



Article

# Planning the Flows of Residual Biomass Produced by Wineries for the Preservation of the Rural Landscape

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Abstract: Circular economy aims to create a system that allows an optimal reuse of products and materials. According to an appropriate planning hierarchy, agricultural and agro-food co-products, by-products and wastes should be primarily employed to re-balance soil fertility, and afterwards valorized as new secondary raw materials used in the same agricultural sector or in different industrial chains (e.g., cosmetics, nutraceuticals, etc.). Finally, only at the end of this process, they could be conveyed to energy production through co-generation. In this paper, different residues generated by the wine production chain have been considered with reference to the Basilicata region (Southern Italy). These biomasses have been quantitatively assessed and qualitatively classified, in order to find the most rational and convenient solution for their valorization from a technical, economic and environmental point of view. From the spatial analysis—elaborated by implementing a Geographic Information System—some thematic maps have been obtained, which allow us to highlight the areas with the highest concentration of residues. In this way, focusing the analysis on these areas, some possible strategies for their management and valorization have been proposed, so as to restore soil fertility and contribute to the sustainable preservation of the rural landscape.

**Keywords:** rural landscape; agricultural biomass; winery pomace; Geographic Information System; soil organic matter; circular bioeconomy

## 1. Introduction

The concept of circular economy is based on the idea of the re-use, renewal and optimal recycling of products and materials [1,2]. Over the last century, industrial and technological development has led to massive economic growth with an exponential increase in resource use [3,4]. At the core of the concept of circular economy is the idea that the value of materials and products is kept as high as possible for as long as possible [5], in order to minimize the need to introduce new materials and energy, thus reducing the environmental pressure related to their use [5].

The urgent need to transform the linear model into a circular one is particularly important in the case of agriculture. Primary production—including agricultural productions (vegetal or animal), forest utilization, agro-food industries, as well as fisheries and aquaculture—still represents the main sector for the simultaneous production of goods necessary for human life (food, textiles, energy, etc.) as well as for the sustainable renewal of natural components (air, water, soil, ecological systems, etc.). The valorization of biomass generated into the primary sector can therefore play a significant role for

the success of a circular bioeconomy. This concept may be conceived indeed through the intersection of bioeconomy—i.e., the production of renewable biological resources and the conversion of these resources and waste streams into new added-value products, such as food, feed, bio-based products and bioenergy [1,6]—and circular economy, based on the use of recycled materials to prolong their useful life and on the transformation of waste into new resources.

Some authors [7] have proposed a system for the spatial analysis of residual biomass from the primary sector. These biomasses have been identified and inserted in a Geographical Information System (GIS), with the aim of their sustainable valorization in the rural areas in a region of Southern Italy. These areas have been considered within the potential installation of biomass and/or biogas plants, taking into account transport costs, management costs and the suitability of the land for soil fertility depletion, in the context of an "Energyscape" approach [8]. These residues can be considered as an important source of energy, as they can be used as an alternative fuel in energy installations [9]. On the other hand, they are important factors in restoring the level of soil organic matter (SOM), so as to preserve soil fertility over time. In addition, more value-added options could be explored for the reuse of these residues and their possible exploitation in different production chains (cosmetics, nutraceuticals, etc.), as well as in the construction sector, for example, as a natural additive that could be incorporated in clay bricks to increase their technical performance [10]. In this context, due account should therefore be taken of the need to respect a hierarchical process, giving priority to restoring of soil fertility, as well as other secondary possibilities offered in the agricultural sector itself, which can contribute to its sustainable development [11].

Soil is a fundamental component of agriculture, as it forms the basis of all chemical and physical processes. In recent decades, inadequate soil nutrient substitution has led to the depletion of the natural soil base available for food production [12]. Sustainable agricultural production incorporates the idea that natural resources are used to increase agricultural production and income without depleting the natural resource base [13]. In addition, SOM in soils improves soil structure and increases their ability to retain water [12,14].

