Article

Evaluation of Land Potential for Use of Biosolids in the Coastal Mediterranean Karst Region

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Abstract: The aim of this study was to evaluate the potential of agricultural land in the coastal Adriatic Karst region (Šibenik region, Croatia) for biosolids application by integrating spatial data from different sources: digital maps and remote sensing, parcel identification system, GIS field observations and measurements focusing on specific land and soil properties. Due to the rapid development of the wastewater treatment industry, excessive accumulation of sewage sludge (SS) in wastewater treatment plants is a growing problem worldwide. Management options for land application of biosolids require a comprehensive characterization of both SS and SS-amended soils. The assessment of agricultural land in the study area for SS disposal was based on EU and national legislation. The evaluation revealed that agricultural land in the study area accounts for only 10% of the total area (25,736 ha), but only a quarter of the existing land (6065 ha) is suitable for biosolids application. Furthermore, the data indicate that the sewage sludge can be safely applied to the soil in terms of soil metals according to the Croatian legislation. The short-term potential of the soil to sustain this ecosystem service, namely soil improvement with biosolids, should be used to determine the inherent long-term potential based on resistance to soil degradation and resilience. However, caution is needed and the long-term effects should be investigated before biosolids are continuously used for soil application.

Keywords: ecosystem services; land evaluation tools; spatial information; soil attributes; land degradation; soil heavy metals

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1. Introduction

In the context of integrated land and water management, the generation of wastewater and the growing quantities of biosolids from local wastewater treatment plants are issues of high importance worldwide [1]. The term biosolids is used to refer to the nutrient-rich organic materials generated during the treatment of domestic wastewater in a wastewater treatment plant during the primary (physical and/or chemical), secondary (biological) and tertiary (in addition to secondary) treatment processes of the wastewater [2]. The terms “biosolids” and “sewage sludge” (SS) are often used interchangeably [3]. Anaerobic digestion and composting are the two most widespread SS stabilization processes at wastewater treatment plants (WWTP), which play a key role in SS recycling and utilization [4].

Operating methods and regulatory framework to ensure the proper application of biosolids for protection of the environment and human health have been defined worldwide and applied at both international and national levels [3,5,6]. In the EU, there is a general impression that the Sewage Sludge Directive (SSD) issued in 1986 [7], although it has well fulfilled its objective of ensuring the diverse and safe use of sewage sludge, needs to be revised to meet the new challenges: emerging pollutants, digitalization and the circular

economy [8]. Current national regulations on this issue differ between EU member states, resulting in a fragmented legal framework in which sewage sludge is still classified as waste and managed in a coherent manner [9]. The differences in the legislation strictness at the national level can be reasonably explained by the different geomorphology, marked climatic differences and variability of soil types within the country. However, few countries have enacted regional legislation to regulate the use of sewage sludge in different areas, such as in Italy [10].

Croatian Adriatic coastline is a region of exceptional karst landscape, and the main drivers of the regional economy are agriculture and tourism. The population density in the region is rather low (36.65 inhabitants per square kilometer of land area), but since it is an attractive tourist region, the production of SS in sewage treatment plants is continuously growing [11]. The Šibenik region (Middle Eastern Adriatic) has in the past been confronted with the serious problem of eutrophication of the Krka River Estuary and the coastal marine environment, which has been receiving untreated industrial and domestic wastewater for years [12].

The discharge of SS into the Adriatic Sea was stopped in 2008 after the construction of a wastewater treatment plant, and the treated wastewater is discharged into the nearshore marine environment SE of Zlarin Island through a 5000 m submarine pipe system [13]. Nowadays, most households, industrial and municipal facilities in the Šibenik region are connected to the WWTPs, and eight plants are currently in full operation. After the installation of the solar SS drying plant, the annual production of biosolids that should be managed is estimated at two thousand tons of dry matter per year.

However, the end-use scheme to solve the problem of increasing production of biosolids in the agglomeration has not yet been defined. Currently, the end use of biosolids varies worldwide and is determined by the quality of the product and the available options for beneficial use or disposal [14]. While biosolids can be used as a valuable resource for organic matter, nutrients and trace elements, land application appears to be a preferred management option compared to conventional disposal, which is now limited [15].

For the application of biosolids to land, European Union provides a legal framework at Council Directive (86/278/EEC). However, Hudcová and Vymazal [16] pointed out that this directive, being outdated, does not meet the current requirements for ensuring the safety of SS application in agriculture. Croatia, as an EU member state, has developed national legislation based on this directive [17]. Besides the restriction on the choice of crops, one of the important points in the sewage sludge regulation is the setting of limits for heavy metal concentrations, pathogens and organic compounds, as well as sludge and soil analysis and their frequency.

Biosolids are the most researched and regulated organic material applied to land, and information sources agree on the significant potential of such a practice. Although the current regulation on the reuse of biosolids to agriculture in Croatia is in line with the EU Sludge Directive (86/27/EEC), there are important differences between the various regulations, especially in relation to the permitted uses. In addition, scientific support is needed for the development of national guidelines to complement a future regulation on the application of biosolids, taking into account regional specificities.

Karst environments are considered to be highly fragile environments in the world, extremely vulnerable to a range of degradation events [18]. Moreover, karst aquifers are characterized by rapid recharge via karst features and extensive surface water-groundwater interaction. Due to the carbonate environment and the exposure of bare rock, there is generally no true water flow system at the surface as water rapidly infiltrates into the subsurface through the network of karst fissures and conduits in the rock mass [19]. This fact is crucial for understanding the risk that the application of sewage sludge and the leaching of potentially toxic substances contained in the sludge may have on karst aquifers.

The use of SS as a soil amendment has many benefits when it comes to improving the nutrient status of depleted Mediterranean soils [20,21] and, therefore, the application of SS

as an organic fertilizer has increased in recent years [22,23]. Restoration of degraded soils with organic waste could also be a viable method to minimize erosion in such areas [24].

