropriate way of mapping the specific parameters, without creating secondary effects (e.g. bull's eye effect around.

The interpolation method used in order to proceed with the surface analysis was Inverse Distance Weighting (IDW). IDW calculates cell values by averaging the values of sampling points in the vicinity of each cell based on distance. Consequently, the closer a point is to the centre of the cell being calculated, the more influence or the more significant weight it has in determining the final output value. This method assumes that the values of the chemical feature being mapped decrease in influence with distance from the sampled locations.

Furthermore, IDW offers the potential to the user to define the power of known values as an individual parameter.

The power controls the significance of measured values on the interpolated cells, based on their distance from the sampling points.

By defining a high power, more influence is given to the nearest points and the resulting surface will have more local detail.

On the contrary, a lower power will emphasize the points located further away. In this project, all the interpolated surfaces were created by defining a power of 2 for the known values.

It has to be mentioned that surfaces could only be interpolated for more than 4 known values. In cases where the measured values were less than 4 it was not possible to interpolate surfaces and therefore no results were produced.

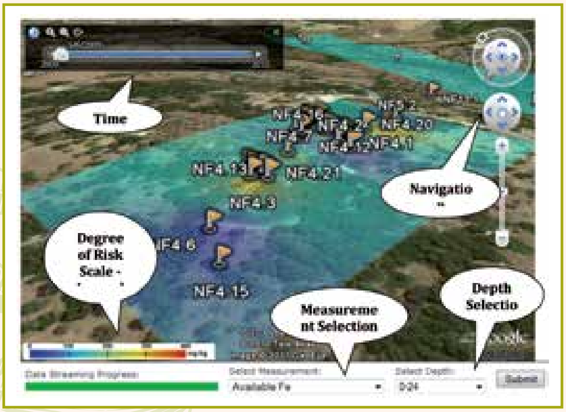
There are several tools and techniques in order to present the interpolated surfaces via an understandable and easy to use interface. In order to present these interpolated surfaces (i.e. images actually) inside the website of PROSODOL, so at the end a web based application to be developed in order to be used and handled by the public, integration of the entire interpolation process results into a Map API such as Google API, or Google Earth API, flash maps techniques (Flash Builder software) was performed, so that users can view the interpolated surface area images simultaneously above a topographic/satellite map provided by those APIs.

After consideration of the time based functionality, which users can view the chemical parameters diffusion in a subsurface over time, testing and using several of the above techniques, Google Earth API was the most appropriate to use, which handles this possibility by integrating time tagged images via EXtensible Markup Language (XML), a language designed to transport and store information data.

ArcGIS Desktop was chosen for creating the interpolated surface maps using the ArcGIS ModelBuilder Tool. ModelBuilder is an application in which user can create, edit, and manage models. Map 7 presents the interface of the map application.

These maps could be used by regional/local authorities as a powerful tool for screening the status of soil health at OMW disposal areas while at the same time could be used as database, in which past and future soil quality data could be stored and used for evaluation, decision making and management.

The application could be easily developed and installed, however, it requires an extensive and detailed initial screening of the areas and periodical monitoring in order to feed the database with data.



Map 7. The interface of the map application.

The software offer to the user a series of choices, like

- measurement selection : the user selects the name of the chemical parameter in order to see the corresponding interpolated surface map.
- depth selection : the user can choose the depth of soil for which she/he wishes to see the value of the selected chemical parameter. Then the user can submit the information provided, and the map application starts to stream the data needed and present the corresponding interpolated surface map.
- navigation controls : the user may navigate inside the map through the navigation controls, in any direction, angle, pan and zoom, giving her/him the freedom of any view perspective.
- time slider : once time is introduced, a time slider appears, allowing the end user to manipulate time. An animation of the interpolated surface area map can be viewed through different time periods on the Google map.
- degree of risk scale - legend : when the interpolated surface map is loaded the corresponding scale of the risk degree of the selected chemical parameter is shown.

The 3D map application was designed in such a way, that the end user can easily and effectively use, and retrieve the surface interpolated information needed.

The simpler tool for individuals

The system is targeting towards public or private users that need to monitor measurement results of targeted areas in order to evaluate the degree of risk in the vicinity of the waste disposal areas.

The system has been implemented in four languages (English, Greek, Italian and Spanish) and it is available for free download from the Web site of the project (http://www.prosodol.gr/homepage/menu/downloads).

The application tool requires the periodical measurement of the proposed soil quality indicators, then the users can enter measurements at various time intervals and monitor the fluctuation of the values through the time.

Results are indicated through predefined diagrams that have orange and red flags depending on the degree of alert that needs to signaled to the users. The user can enter more than one OMW disposal area and can also export the data into an excel format and use them for other applications.

The design process of the monitoring application tool consists of three basic principles:

1. Need to specify the most important chemical parameters for evaluating the degree of risk of the waste disposal areas
2. Need to specify the limits and range of risk zones (range of values), such as the red and the orange risk zones.
3. Design an interface that satisfies specific user needs, such as inserting, editing, searching functionalities of waste disposal areas measurements, as well as graphically presenting the risk assessment results.

The whole application was designed and implemented with the Microsoft Visual Studio (2010) software, enhanced with the capability of monitoring one or even more measurements on different waste disposal areas, capable of use for a larger scale services, than private use only.

The tool permits the monitoring of eleven soil chemical parameters, i.e. the soil quality indicators plus total Ni, Cr and Mo.

The three heavy metals were included because, as already discussed, in case of inferior quality steel equipment of olive oil mills there is a potential risk of soil pollution.

The limits and the values range of the risk zones (red - high risk, orange - moderate risk) are denoted in the specific columns of Table 3.

A user, in order to evaluate the degree of risk in the vicinity of a waste disposal area, needs to assign values to some or all of the above chemical parameters, and evaluate graphically afterwards the results upon a XY point diagram, where the red and orange risk zones are also presented (Fig. 2).

The inserted chemical parameters values, which are stored in the systems database, can be viewed graphically on a XY diagram, presenting in the most meaningful way if the corresponding measurement exceeds or is inside the parameter limits (risk degree).

The application tool has an extra functionality of exporting measurements in Excel File format (Fig. 3). The user not only can search measurements with certain criteria and needs, but also can export the search results information (measurements found) in an Excel File Format.

The tool has also another extra functionality, the one of measurements printing report.

The user not only can search measurements with certain criteria, but can also print the search results (measurements found), actually all the visualized graph chemical parameters information on a paper.

For the easy download and use of the application tool, a detailed and useful manual was developed in four languages and is available from the web site of the project.

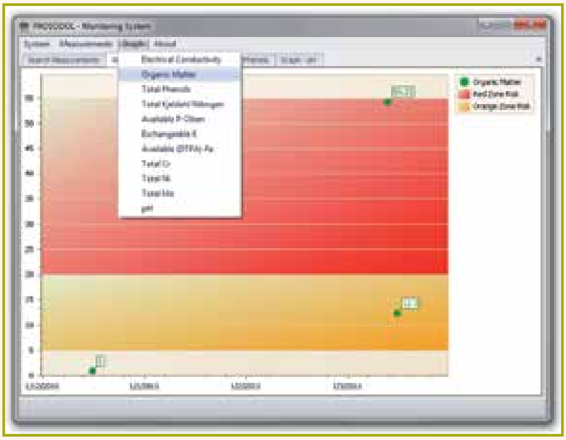


Figure 2. Visualization of the selected chemical parameter.

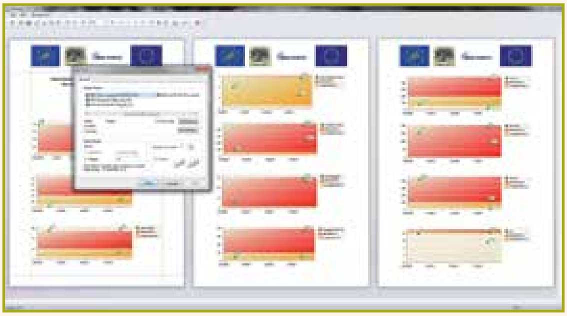


Figure 3. Print Search Results Report

2.3.6 GUIDELINES FOR PERIODICAL WATER BODIES MONITORING

Neighboring water bodies is recommended to be monitored periodically. Water samples should be collected from all 1st, 2nd and 3rd catchment order discharge and in the groundwater both before and after rainfalls. This way a database could be developed where the ground and surface water will be recorded spatially and temporarily. Water would be analyzed for electrical conductivity, pH, polyphenols, K and other suitable parameters, which are considered water quality indicators for OMW application.

2.3.7 CODE OF GOOD PRACTICES FOR SOIL MANAGEMENT

A Code of Good Practice should be developed which gives advice in relation to soil management practices, which should be adopted by all sectors, which have the potential to impact on soil quality. The Code should be a practical guide that will assist owners of the OMW disposal areas, farmers who may use OMW for soil fertilization or land managers to protect the environment in which they operate. The Code should describe key actions that the main actors can take to protect and further improve the quality of soil, water, and air and to meet legal obligations. The Code should not be just a manual on how to manage such areas but should assist on selecting the appropriate actions for the specific conditions (environmental, climatic, social, financial). Such a Code entitled "Good Practices for the agronomic use of olive oil mills wastes" has been developed within the framework of the PROSODOL project and is available on the web site of the project. It includes guidelines for soil sampling, use of OMW for crops irrigation (technical and financial aspects), for OMW disposal on soil, for periodical soil monitoring, soil remediation techniques, composting and existing legislative framework.

2.4. SOIL REMEDIAL TECHNIQUES

2.4.1 CONTAMINANTS FATE AND TRANSPORT

One of the main PROSODOL project's objectives was to develop and implement cost effective soil remedial actions that will remediate or, at least, protect soils from further degradation. It should be, however, highlighted that the development and implementation of soil remedial actions, appropriate and specific for OMW disposal areas, have been never implemented and demonstrated before and thus, the selection among available soil remedial methodologies was not existed as an option and, most significant, there was no possibility to compare the obtained results with results obtained from other already implemented and demonstrated methods. Therefore, all potentially applicable soil remedial methods were recorded and evaluated.

It was, however, clear that a soil remediation and protection plan suitable for OMW disposal areas, should include methodologies for polyphenols reduction and retention or immobilization of inorganic constituents. Therefore, for the reduction of polyphenols concentration in soil, in situ-bioremediation was selected since it targets to the biodegradation of organic pollutants in soil by taking full advantages of the natural biodegradation process of organic molecules by soil microorganisms (Thomas and Ward 1989; Cauwenberghe and Roote 1998; Cookson 1995). For the reduction of inorganic soil constituents, the use of natural zeolite, clinoptilolite, as soil amendment was considered the most suitable for this case because of the already well-known properties of natural zeolites to attract, retain and slowly release many inorganic cations, such as K+, Na+, Fe3+, Cu2+ and others. Moreover, the method is of very low cost and very easy to be implemented, even by no qualified personnel (Doula et al., 2011; 2012).

2.4.2 MECHANISM AND PATHWAYS OF CONTAMINANTS

The mechanisms and pathways, by which the contaminants originated by OMW could be released from their current locations, move through environmental media, and potentially impact human and ecological receptors have been evaluated (USACE, 2005). Potential release mechanisms include wind erosion; surface water runoff, erosion and deposition, and water infiltration through soil layers. These release mechanisms may enable contaminants mobility and migration from their current locations to adjacent media (e.g., from surface soil to subsurface soil and bedrock). However, wind erosion; and erosion-deposition are not considered to be significant mechanisms for contaminants releases from the site.

Surface water runoff following a rain or leaching through soil are the two mechanisms that are considered as significant pathways for contaminants (both organic and inorganic) transport from the pilot area (as well as other similar areas under Mediterranean climatic conditions). Wind erosion could also be considered as a potential transport pathway, but mainly for soils consisting of loose materials (e.g. sandy soils). Water that infiltrates into surface soils could remain fixed in the unsaturated vadose zone soils or percolate to groundwater. Water percolating through contaminated soil could result in the dissolution of water-soluble compounds, which could be transported to groundwater.

This constitutes a real threat for the mitigation of polyphenols, however, these molecules can not easily transport through the soil profile due to their size and most likely remain at the upper soil layer. Surface run-off may result to polyphenols transport and thus in contamination of neighboring systems, mainly during the periods when wastes are disposed on soil. Indeed The dissolution of the inorganic OMW constituents is possible however, the process is strongly depended on soil pH, soil clay content, texture, Cation Exchange Capacity, a.o. Transport through groundwater is a potential mechanism by which contaminants could move from the site and impact human and ecological receptors in the long term, however, mainly when the level of the groundwater table is high. High clay content, in general, protects soil from the very high organic and inorganic load of the wastes, while high pH and CaCO3 content from wastes' acidity.

