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Estimation of energy and emission properties of waste from medicinal and aromatic plants

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This study evaluates the feasibility of utilizing post-extraction waste from medicinal and aromatic plants - lavender, thyme, yarrow, wormwood, sandy everlasting, oregano, costmary, tarragon, sage, and hyssop - as a feedstock for bioenergy production. Key fuel properties of the residual biomass were assessed, including heat of combustion, higher heating value (HHV), ash content, volatile matter, and moisture content. Elemental analysis was performed to determine the concentrations of carbon, hydrogen, nitrogen, and sulfur. The results demonstrate that these residues possess high potential as solid biofuels, characterized by favorable composition: high carbon (45.4–49.8%) and hydrogen (3.9–8.2%) contents, low ash (6.21–18.42%), nitrogen (1.78–3.16%), chlorine (0.022–0.586%), and sulfur (0.033–0.294%) contents, and high HHV (16.89–19.23 MJ/kg). Compared to coal, combustion of this biomass could significantly reduce emissions, with estimated reductions of up to 32% for CO and 31%CO_2, 40% for NO_x, 99% for SO_2, and 67% for particulate matter, depending on the specific biomass type.

Keywords: medicinal and aromatic plant waste, biofuel, elemental analysis, emission factors

INTRODUCTION

In recent years, alternative energy sources characterized by low emissions, including greenhouse gases and dust particles, have been increasingly sought after [1, 2]. Waste plays a crucial role in the European Commission's strategy for energy security and greenhouse gas (GHG) emission reduction. As a renewable energy source, the use of waste biomass has been expanding worldwide, primarily for household heating as an alternative to fossil fuels [3, 4]. The demand for plants with high productivity, low nutrient requirements, and valuable biological composition for various bioeconomic applications is of utmost importance. Perennial plants, in particular, offer advantages by reducing production costs and energy inputs compared to annual field preparation and planting. Additionally, such a biomass production system can enhance ecosystem services and support soil conservation, aligning with the concept of "low energy consumption and high output" regarding energy investments and additional costs. Essential oil crops are primarily cultivated for their essential oils [5-7]. The area dedicated to essential oil production in the European Union is steadily increasing, reaching approximately 80,000 hectares [8]. In recent years, global trade in medicinal and aromatic plants has grown by 10–12% annually [9]. Consequently, waste generation in this industry has also increased, reaching up to 30 million tons annually [10]. A substantial amount of waste biomass is generated during harvesting, pre-processing, drying, collection of harvested crops, and plant feedstock processing. After essential oil extraction, solid residue remains, which can be utilized for fuel energy production through pyrolysis, gasification, or hydrothermal carbonization processes [11]. Despite containing valuable substances, these residues are often discarded through stockpiling, landfilling, or open-field burning, leading to resource waste and significant environmental pollution [10, 12]. Despite the strong economic potential of utilizing plant biomass waste, limited research has focused on the calorific value of these residues [8, 13-14].

This study aims to evaluate the energy potential of residues obtained after steam distillation of essential oils from lavender, thyme, yarrow, wormwood, sandy everlasting, oregano, costmary, tarragon, sage, and hyssop. Additionally, it examines the emission factors of toxic exhaust components and explores the feasibility of using these residues for energy production

EXPERIMENTAL

Aromatic plants, including lavender, thyme, yarrow, wormwood, sandy everlasting, oregano, costmary, tarragon, sage, and hyssop, were collected from southern Bulgaria at the flowering stage. After

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essential oil extraction *via* steam distillation, the plant residues were dried in an oven at 105°C and ground using a laboratory grinder. The physicochemical characterization of the plant residues was conducted based on calorific value, ultimate analysis, and proximate analysis.

The heating value was determined following the ISO standard (BDS EN ISO 18125) [15] using an IKA C6000 oxygen bomb calorimeter (IKA Werke GmbH, Germany).

The samples were analyzed according to standard methods: moisture content [16], ash content [17], volatile matter [18].