Considering the ecosystem service of agricultural land as SS amendment in addition to agricultural production, SS sustainable management and environmental protection, and based on the current legislation defining the conditions for the application of SS on agricultural land, this study was aimed: (i) to provide the methodology of rapid evaluation of agricultural land for biosolids application by integrating different spatial information such as digital maps and remote sensing, parcel identification system, GIS field observations and measurements, and (ii) to assess the potential of agricultural land within the study area for SS use as soil amendment, focusing on specific soil and land properties, including pedo-geochemical characterization of agricultural land.

2. Materials and Methods

2.1. Study Area and Land Resources Inventory

According to the Croatian regional administrative system, the study area belongs to Šibensko-kninska County, located in Central Dalmatia (Figure 1). The region covers 2984 km² with a population of 109,375 inhabitants [25]. As a coastal area on the Adriatic Sea, attractive for its exceptional natural resources and cultural heritage, the county is a popular tourist destination and the number of tourists is higher than the number of inhabitants during the summer months.

Figure 1. Geographical setting of the study area, surface soil sampling points and GIS sample point identify window (turquoise dot).

Geospatial tools combined with targeted field studies were used for the land resources inventory. Information on spatial distribution and classification of soils and land use was processed. Soil and land use mapping were used to identify different site-specific soil

management practices and agricultural land was found to occupy about 10% of the total land area (5736 ha). Different types of agricultural land use were identified using remotely sensed data and land parcels database, particularly Croatian agricultural parcel identification system (ARKOD), which is a national agricultural parcel identification system [26]. The 1:25,000 digital soil map was used to identify dominant soil types in the study area.

Soil survey and sampling were carried out according to the square mesh sampling grid with sampling points spaced at 8×8 km using 1:25,000 scale topographic maps, apart from the Petrovo *polje* area where the sampling distance was 1 km (Figure 1). The composite samples were made up of 10 soil subsamples taken in a cross pattern, with a 5 m distance between subsamples. The observation sites were spatially referenced using GPS and data were stored in different GIS layers. A site survey, carried out during the sampling process, provided site-specific information related to land use and other human activities near the sampling points.

2.2. Laboratory Methods

Soil samples (average weight 2 kg) were air-dried, and, after homogenisation, half of each sample was sieved through a 2 mm mesh screen and stored in polyethylene bottles as a backup sample.

The other half was sieved through a $0.5 \mu\text{m}$ mesh and used for determining soil concentrations of heavy metals after microwave (MARSXpress system, CEM) assisted aqua regia digestion [27]. Elements' concentration (Cd, Cr, Cu, Ni, Pb and Zn) in soil digests were determined by inductively coupled plasma optical emission spectroscopy (ICP-OES) (Vista-MPX AX, Varian, Inc., Palo Alto, CA, USA). Concentration of Hg was measured on Flow Injection Mercury System (PerkinElmer FIMS 400 with AS-91 Autosampler) by mercury cold vapour generation and interference-free using SnCl_2 as reducing agent. For each sample, soil pH was determined using a 1:5 soil weight/water volume ratio, organic carbon (OC) was determined by sulfochromic oxidation and available K_2O and P_2O_5 by the ammonium lactate method [28]. Total nitrogen was determined by the Kjeldahl method [29]. The EC and pH were measured using a MettlerToledo MPC 227 conductivity/pH meter. Sodium and potassium concentrations were determined by atomic emission spectrometry (AAS PerkinElmer 3110). All concentrations were calculated on the basis of dry weight (d.w.) of the samples (105°C , 24 h). The total amount of polycyclic aromatic hydrocarbon compounds in the sample was determined by liquid chromatography (HPLC). The congeners of polycyclic biphenyls (PCB-28, PCB-52, PCB-141 and PCB-180) and total polychlorinated biphenyls (PCBs) were determined by gas chromatography.

Biosolids characterization methods, including the SS stabilization methods, were described in detail by Černe et al. [23].

Quality control consisted of reagent blanks, duplicate samples and several referenced soil and sediment samples of a similar matrix. The maximum allowable relative standard deviation between replicates was set to 10%.

2.3. Soil Types

A 1:25,000 digital soil map of the Šibensko-kninska County was used to identify dominant soil types in the study [30]. According to the World Reference Base [31], the dominant soil types of the study area are classified as Rhodic Cambisols clayic, Chromic Cambisols loamic and Rendzic mollic Leptosols loamic. Related to parent material and relief, Rhodic Cambisols clayic can be additionally specified as leptic, skeletal or colluvic, and Chromic Cambisols as leptic, colluvic or calcaric.

2.4. Data Processing and Mapping

The statistical analyses were conducted by using the Statistical Analysis Software [32]. Thematic soil maps, needed to evaluate the potential of agricultural land in the study area, were generated with the Geostatistical Analyst extension in the ArcGIS 10.8 program [33]. Interpolated maps of heavy metals and potentially toxic elements, PAHs and PCBs in

surface soil layer were produced by applying Inverse Distance Weighting (IDW) method. This method assigns weights in an averaging function based on the inverse of the distance (raised to some power) to every data point located within a given search radius centered on the point of estimate [34].

2.5. Legislation

Specific legal rules for sludge treatment, disposal and usage are defined at both the EU (Directive 91/271/EEC, Directive 86/278/EEC) and at national level. Agricultural usage of sewage sludge has been regulated in Croatia by the Directive on sewage sludge usage in agriculture, as shown in Table 1 [21]. Table 2 shows limit values of Cd, Cu, Ni, Pb, Zn, Hg and Cr in sludge-treated agricultural soil (mg kg^{-1} dry soil) [17].