# 2. Residual Biomass Produced by Wineries

Mostly in the agro-industrial sector—such as wineries, olive oil mills, cheese factories, flour mills, etc.—large quantities of secondary products are usually available, and they can be transformed into a new resource. In the case of residual biomass coming from wineries, there are currently different possibilities of re-utilization of residues which, depending on the relevant industrial process, may have a very different environmental impact. Table 1 shows the main residual materials classifiable as by-product and the processing yields of grape [15].

**Table 1.** Wine production and relevant residues from the transformation of 100 kg of grape.

Exit (kg)	Residues Classifiable as by-Products (			
	Virgin (5.4) and exhausted pomace (4.6)			
Wing (77 Va)	Grapeseeds	5		
vviile (77 Kg)	Stalks			
	Dregs	5		
	Exit (kg) Wine (77 Kg)	Virgin (5.4) and exhausted pomace (4.6) Wine (77 Kg) Grapeseeds Stalks		

From each one of the winemaking phases, different types of by-products are generated. The stalks are the first to be released. They correspond to the woody part of the bunch and are eliminated during de-stemming. Subsequently, in white vinification, the pressing releases a virgin, unfermented pomace, characterized by high concentrations of sugars used for processing into alcohol. In red vinification, the pomace produced is composed of grape seeds, grape skins and stems residues. It is characterized by a good level of sugar fermentation. The scum of wine is the muddy residue that is deposited in the containers, after fermentation, during storage or after authorized treatments, as well as the residues

obtained by filtration or centrifugation of this product. This consists mainly of yeast cells produced during alcoholic fermentation, bacteria, tartaric salts, plant cell residues and ethanol [16,17].

The main residue of the supply chain is the pomace, which represents about 10%–30% of the mass of grapes crushed. It consists mainly of unfermented sugars, alcohol, polyphenols, tannins, pigments and other valuable products. The size of the winery and the winemaking methods used directly influence its quantity and quality [18]. Together with the residual sugars, it has other physical (e.g., pH, moisture, etc.) and chemical (e.g., lignocellulose, polyphenols, ash, etc.) characteristics, which are important to consider when pomace is used as raw material (Table 2).

Parameter	Value	Reference	Parameter	Stalks	Pomace	Dregs	Reference
Ph	$3.6 \pm 0.2$		рН	4.4	3.8	4	
Moisture	$73.6 \pm 2.6\% \text{ w/w}$	[19]	Organic Substance (g/kg)	920	915	759	
Reducing sugars	$1.5 \pm 0.3\% \text{ w/w}$		Oxidizable organic carbon (g/kg)	316	280	300	
Ash	$4.6 \pm 0.5\%$ w/w		Water soluble carbon (g/kg)	74.5	37.4	87.8	[21]
Cellulose	20.8 % w/w		Total nitrogen (g/kg)	12.4	20.3	35.2	
Hermicelluloses	12.5 % w/w		P (g/kg)	0.94	1.15	4.94	
Tannins	13.8 % w/w	[20]	K (g/kg)	30	24.2	72.8	
Proteins	18.8 % w/w		Ca (g/kg)	9.5	9.4	9.2	
Ash	7.8 % w/w		Mg (g/kg)	2.1	1.2	1.6	
			Fe (mg/kg)	128	136	357	
			Mn (mg/kg)	25	12	12	
			Cu (mg/kg)	22	28	189	
			Zn (mg/kg)	26	24	46	

Table 2. Physical and chemical properties of grape pomace and other by-products.

In the specific case of pomace, the solid waste produced as a result of the treatment processes carried out must be disposed of safely, to avoid environmental hazards and bad odors [22].

The entire wine supply chain does not just include the production of grapes, their transformation and marketing alone, but also the series of processes that concern the transformation of waste, such as those destined to distilleries. This recovery phase is therefore a fundamental link for the realization of a virtuous system, able to enhance every single phase of the manufacturing process and thus reduce its environmental impact [23].

The concern regarding increases in GHGs has led many companies towards sustainable wine production practices [24–26]. There are five main critical points in the wine industry that could be an environmental problem, as shown in Table 3. The main environmental impacts are due to the use of pesticides and fertilizers (41%), to which must be added a 32% due to transport [26,27]. Knowledge is the first step towards sustainable production: knowing where organic waste comes from in order to manage it correctly and to reuse it for alternative purposes [25,28,29].