2.4.3 OBJECTIVES OF THE REMEDIAL STRATEGY

Considering the above, the objectives of the remedial strategy would be:

1. *Reduction and stabilization of soil total organic matter below 5% (Ilaco, 1985; MAAF, 1988)*
2. *Reduction of soil total polyphenols at concentrations equal to background levels of the area, i.e. 57 mg/kg (Swartjes, 1999; Kavvadias et al., 2010, 2011)*
3. *Reduction of soil electrical conductivity and stabilization under threshold of salinity, i.e. <4mS/cm (Ilaco, 1985; MAAF, 1988)*
4. *Reduction stabilization of soil total nitrogen below 3,0mg/g, i.e 0,3% (Ilaco, 1985; MAAF, 1988; Brady, 1990; Tisdale et al., 2003)*
5. *Reduction of exchangeable K and Mg below 1,2cmol/kg and 2,2cmol/kg, respectively, or immobilization on reactive media (Ilaco, 1985; MAAF, 1988)*
6. *Reduction of available Fe and Cu below 50mg/kg (Abreu et al., 2005; Mitra et al., 2009; Kavvadias, 2011) and 3,0mg/kg (Ilaco, 1985; MAAF, 1988), respectively, or immobilization on reactive media*
7. *Reduction of available P (Olsen P) below 28mg/kg (Carrow et al., 2004) or immobilization through precipitation*
8. *Reduction of available B below 1.5mg/kg (Carrow et al., 2004) respectively.*

2.4.4 IN SITU BIOREMEDIATION (BIOPILING)

The method applied was the bioremediation and in specific biopiling, but with some changes in order to be conformed to the local conditions and the specific characteristics of the area under treatment (Schulz-Berendt, 2000).

Bioremediation is a process in which microorganisms metabolize contaminants through oxidative or reductive processes.

As such, it uses relatively low-cost and simple techniques, which generally have high public acceptance and can often be carried out on site.

However, bioremediation is not always suitable for a given problem and detailed study of local soil conditions are required in order to identify if the organic contaminants could be biodegraded by soil microorganisms and if the residual contaminant levels after bioremediation implementation are acceptable (Vidali, 2001).

Under favourable conditions, microorganisms can completely metabolize organic contaminants and convert them into non-toxic by-products, such as carbon dioxide and water or organic acids and methane (USEPA 1991).

Generally, bioremediation can be used in any soil type with adequate moisture content, although it is difficult to supply oxygen and nutrients into low permeability soils.

It should be noted, however, that very high contaminants concentrations may be toxic to microorganisms and thus may inhibit their activity. In such cases of heavy contaminated sites, bioremediation may not be the best remediation option.

Therefore, prior implementation, a feasibility investigation is needed to determine if biodegradation is a viable option for the specific site, soil type and contaminant conditions (Aggarwal et al. 1990). For the determination of the bioremediation potential of a site contaminated with organic wastes, treatability studies are required to provide specific information regarding the potential rate and extent of bioremediation, the fate and behavior of organic pollutant in surface soil and deeper vadose zone. Treatability studies include studies in field and in laboratory. A flowchart for determining the bioremediation potential of an OMW contaminated site is presented in Scheme 1.

The application of bioremediation included many stages (Scheme 1), which, in general, include:

1. Feasibility studies to identify the bioremediation potential of the site of interest
2. Implementation of the bioremediation technology, and
3. Effectiveness monitoring

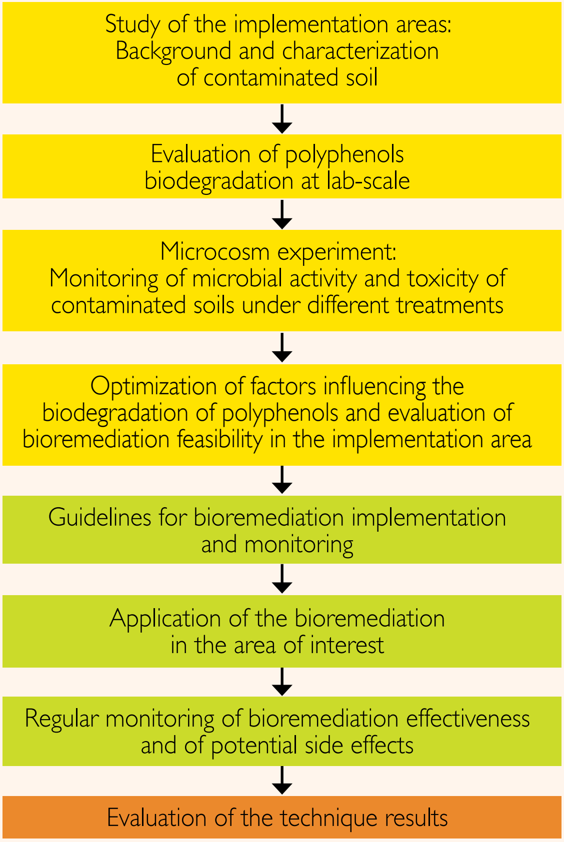
During the 1st stage, laboratory studies were performed in order to determine the optimum conditions for microbial activity and if these present in the area of interest. Therefore, the existence of microorganisms capable to biodegrade polyphenols was determined. If such microorganisms were not existed, then they should be artificially added. Thereafter, the conditions (i.e. soil aeration, moisture, nutrients' concentration) that ensure the optimum microbial activity were determined. For this purpose, the microbial activity and some soil properties under various conditions of aeration, moisture and nutrients' levels were tested in soil samples collected from the targeted area.

The required determinations that were performed during the feasibility studies were:

- *Separation, identification and quantification of polyphenols (using HPLC)*
- *Water soluble polyphenols*
- *Microbial biomass carbon*
- *Water soluble carbon fraction*
- *Soil pH, electrical conductivity and all other soil parameters*
- *Phytotoxicity tests by conducting germination tests*
- *Exotoxicity tests*
- *Soil enzymatic activities by identifying Urease and Dehydrogonase activity*

Soil samples were analyzed for the above parameters before and after the application of the different conditions of aeration, moisture and nutrients' level at lab scale. Thereafter, a series of lab experiments were carried out in order to identify the optimum treatment methodology to be applied at a pre-selected pilot area.

Several scenarios were examined in order to select the optimum one for the specific disposal area :
I No treatment (natural attenuation);
II) Soil aeration;
III) Soil aeration plus nutrient addition;
IV) Soil aeration plus microorganisms and enzyme inoculation;
V) Soil aeration plus compost addition.



Scheme 1: *Flow chart for determining the bioremediation potential of OMW disposal sites.*

The soil parameters that were measured, besides of polyphenol concentration, were: pH, electrical conductivity, enzyme activities, assay of phytotoxicity effect (germination seed experiment) and ecotoxicity, measured by luminescent bacteria.

The feasibility studies identified the optimum conditions for the implementation of the bioremediation at the pilot area and a set of instruction on how to implement the technology was delivered.

In particular, the scenario ii) (aeration) was identified as the optimum one, since the nutrients and moisture content of the soil under remediation were adequate to provide microorganisms the appropriate conditions for their activity.

Thus, the proposed remedial scenario foresaw: "In the beginning of the field-works, the area should be homogenized with the use of field machines (e.g. tilling machines, excavators, mechanical shovel) until 25cm depth, while large and medium stones it would be better to be removed.
One subplot should be used as control area, while after area configuration, soil samples should be collected at time zero (T0) and every 15 days throughout the bioremediation treatment (6 months). The area should be tilled every 15 days to ensure aeration while special care should be taken to maintain soil moisture at sufficient level for the enhancement of microorganisms activity".

Bioremediation was applied between 5th November 2010 and 14th May 2011 in an area of 30m x 25m (i.e. 750m2), where untreated OMWs from a 3-phase mill were uncontrolled disposed on soil surface for more than 15 years. Considering that soil was treated until depth of 25cm, the total treated soil volume was ***187.5m³.***
The two following steps, namely implementation and effectiveness monitoring were carried out simultaneously since after each treatment (i.e. tillage-soil aeration) soil samples were collected and analyzed to record bioremediation progress.
During the implementation, the soil of the area was homogenized with the use of an excavator until 25cm depth, while large and medium stones were removed (Photos 4-9).
A small area at the upper part of the field was used as control area since no wastes are disposed there. After area configuration, soil samples were collected (one control and one mixed sample from the main experimental area).

According to the instructions the area should be tilled every 15 days until 25cm soil depth and this was carried out by using a small tilling machine. Thus, mixed soil samples were collected from the area on 5/11/2010 (first day of implementation); 23/11/2010; 27/12/2010; 18/1/2011; 12/2/2011; 14/3/2011; 15/4/2011 and on 14/5/2011. The samples were transferred to the lab and analyzed for texture classification, saturation percentage (%SP), electrical conductivity, total salts, pH, organic matter, calcium carbonate, total nitrogen, water soluble Na, exchangeable Na, exchangeable K, exchangeable Ca, exchangeable Mg, available P (Olsen), available-DTPA Fe, available-DTPA Cu, available-DTPA Mn, available-DTPA Zn, available B, total polyphenols, NH_4^+, NO_3^-, Cl^-, PO_4^{3-}, SO_4^{2-}, ESP and SAR. In May 2011 it was confirmed that soil quality, as far as the polyphenols content, was significantly improved. Thus, it was decided that the pilot phase was successfully accomplished.
After the completion of bioremediation implementation, results indicated that the method could be effective in reducing soil polyphenols, total nitrogen, available iron and available boron, although the latter was not exceeded the threshold value before the remediation. Soil analyses revealed that the total polyphenols content was significantly reduced (Table 4) during the bioremediation implementation. The initial very high value was reduced by 72.6% while the final polyphenols concentration was very low, lower than the control sample of the area.
There was a gradual decrease of total N throughout the sampling period. The soil bioremediation procedures seem to enhance N mineralization. The initial N content was significant high, considering the threshold of 3.0mg/g above which, a soil is characterized as very rich in nitrogen and also the mean value of the control samples of the pilot area, which is 2.3mg/g. The final N values are considered satisfactory and the soil could be characterized as containing the sufficient amount of total nitrogen. Available iron, although very much reduced, however the final value was not lower than the threshold of 50mg/kg. However, this result is accepted and the method is considered as effective in reducing available Fe.
On the contrary, the method was not effective in reducing at acceptable values the soil organic matter, exchangeable potassium and magnesium, available cooper and phosphorous. In specific, in-situ bioremediation seems to have no effect on available Cu concentration and on the exchangeable Mg, while the reduction in the concentration of exchangeable K and available P, although substantial, yet the values of these two parameters are still very high compared to the accepted thresholds.






Photos 4-9. *Pilot area configuration prior the implementation of in-situ bioremediation*

Soil Parameter	Target value	Blank sample	Value before treatment	Value after treatment
Organic Matter, %	5.0	4.3	6.4	6.02
Total polyphenols, mg/kg	57	57	117	32
Electrical Conductivity, mS/cm	4.0	0.67	1,89	0,67
Total Nitrogen, mg/g	3.0	2.3	4.4	3.0
Exchangeable Potassium, cmol/kg	1.2	0.6	7.8	4.1
Exchangeable Magnesium, cmol/kg	2.2	2.9	4.0	3.6
Available Iron, mg/kg	50	46	106	67
Available Copper mg/kg	3.0	2.6	4.6	4.4
Available Phosphorous, mg/kg	28	16	113	77
Available Boron, mg/kg	1.5	0.2	0.6	0.3

Table 4. *Soil parameters values before and after bioremediation implementation in relation to the remedial objectives.*

2.4.5 COSTS GENERATED FROM THE IMPLEMENTATION OF IN SITU BIOREMEDIATION (BIOPILING)

In general, there is not a common rule or general guidelines for the implementation of the bioremediation technique since the conditions and the methodology are depended on the specific site and soil characteristics.
The calculation of the cost was based on the costs generated during the treatment of an area of 750m^2 until 0,25cm depth which was extrapolated to a field of 1acre while the final cost is given in €/m^3 of treated soil.
For the specific pilot area, the scenario ii) was selected as being the optimum one. However, for other areas, one of the other four scenarios (i, iii, iv and v) could be potential. Although the cost of feasibility study would be the same, the cost of implementation step would be significant different.
In specific, and for a general application of bioremediation feasibility studies, the required experimentations are

- *Total Organic Carbon and total Nitrogen (8€/sample)*
- *Elemental analysis by ICP-OES 5€/sample*
- *Nitrates, sulfates, phosphates etc by ionic chromatography 5€/sample*
- *Physicochemical, biochemical and microbiological assay 80€/sample*

Total cost/sample = 98€ excluding VAT

which will be carried out under different conditions of moisture, oxygen and nutrition content.

Similarly, for the specific pilot area, the existence of native microorganisms which were capable to biodegrade polyphenols at a satisfactory rate and extent was confirmed. However, for other cases it would be possible to transfer such microorganisms from another area to the targeted area, which causes additional cost during the implementation step.
There is also the possibility, depending on local conditions, to use specific soil covers (e.g., plastic or geofabric/textile) to protect the implementation area. The cost for the implementation of in-situ bioremediation is included in the following Table 5.

Activity (for the specific remedial scenario at a field of 1 acre)	Mean cost in €
Feasibility studies	98.00
Area protection (metallic fencing)	1,000.00
Area configuration (8 hours are needed for 1acre)	
- machine for area configuration (for 1day, 8 hours for 0.50€/m2)	500.00
- labor cost (2 workers for 1day for 0.50€/m2 /worker)	1,000.00
Soil aeration every 15 days for 7 times (each time four hours are charged for tilling 1 acre)	
- small tilling machine (hours for 0.50€/m2))	1,750.00
- labor cost (2 workers for four hours each)	3,500.00
Soil analysis, 7 times x 2* samples x 27 soil parameters	
(mean cost of each parameter=4.00€)	1512.00
Annual Monitoring of soil quality for the next 10 years	
(analysis of 2* soil samples annually x 27 soil parameters)	2160.00
Total cost (VAT excluded)	11,520.00
Cost per remediated cubic meter (VAT excluded)**	**46,08**

Table 5. *Cost for the implementation of in-situ bioremediation*

**one blank soil sample and one collected from the treated area*

*** 1000m2 x 0,25m depth = 250m3 were treated*

2.4.6 CLINOPTILOLITE AS SOIL AMENDMENT

Zeolites are highly porous aluminosilicates with different cavity structures. Their structures consist of a three dimensional framework, having a negatively charged lattice. The negative charge is balanced by cations, which are exchangeable with certain cations in solutions (Fig. 4).