Table 1. Limits of Cd, Cu, Ni, Pb, Zn, Hg and Cr, and of polychlorinated biphenyls (PCBs), for sludge used in agriculture (mg kg^{-1} d.w. of sewage sludge) [17].

		Limit Value (mg kg^{-1} d.w. of Sewage Sludge)
Heavy Metal	Cadmium (Cd)	5
	Copper (Cu)	600
	Nickel (Ni)	80
	Lead (Pb)	500
	Zink (Zn)	2000
	Mercury (Hg)	5
	Chromium (Cr)	500
PCBs	2,4,4'-Trichlorobiphenyl	0.2
	2,2',5,5'-Tetrachlorobiphenyl	0.2
	2,2',4,5,5'-Pentachlorobifenyl	0.2
	2,2',3,4,5,5'-Hexachlorobifenyl	0.2
	2,2',3,4,4',5,5'-Heptachlorobifenyl	0.2

Table 2. Limits of Cd, Cu, Ni, Pb, Zn, Hg and Cr in sludge-treated agricultural soil (mg kg^{-1} d.w. of soil) [17].

Parameter	Limit Values (mg kg^{-1} d.w. of Soil for Soil Heavy Metals Concentration)		
	$5.0 < \text{pH} < 5.5$	$5.5 < \text{pH} < 6.5$	$\text{pH} > 6.5$
pH_{KCl}	$5.0 < \text{pH} < 5.5$	$5.5 < \text{pH} < 6.5$	$\text{pH} > 6.5$
Cadmium (Cd)	0.5	1	1.5
Copper (Cu)	40	50	100
Nickel (Ni)	30	50	70
Lead (Pb)	50	70	100
Zink (Zn)	100	150	200
Mercury (Hg)	0.2	0.5	1
Chromium (Cr)	50	75	100

3. Results

3.1. Land Resources and Agricultural Land Use

Based on the current national legislation and on the data of the Croatian Agricultural Parcel Identification System (ARKOD) from 2017 [26], 25,736 ha of land may be considered for biosolids application in the study area (Figure 2). Further analysis of land use and parcel numbers per specific land use was carried out as shown in Table 3. According to this, the average parcel size is 0.65 ha. The largest area of agricultural land in the study area is occupied by karst pastures (16,871 ha), divided into 6820 parcels. In addition, olive groves are planted on 2662 ha, being divided into 14,404 parcels, giving an average parcel size of only 0.18 ha. The spatial distribution of parcels in relation to each land use category is shown in Figure 2. Obviously, such a pattern of parcel size and land use diversity may pose a serious constraint for the management of agricultural use of biosolids.

Figure 2. Agricultural land use pattern. Enlarged area presents agricultural parcels' spatial pattern in Petrovo polje.

Table 3. Utilized agricultural area by categories (ha) and number of parcels within the land use category (Šibenik region, Croatia).

Agricultural Land Categories	Land Area (in Hectares)	Number of Parcels
Arable land	1752	5108
Greenhouses	2	55
Meadows	2762	6695
Karst pastures	16,871	6820
Vineyards	951	3289
Abandoned vineyards	19	7
Olive groves	2662	14,404
Orchards	221	1074
Perennial crops	476	2358
Other	20	20
Total	25,736	39,830

3.2. Soil Properties

The basic statistics of the soil properties of the study area are summarized in Table 4. The soils are alkaline, with an average pH of 7.89. The average CaCO_3 content is 17.1% and the average soil organic matter content is 29.7 g kg^{-1} , ranging from 11 to 117 g kg^{-1} . The ranges of bioavailable nutrients concentrations were rather wide, which can be explained

by a number of interrelated factors, such as soil types' variability, including natural nutrient supply capacity, variation in land use and inconsistent fertilization practices.

Table 4. Basic statistics on soil properties, heavy metal concentrations, polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) (N = 68). Threshold values set by legislation are given.

Variable	Symbol	Unit	Descriptive Statistics							Threshold Value	
			Mean	Median	Std. Dev.	Min	Max	Skewness	Kurtosis	NN 71/19 ^a	EU ^b
pH	pH		7.89	7.89	0.25	7.23	8.33	−0.132	−0.63		
Electrical conductivity	EC	dS m ^{−1}	0.16	0.15	0.05	0.04	0.42	1.99	9.41		
Calcium carbonate	CaCO ₃	%	17.1	11.0	16.9	0.83	59.8	1.09	0.10		
Organic matter	OM	g kg ^{−1}	29.7	26.7	16.7	11.0	117	2.54	10.2		
Clay		%	32.4	35.0	10.8	6.00	51.0	−0.74	−0.02		
Available phosphorus	P ₂ O ₅	mg 100 g ^{−1}	30.4	6.25	79.4	0.05	618	6.22	44.7		
Available potassium	K ₂ O	mg 100 g ^{−1}	48.5	33.9	34.2	9.54	186	1.66	2.97		
Cadmium	Cd	mg kg ^{−1}	1.52	1.36	1.07	0.08	8.43	4.01	24.9	1,5	20–40
Chromium	Cr	mg kg ^{−1}	67.1	67.9	27.5	13.2	129	0.22	−0.46	100	/
Copper	Cu	mg kg ^{−1}	96.0	42.6	162	9.01	1165	4.75	27.8	100	1000–1750
Mercury	Hg	mg kg ^{−1}	0.10	0.06	0.18	0.00	1.39	6.14	43.1	1	16–25
Nickel	Ni	mg kg ^{−1}	46.6	44.9	20.3	11.1	126	1.20	2.97	70	300–400
Lead	Pb	mg kg ^{−1}	37.1	32.6	25.5	7.12	172	2.79	11.4	100	750–1200
Zinc	Zn	mg kg ^{−1}	90.0	88.5	33.0	24.7	208	0.60	1.38	200	2500–4000
Polycyclic aromatic hydrocarbon	PAH	mg kg ^{−1}	0.21	0.05	0.39	0.05	1.01	2.45	6.00	0.2	
Polychlorinated biphenyl	PCB	mg kg ^{−1}	0.10	0.10	0.00	0.10	0.10	1.37	−3.33	0.2	

^a NN (71/19)—Maximal permitted concentrations as defined by Croatian Government regulations (mg kg^{−1}); ^b EU Directive 86/278/EEC—Council Directive on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture.