Industry: Process Steps Environmental Risk	co. Correlation between while may	abily process and environmental flow (adapted from	
	Wine Industry: Process Steps	Environmental Risk	

Glass bottles and paper labels

Electricity, water, sulphur dioxide and sodium hydroxide

Fuel

Effluents, wastewater and grape pomace

**Packaging** 

Vinification

Transport

Waste management

Table 3. Correlation between wine industry process and environmental risk (adapted from [27,28]).

Problems associated with waste generation in the wine industry are of special relevance during the grape harvest, a very short period of time, usually located between September and October in the Mediterranean area [30]. Annually, the grape production requires big amounts of chemical fertilizers.

Sustainability **2020**, 12, 847 4 of 13

The possibility of recovering organic wine wastes to soil vineyards may be considered as a sustainable strategy for waste management.

The reuse of vinification residues could anyway find a second life in different areas. Portilla-Rivera et al. (2007; 2010) [31,32] used the pomace as a carbon source for lactid acid fermentation and for the production of bio-emulsifiers, while Ping et al. (2011) [33], Thorngate and Singleton (1994) and Karleskind (1992) [34,35] used the polyphenols present in the pomace as dyes for the food industry and as antioxidants. Other authors have proposed the use of pomace for phytopathogenic control [16,36] or have used the vinification dregs as a fundamental raw material for the production of ethanol and tartaric acid [37,38]. Diaz et al. (2002) [39], Mustin et al. (1987) [40] and Ferrer et al. (2001) [41] have used stalks mainly as a compost for the restoration of soil fertility with consequent spreading in the soil.

However, the potential uses of cellar and distillery waste as raw materials depend heavily on the market value of the products obtained [42]. In this paper, a further specific analysis is presented focusing on by-products generated by the wine sector with reference to a study area, within which there is a strong correlation between agriculture and human actions aimed at the sustainability of the rural landscape [43].

### 3. Materials and Methods

The analysis on the residues of the wine industry has been performed using a Geographical Information System (Quantum GIS-v. 3.4) applied to a study area, whose data concerning the main characteristics of biomass coming from wineries have been considered. In addition, a relevant analysis of the fertility of soils in a sample study area has been performed as well, through some experimental tests on Soil Organic Matter.

## 3.1. Study Area

The study area consists of the total land of the Basilicata region (Italy) (Figure 1). This region has a geographic surface of 10,073.11 ha [44]. It is a mostly rural territory; the population was 562,869 inhabitants in 2017 [45], with a quite low regional population medium density (56.3 inhabitants per km²) [46]. The territory of Basilicata shows wide morphological differences, mainly in elevation, ranging from the sea level to over 2200 m a.s.l. Despite the diversity of the various areas, the climate of the region can only be defined as continental with Mediterranean characteristics in coastal areas; while in inland areas, especially in winter, the mildness is immediately replaced by a harsh and humid climate. It is characterized by seasonal variations in temperature (hot summers and very cold winters). The territory is mainly mountainous (47%) and hilly (45%) with a modest percentage of flat land (8%). The hills are clayey and subject to erosion phenomena that give rise to landslides. The area used for agriculture is 472,833 ha (accounting for 46.9% of the regional surface area), of which 58.6% is used for arable crops and 11.7% for woody crops. The 29.7% remaining surface area is characterized by permanent grassland, especially in mountain areas [46].

In the year 2013, the regional surface covered by vineyards was equal to 5310.43 hectares [46]. The area potentially producing D.O.C. (Denominazione di Origine Controllata–Product of a Controlled Origin) wines is 32% of the total area covered with grapes (1699.33 hectares).

Assuming an annual production of 8 t/ha of grapes, and calculating the territory dedicated to the whole amount of production of wine grapes (1699.33 ha), the quantity of exhausted pomace usable has been estimated [11]. Moreover, the percentage of the total exhausted pomace considered in this case is the 4.6% of production of wine grapes, which means a pomace production of about 625.35 t/year.