Total carbon, hydrogen, nitrogen, and sulfur contents were determined by dry combustion using a Vario Macro CHNS analyzer (Elementar GmbH, Germany) [19].

Using the results from the ultimate analysis, emission factors for CO, CO_2, NO_x, SO_2, and dust emissions were estimated through equations (1) – (7) [20]:

CO emission factor

$$\mathrm{CO} = \frac{28}{12} x\ \mathrm{Ec}\ x\ (C\_CO/C) \qquad (1)$$

where: CO – carbon monoxide emission factor (kg/kg); 28/12 – molar mass ratio of carbon monoxide to carbon; Ec – emission factor of chemically pure coal (kg/kg); C_CO/C – proportion of carbon emitted as CO (for biomass: 0.06).

Emission factor of chemically pure coal

$$Ec = c.uc \qquad (2)$$

where: c – carbon content in biomass (kg/kg); uc – proportion of carbon oxidized during combustion (for biomass: 0.88).

CO_2 emission factor

$$\mathrm{CO_2} = \frac{44}{12} x \left(Ec - \frac{12}{28}\ x\ CO - \frac{12}{16}\ x\mathrm{ECH_4} - \frac{26.4}{31.4}\ x\ ENMVOC\right) \qquad (3)$$

where: CO_2 – carbon dioxide emission factor (kg/kg); 44/12 – molar mass ratio of carbon dioxide to pure coal; 12/28 – molar mass ratio of carbon to carbon monoxide; 12/16 – molar mass ratio of carbon to methane; ECH_4 – methane emission factor; ENMVOC – non-methane VOC emission index (for biomass: 0.009).

Methane emission factor

$$\mathrm{ECH_4} = \frac{16}{12} x\mathrm{Ec} \times (\mathrm{C\_CH4/C}) \qquad (4)$$

where: ECH_4 – methane emission factor (kg/kg);16/12 – molar mass ratio of methane to carbon; C_CH_4/C – proportion of carbon emitted as CH_4 (for biomass: 0.005).

NOx emission factor

$$NOx = \frac{46}{14} \times Ec \times N/C \times (N\_NOx/N) \qquad (5)$$

where: NO_x – emission factor for nitrogen oxides (kg/kg); 46/14 – molar mass ratio of nitrogen dioxide to nitrogen (NO in air oxidizes to NO_2); N/C – nitrogen to carbon ratio in biomass; N_NOx/N – proportion of nitrogen released as NO_x (for biomass: 0.122).

Emission factor of SO_2

$$\mathrm{SO_2} = \frac{2S}{100} \qquad (6)$$

where: SO_2 – sulfur dioxide emission factor (kg/kg); 2 – molar mass ratio of SO_2 to sulfur; S – sulfur content in the fuel (%).

Dust emissions

$$Edust = 1{,}5 \times A \times \frac{100 - \eta 0}{100 - k} \qquad (7)$$

where: Edust – dust emission factor (kg/Mg); 1.5 – coefficient denoting 15% of ash released as volatile dust; A – ash content in the fuel (%); η_0 – dust removal efficiency (for biomass: 20%); k – combustible component content in the dust (for biomass: 5%).

RESULTS AND DISCUSSION

Proximate and ultimate analysis

The proximate and ultimate analysis results for essential oil plant waste are presented in Tables 1 and 2. According to CEN/TS 14961 [21], biomass for solid biofuels is categorized into three main groups: woody biomass, herbaceous biomass, and fruit biomass. Essential oil plant waste falls under the herbaceous biomass group - specifically, agricultural and garden herbs. The key parameters of biomass waste were compared against the requirements of ISO 17225-6 standard [22] for herbaceous biomass and data from other studies.

Heating value and energy potential: The lower heating value (LHV) is a key parameter for assessing the suitability of biomass as a biofuel, as it reflects the usable energy released during combustion, excluding the latent heat of vaporization of water. According to ISO 17225-6 [22], the minimum acceptable LHV for solid biofuels is 14.5 MJ/kg. The LHV of the tested plant wastes ranged from 15.39 MJ/kg (costmary) to 17.86 MJ/kg (wormwood), indicating that all samples meet or exceed the standard requirement and possess good energy potential.