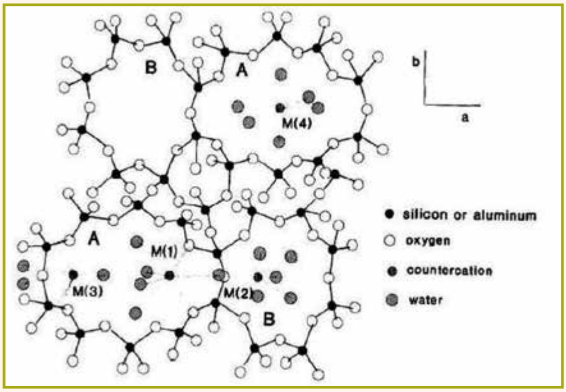
Zeolites consist of a wide variety of species, more than 40 natural species. However, the most abundant and frequently studied zeolite is clinoptilolite, a mineral of the heulandite group. Its characteristic tabular morphology shows an open reticular structure of easy access, formed by open channels of 8–10 membered rings. Clinoptilolite has been shown to have high selectivity for certain pollutants. The characteristics and environmental applications of zeolites have been extensively studied (Inglezakis, 2003; Doula et al., 2011, 2012). High ion-exchange capacity and relatively high specific surface areas, and more importantly their relatively cheap prices, make zeolites attractive adsorbents. Their price is about 120-150€/tn, depending on the quality of the mineral. Zeolites are becoming widely used as alternative materials in areas where adsorptive applications are required. They have been intensively studied recently because of their applicability in removing trace quantities of pollutants such as heavy metal ions and phenols. Adsorption has been investigated to remove phenol from an aqueous solution by using zeolites as adsorbents and adsorption properties were compared to those of an activated carbon (Doula et al., 2012).

The use of clinoptilolite as soil amendment was applied between 5th November 2010 and 10th July 2012 in an area of 8.5m x 12m (**i.e. 102m^2**) where untreated OMWs from a 3-phase mill were uncontrolled disposed on soil surface for more than 15 years. Considering that soil was treated until depth of 25cm, the total treated soil volume was **25.5m^3**. **It should be highlighted that the area during the treatment with zeolite continued to accept surface disposal of wastes.**

The methodology included three stages:

1. Complete physicochemical characterization of the area of interest
2. Area configuration and application of zeolite on soil
3. Effectiveness monitoring

During the 1st stage, the area was monitored regarding soil parameters and soil quality for 1,5 years during which soil samples were collected every two months and analyzed for texture classification, saturation percentage (%SP), electrical conductivity, total salts, pH, organic



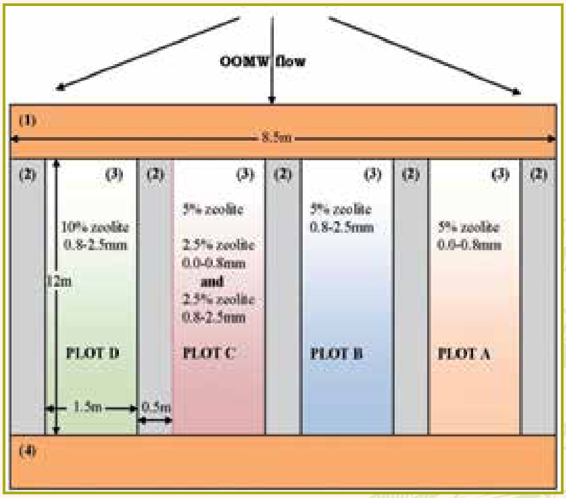
matter, calcium carbonate, total nitrogen, water soluble Na, exchangeable Na, exchangeable K, exchangeable Ca, exchangeable Mg, available P (Olsen), available-DTPA Fe, available-DTPA Cu, available-DTPA Mn, available-DTPA Zn, available B, total polyphenols, NH_4^+, NO_3^-, Cl^-, PO_4^{3-}, SO_4^{2-}, ESP and SAR.

After having identified the threats to soil quality and the parameters that are mostly affected by the disposal of OMWs, application of zeolite on soil at two ratios and two grain sizes was carried out on 5 November 2010. For this, the area was configured in order zeolite to be added as soil amendment and it was divided into four sub-areas (Photos 10-17 and Scheme 2).

Clinoptilolite was added as dust with particles diameter <0.8mm, and of larger size (particles diameter of 0.8mm-2.5mm). After clinoptilolite application, the area was tilled until 25cm depth with a small tilling machine.

For monitoring methodology effectiveness, soil samples were collected and analyzed every 2 months. In specific, soil samples were collected and analyzed on 18/1/2011; 23/3/2011; 17/5/2011; 28/8/2011; 8/11/2011; 24/1/2012; 29/5/2012 and 11/7/2012.

Photos 10-13. *Configuration of area where zeolite was applied as soil amendment*



(1) Blank zone without zeolite which will be affected by the surface disposal of OMW.
(2) Buffer zone between the experimental plots
(3) Experimental plots with different zeolite content
(4)Zone after the zeolite-zones where "treated" leachates from zones (3) will arrive. Soil analyses results from this Zone will be periodically compared with the results from Zone (1).

Scheme 2: *Experimental plots and description of the applied experimental strategy for the application of clinoptilolite.*




Photos 14-17. *Zeolite application on soil.*

Table 6 includes the values of the soil parameters measured during the treatment in relation to the remedial objectives. Since the area continued to accept wastes disposal during the treatment, a range of values for all parameters is given in Table 6 and not absolute values. In general, and by considering that the soil stress due to wastes disposal continued during the treatment, we may conclude that the results of this type of soil remediation are very much satisfactory and that the method succeeded to protect to a great extent the quality of soil.

Although the final values of soil organic matter are higher than the target value, thus, the use of clinoptilolite as soil amendment stabilized and maintained soil organic matter (OM) values at constant values (between 5.6 and 7.1% which are very close to target values). This is owed to the improvement of soil aeration and thus to the enhancement of soil microorganism activity to biodegrade soil organic matter. The effect of zeolite on total nitrogen content is similar to that on OM content and due to the same reasons, however, the final values of total nitrogen are considered unacceptable related to the target value.

Exchangeable K and available metal Fe were significantly increased in soil. The increase is owed to the retention of these elements from clinoptilolite. However, the increase is not attributed to the increase of these elements in soil particles but in zeolite framework. Consequently, this increase ***does not lead*** *to extent K and Fe* *leaching but to slow release from zeolite to soil solution contributing thus to the improvement of soil quality and to the prevention of nearby systems overloading. On the contrary, and same as during bioremediation, no effect on available Cu was recorded.*

Regarding soil electrical conductivity, its values were decreased due to the retention of ions within zeolite framework; the EC of soil was lower than the target value of 4mS/cm. Thus, despite the increase in exchangeable K and available metals contents in soil, these amounts do not increase soil electrical conductivity because ions are held in/on the zeolite framework.

Total polyphenols were reduced, however not as such extent to satisfy the target value.

Available phosphorous was decreased as far as its higher concentration is concerned; however the final values are unacceptable. Exchangeable Mg was also significantly decreased and stabilized at lower values than the initial ones but it concentrations remain almost double than the target value. No significant effect was recorded regarding the presence of available B in soil, which remained higher than the target value after the treatment.

No significant difference was obtained from the different ratios and different grain sizes of clinoptilolite, thus it is proposed that the use ***of up to 5% zeolite*** *on soil could result in substantial improvement and protection of soil quality, as far as the above mentioned parameters.*

Soil Parameter	Target value	Blank sample	Value before treatment	Value after treatment
Organic Matter, %	5.0	4.3	6.0-29.0	5.6-7.1
Total polyphenols, mg/kg	57	57	54-118	32-94
Electrical Conductivity, mS/cm	4.0	0.67	1.40-6.93	1.08-2.91
Total Nitrogen, mg/g	3.0	2.3	4.0-17	4.0-7.0
Exchangeable Potassium, cmol/kg	1.2	0.6	7.0-12	13-20
Exchangeable Magnesium, cmol/kg	2.2	2.9	7.1-11	5.1-5.4
Available Iron, mg/kg	50	46	119-202	123-333
Available Copper mg/kg	3.0	2.6	3.9-8.5	4.6-8.5
Available Phosphorous, mg/kg	28	16	162-591	171-387
Available Boron, mg/kg	1.5	0.2	1.0-3.0	0.9-3.0

Photos 10-13. *Configuration of area where zeolite was applied as soil amendment*

2.4.7 COSTS GENERATED FROM THE USE OF ZEOLITE AS SOIL AMENDMENT

The calculation of the cost was based on the costs generated during the treatment of an area of 102m2 until 0,25cm depth which was extrapolated to a field of 1acre while the final cost is given in €/m3 of treated soil.

Prior to clinoptilolite application, preliminary land configuration activities should be carried out. In particular, the disposal area should be homogenized with the use of light field machines (e.g. tilling machines, excavators, mechanical *shovel) up to almost 25cm soil depth. Stones should be preferably removed.*

The addition of clinoptilolite at 5% w/w is considered appropriate for OMW disposal areas considering that no OMW disposal will further occur.

For 5% zeolite content the amount of clinoptilolite to be added is almost 150tn/ha or15kg/m2. It is recommended that clinoptilolite should be of small grain size in order to be more effective and well distributed. Thus, it could be applied as dust with particles diameter <0.8mm, or of larger size (particle diameter of 0.8mm-2.5cm). Very small grain size, although more effective, is difficult to be distributed because of the dust produced during the application. However, it may be considered the possibility to use a mixture of clinoptilolite (in dust form and grain size of 0.8mm-2.5cm). Clinoptilolite should be distributed homogeneously and very well tilled.

After application, it is possible that periodical irrigation, in order to avoid excess sodium leaching would be necessary. The amount of applied water should be defined based on total cumulative net infiltration (subtracted estimated evaporation and adding precipitation), considering water parameters (chemical properties) and soil properties (e.g. soil bulk density, moisture content, electrical conductivity, texture, exchangeable cations). This management intends to achieve high leaching efficiency (i.e. remove the maximum salt possible per unit of leaching water) by using intermittent leaching with continuously unsaturated conditions on the soil surface, minimizing some surface ponding.

Soil quality should be monitored annually. Especially, after the zeolite applications, soil samples should be analyzed for SAR and ESP every two months and for the first six months period after application.

In case that values of SAR and ESP exceeded the upper limits (13cmol/kg1/2 for SAR and 15% of ESP) the area should be periodically irrigated.

The cost for the application of this remedial method is shown in Table 7.

2.4.8 COMBINATION OF THE BIOREMEDIATION AND ZEOLITE ADDITION

Considering the results obtained relative to the final quality parameters of treated soils, it is recommended that a combination of the methods would be more effective. The combination includes two stages:

1st stage

Implementation of bioremediation. The treatment is anticipated to reduce significantly the polyphenols content, to reduce and stabilize total nitrogen content as well as to reduce available iron and boron.

2nd stage

Addition of clinoptilolite at the bioremediated area. The treatment is anticipated to stabilize and reduce soil organic matter, reduce the electrical conductivity and stabilize exchangeable potassium and iron because of the zeolite property to keep them on/in its 3D framework.

An overall reduction of available phosphorous and boron is anticipated due to both the remedial actions.

In case of the implementation of the 2-methods combination the cost would be the sum of the respective cost, however reduced by 1,000€/acre for the metallic fencing (area protection) and by 2,160€/acre for the annual monitoring of soil quality which is charged once.

Thus the final cost would be 60.81€/m3 of treated soil.

Activity (for the specific remedial scenario at a field of 1 acre)	Mean cost in €
Initial monitoring of the area For flat and homogenous areas: 2 soil samples* x 27 soil parameters (mean cost of each parameter=4.00€)	108.00
Area protection (metallic fencing)	1,000.00
Zeolite purchase (15 tons; 120€/tn)	1,800.00
Area configuration (8 hours are needed for 1acre) - renting an excavator or other machine for area configuration for one day (i.e. 8 hours for 0.50€/m2) - labor cost (2 workers for 1day for 0.50€/m2 /worker)	500.00 1,000.00
Zeolite addition on soil - labor cost and small tilling machine (1 worker for 4 hours)	250.00
Effectiveness monitoring - Chemical analysis of two soil samples annually for 27 parameters for 10 years - ESP and SAR , 3 analysis during the first 6 months	2,160.00 24.00
Total cost (VAT excluded)	6,842.00
Cost per remediated cubic meter (VAT excluded)**	27.37

Table 7. *Costs for the application of clinoptilolite as soil additive (estimated using a pilot area of 1 acre)*
**one blank soil sample and one collected from the treated area*
*** 1000m2 x 0,25m depth = 250m3 were treated*

2.5 COMPOSTING OLIVE MILL WASTES

Composting is the most commonly used method applied for the recycling and transformation of organic wastes into fertilizers-soil amendments. In the case of OMWs, it is possible to mix directly with manure from seep, cattle, horse, chicken or other suitable Nitrogen sources, as well as with other raw materials such as straw, leaves, prunings, etc.