Sustainability **2020**, 12, 847 5 of 13

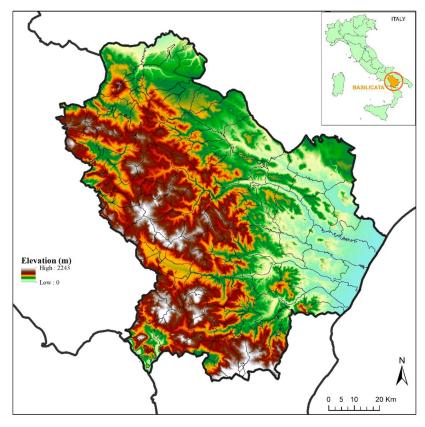


Figure 1. Study area (Basilicata Region).

## 3.2. Analysis of the Soil Organic Matter

Starting from an analysis of the content of organic matter present in the soils of the Basilicata region (Figure 2) and observing that in the north-east part of the region there are smaller quantities of organic matter, the study area has been restricted to this territory, also considering that here the largest distribution of wineries is concentrated. This area is one of the most important areas of the Basilicata region for the production of wine ("Monte Vulture"). In this area, the warm winds of Adriatic influence contribute to the development of vineyards and the presence of an ancient volcano, but now extinct, gives the wine its rare and much appreciated characteristic taste [11]. The DOC wine par excellence is the "Aglianico del Vulture", to which the lava matrix of the soil gives particular organoleptic characteristics different from other titles of Aglianico. It is cultivated up to 800 m of altitude, although the optimal conditions are between 200 and 600 m. In this area, this type of vine has found its suitable territory, thanks to the composition of the soil and the complex orography that determines a wide variety of climates depending on altitude, slope and exposure.

In order to confirm the data shown on the map, some samples were taken from land in vineyards in the area. The survey has been carried out in n.5 specimens soils, using a Global Positioning System (GPS), so as to detect the exact location of soil sampling pits (Figure 3), with the aim to identify physical and chemical properties of the soil units. The coordinates (latitude and longitude) of these five points have been recorded and imported into the Quantum GIS-v. 3.4.

The collected soil samples were air dried in shade, crushed and sieved for the laboratory analysis. The laboratory analysis on these five have included: total nitrogen, organic matter, total phosphorus and potassium, while the information about particle size distribution and soil pH have been obtained by consulting the soil maps for the Basilicata region [47].

Sustainability **2020**, 12, 847 6 of 13

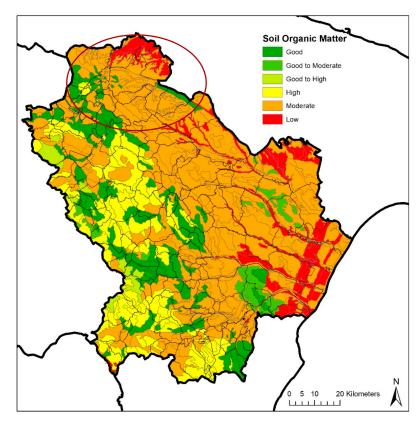
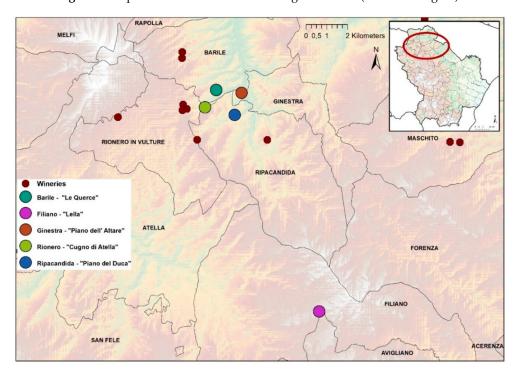


Figure 2. Map of the concentration of soil organic matter (Basilicata Region).



**Figure 3.** Location of wineries and sampled soils in the study area: EPSG 32633—40°56′01″ N 15°41′39″ E (Barile—"Le Querce");  $40^\circ50'13''$  N 15°45′08″ E (Filiano—"Lella");  $40^\circ55'56''$  N 15°42′31″ E (Ginestra—"Piano dell'Altare");  $40^\circ55'34''$  N 15°41′16″ E (Rionero—"Cugno di Atella");  $40^\circ55'22''$  N 15°42′17″ E (Ripacandida—"Piano del Duca").