The mean value of the total trace metal contents in the soils followed a descending order: Zn > Cu > Cr > Ni > Pb > Cd > Hg (Table 4). Of all the heavy metals examined in the study, only Cr and Zn showed normal distribution (skewness < 1). The mean value of Cd content was 1.52 mg kg^{−1} and ranged from 0.08 to 8.43 mg kg^{−1}. The phenomenon of anomalously high Cd concentrations in soils developed on limestone has been described previously [35,36]. The mean value of Cu content was 96 mg kg^{−1}, but the median value was more than twice lower, amounting to 42.6 mg kg^{−1}.

Considering that a median value can represent a baseline, the large number of samples with anomalous Cu contents was evident, caused by application of copper salts as fungicides in vineyards and orchards over decades [37]. The mean value of Cr content was 67.1 mg kg^{−1}, with a range from 13.2 to 129 mg kg^{−1}. In the Croatian Karst region, with soils developed from carbonate bedrock, the median value for Cr was 121 mg kg^{−1} [36]. Higher Cr content is an intrinsic property of heavier-textured soils, such as Terra Rossa's appearance in the study area. Median and average values of Ni content in the soils of the study area are very close at 44.9 and 46.6 mg kg^{−1}, respectively. The maximum Ni content determined in the study was 126 mg kg^{−1}. In current national legislation, the permitted limit for Ni in agricultural soils in Croatia is set at 50 mg kg^{−1} dry weight and the accumulation of this metal in soils receiving wastes such as sewage sludge can give rise to potential hazards for human, animal and plant health. The highest soil Pb concentration of 172 mg kg^{−1} was found near the military facility, and the accumulation of Pb in surrounding soils may be associated with ammunition use or disposal. The median value for Zn reported in this study is 90 mg kg^{−1}, which is lower than the baseline value for the

Coastal Croatia of 108 mg kg^{-1} . The Zn concentrations of the soils follow a normal distribution (skewness = 0.60). The interpolation maps for the studied metals are shown in Figure 3.

Figure 3. Interpolated maps of soil heavy metals.

3.3. Land Parcels Suitable for Biosolids Application

Based on the criteria set by legislation and the land use inventory, the land parcels suitable for biosolids application were identified (Table 5), and a total of 6032 ha of land was available. However, the land is divided into 26,233 parcels, which is another significant constraint to the sustainable biosolids management and use in agriculture in the study area, as shown in Figure 4.

Table 5. Agricultural land considered as potentially available for biosolids application.

Agricultural Land Use	Area (in Hectares)	Number of Parcels
Arable land	1752	5108
Vineyards	951	3289
Olives	2662	14,404
Orchards	221	1074
Combined permanent crops	476	2358
Total	6032	26,233

Figure 4. Agricultural land parcels suitable for biosolids application. The enlarged section shows the differences in size and spatial distribution of these parcels in Petrovo *polje*.

The next set of criteria applied in land suitability assessment was related to the potentially toxic elements and organic compounds in soils receiving biosolids. The limitation was found mainly regarding the concentration of Cd in soils that exceeds the highest permissible concentration (Table 4). Sites where the concentrations of heavy metals and organic residues in soil exceed the limits are excluded from further inventory.

4. Discussion

Efficient land use depends primarily on land availability and soil quality. The land use inventory undoubtedly plays an important role in comprehensive planning and can be used to distinguish spontaneous or natural drivers of land use change from those stimulated by public policies. Soil survey information is widely applied in Croatia. In recent years, the supply of suitability maps has increased, reflecting the spread of environmental and agricultural interest [38,39]. Every decision on the application of any measures in the environment related to soil quality and management, whether statutory regulations or practical actions, must be based on reliable and comparable data on the status of this part of the environment in the given area [40,41].

The priority target pollutants in biosolids used in agriculture may depend on several specific conditions, such as regional activities (socio-economic conditions and demography, industry, land use and climate), properties of the receiving soil, land geomorphology and hydrology [42]. Recent studies provided evidence for the accumulation of microplastics in agricultural soils over time after biosolids application [43,44]. Moreover, a new debate has been recently arisen in various research groups regarding the possible detection of SARS-CoV-2 virus in wastewater samples [45]. As the broad and fragmented EU legislative framework cannot in detail regulate specific regional conditions, new findings put pressure on policymakers to accustom their national legislation accordingly.

Recently opened, the re-evaluation of the EU Sewage Sludge Directive (86/278/EEC) is expected to make progress in addressing new contaminants in biosolids as well as the management options to reduce environmental harm [5]. Significant differences in quality and quantity of produced biosolids, as well as the possibilities for their use in agriculture, are evident even on the national level. In 2018, for Croatia, the total production of 19.23 thousand tons of dry SS was reported, of which 1.548 thousand tons were disposed on agricultural land [46]. In addition, a recent study [23] provided a comprehensive physico-chemical characterization of stabilised sewage sludge collected from different WWTPs in Croatia.

As such, considering the complexity of the soil system, the legal regulation of the maximum amount of biosolids that can be spread on land is certainly extremely demanding. The authors of [10] analysed the current legislation for the application of biosolids on agricultural land in Europe and found significant differences in many aspects of regulation among countries, including the maximum amount of biosolids spread on land, the soil where the use of biosolids is prohibited and the treatment requirements. Regulations that ensure the safe and responsible management of biosolids have been continuously upgrading. Nevertheless, it seems that the legislation can hardly follow the momentum of occurrence of new classes of water contaminants, or so-called contaminants of emerging concern [47].