Composting is a controlled aerobic and thermophilic decomposition, the natural breakdown process of organic residues. Composting transforms raw organic waste materials into biologically stable, humic substances that make excellent soil amendments. Compost is easier to handle than manure and other raw organic materials, stores well and is odor-free, while the high temperatures reached during the process ensure the sanitization of the final product (Wornel and Vesilind, 2012). The currently available compost systems can be generally classified into two broad categories the "windrow" and the "in-vessel" composting systems. Windrow composting is one of the least expensive options but has a much higher turn-over period - 6 months compared to 8 weeks for some of the other technologies (Pisarek, 2012). The main feature of windrow technology is the accumulation *and formation of the organic substrate into piles. Typically, the piles are usually shaped into more or less elongated windrows with specified width and height. Windrow systems are further subdivided on the basis of the aeration method of the substrate into "turned windrow" and "forced air windrow or static pile". In the windrow composting process, the mixture to be composted is stacked in long parallel rows or windrows. The cross section of the windrows is usually trapezoidal or triangular, mainly depending on the characteristics of the equipment used for the agitation or aeration of the piles.*

The turned windrow method is the one that traditionally and conventionally has been associated with composting. The term "turned" applies to the method used for aeration. Aeration of the windrow is achieved by agitation of the substrate using tractors with front-end loaders or any other appropriate machinery, which tears down the piles and reconstructs them. In operations in which the turning is carried out mechanically, the pile configuration that results will obviously be the one imparted by the machine. Ideally, the windrow should be about 1.5–2.0m high (Diaz et al., 2002). In situations in which it is practical to perform the turning manually, the height should be roughly that of the average laborer. The height for mechanical turning depends on the design of the turning equipment — generally, it is between 1.5 and 3.0 m (Diaz et al., 2002). With manual turning, a width of about 2.4-2.7 m is considered to be suitable, whereas the width of the pile with mechanical turning depends upon the design of the mechanical equipment (usually 3.0 to 4.0 m) (Diaz et al., 2002). The windrow length could be up to about 100 m (Haaren, 2009).

Field pilot composting was implemented during the PROSODOL project; composting started in November 2011 by preparing 6 small composts using OM sludge from the evaporation ponds, straw, caw manure, fresh and dry leaves, and different ratios of zeolite dust (0.00-0.80mm). The composition of the composts is presented in Table 8. The temperature is measured weekly and the composts were turned over in order to be well aerated. After the evaluation of the results and the chemical analysis of the composts the most appropriate composition was selected and proposed.

	OOM sludge (kg)	Fresh leaves (kg)	Dry leaves (Kg)	Caw manure (kg)	Straw (kg)	Zeolite (kg)
Compost 1 (5% zeolite)	60	6	6	12	24	5,7
Compost 2 (10% zeolite)	60	6	6	12	24	12
Compost 3 (20% zeolite)	60	6	6	12	24	27
Compost 4 (30% zeolite)	60	6	6	12	24	46
Compost 5 (Blank)	60	6	6	12	24	0
Compost 6	72	18	18	36	30	0

***Table 8.** Composition of the 6 compost prepared at small scale*

From the tested ratios, the compost 2 was selected and proposed for field application. The chemical analysis of the produced compost revealed (Table 9) that the material is rich in inorganic nutrients as well as in organic matter. Moreover, the presence of zeolite ensures the slow release of the nutrients according to plants needs.

Parameter	Value
Electrical Conductivity, mS/cm	1.70
pH	8.20
Organic Matter, g/kg	244
Total Nitrogen, g/kg	14.5
CaO, g/kg	97.9
MgO, g/kg	10.0
K2O, g/kg	6.03
P2O5, g/kg	14.4
C/N	16.7
Ni, Cr, Pb, Cd, Mo, mg/kg	n.d.

Table 9. *Some chemical parameters of the Compost No 2 (ref. Table 7)*

2.5 COMPOSTING OLIVE MILL WASTES

PROSODOL focused on the development and demonstration of low-cost OMW pretreatment techniques with the use of various reactive agents. These reagents were used to remove solids, add alkalinity, remove some of the toxic load and degrade organic contaminants so that the main treatment that follows becomes easier or disposal to land as fertilizer is feasible.

Various materials were used in lab scale in pretreatment experiments to investigate sorption of organic contaminants, increase pH, initiate precipitation of metals in stable forms and/or remove solids from OMW. Most of these materials are low cost, by-products of other processes and are abundant in Mediterranean countries. For the pretreatment of OMW the materials used include magnesite by-products, natural zeolite, limestone, two different types of soils, goat manure (GOM), zero valent iron (ZVI) and activated carbon (AC). GOM and ZVI show promising results in terms of phenol removal and pH increase.

GOM is a commercial biological organic fertilizer with trade name "Viol-Li Natural Organic Fertilizer" mixed with soil. It is characterized by a pH=7 and contains 47% organic matter, $21*10^{12}$ microorganisms/g and traces of Ca, Mg, Fe, Mn, Cu, B, Mo and Zn.

ZVI can be obtained as by-product from various operations involving machining of iron parts. ZVI used was purchased from Gotthart Maier company, Germany. Table 1 shows its chemical analysis and some main properties.

Having performed a series of experiments in laboratory and evaluating the obtained results, the following steps are proposed to be considered in order to remove oil/solids from OMW with the use of low-cost and environment friendly materials:

Oil/fat removal

- *Centrifugation of OMW for five min to concentrate and float oils (3-6% w/w) which are collected and removed from the surface; solids and other impurities precipitate and are also removed (about 15% w/w). It is known that the presence of oil and suspended solids at the surface of OMW when disposed in evaporation ponds hinders biodegradation, since it reduces the effect of the solar energy and prevents oxygen diffusion.*

• Addition of saw dust at the surface of OMW in order to remove remaining oil phases and solids. The optimum amount of saw dust added at the surface of OMW reaches 4.4 g/L and almost one hour is needed for the oil phases to be adsorbed on saw dust. The weight of oil and paste removed after centrifugation is estimated at 10-20 kg per kg of saw dust.

• Addition of coagulants ($AlCl_3$, $Al_2(SO_4)_3.18H_2O$, $FeCl_3$ and $FeSO_4.7H_2O$) in OMW in various concentrations (2 to 20 g/L) to enable removal of solids and/or precipitation of other contaminants. Addition of 7 g/L $FeSO_4.7H_2O$ or $AlCl_3$ was found to be the optimum concentration that results in OMW coagulation after mixing for about 5 min; then centrifugation for 5 min is carried out to precipitate solids. No coagulation is seen when $Al_2(SO_4)_3.18H_2O$ is added in concentrations up to 20 g/L. Addition of $FeCl_3$ in various concentrations up to 10 g/L results in a very dark solution with a pH of 2 and no coagulation is seen.

Phenols and COD removal

After the removal of oil and solids, various tests have been carried out to remove phenols and COD. The most important results are seen in the following sections.

• Addition of $Ca(OH)_2$

After the addition of the optimum amount of coagulants (7g/L $AlCl_3$ or 7g/L $FeSO_4.7H_2O$), $Ca(OH)_2$ could be added in OMW (using concentrations between 5 and 20 g/L) and mixed for 5-10 min to provide alkalinity in the system. After the addition of 7 g/L AlCl3 as coagulant, pH of OMW increases gradually with increased concentration of $Ca(OH)_2$ and reaches almost 6.5 when 10 g/L $Ca(OH)_2$ are added; however phenol and COD removal is limited and does not exceed 7 and 3%, respectively. Liming of OMW after the addition of 7 g/L $FeSO_4.7H_2O$ as coagulant improves substantially the quality of OMW; pH increases gradually and reaches 10.5 (addition of 10 g/L $Ca(OH)_2$) and 12.6 (addition of 20 g/L $Ca(OH)_2$). The optimum phenol and COD removal reaches 65 and 31%, respectively, when 10 g/L $Ca(OH)_2$ are added.

• Addition of ZVI and H_2O_2

Experimental results have shown that when 20 g/L ZVI and 20 or 30 % v/v H2O2 are used then 65% of phenols are removed after 24 hours. Comparable results (60% phenol removal) are also seen when 10 g/L ZVI and 20% v/v H_2O_2 are used. The lowest phenol removal that does not exceed 50% is seen for 10 g/L ZVI and 30% v/v H_2O_2. COD removal is limited in all cases and does not exceed 7%. It is therefore concluded, that addition of 10 g/L ZVI and 20% v/v H_2O_2 in OMW results in phenols and COD removal 60 and 7%, respectively.

• Addition of $Ca(ClO)_2$

$Ca(ClO)_2$ was added in OMW in various concentrations (3-30 g/L), after treatment with 10 g/L lime (experiment 1) or after coagulation with 7 g/L $FeSO_4.7H_2O$ and then treatment with 10 g/L lime (experiment 2). It is observed that the addition of adequate amount of $Ca(ClO)_2$ results in very quick precipitation of suspended solids. When the concentration of $Ca(ClO)_2$ increases from 3 to 30 g/L, phenols removal increases accordingly, from 5 to 98%, after 10 min of mixing; phenols removal from 0.5 to 97% is also seen for experiment-2 treatment, respectively. Thereafter and until the end of the experiment after 120 hours, phenol removal percentages do not practically improve. After 10 min from the addition of the optimum amount of $Ca(ClO)_2$ (30 g/L) the concentration of free chlorine is 3 mg/L while the concentration of combined chlorine which is associated with the formation of chloramines reaches 97 mg/L. However these values gradually decrease with time and after four days drop to 0.6 and 1.1 mg/L, respectively, indicating that chloramines are destroyed. The total nitrogen concentration decreases slightly from 250 to 200 mg/L, before and 10 min after addition of 30 g/L $Ca(ClO)_2$, respectively, and remains at that level also after four days. It is known that excess use of chlorine destroys chloramines oxidizing them completely to N_2 (Boukhoubza et al., 2009). It is also noted that addition of 30 g/L $Ca(ClO)_2$ results in significant decolourization of OMW, due to minimization of phenols concentration, as also reported by Inan et al. (2003). However, the decolourization effect is temporary and after about one hour OMW obtains again a darker colour.

Goat manure as a substrate

Goat manure has been tested as a potential substrate in evaporation ponds for the prevention of OMW leakage by studding different treatment methodologies. Results evaluation revealed that goat manure can be used effectively as substrate in evaporation ponds. When OMW are applied after centrifugation for 5 min, surface addition of saw dust (4.4 g/L) for one hour and again centrifugation for 5 min, contaminants are adsorbed and quality of the leachates infiltrating towards deeper soil horizons and groundwater is improved. It is anticipated that phenols and COD will be removed by 90% and 25%, respectively.

Based on the experimental results, different OMW pretreatment options are proposed as shown in Figure 5.

The most important steps of the proposed methodology include:

- *Centrifugation of OMW for 5 min prior to disposal in ponds apart from removal of oil and solids, accelerates biodegradation of organic contaminants. Oils collected can be centrifuged again for the production of refined oil which can be further used in other applications, such as lubricant, preservative, cosmetic etc.*
- *Addition of saw dust at the surface of OMW (4.4 g/L); after one hour oil phases are adsorbed on saw dust and removed from the surface while mixture of saw dust and paste precipitates and removed after centrifugation for 5 min. These mixtures can be used as compost (probably with co-treatment and stabilization with the heavy fraction of OMW removed in the following steps).*
- *Addition of 7 g/L $FeSO_4.7H_2O$ in OMW, mixing for 5 min to enable coagulation and centrifugation for 5 min to remove solids and/ or precipitate other elements; TS content is further decreased by 15%.*
- *Addition of 10 g/L $Ca(OH)_2$ in OMW and mixing for 10 min provides alkalinity to the system (pH increases from 4.7 to around 8) and also results in significant removal of phenols and COD (35-65% and 15-30%, respectively). Precipitates collected after 5 min centrifugation may be mixed with straw (an excellent lightweight soil improver) and digested to produce compost, which can be used as soil amendment; in this case phytotoxicity tests should be carried out.*
- *Addition of ZVI (around 10 g/L) and 20% v/v H_2O_2 (30% w/w) will result in further phenol removal reaching 60%. Iron chips are cheap by-products of metal finishing operation and can be used, recovered and re-used (4-6 times) until exhaustion.*
- *Addition of 30 g/L $Ca(ClO)_2$ in OMW and mixing for 10 min results in further phenol removal to a cumulative percentage of 98% and in decolourization of OMW.*
- *After centrifugation for 5 min, surface addition of saw dust (2-4 g/L) settling for one hour and finally centrifugation for 5 min, OMW may be disposed of in an evaporation pond with a goat manure substrate of 20 cm width. The liquid produced after filtration will have a pH of around 6.5, while 90% and 25% phenols and COD removal, respectively, is anticipated after 8 days.*