Sustainability **2020**, 12, 847 7 of 13

#### 4. Results

Figure 4 shows the distribution at the municipal level of grape pomace, the main residues of the wine supply chain. From the analysis of this map, it is possible to identify the distribution of this resource over the whole regional territory, expressed in tons, as well as to highlight the areas with the highest concentration of this residue. The resulting map highlights two areas where the concentration of this residue is at the greatest level: one is in the northeastern area, and another near the coast of the Ionian Sea. The data are represented at municipal level given the availability of the same at this level of detail.

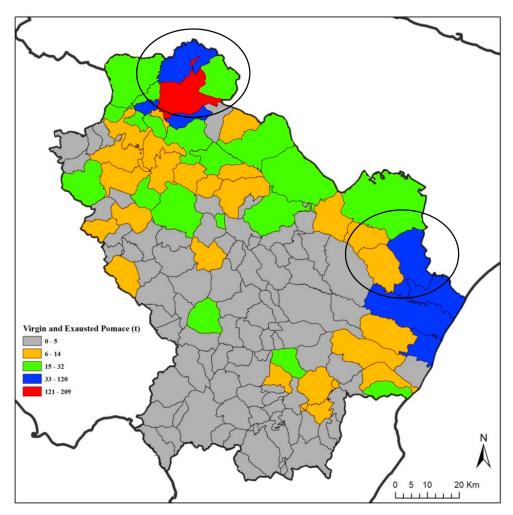


Figure 4. Availability of the pomace (virgin and exhausted), the main residue of wine supply chain.

On the basis of the present analysis, it is possible to plan a suitable system aimed at the collection and subsequent recovery of these pomace, mostly if concentrated into the two main areas of the region. The re-use of these by-products needs to be planned, also considering the spatial reference, in order to facilitate and/or solve environmental issues in the process associated with their disposal.

These data represent the starting point for the implementation of a strategy aimed at the reuse of these residues respecting a hierarchical process that privileges, above all, the restoration of soil fertility and then continues towards other forms of valorization. This strategy is represented by a virtuous and circular system based primarily on a good organizational model that provides for participation and collaboration between different actors: agricultural production activities and agro-food industries that have the task of giving a second life to these residues, associations and public bodies for the construction of composting plants (for the production of compost) and distilleries (to eliminate the

Sustainability **2020**, 12, 847 8 of 13

alcohol component still present in the residues following their production)—and a careful analysis of the entire network based on distances, accessibility, viability and amount of biomass usable.

The final objective will be to obtain a sustainable process from an economic, environmental and agronomic point of view and maximize all the advantages deriving from the reuse of these residues within the company itself, with a view to a circular bioeconomy, i.e., minimal environmental impacts and the reduction in costs related to the transport of residues, minimum production of waste within the company, improvement of soil properties following the enrichment of the soil with natural organic matter with consequent advantages on the sustainability of the rural landscape associated with the natural restoration of the soil properties and a reduction in final disposal costs.

Despite all the good intentions and considerations, much should still be done in this direction, especially as regards cultural training. This is the main obstacle to overcome, that is to make the company fully aware of all the advantages that would derive from the effective reuse of these residues. This discussion, applied on a small scale area of Basilicata region (Table 4), could then be extended, on a large scale, to the entire region with even greater advantages [48].

Soil Parameter	Barile "Le Querce"	Filiano "Lella"	Ginestra "Piano dell'Altare"	Rionero "Cugno di Atella"	Ripacandida "Piano del Duca"	Acceptability Range *
SOM (g/Kg)	8.69	24.3	22.24	19.31	15.9	15–20 g/Kg
Total N (g/Kg)	1.4	1.9	1.9	2.6	2.6	1–1.8 g/Kg
Total P (g/Kg s.s.)	1.38	0.367	1.612	2.185	3.002	1.5–2.5 g/Kg s.s.
K (g/Kg s.s.)	7.919	3.288	5.871	8.241	6.150	5.5–8.5 g/Kg s.s.
pН	6.7	8.1	6.8	6.8	6.9	6.5–7.3 (neutral)
Texture	fine clayey	clayey	fine clayey	fine clayey	fine clayey	100–300 g/Kg

**Table 4.** Results obtained on the analyzed soil samples.

According to the results obtained in terms of soil structure, four out of five soils tend to be clayey while only one is completely clayey. The pH is generally comprised between 6.5 and 7.3 (neutral soils) except in a case where the soil is alkaline with a pH value above 8.