Croatian regulation aligned to the EU Directive 86/278/ECC says that the agricultural use of sludge that could cause the permissible levels of heavy metals in the soil to be exceeded is prohibited, and that it is permitted to use a maximum of treated sewage sludge of $1.66 \text{ t}_{\text{DM}} \text{ ha}^{-1} \text{ year}^{-1}$, but it is vague about the frequency of biosolids application on agricultural land. Further restrictions refer to the characteristics of soil receiving biosolids, the composition of the sludge, the food crops produced and the SS treatment method. There is no specific legislation on the regional level in spite of the great regional differences between the continental and coastal parts of Croatia in terms of land availability and soil types, climatic conditions and types of land on which biosolids can be applied.

4.1. Major Challenges in the Coastal Karst Region

A karst environment is characterized by high complexity of relief forms, steep slopes, shallow stony soils and sparse natural vegetation; therefore, karst areas have been subjected to anthropogenic modifications to increase their agricultural potential [20]. A typical feature of Adriatic Dinaride karst is the phenomenon of karst fields (*polje*) [48], on the bottom of which the only arable soil of the karst area accumulates. *Polje's* fertile soils were developed

by fluvial sedimentation and by deposition of the eroded soil material from the karst hills within the catchment. Moreover, *polje* is characterized by periodic inundation and therefore in most of this area's land consolidation was carried out and drainage systems were installed. In the Šibenik region, one of the largest *poljes* of the Dinaric karst, Petrovo polje, is located (Figure 4). Petrovo *polje's* floor spreads on approximately 3200 hectares. From the geomorphological aspect, Petrovo *polje* is an alluvial plain characterized by a few inundation terraces and elevated cascades, which are not flooded regularly. As shown on the zoomed part in Figure 4, drainage, reclamation and land consolidation were done in the past on only 170 hectares, and these parcels are considerably greater than the average parcel size (0.65 ha). Small size parcels scattered across the region cannot be considered as suitable for sustainable biosolids application. Basic criteria in suitability assessment are based on legislation, but each region has to be carefully evaluated, taking into account specific features not covered by the law.

In the coastal region, characterized by semi-arid climate, the progressive depletion of soil organic matter is the major threat to the sustainability of the agricultural production system. The map of soil organic carbon content (SOC) of European soils, from which Croatia was unfortunately excluded, shows that Mediterranean regions of Europe exhibit distinctively smaller values of SOC than those of other regions, with substantial areas having very low OC ($\leq 1\%$) or low OC ($\leq 2\%$, [49]). Soil organic matter (SOM) comprises a wide range of pools, and indices of the quality and potential turnover rates of SOM pools add great value to the sole knowledge of the SOM content of a soil [50].

The decrease in SOM content in the soils of the Mediterranean region can be related to the number of drivers: current dynamic rural transformation and continuous and intensive depopulation, followed by high rate of land and farm abandonment [49]. Additionally, climate change affects disproportionately Croatian regions. In the coastal region, the intensity and duration of anomalous weather events, including extreme precipitation, droughts or high temperature occurrences, become more frequent in the past decades [37]. Dissected karst landscape makes the land prone to erosion by frequent torrential storms, particularly in vineyards and orchards, which promote SOM decrease. The highest annual soil loss rates are observed in the Mediterranean areas, and vineyards are among the land use types with the highest mean soil loss rate (about $9.5 \text{ t ha}^{-1} \text{ year}^{-1}$ vs. $2.5 \text{ t ha}^{-1} \text{ year}^{-1}$ on average for all land use types) [51].

Management practices that favor a slow decay rate of SOM increase C sequestration in soils, but on the other hand the supply of nutrients may be reduced. Therefore, the continuous supplying of carbon in the form of organic matter is needed. Animal waste produced on-farm, such as animal manure and slurries (including their derivatives) is becoming less and less available due to devoid of livestock production. Biosolids application is thus one of a broad range of practices that can be used either separately or combined with other measures for optimizing the SOM content of agricultural soils. As the cost of transportation of biosolids may be reduced due to proximity of wastewater treatment facility, peri-urban agriculture could benefit from these sources. In any case, potential risk in the food chain and the environment should be carefully assessed before biosolids are introduced into the soil.

4.2. Nutrient Value of Biosolids and Heavy Metals Concentrations

Biosolids are nutrient-rich organic material considered as a beneficial soil amendment. The availability of nutrients in the sludge depends though on the wastewater treatment process. Many of the trace metals in biosolids are also present in conventional fertilizers and manure. At the low concentrations and low rates these substances are nontoxic, and the environmental risk is low if good farming practices are. In general, nitrogen and phosphorus are the main nutrients that impact groundwater or surface water quality. Comparing with commercial fertilizers, nitrogen in biosolids is present in a slow-release organic form, which makes it less mobile.

Many studies nowadays address phosphorus recovery from wastewater solids, and besides the struvite precipitation of dissolved phosphate, new technologies have been developed to recover phosphorus from the particulate phase in wastewater solids [52]. The concentrations of phosphorus in stabilized sewage sludge from different Croatian wastewater plants (Table 6) ranged from 3.23–36.1 g kg^{−1}.

Table 6. Mean, range and variability of nutrient concentrations ^a in stabilized sewage sludge collected from different Croatian wastewater plants.

Nutrient	pH	Organic C (%)	Total N (%)	Total K ^b (g kg ^{−1})	Total P ^b (g kg ^{−1})
Mean	6.9	37.0	5.9	3.3	26.2
Range	6.6–7.2	25.8–44.5	3.3–7.7	1.7–4.6	20.7–36.1
Std. Dev.	0.2	7.7	1.8	1.2	6.27

^a Concentrations are on a dried solids basis; ^b *aqua regia* digestion.