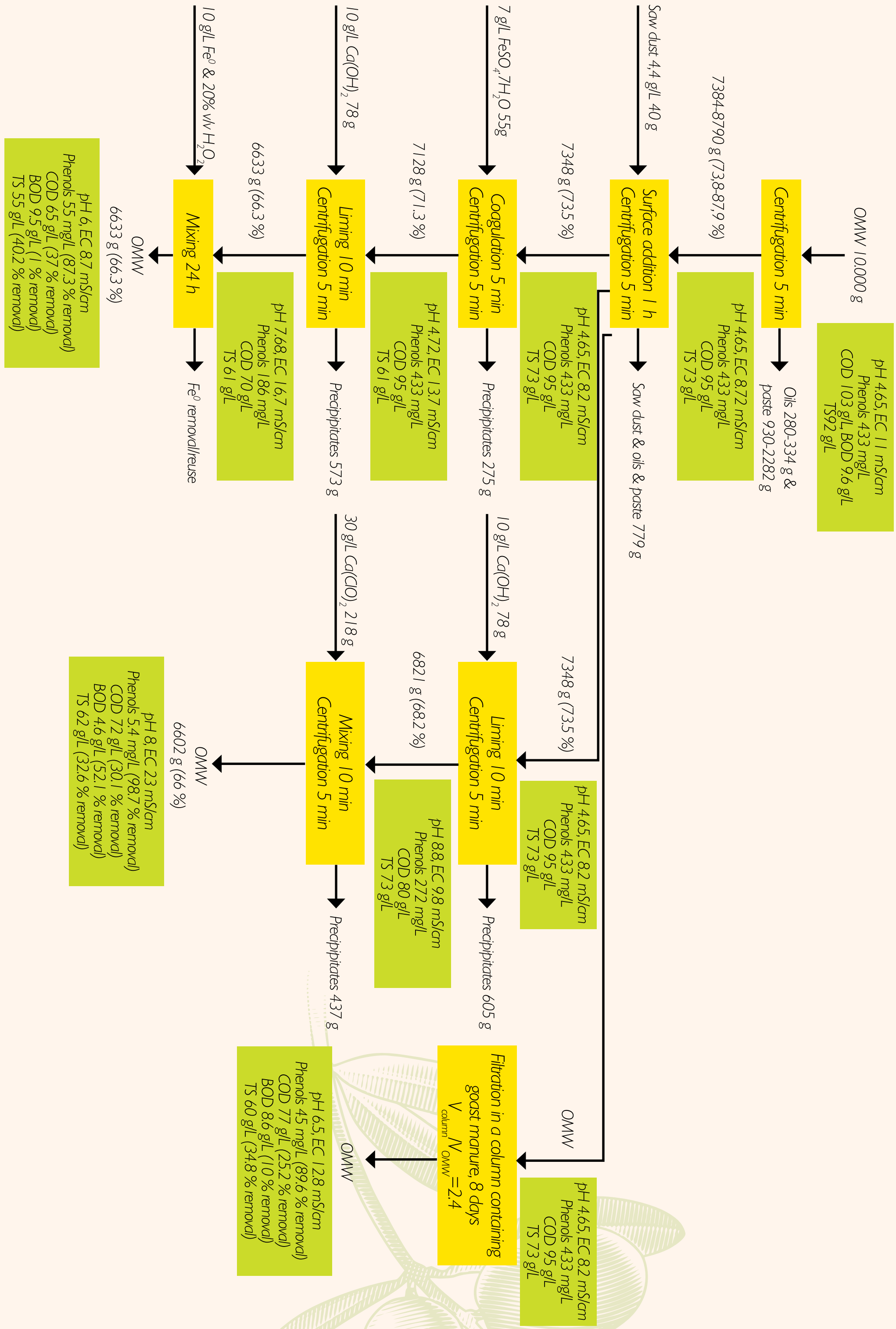


Figure 5. *Flow chart representing different OMW options (note: removal percentages refer to the initial concentrations)*

2.6.1 PILOT APPLICATION OF LOW COST PRE-TREATMENT METHODOLOGY

In the mid of November 2011 all the necessary equipment and accessories for the implementation of pretreatment methodology was transferred to the field. A stainless steel tank of operating capacity of 0.5 m3 was designed, constructed and placed on a metallic base; the tank has two valves on the side and one at the bottom for the collection of liquid pretreated OMW and precipitates, respectively (Photo 18). A stirrer and a motor has been fixed on the top of the tank to mix OMW with various additives (Photo 19). Two pumps are used for the transfer of raw OMW in the tank as well as transfer of pretreated OMW in vessels and then disposal to the bioremediation area.

In detail, the pretreatment methodology in the field includes the following stages to be implemented by taking into consideration that centrifugation which enables fast oil and paste precipitation cannot be applied in field.

***Photo 18.** Tank with operating capacity of 0.5 m3 with two valves on the side and one at the bottom, pump and generator.*

***Stage 1:** OMW were transferred into the pretreatment tank of 0.5 m3 operating capacity and left for almost one day to allow for separation of some oil and paste which are then collected from the surface and removed. A paste (around 10% w/w) precipitates at the bottom and collected at a final stage after decanting or pumping the supernatant solution.*

***Photo 19.** Metallic stirrer placed on the top of the tank*

__Stage 2:__ Saw dust addition (4.4 kg/m3) at the surface of OMW. After almost two hours most oil phases are adsorbed on the saw dust and the mixture of saw dust and oils is removed from the surface. Some precipitates produced (mixture of saw dust and paste) will be collected at a final stage.

__Stage 3:__ Addition of 10 kg/m3 Ca(OH)2 and mixing for 10 min to provide alkalinity to the system (pH is expected to increase from 4.7 to around 8) and partially remove phenols and COD (estimated removal 35-65% and 15-30%, respectively). After about one hour precipitates will be produced and collected. In case phenols are not removed effectively, 10 kg/m3 Ca(OH)2 will be added again once or twice if needed and mixed for 10 min.

__Stage 4:__ Decanting of pretreated OMW and removal of precipitates from the tank.

Stage 5: Transfer of pretreated OMW to vessels of 0.2 m3 capacity with a layer (~20 cm) of goat manure as substrate. Pretreated OMW are filtered and after one week collected.

2.6.2 COSTS GENERATED FROM THE PRE-TREATMENT METHODOLOGY

The pretreatment configuration that is implemented in the field is a low cost process and is expected to result in good quality wastewaters. Table 10 summarizes the cost for construction, operation and implementation of the proposed methodology for the production of 10 m3 of pretreated OMW. A total cost of 2,750 € is estimated; the cost of standard equipment reaches 2,665 € while the cost for the purchase of consumables i.e. saw dust, Ca(OH)2 and goat manure is 85 €.

By taking into consideration that cost for consumables will be substantially reduced when big quantities are purchased, for a typical olive oil mill in Greece that produces around 2,000 m3 of OMW per year the total cost for the pretreatment of OMW is estimated at around 5,000 € for the first year including the purchase of equipment. For the next years a cost of around 2,000 € is foreseen.

Cost category	***Cost in €***
Stainless steel tank of 0.5 m3 operating capacity, base, 3 valves, stirrer, motor and transport/installation cost	1800
Pumps and accessories	250
Generator	315
Various accessories, ie vessels, extension cords, sieves, spoons, nylon layer etc	300
Saw dust, 44 kg/10 m3	10
Ca(OH)2, 100 kg/10 m3	50
Goat manure, 100 kg	25
Total	2750

Table 10. *Indicative cost analysis of the proposed configuration for the production of 10m3 of pretreated OMW*

2.7 A TECHNIQUE FOR COD RAPID MEASUREMENT IN OMWW SAMPLES

In order to characterize OMW the determination of the following parameters is generally required:

- *COD*
- *BOD5 at 20 °C*
- *Total Oily Matters*
- *Tensioactive materials*

All these methods takes averagely long execution times due the particular matrix which is analyzed (OMWW particularly rich in fatty compounds) and the techniques foreseen by the applied method (e.g. BOD5 at 20 °C).

That is the reason why alternative methods have been taken in consideration during PROSODOL which could permit the characterization of the mill waste according to its main components in a more simple and short way. For this, samples were analyzed in a parallel way following traditional methods and verifying innovative approaches.

Accordingly to the official methods it came out that the presence of tensioactive materials has always been very low (in the vicinity of few mg/l). The quantity of tensioactive materials, unlike COD and BOD, is not a good quality indicator of the waste. For such reason it was not possible to identify a correlation between traditional and alternative methods.

Relevant to the other parameters, and specifically about COD (which is a very good global quality indicator), it was possible to find a significant variability among the different parameters analyzed. Therefore, trials were carried out in order to define a technique that could give a simple and rapid quantification of this parameter.

After many experimentations, a methodology was found which correlates COD of the wastes and the measure of their residue at 105 °C.

The technique foresees the following steps:

10 grams waste sample is placed in an aluminium capsule;

the sample is weighed through a thermal balance with halogen heating (OHAUS brand, model MB25);

the sample is heated at a constant temperature of 105 °C till it has reached a constant weight that is electronically verified.

Measurements are inserted in an Excel file and the COD value is calculated by using the appropriate mathematical formula (Fig. 6).

The technique is low time demanding and it can be easily implemented even in the mills itselves. Handling of balance is quite simple since in order to set dry parameters it is enough to push temperature and time buttons, then put in the sample and start measurement. The instrument occupies few space and it is quite cheap.

Results found permit to obtain a mathematical correlation which could express a pollution index of the analyzed waste. Polluting load can be directly measured and monitored during the extraction process of olive oil and countermeasures can be undertaken on-site in order to modify COD parameter according to actual thresholds established by the law.

Determination of COD value in olive mill waste waters
Spread sheet for an easy rough evaluation thant can be used in olive mill waste waters

RECOMMENDED FACILITIES
Thermobalance e.g. Sartorius Mod. MA35 or Ohaus Mod. MB25
alluminium capsules to be used with the correspondent thermobalance
a sample of olive mill waste waters

Phases	
1	Calculate the tare of an alluminium capsule using the thermobalance (each balance has its dedicated capsule)
2	Weight 10 grams net of olive mill waste water pouring it in the alluminium capsule
3	Close the thermobalance cover
4	Set the temperature at 105 °C
5	The thermobalance stops automatically the measurement when the weight of the sample does not change anymore
6	Be sure to read the value indicated in the display as "residue in solid %" after having set up the device accordingly
7	Enter the read value (%) in the yellow box below
8	An indicative COD value is obtained that has an average variability of ±10%

Value expressed in %	COD
2	16937,9
	Note: the value has an average variability of ±10%

Figure 6. *The excel file for the estimation of COD values.*

2.8 USE OF OMW FOR OLIVE TREES IRRIGATION AND FERTILIZATION

One of the main objectives of the PROSODOL project was to investigate if the use of OMWs for olive trees irrigation/fertilization impacts soil quality and if contributes to yield increase. For this, field trials were performed in Albenga, Italy, during which controlled distribution of OMWW and husk took place in a pilot olive orchard.
A pilot scale experimentation site of around 1.500 m2 for the controlled use of OMWW for tree land fertilization was set up (Photos 20-21). The layout of the pilot area is shown in Scheme 3. Around 200 two-years-old olive tree plants belonging to 3 different varieties (Taggiasca, Pignola, Leccino) were transplanted and two different kinds of OMWW (Terre Barone and Maddei) were distributed.

***Photos 20-21.** Overview of the pilot area in Albenga, Italy.*

OOMW SOURCE	BUFFER ZONE	TERRE BARONE	TERRE BARONE	CONTROL	MAFFEI	MAFFEI
TREATMENT NUMBER	1	2	3	4	5	6
SURFACE	260 m2	270 m2	300 m2	360 m2	280 m2	340 m2
PRESENCE OF WELL		X	X	X	X	X

***Scheme 3.** Layout of the pilot area*

A drainage system was set up before plant transplanting, digging trenches of about 1m depth and then positioning 10cm diameter drainage tubes covered by a polyethylene tissue (Photos 22-23). Tubes of 10 meters were placed so to be connected at two opposite sites of the wells (5 totally in correspondence of plots 2,3,4,5,6 - refer to Scheme 3) (Photos 24-25), which have been used to collect leachate for subsequent analysis.
In order to carry out the distribution of OMWW in the pilot area an hanging (50 cm above ground surface) dripline system was set up using Netafim Uniram pressure compensated tubes (normally used in the agricultural sector for the distribution of water or the application of liquid fumigants).
Distribution of OMWs (OMWW and husks) was carried out in 2010 and 2011 according to what is indicated in Table 11. Olive husks were distributed only in 2011. The control treatment is represented by water. Taking into account that amounts of 400-800 m3/ha of wastes distributed on clay soils to adult olive plants did not cause particular phytotoxic effects, amount distributed in the pilot area were chosen in consideration of the following: 1. the soil texture, 2. the young age of plants, 3. the purpose to avoid severe interference with plant correct growth and/or heavy phytotoxic effect which could have significantly compromised

the trials foreseen.
Before distribution, an extensive survey of OMWW was carried out during olive season 2009-2010 at the 2 different olive mills: 1) Terre del Barone located in Borghetto S.S. municipality (Savona province, Liguria Region, Italy) which uses a three phases extraction method; 2) Maffei located in Orco Feglino (Savona province, Liguria Region, Italy) which uses a traditional discontinuous extraction method based on millstone and pressing columns.