The main agronomic characteristic of the soils analyzed is a wide dispersion of their SOM contents, resulting in a general lack of organic matter. The value of the organic matter varies indeed from 8.69 to 24.3 g/kg with an average value of 18.08 g/kg. This contributes to a relatively low total phosphorus content, since many authors [49] have underlined the close correlation of this parameter with organic carbon. Indeed, the total phosphorus content in the study area varies from 0.367 to 3.002 g/kg s.s. with an average value of 1.709 g/Kg s.s., so quite close to the lower limit of the acceptability range. This also happens for Potassium, which is also low, since it varies from 3.288 to 8.241 g/Kg s.s. with an average value of 6.293 g/Kg s.s. The total nitrogen content, instead, appears sufficient, since it varies from 1.4 to 2.6 g/Kg, with an average value of 2.08 g/Kg, so even higher than the upper limit of the acceptability range.

The lack of organic matter prevents the soils to perform their functions properly as it significantly reduces their physical, chemical and biological fertility. These types of soil, generally light in color, do not have sufficient nourishment and are characterized by high fragility, and they do not retain the roots of plants and are more vulnerable to erosion. The continuous removal of organic matter from the soil could be balanced by the intake of nutrients present in the residues of the wine supply chain, in order to close the cycle of organic matter and facilitate the return to the soil of the substances removed from the agricultural activities.

In Table 5, the main average and representative characteristics of a sample of pomace belonging to the grapes produced in the study area are reported. From these results, it is clear that this material can be considered as a good source of organic matter and macro elements such as nitrogen, phosphorus

<sup>\*</sup> Value elaborated from [48].

Sustainability **2020**, 12, 847 9 of 13

and potassium for restoring and improving soil fertility, if it is adequately processed so as to transform into a soil fertilizer.

Pomace	Unit	Value	Soil Test Methods
SOM	g/Kg	915	D.M. 13/09/99 SO GU n. 248 21/10/99 Met. VII.2 D.M. 25/03/02
Total N	g/kg	11.07	D.M. 13/09/99 SO GU n. 248 21/10/99 Met. XIV.3 D.M. 25/03/02
Total P	g/Kg	2.567	D.M. 13/09/99 SO GU n. 248 21/10/99 Met. XV.1
Total K	g/Kg	21.303	UNI EN 13656:2004 + EPA 7000 B 2007

**Table 5.** The main characteristics of the sample pomace.

This inverse proportionality between the values observed for soil and pomace brings with it some very interesting considerations from an agronomic, environmental and economic point of view.

### 5. Discussion

The comparison between the results obtained and the map of spatial distribution of these residues suggests a very important consideration: where the concentration of such residues is high there is a low concentration of organic matter in the soil. Therefore, in order to close the circle of the organic matter, these residues could be reused to compensate the lack of organic matter in the vineyards considered. In fact, most of the cultivated areas are quite poor in organic matter contents and the available phosphorus and potassium is also below the desired level.

At the same time, considering the high content of pomace organic carbon, a limited content of nitrogen and a consequent high carbon–nitrogen ratio, pomace are then suitable for improving the chemical–physical properties of the soil in the medium to long term [50]. Currently, these residues are dumped in landfill and/or incinerated with an increase in the social costs for their disposal, while if properly treated—for example, by eliminating the polyphenolic component, which may be exploited in the cosmetics industry, in which they are widely used—they can be validly reused in agriculture, thus combining the agronomic advantages with the opportunity to recycle residual material.