With regards to the nutrient levels (Table 6), the biosolids are highly elevated in P contents, indicating the potential for being an alternative to P fertilisers. In many studies, researchers have shown that P concentrations in the soils increase with SS amendment [53–55]. A similar trend was also observed in soils amended with SS compost [56,57] or SS biochar [58]. However, repeated biosolids applications may pose a threat to the environment due to potential P release through leaching and the subsequent water eutrophication [59,60]. Nevertheless, our previous study found that the addition of SS-derived compost or biochar to soils did not increase the P mobility due to the high sorption in amended Terra Rossa and Rendzina substrates [61]. From an environmental perspective, the P-based SS application is safer for agricultural use than the dose based on the SOM which may potentially affect the migration of P to deeper soil layers, leading to the ground-water pollution [57]. Table 6 reports that biosolids are a good source of N as well, where aerobically stabilised SS contains more N than reported in the primary and anaerobic SS [23]. Among the macronutrients of agronomic importance, K is less abundant in SS (Table 6) and the long-term use of biosolids as a soil amendment can lead to a K deficiency, resulting in yield decrease [62]. Since SS is relatively deficient in K, the higher application levels are needed to ensure adequate K content for plant growth [63]. Therefore, other studies do confirm an increase in K levels in amended soils when using higher doses of SS compost [64,65] or SS biochar [56,66].

Besides the nutrient contents, the permissible uses of biosolids should be based on heavy metal concentrations (Table 7). The maximum allowable loadings for heavy metals per hectare of land per year are specified in Directive 86/278/EEC as well as the proposed limits based on their working document of sludge for the year 2025 [67].

Table 7. Mean, range, and variability of heavy metals concentration ^a in stabilized sewage sludge collected from different Croatian wastewater plants.

Heavy metal ^b	Cd	Cr	Cu	Hg	Ni	Pb	Zn
	mg kg ^{−1}						
Mean	0.9	55.3	275	1.3	30.7	46.0	1001
Range	0.7–1.3	22.9–109	156–660	0.4–1.9	17.6–52.6	11.8–72.1	801–1254
Std. Dev.	0.2	35.7	185	0.7	13.4	22.1	170
NN (38/08) ^c	5	500	600	5	80	500	2000
EU Directive 86/278/EEC ^d	20–40	/	1000–1750	16–25	300–400	750–1200	2500–4000

^a Concentrations are on a dried solids basis; ^b *aqua regia* digestion; ^c NN (38/08)—Croatian legislation on sewage sludge agricultural use; ^d EU Directive 86/278/EEC—Council Directive on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture.

Considering the metal levels in the studied soils (Table 4), which are one of the limiting factors for SS agricultural use, the results showed that high variability is present for all

metals of concern. From Table 4, it can be seen that average concentrations of Cd, Cr, Cu, Hg, Ni, Pb and Zn remained in accordance with the limit set by current Croatian legislation (Table 2). Further, the heavy metal concentration in soil may be higher than a limit value specified in Table 2. In such cases, the application of biosolids on land should be allowed after considering various additional aspects, such as the uptake of heavy metals by plants/crops, their intake rate by animals, the risk of groundwater contamination or the assessment of the long-term effects on ecosystem biodiversity. Obviously, the evaluation of such possibilities should be done for each specific area or case.

Higher heavy metals concentration, particularly of Cr and Ni, are an intrinsic property of heavier textured soils, although they may originate from anthropogenic sources, too. According to the WRB soil classification system [31] dominant soil types in the agricultural land of the study area are Rhodic Cambisols clayic, Chromic Cambisols loamic and Rendzic mollic Leptosols loamic. Croatian national classification system [68] recognizes these soils as Terra Rossa, Calcocambisol and Rendzina. The term Terra Rossa has been widely applied to red soils overlying limestone and dolomite as a discontinuous layer ranging in thickness from a few centimeters to several meters, particularly in the Mediterranean region, and the Croatian Adriatic Sea coast, as well [69]. Cr abundance in the Terra Rossa matrix is explained as addition from external sources. According to Miko et al. [69], high Cr concentrations in Terra Rossa, but also widely in the karst region, are due to the polygenetic nature of Terra Rossa (e.g., presence of boehmite). It is possible, therefore, that Cr and Ni could be enriched because of weathering of karst bauxite deposits, present all along the Coastal Croatia karst region [70]. Furthermore, the phenomena of anomalously high Cd concentrations in soils developed on carbonate bedrock have been recorded in many studies [71–73]. For soil Cd, which varied from 0.08 to 8.43 mg kg^{−1} d.w. (Table 4), its sorption increases with pH [74] which may be evident in compost-amended soils [64]. The soil organic matter (SOM) which increases with SS compost addition [57,74] was also found as one of the most important factors influencing the phytoavailability of Cd [75]. In the stabilised SS, Cd mobility is low since its major part is associated with Fe and Mn oxides and hydroxides [76]. However, the risk from Cd leaching was reported at neutral to alkaline pH after repeated applications of SS compost [77,78]. The biosolids amendment may possibly increase the mobility of Cd in soils through the formation of Cd-chloro-complexes [79] due to the presence of soluble chloride salts in SS [80]. Nevertheless, a reduction in extractable Cd in the soil was also revealed as a result of SS compost application [81].

Concerning Cr, being in the range from 13.2 to 129 mg kg^{−1} d.w. (Table 4), it was reported that in soils amended with composted SS, the Cr mobility increased under acid rain conditions [82]. In Croatia, aerobic or anaerobic stabilisation are the dominant procedures for sludge treatment, while stabilisation using lime is also a common technology, but it is used only by a few wastewater treatment plants. According to [83], caution should be advised when amending soil with lime-stabilised SS since liming of unstabilised SS can increase the pH to 12, thus promoting oxidation of trivalent Cr to the highly toxic hexavalent Cr.