Photos 22-23. *Digging of the draining system*

Photos 24-25. *Draining tubes and prefabricated wells.*

Treatment	Source (olive mill)	Oil extraction technique	OMWW or olive husks distributed (l or Kg/treatment)	Treatments (num.)	Corresponding volume or mass m3/ha/year or t/ha (rounded)
1	Terre Barone	3 phases (continuous)	1700 (olive husk) only in 2011	1	100 t/ha^
2	Terre Barone	3 phases (continuous)	500	3	80m3/ha/year *
3	Terre Barone	3 phases (continuous)	800	3	120m3/ha/year **
4	Control	-	1000 (water)	3	81,25m3/ha/year
5	Maffei	Traditional (discontinuous)	300	3	50m3/ha/year *
6	Maffei	Traditional (discontinuous)	600	3	75m3/ha/year **

^ no specific limits foreseen by the law
** established by the Italian law*
*** correspondent to the threshold established by the law + 50%*

2.8.1 RESULTS FROM THE USE OF OMWS FOR OLIVE TREES IRRIGATION/FERTILIZATION

The obtained results indicated that the use of OMWs for olive trees irrigation or fertilization is a significant option, if implemented under strict rules. It has to be kept in mind that the recorded values of soil and leachates properties correspond to OMWs distribution which was maximum 1,5 times the amount foreseen by the Italian law.

Effect on soil properties

No acidification of soil at both depths (20 and 50 cm) was recorded after OMWW distribution. The COD and BOD values were constant at 20 cm depth and they tended to decrease at 50 cm depth. The total oily matter remained constant, while exchangeable K, Mg and Ca were all increased during time especially with regard to the starting supply of the soil. Sulphates values tended to be high in all samplings carried out but a general decrease was recorded during time with the exception of the plot treated with high dose of OMWW deriving from the discontinuous extraction technique. Ammonium values were remained almost constant during time at both depths sampled, while nitrates were decreased significantly during time at both depths sampled and in the control plot too. Phosphates content was negligible in the soil sampled from the control plot, while it was measured at constant values in the samples collected at 50 cm depth and at increasing values during time in samples collected at 20 cm depth. High values of available P were measured at both depths sampled. Tensioactives were not detected in almost all cases. Finally, no particular effects were observed in the plot were olive husks were distributed with regards to samples collected in 2011.

Effect on leachates collected in the wells

The availability of leachates is subjected mainly to rainfall volumes, which determine the fill in of the wells. Based on the analysis carried out the data collected the following conclusions were obtained: pH, polyphenols, P, PO43-, K, BOD5 and NH4+ are all parameters characterized by a low variability (at least during the sampling period). Nitrates were strongly affected by the presence of a more superficial groundwater in plots/treatments 5 and 6 (similarly to what happened for soil samples). From a general point of view their concentration tends to decrease during time after OMWW soil spread. The electrical conductivity, as compared to the control plot where water was distributed, it was not significantly affected by OMWW distribution. The concentration of sulphates was high in the control well varying around 195 mg/l averagely. A quite linear increase (from 150 till 380 mg/l) was observed in the wells correspondent to plots treated with OMWW derived from continuous extraction process. More variable concentrations (around 270 mg/l averagely) were observed relevant to plots treated with OMWW derived from discontinuous extraction process. Too variable values were observed for COD; therefore it is not possible to draw clear conclusions.

Effect on trees growth

Olive plants were monitored regarding measures of height and stem diameters. No negative effect in trees growth and development was recorded, except the case of the plants growing at a plot treated with the higher dosage of OMWW coming from a discontinuous extraction system. The conclusion obtained from the comparison between waste-irrigated plants and water-irrigated plants was that the waste irrigated plants were in most plot better developed, confirming the results of many other research studies which reported that the control use of OMWW for olive trees irrigation is beneficial and contributes to better plants growth.

2.8.2 IRRIGATION SYSTEMS AND CRITICAL ASPECTS

It is clear that irrigation means extra costs in terms of design, set up and management of the system in comparison to an olive orchard without any irrigation system. It is also crucial to evaluate costs due to the exploitation of water in areas where it is often lacking.

Irrigation system design: structure and sizing

Different watering systems can be set up:

1. *Drip line hanging on the soil surface*
2. *Dripper plugged into the watering tube*
3. *Sprinklers for micro-aspersion (e.g. on centuries-old plants)*
4. *Buried drip line at 20-30 cm of depth (subirrigation with anti-siphon technology)*

When installing an irrigation system, the following critical aspects should be taken into consideration:

Filtration system: each installation should be provided with a suitable filtration system in order to retain organic and inorganic particles for an overall correct working of the entire system. Such an aspect is even more crucial when OMWW are distributed because of their lipidic nature and of the presence of suspended particles

Maintenance of the irrigation system: in the case of hanging driplines, driplines laying on the soil and sprinklers, troubles and obstructions are easily detectable. With regards to sub-irrigation, besides a precautionary maintenance, it is necessary to verify the correct operation and the flow of the system through water counters.

Economical aspects related to watering systems

Choice regarding the different possible technical solutions to be installed in an olive orchard depends on the investment the user is ready to undertake. Costs related to the materials depend on many factors (technical, technological and agronomic) - e.g. spacing between rows affects the quantity of dripline used, the quality of the water affects the filtration system needed - so that it is not easy to define general cost assessment rules. Indicatively dripline is cheaper then sprinklers and both of them are cheaper than a sub-irrigation system characterized by high laying costs. Thus, it is clear that there are many choices between the irrigation system that will be adopted and for this reason the final investment cost is highly site specific. PROSODOL considered the irrigation system installed at the pilot area, and the relative installation and operating costs were compared to the costs generated by a water-irrigated orchard for the fertilizing of which, synthetic fertilizers are used. Table 12 includes the materials used or proposed for the set up of a complete irrigation system and relative costs.

The irrigation system installed at the pilot area had the following characteristics:

A hanging dripline system at 60 cm above soil surface per each olive row (Photos 26-27) was installed. Uniram (Netafim, Israel) pressure compensated tubes normally used in the agricultural sector for the distribution of water or the application of liquid fumigants were adopted.

Such driplines have the following properties:
- flow: 2 l/h
- distance between drips: 30 cm
- pressure: 1,5 bar

Each plant is provided with 2 drips aligned with plant row roughly at 15 cm apart from olive stem. Driplines are fastened to iron wires (Photo 28).

Two orders of iron wires were tightened between concrete poles, one to give support to dripline, the other one to give support to plants. Distance between poles is 10 m. Each plot (treatment) is provided with a buried plastic tube, which feeds the driplines, and it is connected to the pump for OMWW distribution (Photos 29, 30).

Photos 26-27. Uniram dripline used for OMWW distribution

Photo 28. A detail of the hanging pipeline used for OMWW distribution

Photos 29-30. Valve and connection pipes connected to distribution tubes and driplines.

Filtration

Filtration represents the key aspect during OMWW distribution through dripline in consideration of the composition of such waste. Ease of distribution can vary depending on the thickness and the presence of suspended solids in OMWW. Such parameters may present a very high variability during the olive season related to the characteristics of olive milled. Filtration was realized with simple net filter (1 inch), while in Table 10 advices are given about other possible filters that can be used. Notwithstanding a significant reduction in workability is related to the set up of a filtration system between the feeding tank and the dripline.

It is therefore highly advisable to adopt a filtration system before OMWW collection in the plastic container in order to have OMWW feeding the dripline with the lowest content of suspended solids. Such filtration system should be composed by superimposed filtering elements made in steel which are fed from the top part and which give as output in the lower part cleaned up OMWW. Sediments and solids which accumulate within the filter can be then disposed on the soil together e.g. well mixed together with olive husks in order not to create too concentrated hot spots that could be harmful when kept in contact with the soil for a prolonged time.

In case of OMWW characterized by a significant presence of suspended solids, in order to improve workability it is also possible to think about a different distribution system based on dripline provided with "sip" drippers. In this case dripline is very similar to the one used in the pilot area but water/OMWW dispenser are characterized by higher flows and a distribution mechanism that does not foreseen the presence of an ad-hoc winding water path like it happens within pressure compensated dripline.

2.9 ANALYSIS OF NATIONAL AND EUROPEAN LEGISLATIVE FRAMEWORKS FOR OMW AND SOIL PROTECTION-PROPOSALS TO POLICY MAKERS

An extensive analysis of European Union legislative framework on the subject of olive oil waste management was performed during PROSODOL project, including the relevant regulations of waste, water and soil.The analysis integrated the relevant legislative framework of the partner countries, i.e. Italy, Spain and Greece as well as of Portugal and Cyprus, as well.

In specific, the study includes (a) an analysis of the olive oil industry and the relevant environmental issues; (b) waste management and the relevant EU and national legislation on waste, water and soil; (c) legislative recommendations for olive oil waste management, both statutory and volunteer; (d) legislative recommendations as well as technical specifications and proposed strategies to monitor, protect and improve soil quality at olive oil mills' disposal areas.

*Following this analysis, PROSODOL concluded on the following **statutory** legislative proposals:*

- *Untreated waste/wastewater disposal into the environment should be strictly banned*
- *Irrespectively if is dangerous or not, the waste/wastewater should be treated before any disposal to land/surface waters and specific emission limit values should be defined, especially in the case of land spreading where no statutory standards exist but only application rates in some national legislation.*
- *As olive oil waste is potentially hazardous the legislation should provide statutory limits, especially on phenols, under which the waste is characterized as non-hazardous. The focus should be the categorization as H14 (ecotoxic) and limits as well as tests and monitoring measures should be provided depending on the receiving media, i.e. soil and surface waters.*
- *The legislative act should clearly specify that the waste should be analysed for its physicochemical characteristics, as for example: vegetative trials, germination tests, phytotoxicity tests, growing-on test, etc, testing its toxicity potential regarding plant development and the environment in general. Standard sampling and analytical*

Item	*Specific*	*Cost*	*m.u.*
Dripline	Pressure compensated Flow: 1,5 – 2 l/h	0,3 – 0,6	€/m
Concrete poles		2,50 – 3,50	€/each
Iron wires		1,50 – 2,00	€/m
Wire tightener		1,50 – 2,00	€/each
Solenoid valve		65,00 – 70,00	€/each
Tubes	Polyethylene ø 16 mm (between feeding tube and dripline)	0,42	€/m
	Trichoflex ø 40 mm Trichoflex ø 50 mm (to connect buried PE tubes among plots)	1,20 1,80	€/m €/m
	PE tubes ø 40 mm PE tubes ø 50 mm (buried)	1,90 2,80	€/m €/m
Pump	750 W	150-250	€
Joints	PE valves	2,00	€/each
	PE L tube	0,20	€/each
	Cork ø 40 mm Cork ø 50 mm	4,80 7,00	€/each €/each
	Sleeve coupling	0,16	€/each
	Spheric valve 1" ¼ Spheric valve 1" 1/2	26,00 38,00	€/each €/each
	Fast coupling 1" ¼ Fast coupling 1" 1/2	5,00 6,00	€/each €/each
	PE joint 1" ¼ PE joint 1" 1/2	5,30 6,80	€/each €/each
	Clamp joint ø 40 mm Clamp joint ø 50 mm	2,20 2,30	€/each €/each
	Tap (between clamp joint and dripline)	1,50 – 1,80	€/each
Filters	Net – 1" Net - 1,5"	30,00 40,00	€/each €/each
	Bag filter (steel) Filtering bags	1.635,00 12,00	€/each €/each
	Disc filter (Azud type) ø 1 inch	20,00 – 50,00	€/each
Storage tank	Plastic container 1.000L	500,00	€/each

Table 12. *Overview of the materials used or proposed for the set up of a complete irrigation system and relative costs.*

procedures, harmonized at EU level, could be introduced.

- *There should be a categorization of production industries according to their production capacity and/or waste generation in order to draw specific measures for waste management, i.e. waste/wastewater management facilities within the industry or establishment of collective schemes for smaller units*
- *In case evaporation ponds are used, the minimum requirement should be the use of protective layers (engineered evaporation ponds).*
- *As landspreading is a common and low-cost practice, especially for small production unites, specific regulations should be developed.*
- *In case of landspreading and under the condition that the olive oil waste/wastewater fulfills the requirements of the existing legislation, the OMW could be considered fertilizer and thus, annual dose estimation should follow the general rules of soil fertilization considering soil properties and purpose of use.*
- *On the reuse of treated wastewater for irrigation of agricultural lands, application guidelines should be developed in order to provide a common level of environmental and public health protection*
- *If olive oil waste (OOW) is considered as waste national law should allow it to be treated as municipal waste when produced by smaller olive mills*
- *The EC Commission should provide technical specifications, pursuant to art. 5 of Directive 2008/98, on the conditions for using olive oil waste (OOW) as a by-product regardless of their economic value and regardless of the possible need of a drying phase and/or not removal*
- *National laws should be brought in line with this new concept of by-product namely the part that still provides for the economic value of by-products as a requirement (as in the case of the Italian law)*
- *The regulations should take onto account (a) the use of the land (e.g., agriculture, food products, non-food products, residential/parkland, commercial and industrial), (b) the soil type and (c) the period of reuse.*
- *OMWs are usually discharged in small stream catchments (<10 km2), which are not considered in the Water Framework Directive 2000/60/EC. Therefore, there is a need for including small streams into monitoring and assessment schemes as small streams contribute to the pollution load of the river basin.*
- *Environmental Quality Standards (EQSs) should be set in a EU Directive in the same way that is done for water bodies, at least as minimum requirements per soil type. The threshold for pollutants (as phenols) concentrations in soil could be set in such values as to reflect existing soil maximum background concentrations in natural undisturbed soils.*
- *Emission Limit Values (ELVs) should be provided in national legislation as in the case of Italy and Spain but as the local conditions should be taken into account regional regulations should be also adopted as in the case of Greece*
- *More favorable national laws should be introduced for obtaining permits for facilities producing energy from biomass, especially when they are small*

As regards the volunteer legislation proposals, these could be:

- *Support of technology change to 2-phase process for minimization of waste/wastewater. When utilizing the 2-phase system the fresh water consumption is reduced and also the wastewater streams are eliminated*
- *It would be recommendable to introduce laws that expressly facilitate initiatives for municipalities to build installations in the scope of their local public services, also based on regional agreements with olive mills and with other parties that would significantly contribute to providing biomass for energy production and other uses*
- *National law should expressly provide that, in the absence of adequate private initiative, municipalities are able to build such facilities and operate them within the scope of their local public services*
- *The Integrated Pollution Prevention and Control, Reference Document on Best Available Techniques in the, Food, Drink and Milk Industries, Chapter on Olive Oil industry should*

be amended including the recent advances on waste management in the sector. In the same way, National BREFs on olive oil production should be prepared in the interested countries, covering all industrial units (IPPC and non-IPPC)

- *Promotion of the establishment of collective/ cetralised treatment systems.*

2.10 INTEGRATED STRATEGY OF ACTIONS, MEASURES AND MEANS TO PROTECT SOIL QUALITY, SUITABLE FOR MEDITERRANEAN COUNTRIES

For the promotion of soil protective and remedial actions at OMW disposal areas, PROSODOL proposes a set of recommendations to be included in the national/European legislative frameworks. The recommendation are those derived after evaluation of the project's outcomes and mainly from the soil monitoring actions performed at olive mills waste disposal areas, and their fulfillment is considered necessary for soil quality protection. It is believed that their incorporation as Member States obligations in the legislative framework of the EC or/and of the Med Member States will ensure future effective monitoring of the legal and illegal disposal areas, which in turn will facilitate the sustainable management of these areas.