Table 1. Proximate analysis and heating values of plant waste

Plant waste	M,%	Ash, %	FC, %	VM, %	LHV, MJ/kg	HHV, MJ/kg
Lavender	8.42± 0.08	7.28± 0.03	11.50± 0.10	80.08± 0.21	17.41± 0.01	19.23± 0.02
Wormwood	6.26± 0.04	6.21± 0..03	12.72± 0.11	81.02± 0.10	17.86± 0.04	19.21± 0.05
Sandy everlasting	6.12± 0.06	8.44± 0.15	15.08± 0.15	78.80± 0.40	17.05± 0.01	18.31± 0.01
Oregano	7.48± 0.24	9.00± 0.05	17.82± 0.18	74.70± 0.30	16.76± 0.01	18.30± 0.04
Costmary	7.80± 0.40	18.42± 0.12	17.50± 0.18	74.70± 0.10	15.39± 0.03	16.89± 0.01
Tarragon	7.53± 0.03	12.88± 0.05	17.94± 0.20	74.53± 0.08	16.13± 0.01	17.63± 0.01
Sage	8.23± 0.10	14.57± 0.09	17.03± 0.15	74.74± 0.15	16.42± 0.01	18.10± 0.01
Hyssop	6.86± 0.08	7.20± 0.04	13.34± 0.10	79.80± 0.50	17.31± 0.05	18.75± 0.06
ISO 17225-6	≤12 -15	≤ 6-10	-	-	≥14.5	-
CEN/TS 14961-1 Harvest (July – Oct)	-	6.5 (2.5-10)	-	-	16.6	-

M - moisture, FC - fixed carbon, VM - volatile matter, LHV - lower heating value, HHV- higher heating value

Table 2. Ultimate analysis of tested wastes

Waste	N, %	S, %	Cl, %	C, %	H, %	O, %
Lavender	1.78± 0.01	0.033± 0.005	0.022± 0.001	49.3± 0.2	5.4± 0.2	36.20± 0.10
Wormwood	2.98± 0.12	0.096± 0.012	0.172± 0.008	49.3± 0.4	8.2± 0.5	33.21± 0.10
Sandy everlasting	1.58± 0.02	0.120± 0.002	0.443± 0.002	48.4± 0.3	4.8± 0.7	36.66± 0.12
Oregano	1.67± 0.03	0.062± 0.001	0.115± 0.004	48.6± 0.1	3.9± 0.7	36.70± 0.11
Costmary	2.57± 0.12	0.294± 0.005	0.254± 0.001	45.4± 0.4	5.2± 0.7	28.12± 0.10
Tarragon	3.16± 0.01	0.181± 0.003	0.586± 0.002	46.7± 0.3	8.1± 0.7	28.98± 0.10
Sage	2.46± 0.01	0.152± 0.002	0.043± 0.002	48.3± 0.5	5.6± 0.7	28.92± 0.10
Hyssop	1.80± 0.03	0.278± 0.001	0.054± 0.002	49.8± 0.4	5.8± 0.7	35.12± 0.11
ISO 17225-6	1.5-2	0.2- 0.3	0.1-0.3	-	-	-
CEN/TS 14961-1 Harvest (July – Oct)	1.3	0.1 (0.1-0.2)	0.5 (0.2-0.6)	46	5.7	40

The higher heating value (HHV), which represents the total energy content of the biomass including the energy from the condensation of water vapor, ranged from 16.89 MJ/kg (costmary) to 19.21 MJ/kg (wormwood) and 19.23 MJ/kg (lavender). Lavender and wormwood wastes demonstrated the highest HHVs (19.21–19.23 MJ/kg), making them excellent candidates for biofuel applications. In contrast, the lower HHV of costmary suggests reduced energy efficiency. These findings are consistent with previously reported values for lavender (19.2 MJ/kg) [14] and thyme waste (17.8 MJ/kg) [14], mint waste (15.90–16.64 MJ/kg) [8], miscanthus (17.99 MJ/kg), wood (18.35 MJ/kg), and rapeseed straw (15.97 MJ/kg) [23].