In this perspective there are two main ways to reuse these residues:

- Direct spreading on the ground: this practice is very simple and allows the spreading on lands, in particular vineyards. It is not a very advantageous practice for farms because it is necessary to respect a series of constraints imposed by the legislation (in Italy, the DGR n. 8/5868 of 11/21/2007 and n. IX/2208 of 09/14/2011) linked first of all to the quantity and the extension of the surface. Furthermore, the residues must be appropriately treated before being applied to the soil, since they contain some characteristics that could cause collateral unfavorable effects, such as the alcohol content or the high content of polyphenols. These elements could give the soil too much macronutrients, thus giving rise to a general negative effect.
- 2. Mixed with other agricultural residues to produce compost: this practice is much more used and convenient for wineries or farms as it contributes to the improvement of the content of organic matter. It represents a technically and economically advantageous process, with low environmental impacts. This organic matter increases microbial biomass and helps to maintain the beneficial bacterial and fungi populations. In addition, there are economic advantages, since the use of residues involves lower costs than those related to conventional materials [51,52]. Particularly, the compost obtained is recommended for application to the soils, because the nitrogen is gradually released, leaving the soil time to absorb it.

The increase in organic matter in the soil, following the reuse of the considered residues, generates many benefits:

Positive consequences on the concentration of many macro nutrients, the mineralization of the
organic substance causes the release of the contained macronutrients, which can then be absorbed
and used by the soil.

Sustainability **2020**, 12, 847 10 of 13

• The improvement of the structure of the soil and maintenance of the pH at values close to neutrality. Neutral soils are the most suitable for agriculture because most agricultural species adapt optimally to pH values between 6.5 and 7.5.

- The conservation of soil biodiversity and limitation of erosion since the organic fraction is a source for a wide range of organisms [53].
- Mitigation of climate change, in this context, the recycling of organic residues allows to combat carbon emissions into the environment (e.g., CO<sub>2</sub>), co-responsible for the greenhouse effects. Globally, soil stores about twice as much carbon in the atmosphere and three times as much carbon as vegetation. Therefore, agricultural soils are a huge storage tank for organic carbon [50].
- Preservation of the rural landscape, thanks to the natural restoration of soil fertility, without
  introducing chemical/artificial fertilizers, which may transform, in the long time, the main features
  of the soil, then the consequent vegetation and associated ecosystems.

#### 6. Conclusions

Land use for agricultural purposes is leading to unprecedented changes in the rural landscape, ecosystems and environment. Rural landscapes are changing due to the intensification of agriculture, which exploits the soil and changes its state and natural functions, bringing with it environmental problems such as climate change, loss of biodiversity and pollution of water, soil and air [54]. To overcome this deficiency and at the same time to complete the cycle of organic matter, the continuous removal of nutrients for agricultural purposes and, in general, for the growth of plants, should be compensated by the contribution of macronutrients.

The results obtained from the analyses and elaborations obtained have been presented in the form of maps, in order to identify the spatial distribution of biomass in Basilicata region. They represent a powerful tool to help planners' in landscape analysis [55] and support policy makers in their decisions based on the information derived from these databases [56].

The laboratory results have shown that where the availability of residues is high, this corresponds to a low concentration of organic matter in the soil. In fact, most of the cultivated areas are quite poor in organic matter contents and the available phosphorus and potassium is also below the desired level. This is probably due to the continuous tillage of the soil for agricultural purposes, which requires a continuous extraction of macronutrients from the soil, without a simultaneous supply of nutrients from natural and artificial sources. The reuse of residues generated by agricultural activities could be an excellent strategy in the perspective of a circular bioeconomy, because it would transform waste into a precious resource. This virtuous system, based on the one hand on the collaboration between productive agricultural activities, agri-food industries and entrepreneurs—and on the other, on the analysis of economic and environmental feasibility—would bring numerous benefits, essentially related to the costs of non-disposal, ease of transport and waste management, improvement of soil properties and better sustainability of the rural landscape associated with the natural restoration of soil fertility.

**Author Contributions:** The five authors collaborated to produce this paper. C.M. and D.S. proposed and developed the research design, methodology, manuscript writing, and data analysis. G.G. allowed the collection of samples to be analyzed in the laboratory, A.D.P. contacted the laboratory for samples analysis, P.P. supervised the work, provided additional comments on the results and interpretation, reviewed and approved the final version. All authors have read and agreed to the published version of the manuscript.

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