Moreover, PROSODOL proposes a set of technical standards which could be utilized either as Best Available Techniques for Soil Monitoring and Soil Quality Improvement or as Annexes in future Directives and legislative acts, which will assist national local/regional/governmental authorities to implement strategies to monitor, protect and improve soil quality at olive oil mills' waste disposal areas.

2.10.1 STATUTORY LEGISLATION PROPOSALS FOR SOIL PROTECTION

In all Mediterranean European countries, regardless if specific laws are existed or not, the uncontrolled disposal of olive oil mills' wastes is not permitted. Thus, prior soil disposal, the mills waste should be pre-treated according to guidelines described in the national legislative framework. If there is no legislative framework, then a minimum required measure could be the treatment of wastes with lime in order to increase pH and reduce the organic load and the total solids.

The following six measures are proposed to be included in the European legislative framework as well as in the national frameworks of Mediterranean olive oil productive Member States:
(1) Recording Olive Oil Mills Waste disposal areas
(2) Characterization of disposal areas-Risk assessment
(3) Evaluation of risk level
(4) Defining the conditions of OMW soil disposal
(5) Adoption of soil quality indicators
(6) Monitoring soil indicators-Evaluation of the results

These measures are considered as being efficient for maintaining soil quality and sustainability.

(1) Recording Olive Oil Mills Waste disposal areas
Each country should identify the OMW disposal areas in its territory and record them in a national inventory. The inventory will contain all licensed disposal areas and as many as possible non-licensed ones. Local inventories should be created as a first step under the responsibility of local or regional authorities, which afterwards will be integrated into a national inventory under the responsibility of governmental agencies. GIS mapping of the disposal areas and the establishment of a digital database is strongly recommended.

(2) Characterization of disposal areas-Risk Assessment
As a second step, governmental and local authorities should proceed to complete and

detailed characterization of the disposal areas and to the performance of risk assessment studies.

Recorded OMW disposal areas should be characterized considering location, hydrogeology, physiography, geomorphology, land use, soil structure, texture, water permeability, coefficient of hydraulic conductivity (saturated or unsaturated), porosity, presence and depth of impermeable soil layers. Additionally, the collected data may include, history of the site, extent and types of contaminants that may exist, hydrogeological and hydrological regime for the broader area, known/anticipated presence and behavior of receptors, sampling of soil and groundwater: comparison with generic guideline values or quality standards, sampling of soil and groundwater: site-specific modeling of fate, transport and exposure and comparison with toxicological values, and other parameters which may be considered necessary for the complete characterization of the area. Such a characterization will permit the performance of the risk assessment study of the area and the identification of the sites, which pose risk to human health and to the environment.

Indicatively, a risk assessment study could comprise:

1. *Preliminary investigation (desk study, site reconnaissance and sometimes limited exploratory investigation). The goal of this preliminary stage is to assess whether potentially contaminating activities have taken place on the site, whether soil and/or water pollution is suspected, and in some cases to confirm the existence of pollution. In short, this phase focuses on hazard identification.*
2. *Detailed investigation. The aims at the main site investigation stage are (a) to define the extent and degree of contamination, (b) to assess the risks associated with identified hazards and receptors and (c) to determine the need for remediation in order to reduce or eliminate the risks to polluted or actual receptors.*
3. *Supplementary or feasibility investigations to better define the need for and type of remedial action or monitoring. The aim may be to assess the feasibility of various remediation techniques; this may include more detailed physical and chemical characterization of soils and laboratory studies on soil or groundwater treatability. Supplementary investigations may also be designed to improve understanding of the nature, extent and behavior of contaminants.*

The risk assessment, however, should not be limited to toxic constituents, like the polyphenols, which may pose threat to human and animal health but to consider also the potential progressive soil degradation due to the presence in OMW of other less hazardous or non-hazardous constituents, like nutrients and other inorganic waste's constituents. This factor is often underestimated and the majority of risk assessment studies focus on the toxicity, which may be caused to soil and to humans from polyphenols. Thus, if land distribution is planned the organic load and the toxic substances (polyphenols) should not be the only issues of concern. Specific care should be taken also for inorganic constituents (e.g. K, Cl^-, NO_3^-, SO_4^{2-}, P, Mg, Fe, Zn and others), since the very high concentrations disposed on soil change drastically its quality properties, while their concentrations in soil as well as, the soil electrical conductivity remain high even many years after the last disposal. For this, the performance of a complete soil physicochemical analysis and identification of the organic and the inorganic soil constituents are strongly recommended. Determination of phytotoxicity potential is also recommended.

The risk for each potential pathway is considered to be a combination of the probability that a hazard will reach the target (e.g. high polyphenols concentration in soil due to OMW disposal) and the magnitude of harm if the target is exposed to the hazard (e.g. phytotoxicity). The probability that a contaminant will reach a target in sufficient concentration to cause harm may be assessed qualitatively according to the scale: ***high*** *(certain or near certain to occur),* ***medium*** *(reasonably likely to occur),* ***low*** *(seldom likely to occur) or* ***negligible*** *(never likely to occur). The magnitude of harm is assessed as:* ***severe*** *(human fatality or irreparable damage to the ecosystem),* ***moderate*** *(e.g. human illness or injury, negative effects on ecosystem function),* ***mild*** *(minor human illness or* *injury, minor changes to ecosystem) or **negligible** (nuisance rather than harm to humans and the ecosystem). The qualitative level of risk associated with each pollutant pathway is then assigned by the combination of the aforementioned probability with the magnitude of harm. Thus, having identified all the crucial parameters the risk should be rated according to 13 (Modis et al., 2008).*

	Magnitude			
Probability	Severe	Moderate	Mild	Negligible
High	High	High	Medium/Low	Near zero
Medium	High	Medium	Low	Near zero
Low	High/Medium	Medium/Low	Low	Near zero
Negligible	High/Medium/Low	Medium/Low	Low	Near zero

Table 3. *Risk assessment rating*

(3) Evaluation of risk level

The third step is to evaluate the level of risk of the suspicious areas and exclude for further future disposal of all areas under high risk. For these areas a remediation plan should be developed and implemented immediately. For areas under medium risk, further assessment of the threat type and potential extent is strongly recommended in order to decide the conditions of waste disposal or the design and implementation of remediation actions. For these cases, decisions should be taken considering data collected during the risk assessment study is proposed. For areas under low or near zero risk, a management plan for the safe disposal of OMW should be developed and implemented under the supervision of local authorities and the responsible governmental agencies.

(4) Defining the conditions of OMW soil disposal

It is very likely, some areas, although being of low or negligible pollution/degradation risk, to be inappropriate to accept OMW soil disposal due to their specific characteristics. In order to ensure safe disposal of OMW, soil and land data have to be considered in combination with bioclimatic conditions and management practices. The ultimate goal should be to apply or dispose OMW to land in such a way, that the soil either filters the potential toxic elements effectively, or electrochemically absorbs them or decomposes them in order that a clean solution passes through the soil body. The soil should not be overloaded with inorganic constituents and must maintain all its functions and its absorption capacity to ensure a sustainable system.

The decision of land distribution is proposed to be taken considering appropriate suitability criteria, as presented in Paragraph 2.3.3 Considering the specific properties of soils at disposal areas, local particularities and the limitations of Table 2 the following steps should be followed in order to adopt and implement safe disposal or application of OMW.

Step 1: Definition of suitable or unsuitable soils for OMW disposal

Soils with the potential to receive or soils that should be excluded from OMW disposal/distribution/application are identified based on permanent physical and/or chemical characteristics (Table 2). Moreover, prior the final decision and in complementarity with the parameters of Table 2, the presence of toxic soil conditions should be assessed by using the standard methods for the determination of (a) nitrogen mineralization and nitrification in soils and the influence of chemicals on these processes (ISO 14238:1997); (b) the effects on earthworms (ISO 11268-1:1993); (c) the chronic toxicity in higher plants (ISO 22030:2005); and (d) soil biomass or soil respiration (ISO 14240-1:1997).

The selection among these standard methods should be based on several factors, such as current soil quality condition, past, present and future use of the area, amounts of produced waste and treatment level. Olive mills' wastes should also be analyzed in terms of BOD_5, COD_5, pH, total solids, total suspended solids, total volatile solids, ash, total organic carbon, total nitrogen, total phosphorous, electrical conductivity, total sugars, fats and oils, total phenols, potassium, sodium, calcium, magnesium, total sulfur, total chlorine, iron, manganese, zinc, copper, nickel, chromium and molybdenum.

Step 2: Estimation of the maximum permitted OMW amount

The soils that are suitable for OMW application should be further studied in order to define the maximum permitted amount (or the maximum amount they can afford) of OMW based on their physicochemical properties and on OMW composition and considering legally applied thresholds for these properties (see paragraph 2.3.3).

Step 3: Estimation of annual permitted application of OMW

The annual distribution rate and timing of wastes application should be determined, regardless if wastewater or solid OMW (i.e. dry husk, wet husk, composts from all mills by-products), or wastes originated from 2-phase systems are to be distributed (see paragraph 2.3.3).

Step 4: Time of OMW application

In case of olive mills' wastewater or solid waste use for land spreading, the time of application has to be defined considering the annual rainfall rate, intensity and distribution throughout the year and the temperature, in relation to water balance, soil properties and processes, microbial activity and OMW decomposition. The background philosophy is to apply OMW at periods where rainfall induced leaching of the soil water is not expected.

Step 5: Soil Monitoring

The next step is the periodical monitoring of the impact of OMW application on soil, on water bodies and the environment under the specific bioclimatic conditions of the Mediterranean areas through a systematically planned sampling scheme combined with different eco-biotoxical test (see paragraph 2.3.3).

2.10.2. TECHNICAL RECOMMENDATIONS AND GUIDELINES FOR SOIL QUALITY PROTECTION

Apart from the aforementioned, statutory measures, which ensure sustainable management of OMW disposal areas, there are also other measures that can be optionally adopted in order to facilitate local and regional authorities to perform continuous control as well as, to select and apply the appropriate soil remedial technique, in case that remediation is required. These recommendations can be utilized either as Best Available Techniques for Soil Protection or as Annexes in future Directives and legislative acts.

The proposed optional measures are of two kinds:
(1) Measures for continuous monitoring of OMW disposal areas, and
(2) Soil remedial technologies, appropriate for OMW disposal areas.
The adoption of these measures in combination with the statutory ones ensures the integrated control of the OMW disposal areas, but mainly the future protection and improvement of soil quality. As such, they are considered to be fully harmonized with the EC requirements for soil quality protection as these are described in the Thematic Strategy for Soil Protection.

(1) Measures for continuous monitoring of OOMW disposal areas

Three measures are proposed; the two of them could be adopted by local and regional authorities *since their development and application require the contribution of qualified personnel and scientists. The third one is suitable for use by individuals (e.g. disposal areas' owners) but also by local authorities, as it is simpler and does not require specific knowledge and qualifications.*

Suitability of OMW disposal-Soil maps

To facilitate the implementation of the proposed measure No 4 (i.e. defining the conditions of OOMW soil disposal), the introduction of a Geographical Information System is proposed as necessary to define the application of OMW to agricultural or other type of lands because of the importance of spatial accuracy in the application. Such an illustration will further facilitate decision making while at the same time a very useful GIS maps database will be created. For the GIS maps creation, it is necessary to include information on land, soil and OMW properties, processes and composition; climate variability; land use and management; and possible environmental risks. The land suitability system for spatially manipulating soil and land data that is proposed, has been designed and developed in Soil Science Institute of Athens (SSIA)-ELGO-DEMETER for other Greek areas in the past and it was adapted to the peculiarities of OMW disposal (see details and description in paragraph 2.3.3).