Moisture content and ash content: Moisture content significantly influences combustion efficiency, as high moisture reduces the net energy output by requiring additional energy to evaporate water during combustion. The moisture content of the tested biomass ranged from 6.12% (sandy everlasting waste) to 8.23% (sage waste), well within the ISO 17225-6 standard limits of 12–15% [22]. These low moisture levels contribute positively to combustion performance.

Ash content is another critical factor in determining biomass fuel quality. Excessive ash can lower combustion efficiency, increase particulate emissions, and cause operational issues such as slagging and fouling in combustion systems. In this study, ash content varied from 6.21% (wormwood) to 18.42% (costmary). The ash content in wormwood, sandy everlasting, oregano, and hyssop wastes falls within the acceptable ISO 17225-6 range (≤6–10%) [22]. Moisture and ash values for lavender, wormwood, sandy everlasting, oregano, and hyssop meet standard criteria, supporting their suitability as solid biofuels. These results align with earlier findings for lavender waste (6.7–7.8%) [13] and mint waste (7.23–10.29%) [8].

Fixed carbon and volatile matter: Fixed carbon content is a desirable property in biomass fuels, as it significantly influences combustion stability and duration of burning. Although ISO 17225-6 [22] does not define standard limits for fixed carbon, values in this study ranged from 11.5% (lavender waste) to 17.82% (oregano waste) and 17.94% (tarragon waste). The higher fixed carbon content in oregano and tarragon suggests superior combustion stability. Similar fixed carbon values have been reported for mint waste (9.40–15.77%) [8] and lavender waste (15.3%) [13].

Volatile matter is another critical factor in biomass combustion. A higher volatile matter content typically results in easier ignition and more intense combustion. In the current study, volatile matter ranged from 74.53% (tarragon waste) to 81.02% (wormwood waste), aligning with values reported for mint waste (64.99–70.36%) [8], Miscanthus (72.5%), and rapeseed straw (73.5%) [23]. The balance between fixed carbon and volatile matter suggests that the tested plant wastes possess stable combustion characteristics, making them suitable for energy applications.

Table 2 presents the ultimate analysis results of the tested plant wastes, including carbon (C), hydrogen (H), oxygen (O), nitrogen (N), and sulfur (S) content. These values are compared to ISO 17225-6 requirements [22] and to results from related studies.

Carbon and hydrogen are key energy-contributing elements in biomass. During combustion, these elements undergo exothermic oxidation reactions, producing CO_2 and H_2O, thereby determining the gross calorific value of the fuel [24]. The total carbon content in the tested samples ranged from 45.4% to 49.8%, with hyssop waste having the highest concentration. These values are consistent with previous findings for mint waste (44.82–47.05%) [8], lavender (45.4–48.1%) [13], willow biomass (50.84%) [25], and wood (49.80%) [26].

Hydrogen content ranged from 3.9% (oregano waste) to 8.2% (wormwood waste). This range is comparable to that reported for mint waste (5.54–5.76%) [8], lavender waste (5.8–6.77%) [13], willow biomass (5.86%) [25], and wood (6.30%) [26]. These values also closely match the hydrogen content found in cereal and forage straws, such as wheat, barley, flax, and timothy (6.1–6.4%).

Oxygen content, on the other hand, tends to negatively influence the energy value of biomass. In the tested samples, oxygen content ranged from 28.12% (costmary waste) to 36.70% (oregano waste), which is in line with values observed in mint waste (29.14–36.09%) [8] and lavender waste (37.8%) [13].