Copper is one of the major toxic metals, and a highly reactive one, as well. Elevated levels of Cu in agricultural soils result from the use of Cu-containing compounds to control plant diseases and from application of manure or sewage sludge. These applications may lead to gradual accumulation of Cu in the soil and thereby increase Cu toxicity toward crops and beneficial microorganisms [37]. In the case of Cu levels which fall within limits from 9.01 to 1165 mg kg^{−1} (Table 4), [64] reported that higher doses of SS compost may reduce the plant available Cu due an increase in soil pH. Similarly, [84] revealed that higher Cu leaf accumulation was found in less-amended soils, while lower in more fertilised soil. At low pH, Cu solubility increases in amended soils until a threshold pH value is achieved (pH of 5.5), at which Cu precipitates [85,86]. In soils amended with SS biochar, the bioavailability of soil metals decreases due to rise of soil pH and by sorption to the biochar surface [87]. However, [66] found that Cu concentrations in the radish leaves increased with the application level. It was also pointed out, that in agricultural soils with a long history

of biosolids application the soluble Cu can be released through precipitation [74]. Most of the vineyards in the study area are planted on the hilly karst area on the steep slopes and shallow stony soils, managed in traditional way (Figure 5A), or on karst terraces reclaimed by stone crashing (Figure 5B). Only vineyards planted in karst *poljes* are considered as suitable for biosolids use, such as in Petrovo *polje*, shown on Figure 4.

(A)

(B)

Figure 5. Photographs of (A) Vineyards and olives plantations on karst steep slopes within stony walls (Location Primošten); (B) Grapevine and olives plantations on karst plateau reclaimed by stone crushing (Location Jadrtovac, Šibenik).

Even though Hg concentrations remained below the regulation limit (Table 4), the potentially elevated Hg levels in SS arising from possible Hg sources such as old paints and pigments, wood preservatives, mercury thermometers (nowadays forbidden in most EU countries), pesticides and glass mirrors [88] need to be carefully considered before SS is continuously used as an amendment of soil.

As regards the Ni varying from 11.1 to 126 mg kg⁻¹ d.w. (Table 4), ref. [89] revealed that lime-stabilised SS may pose environmental hazard since liming can enhance the solubility of Ni, making biosolids unsuitable for agricultural use. Interestingly, for soils with repeated applications of SS compost, it was found that the loss of Ni through leaching was low due to its immobilisation [74].

Regarding Pb, the minimal risk is associated with low mobility of Pb in biosolids attributed to its higher fraction adsorbed to the silicate lattice or crystalline Fe and Mn oxides [76]. For instance, [84] found that transfer of Pb to edible parts of Chinese cabbage (*Brassica rapa* L. subsp. *pekinensis* (Lour.)) was limited when amending soil with SS-derived compost or biochar. However, [82] observed that the application of SS compost to a flower garden and abandoned phosphate mine soils enhance the mobility of Pb.

An increased mobility of Zn in biosolids [76] means that health hazard over its phytoaccumulation should be taken into account when using SS as a fertiliser for soil [84,90,91]. According to [91], amendment rates higher than 150 tonnes/ha of SS compost should not be used, in order to prevent the potential Zn loading in the surface soil. In soils treated with SS compost, the high preference of Zn for sorption on the Fe and Mn oxides and hydroxides was reported at high pH [90]. In earlier study, a reduction of Zn concentrations in lettuce (*Lactuca sativa* L., “Tanya”) compared to the control was documented at increased pH for the same agronomic practice [92]. In the case of SS biochar, [93] observed that the amounts of DTPA-extractable Zn increased in amended soils. In a similar study [94], an increase in the bioavailable fraction of Zn was found, while [21] revealed that the risk of Zn leaching is low.

4.3. Karst Hydrology and Environmental Risk

Increasing water demand and urbanisation in the coastal regions of the Mediterranean are already exerting pressure on coastal and alluvial aquifers, which are being degraded by over-pumping [95], groundwater pollution, alluvial and marine sediment quality [96,97], seawater intrusion [98], and intense and continuing interactions with various types of human activities [99]. Under such conditions, it is necessary to determine and understand the extent of all anthropogenic impacts on the natural and cultivated karst environment prior to the application of biosolids.

Because soils vary across the landscape, each soil type contains unique trace element concentrations based on its parent material and other soil-forming factors that may have added or removed these elements from the soil. High background concentrations of trace elements characteristic of soils developed on limestone could result in mobilization and release to surface and subsurface waters and subsequent incorporation into the food chain. Soil properties such as organic matter, type and amount of clay, pH, and cation exchange capacity (CEC) affect the quantity of trace elements available for mobilization and release or sorption in a soil.

In karst catchments, groundwater and surface water are hydraulically connected by numerous karst features, which can increase the risk of contaminant transport from soil to groundwater, surface water, and lacustrine sediments or even coastal marine sediments. Fluvial systems can be strongly influenced by human activities and can act as a carrier and/or receptor of contaminants [96,97,100].

Specific operational methods can be proposed to ensure the proper application of biosolids to protect the environment and human health in the Mediterranean karst region by defining the available land and land use, soil properties, possible geomorphological constraints (slope, rock outcrops and fractures, stoniness and underground karst formations, groundwater depth), as well as biosolids characteristics, application rates and placement

methods. Leaching of nutrients and potentially toxic substances is often cited as a major problem in the proper application of sewage sludge. In [100], it is stated that mismanagement of biosolids land application can lead to leaching of $\text{NO}_3\text{-N}$ into groundwater and accumulation of P in surface soil, which can increase the risk of runoff/erosion loss of P and cause eutrophication [100].