Monitoring soil quality-Development of maps of soil constituents distribution vs time and depth

To facilitate the implementation of the proposed statutory measure No 6 (i.e. monitoring soil indicators-evaluation of the results), the development and use of maps of soil constituents' distributions vs. time and depth is proposed. Through this tool, local and regional authorities will have the opportunity to screen disposal areas rapidly, identify potential risky conditions, carry out systematic monitoring of the areas of interest and facilitate decision making on the appropriate measures to be taken at field or municipal scale. The proposed tool integrates the continuous monitoring of the OMW disposal areas into the regular activities of local/regional authorities and thus, allows the proper and continuous monitoring of such areas (see paragraph 2.3.5).
However, this indeed requires the cooperation of the owners of the disposal areas, since repeated soil samplings at various sites are necessary for maps creation and update. The proposed application tool uses interpolation surfaces that indicate the distribution of the different physical and chemical parameters in the area of interest, so the user can rapidly obtain an idea of the possible diffusion of the chemical parameters and the degree of risk in the vicinity of the waste disposal areas. This, potentially, allows also the establishment of an Operational Centre, which could be located, for instance, in cooperation with the Environmental Protection Office of the Local Government (District) in the premises of a Municipality, and can undertake the continuous monitoring of areas under risk and the scientific and consulting supporting of the owners.
The design of the particular software package needs to monitor a number of private fields that are spread around and make queries based on various spatial and chemical attributes. Thus, it is proposed that, for each OMW disposal area, one initial mapping should be carried out by performing soil sampling from various sites and for at least 4 times (e.g. every two months). The sampling sites will be decided according to the generally accepted soil sampling rules and a qualified person should be present and undertake the overall control. The collected soil samples should be analyzed for the parameters, proposed as suitable indicators for OMW disposal areas (paragraph 2.3.1). The maps that will be created should be used for no more than 5-8 years. After this period, the maps should be updated by repeating the sampling procedure. In the meantime, soil quality could be assessed by using the tool of the paragraph 2.3.5, i.e. the simpler one that does not require specific skills and can be carried out by the areas' owners annually.

Monitoring soil quality- Software for soil monitoring by land users and polluters

A simpler version of the application tool, mainly addressed to individuals, such as mills' owners, disposal areas owners and farmers who may use OMW for irrigation/fertilization or just land distribution is proposed as an additional optional measure, which provides interested individuals with the potential to monitor soil quality of their property periodically, identify potential risks on time and take the appropriate measures in cooperation with local responsible authorities (see paragraph 2.3.5). This tool, although not proposed as statutory measure, however it would be very useful to be included in the annual reporting obligations of the disposal areas' owners to the local/regional authorities. Thus, the owners could submit the outputs of the tools to the local responsible authority annually, and obtain specific advice on the management of their property.

(2) Soil Remediation

In accordance with the Thematic Strategy for Soil Protection (COM 2006, 231 final) actions and means should be oriented to ensure sustainable use of soil. In the light of the above, Member States should proceed to restoring actions in areas that have been identified to be under high risk of soil degradation and conform with the requirement expressed in the Strategy: "restoring degraded soils to a level of functionality consistent at least with current and intended use, thus also considering the cost implications of the restoration of soil".

In addition, areas under medium risk could be also subject to remediation, but this decision will be made by the responsible local or regional authorities considering many factors and not only the rate of the soil risk assessment.

So far, no specific technique has been developed for the remediation of OMW disposal areas. In the framework of PROSODOL project, two methods were developed and implemented in a pilot area in Crete Island, South Greece.

The evaluation of the results revealed that both of them are suitable for OMW disposal areas, if applied properly and under scientific and technical control by qualified persons.

These techniques are (a) bioremediation and (b) application of the natural zeolite, namely clinoptilolite (see details and description in paragraphs 2.4.4 – 2.4.8).

It should be, however, underlined that although efficient, the techniques target to different soil properties and contaminants and their application depends on the specific problem recorded at the targeted areas.

Thus, bioremediation targets to organic pollutants, such as polyphenols, while the application of zeolite targets to the inorganic soil constituents.

Considering these, it is very likely to apply both techniques at the same waste disposal area, however, starting from bioremediation.

The successful application of the two methodologies is depended on the exact adoption of the guidelines, as proposed in the following, and the periodical monitoring of their effectiveness, as far as the soil quality is concerned.

Apart from these two options, more technologies for soil remediation were evaluated regarding their implementability, remedial effectiveness and implementation costs (Table 14).

The selection between the technologies should consider that all the three criteria (i.e. implementability, remedial effectiveness and implementation costs) are highly site depended. Therefore, it is very likely to conform the technologies to the areas under treatment and thus, the costs presented in table 15 would change respectively.

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Remedial Action	Effectiveness	Implementability	Cost €/m^3
No Action	Not protective of human health and the environment; does not meet unrestricted or restricted use criteria	Technically and administratively feasible; availability of equipment and services is not applicable	0.00
Limited Action	Protects human health and partially protects the environment; does not meet unrestricted use criteria and may meet restricted use criteria	Technically implementable, although administrative feasibility could be difficult	13.07
Soil covering -reclamation cover -single layer infiltration reducing cover	Protects human health and partially the environment; the protection of groundwater is in generally safeguarded, polyphenols is anticipated to be reduced only due to natural processes as well as the reduction of all the increased soil parameters. Long time for area recovery, however this is not guaranteed.	Technically and administratively feasible; availability of equipment and services	6.20 22.00
In-situ immobilization of contaminants (zeolite addition)	Protects human health and the environment; the protection of groundwater is in generally safeguarded, polyphenols is anticipated to be reduced however not at that level to meet the remedial objective. Inorganics are "immobilized" by the zeolite however, the method is not effective in reducing copper and phosphorous. It is proposed to be combined with bioremediation technologies. It requires short time (2-3 months) to meet the remedial objectives.	Technically and administratively feasible; availability of equipment and services, although this need is very much limited. It can be implemented by non skilled personnel	27.37
In situ land treatment (biopiling)	Protects human health and the environment; the protection of groundwater is safeguarded, polyphenols are reduced and meet the remedial objective, however this conclusion is site specific. The technology is effective in reducing also K, N, Fe, B at levels that meet the remedial objective. There is also a reduction in other inorganics however not at that level to meet the remedial objectives. It requires short time (4-6 months) to meet the remedial objectives.	Technically and administratively feasible; availability of equipment and services, although this need is very much limited.	46.08
Ex-situ soil composting	Protects human health and the environment; the protection of groundwater is safeguarded, polyphenols could be reduced to meet the remedial objectives, however the reduction of the inorganics at acceptable level is not guaranteed and specific treatment may be required. The method could be combined with the zeolite addition, which can be effective in reducing the toxicity of the inorganics or planting. It requires short time (almost 6 months) to meet the remedial objectives.	Technically and administratively feasible; availability of equipment and services, specific machine is required (turning machine)	57.41
Ex-situ landfarming	Protects human health and the environment; the protection of groundwater is safeguarded, polyphenols could be reduced to meet the remedial objectives, however the reduction of the inorganics at acceptable level is not guaranteed and specific treatment may be required. The method could be combined with the zeolite addition, which can be effective in reducing the toxicity of the inorganics or planting. It requires short time (almost 6 months) to meet the remedial objectives.	Technically and administratively feasible; availability of equipment and services, it required large implementation area	89.43

Table 14. *Evaluation of the selected technologies for the treatment of soils that accept OMWs disposal (for 1 acre of contaminated soil)*

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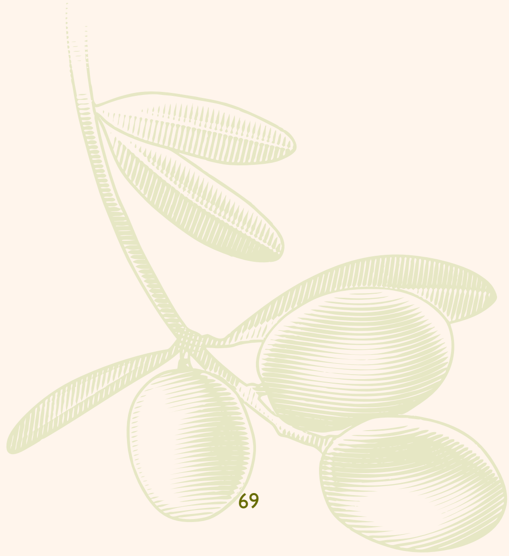
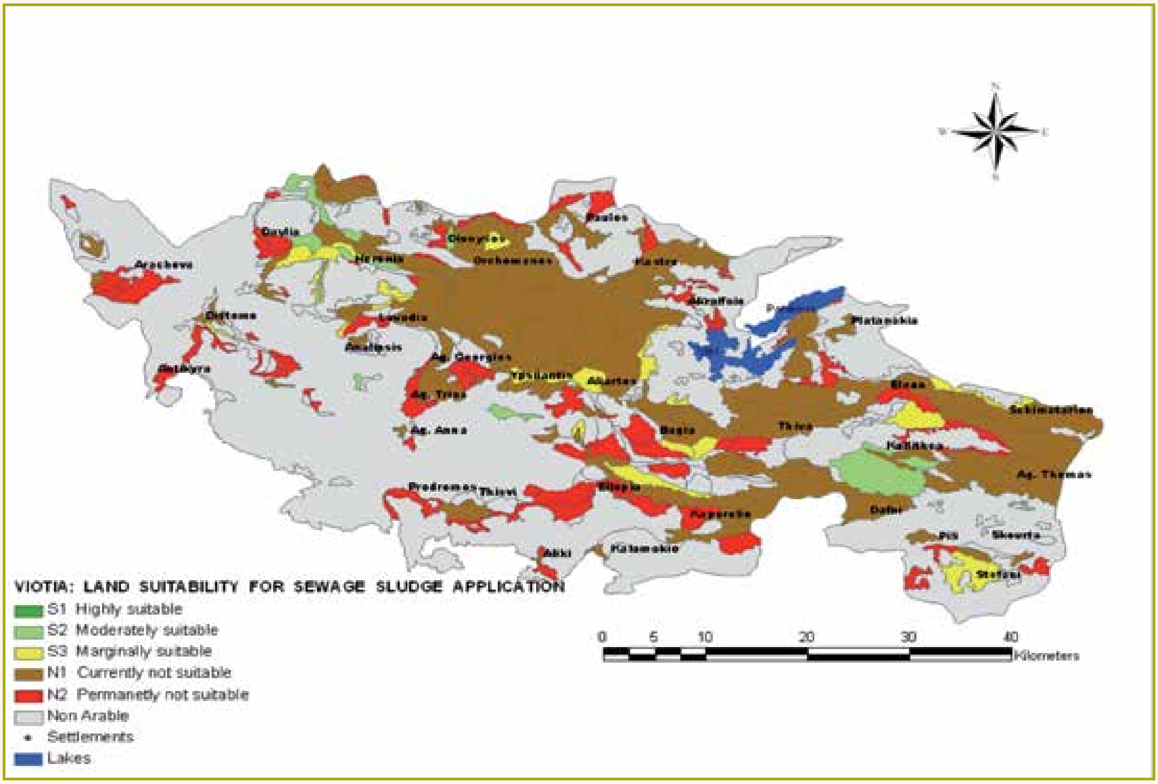
Soil properties	normal/average range	high	very high	excessive	Comments
pH	6-8 [1]				
Electrical Conductivity (EC)	2,0 mS/cm			> 4,0 mS/cm*	*soil quality threshold
Organic Matter (OM)	> 3,4%	> 5%			
Total Nitrogen (N)		> 0,3 %			
Available Phosphorous (P)	12-28 mg/kg	40-50 mg/kg >33-36 mg/kg			
		>59 > 60 mg/kg			
Exchangeable Magnesium (Mg)	1,2-2,2 cmol/kg				* - Nutrient imbalances due to Mg antagonism
			> 2,2 cmol/kg*		- Adverse effect on soil quality
Exchangeable Potassium (K)	0,26-0.60 cmol/kg		>1,2 cmol/kg	> 2,0 cmol/kg	
Exchangeable Calcium (Ca)	2,5-3,8 cmol/kg		> 20 cmol/kg		
Extractable Ammonium (NH4+)	28-280 mg/l				
Phenols	target value: 0,05 mg/kg				< target value: clean soil; < target value < intervention value: slightcontaminated soil; > intervention value: contaminated soil
	intervention value: 40 mg/kg				
Hot water soluble Boron (B)	0,5-1,5 mg/kg	1,6-3,0 mg/kg		> 3 mg/kg	For medium to heavy soils in texture
				> 2-3 mg/kg* > 5 mg/kg*	* phytotoxicity
Available Copper (DTPA-Cu)		> 3,0 mg/kg	1.6-15 mg/dm3	>20mg/kg*	* potential phytotoxicity
Available Iron (DTPA-Fe)			> 50 mg/kg 25-60 mg/dm3		
Available Manganese (DTPA-Mn)			> 50 mg/kg 10-50 mg/dm3		
Available Zinc (DTPA-Zn)			> 8,1 mg/kg-1 2,4-15 mg/dm3	> 130 mg/dm3	

ANNEX 1: Critical levels of some main soil properties (Kavvadias et al.,2010)





ANNEX 2: Land suitability map for the distribution of wastes for Viotia prefecture, Greece

NOTE

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