The elemental composition of biomass is crucial for estimating heating values, combustion air requirements, and composition of flue gases [27]. It also plays a key role in assessing the environmental impact of biomass combustion. Nitrogen and sulfur, in particular, are undesirable in biomass fuels due to their role in generating harmful emissions. During combustion, nitrogen contributes to nitrogen oxides (NO_x) formation, while sulfur leads to sulfur oxides (SO_x) emissions, which are associated with particulate pollution, acid rain, and equipment corrosion [24].

Nitrogen content in the tested biomass ranged from 1.78% (lavender waste) to 3.16% (tarragon waste). Only the wastes of sandy everlasting, oregano, lavender, and hyssop had nitrogen levels below the 1.5–2.0% threshold set by ISO 17225-6 [22], making them more suitable for combustion from an environmental standpoint. For comparison, nitrogen content in mint waste has been reported in the range of 0.23–0.70% [8], and in lavender at approximately 1.3% [13].

Sulfur content in biomass is generally low but still influences emission profiles. The tested samples exhibited sulfur levels ranging from 0.033% (lavender waste) to 0.294% (costmary waste), remaining within the acceptable limits (0.2–0.3%). These values are consistent with reported sulfur contents in lavender waste (0.1%) [13] and mint waste (0.0–0.19%) [8]. A low sulfur content is advantageous, as it minimizes SO_2 emissions and reduces environmental risks.

Chlorine content in the tested plant wastes ranged from 0.022% in lavender waste to 0.586% in tarragon waste, remaining within the acceptable limits for solid biofuels. A lower chlorine concentration is advantageous, as it decreases the risk of corrosion in combustion systems and limits the formation of harmful emissions, such as hydrogen chloride and dioxins.

Emission factors

Determining emission factors is essential for estimating pollutant levels released during fuel combustion. These factors depend on the physicochemical characteristics of the biomass used. Table 3 presents the emission factors for carbon monoxide (CO), carbon dioxide (CO_2), nitrogen oxides (NO_x), sulfur dioxide (SO_2), and particulate matter (dust) for the investigated plant wastes, with coal data provided for comparison.

Significant differences were observed in CO emissions among the plant wastes. The highest CO emission rate was recorded for lavender and wormwood waste (60.74 kg/Mg), while sage waste had the lowest (55.44 kg/Mg), reflecting a variation of up to 5%.

NO_x emissions ranged from 2.44 kg/Mg for tarragon waste to 11.15 kg/Mg for hyssop waste. Compared to hard coal, only tarragon and sage waste exhibited lower NO_x emission levels.

The CO_2 emission factor was highest for lavender and wormwood waste (1487.07 kg/Mg) and lowest for sage waste (1357.34 kg/Mg).

SO_2 emissions ranged from 0.067 kg/Mg for lavender waste to 0.588 kg/Mg for costmary waste. Dust emissions were highest for costmary waste (23.27 kg/Mg) and lowest for tarragon waste (7.83 kg/Mg).

The emission factors observed are comparable to those reported for other plant-based materials such as mint waste [8], *Eucalyptus globulus* [28], and larch needles [29]. Overall, the studied plant wastes exhibit emission profiles similar to those of various biomass sources, with particularly low SO_2 emissions. CO and CO_2 emission levels are close to those reported for mint [8], larch needles [29], and hazelnut husk and leaves [29–30]. In terms of NO_x, the studied wastes show significantly lower emissions compared to straw pellets, sunflower stalks, corn stalks, and wood pellets [31], as well as *Eucalyptus globulus* [28].

Wastes from hyssop and costmary showed elevated dust emissions, comparable to those from mint (9.14–13.0%) [32], tree leaves (10.80%) [29] and hazelnut waste (10.95%) [30]. High particulate emissions are attributed to the elevated ash content in these materials. Therefore, technological measures such as installing additional dust filters should be considered to mitigate particulate emissions.