One of the major concerns regarding biosolids land application is the accumulation of pollutants (including heavy metals, pathogens and organic pollutants) in the soil. The behavior, mobility and bioavailability of heavy metals in soil are controlled by a number of complex physicochemical processes [101]. The mobility of metals can be greatly reduced in calcareous Mediterranean soils due to high soil pH. Soil pH is a key variable in the redistribution of heavy metal distribution in biosolid-applied soils. In addition, low rainfall and high evapotranspiration can lead to metal accumulation in the first few centimeters of soil [102]. OM also plays an important role in the retention of heavy metals in the soil surface horizon [103]. The ability of soils to adsorb metal ions from aqueous solution is of particular interest and has implications for both agricultural issues such as soil fertility and environmental issues such as remediation of polluted soils and waste disposal [104]. The metal-soil interaction is such that when metals are introduced at the soil surface, their mobilization does not occur on a large scale unless the metal retention capacity of the soil is overloaded or metal interaction with the associated waste matrix enhances mobility.

4.4. Legislation: National or/and Regional Requirements

At the EU level, national laws have been created to implement EU directives and other related acts. Although information sources agree on the significant potential of biosolids recycling, one of the main barriers identified in biosolids land application is the lack of harmonization in the national regulatory framework. The legislation on sewage sludge, biosolids and their final use possibilities have been continuously changing and modifying. At the same time, management of growing quantities of sewage sludge collected on wastewater purification plants are becoming an acute issue of sustainability. In EU-28, about 50% of sewage sludge is spread on agricultural soils, and about 8% in Croatia [105]. However, there is a gap in reporting all the options in accommodating sewage sludge. Additionally, there are important divergences among the different regulations in Croatia, primarily regarding the permitted uses. For example, in 2016, disposal of sewage sludge in landfills for final disposal was prohibited [17]. Furthermore, in 2019, Ordinance on agricultural soil protection [106] put additional restrictions on agricultural use of biosolids, which calls for the control of legislative compliance. Therefore, the other governance instruments are needed for the clear division and delegation of responsibilities and competences in the administrative set-up for sludge management. Additionally, stakeholders should be provided with information on land potential before soliciting input on desired land use.

5. Conclusions and Perspectives

In this study, the potential of agricultural land in the coastal Adriatic Karst region (Šibenik region, Croatia) for biosolids application was evaluated by integrating spatial data from different sources: digital maps and remote sensing, parcel identification system, GIS field observations and measurements focusing on specific land, soil and biosolids properties.

Spatial analysis of the land resources identified only 6065 ha of 25,736 ha of agricultural land suitable for biosolids use. However, suitable land is divided into 26,288 parcels, dominantly occupied by olive orchards.

Furthermore, concentrations of certain heavy metals in soils in the Dinaric karst, in particular Cd, Cr and Ni, are inherently higher of the average for soils in general. Therefore, each load of material rich in heavy metals may lead to their accumulation in soil in concentrations that are harmful for the environment and humans.

pH is the key soil property that control the uptake of available elements into plants. High pH of soils in the study area, which is 7.89 in average, considerably reduces the bioavailability of potentially toxic elements. However, it is well known that the long-term

application of biosolids produced from sewage sludge may decrease soil pH, and thus enhance the uptake of toxic elements into plants.

Continuous increasing of the sewage sludge production imposes needs for diversification of sludge treatments and recycling methods. In case of the studied area, limited resources of land suitable for biosolids use were identified. Furthermore, the small and spatially scattered parcels considerably reduce the feasibility of biosolids land application.

Common international and national environmental standards for biosolids have largely been developed to date, but specific regional land features should certainly be considered. Basic criteria for assessing suitability are based on legislation, but each region must be carefully evaluated, taking into account specific characteristics not covered by legislation.

Comparatively small landscape units and catchments may seem marginal on a global scale in terms of impact on the aquatic environment. However, when dealing with the fractured karst geomorphology and aquifers, coastal floodplains and wetlands, and inland waters that rapidly communicate with seawater, such impacts quickly spread to the Adriatic Sea and beyond to the entire Mediterranean Sea. Therefore, regional studies limited to the specific and often small coastal regions, including islands, such as this one, can provide valuable input for the development of biosolids management systems on a regional scale.

The main challenges in biosolids land application in the Mediterranean karst region are related to the protection of the environment. Disposal on agricultural land was identified as a marginal option in this study, mainly due to the low availability of agricultural land and the small plots scattered throughout the karst area. Anyway, there are many other practical options, such as improving forest soil quality, reforestation and forest fires reclamations activities, maintenance of tourist areas and recreation zones, and many others. In any case, an assessment of both short-term and long-term impacts of biosolids amendment is recommended before their application to land in karst regions. In this study the spatial aspects of the problem were analyzed and to land suitable for biosolids application first was identified, while respecting the limits set by legislation.

The next recommendation is that the regional approaches in biosolids management in the Mediterranean karst region must be applied based on the uniqueness of landforms, hydro-geomorphology and climatic conditions, soil types and land use. Surface roughness, steep slopes and a complex fracture matrix system create complex pathways and velocities for solute migration. Selecting optimal activities for biosolids land application can reduce the risk of pollution distribution through heterogeneous groundwater flow.

Local and regional authorities should be provided with science-based information to identify the policies and methods for diversification of SS and biosolids management. The methodology for evaluating land potential for biosolids application combines data from different sources:

- qualitative and quantitative control of biosolids based on current legislation;
- environmental monitoring programs, mapping of land resources, land use and vulnerable areas, and the development of GIS-based models.

Ultimately, the results could help decision makers, managers of local wastewater treatment facilities, and landowners set realistic goals for efficient and sustainable application of biosolids on agricultural lands to ensure food security and land and water degradation neutrality in a fragile karst landscape.

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