Table 3. Emission factors (kg/Mg) for analyzed wastes and coal

Waste	CO	CO_2	NO_x	SO_2	Edust
Lavender	60.74± 0.30	1487.07± 7.33	6.29± 0.10	0.067± 0.005	9.20± 0.10
Wormwood	60.74± 0.30	1487.07± 7.33	10.51± 0.18	0.192± 0.015	7.84± 0.10
Sandy everlasting	59.63± 0.29	1459.92± 7.08	5.57± 0.09	0.240± 0.015	10.66± 0.15
Oregano	59.88± 0.28	1465.95± 7.08	5.89± 0.09	0.124± 0.010	11.37± 0.15
Costmary	55.93± 0.27	1369.41± 6.55	9.07± 0.19	0.588± 0.050	23.27± 0.30
Tarragon	57.29± 0.28	1402.60± 6.78	2.44± 0.06	0.073± 0.008	7.83± 0.10
Sage	55.44± 0.27	1357.34± 6.50	3.28± 0.06	0.284± 0.040	11.79± 0.10
Hyssop	57.53± 0.28	1408.63± 6.78	11.15± 0.25	0.362± 0.040	16.27± 0.15
Hard coal	82.01± 0.64	1969.00± 15.15	4.09± 0.08	5.200± 0.100	23.57± 0.50

The emission assessment method used in this study provides an effective means of evaluating the environmental impact of plant waste for energy production without the need for advanced analytical equipment. This approach offers a practical tool for rapid estimation of biofuel emissivity, an aspect often overlooked in biomass suitability assessments.

Differences in emission factors among the tested wastes may influence their selection for energy use, especially when environmental performance and combustion efficiency are priorities. Among the tested materials, tarragon waste demonstrated the lowest overall emissions, while lavender and wormwood waste exhibited the highest CO and CO_2 emissions. Therefore, utilizing tarragon waste for energy purposes may offer advantages in reducing greenhouse gas (GHG) emissions.

Environmental impact of essential oil crop waste combustion

The use of waste from essential oil crops as biofuel results in substantial emission reductions compared to coal combustion [20]. Depending on the type of waste, emission levels were reduced by: 25.9–32.4% for carbon monoxide (CO); 24.4–31.1% for carbon dioxide (CO_2);19.8–40.2% for nitrogen oxides (NO_x); 88.7–98.7% for sulfur dioxide (SO_2); and 66.8% for particulate matter (dust).

These reductions confirm the significant environmental benefits of utilizing such biomass wastes as alternative fuels.

CONCLUSIONS

The study revealed significant differences in the calorific values and emission characteristics of the tested plant wastes. Lavender (17.41 MJ/kg; 19.23 MJ/kg), wormwood (17.86 MJ/kg; 19.21 MJ/kg), hyssop (17.31 MJ/kg; 18.75 MJ/kg), sandy everlasting (17.05 MJ/kg; 18.31 MJ/kg), oregano (16.76 MJ/kg; 18.30 MJ/kg), and sage waste (16.42 MJ/kg; 18.10 MJ/kg) exhibited relatively high lower and higher heating values, indicating good potential as biofuels. In contrast, costmary (15.39 MJ/kg; 16.89 MJ/kg) and tarragon (16.13 MJ/kg; 17.63 MJ/kg) wastes showed lower energy content.

The chemical composition of the plant materials significantly influences both energy potential and emission factors. For instance, costmary waste had the highest ash content (18.42%), which adversely affected its calorific value.

Emission factors varied depending on the waste type. Wormwood and lavender waste produced the highest emissions of CO (60.74 kg/Mg) and CO_2 (1487.07 kg/Mg), while hyssop waste showed the highest NO_x emissions (11.15 kg/Mg). Costmary waste generated the most particulate emissions (23.27 kg/Mg), likely due to its high ash content.

When selecting appropriate biomass for energy production, it is essential to consider both energy efficiency and environmental impact. If maximizing energy output is the primary objective, lavender, wormwood, hyssop, sandy everlasting, and oregano waste are suitable candidates. However, if minimizing environmental impact is prioritized, tarragon waste is the preferable choice due to its low emissions profile.

Ultimately, the selection of biomass for energy use should be guided by specific local contexts, including regulatory frameworks, environmental standards, and energy demands.

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