

QUANTIFICATION AND MATERIAL CHARACTERIZATION OF ACCUMULATED RESIDUES AS A BIOFUEL IN MOIST TEMPERATE FORESTS OF PAKISTAN

SANIA KHAN¹, MUHAMMAD IRFAN ASHRAF^{2*}, MUHAMMAD UMAIR³, MUHAMMAD ATIF MAJEED⁴, OYTUN EMRE SAKICI⁵, MUHAMMAD ARIF GOHEER⁶ AND SAMINA JAHANDAD⁷

¹Department of Forestry & Range Management, PMAS Arid Agriculture University, Rawalpindi, Pakistan

²Department of Forestry, Faculty of Agriculture, University of Sargodha, Punjab, Pakistan

³Department of Energy Systems Engineering, PMAS Arid Agriculture University, Rawalpindi, Pakistan

⁴Pakistan Forest Institute, Peshawar, Pakistan

⁵Faculty of Forestry, Kastamonu University, Kastamonu, Turkiye

⁶Global Climate-Change Impact Studies Centre (GCISC) Islamabad, Ministry of Climate Change & Environmental Coordination (MoCC & EC), Government of Pakistan

⁷Hydrocarbon Development Institute of Pakistan, Islamabad, Ministry of Energy, Government of Pakistan

*Corresponding author's drirfanCanada@gmail.com; mirfan.ashraf@uos.edu.pk

Abstract

Energy security had become a serious global concern after the COVID-19 pandemic. The Russian-Ukraine war has further aggravated this issue by disturbing fuel supplies all over the world. This has created the scarcity of fuels leading to a drastic price hike in the international markets resulting in a critical setback to the economies of the developing countries. Under the current scenario, identification and development of alternate and renewable energy resources are exceedingly essential. Biomass is an important renewable energy resource that is also environmentally friendly. In the present study, material characterization of accumulated residues as a biofuel was conducted in the moist temperate forests of Pakistan. The residues associated with tree species i.e., *Abies pindrow* (Fir), *Cedrus deodara* (Deodar) and *Pinus wallichiana* (Blue Pine) were investigated. The collected residue samples were subjected to proximate, ultimate, and physical analyses according to conventional protocols. These include estimation of calorific value, ash content, volatile matter, carbon and sulfur content, and bulk density. Among three types of forest residues (species), blue pine showed the highest and deodar showed the lowest calorific value. Ash content was highest in the fir residue samples. The residue of the investigated species showed comparatively lower carbon content than that of typical fossil fuels. High volatile matter was observed in the residue of deodar species. It is worth mentioning that sulfur content was not detected in any of the samples analyzed. The absence of sulfur in forest residues hinders production of sulfur oxides (SO_x) during burning and ultimately making them environment-friendly fuel. Annual residue accumulation was highest for fir species and lowest for blue pine species.

Key words: Biomass; Feedstock; Pellets; Briquettes; Forest Residue; Value-addition.

Introduction

Energy sector plays a vital role in the economic development of a country. Recent decade witnessed a manifold increase in the energy demand as well as the record high prices in the history of fossil originated fuels. This created a global energy crisis in the years 2021 and 2022. Further, the energy crisis worsened due to the Russian-Ukraine war that began in February 2022. Russia, the largest fossil fuel exporter stopped all exports leading to the energy crunch (Anon., 2022). Three fundamental drivers of increased energy demand are the intensification of economic activities, elevated population growth and rapid technological transformation in the world. The world population is growing annually at the rate of 1.05% and energy demand is expected to increase by 1.46% annually by the end of year 2035 (Worldometer, 2022; Enerdata, 2024). In year 2021, the International Energy Agency reported that 75 million people around the world might no longer be able to afford electricity due to the rising fuel prices (Anon., 2021).

Pakistan is an energy deficient country. The rise in global fuel prices, devaluation of the Pakistani currency and recent global economic recession are propelling the state towards energy insecure country (Malik *et al.*, 2019). Pakistan imports almost one third of its energy needs in the form of oil, coal and gas, which puts a lot of pressure on

the national economy (Malik *et al.*, 2019). In Pakistan, current energy demand is 80.62 Mtoe, out of which only 46.56 Mtoe is indigenous, whereas remaining energy is imported. This energy demand is expected to rise by 87.9 Mtoe in 2025 and 99.2 Mtoe by 2030 (Anon., 2021). Energy supply by source in Pakistan includes oil: 25%, gas: 35%, coal: 14%, nuclear: 3% and renewables: 22% (Anon., 2022). Bioenergy contributed 80% to this renewable energy production due to the dependency of countryside inhabitants on the fuelwood (Anon., 2022; Anon., 2023).

Although fossil fuels are limited and non-environment-friendly source of energy, they are widely being utilized around the world. They contribute a major proportion in global energy production i.e., 84.3% of the total (Ritchie *et al.*, 2020). Fossil fuel burning produces a huge amount of CO₂ and other greenhouse gases. In year 2020, about 35 billion metric tons of CO₂ have been accumulated in the atmosphere (Friedlingstein *et al.*, 2020). These non-renewable sources are expected to be at the verge of extinction if their depletion continues at the same pace. On the contrary, renewable energy sources are sustainable as well as environment-friendly (Anon., 2020). These sources are not commonly used, but are more economical and frequently available (Arsent'eva, 2014). Globally, efforts are being made to reduce dependency on fossil fuels due to multiple motives and advantages. The Government of Pakistan is taking effective steps to reduce the pressure on

fossil fuel resources particularly by promoting renewable energy sources (Anon., 2022). Strategies and plans are being developed for escalating utilization of renewable resources in energy mix by 30% up to year 2030 (Anon., 2024). Renewable energy sources have a great potential to reduce energy costs and fossil fuels dependency (Enerdata, 2024). These sources help in mitigating the environmental issues that are a global priority. Economic and sustainable development can be achieved by promoting natural renewable sources, including biomass (Tursunovich, 2022).

Biomass is the best alternative and emerging source of bioenergy as it has the potential to overcome energy crisis of Pakistan (Khan *et al.*, 2022). Biomass includes plant materials such as wood, crops, agriculture and forest residues as well as animal and food processing wastes.

Forest residues were previously considered waste. However, in the recent era, biomass is regarded as a valuable ecosystem service and is being utilized extensively (Bout *et al.*, 2019). These residues consist of leaves, tiny branches, stem portions and tumbled bark on the forest floor because of miscellaneous circumstances and processes (Sahoo *et al.*, 2019). Forest residue might be a threat if it remains on the forest floor for a long time and can cause wildfire under dry and hot weather conditions. Globally, about 350 million hectares forest area is affected every year by forest fire (Anon., 2020). An eco-friendly alternative to this waste is its removal that could be its utilization as a value-added product (Tyurin *et al.*, 2020). These residues are available widely in the forest area and near forest-based industries. In Europe, around 40 million tons of residues are produced annually from forest-based industries (Gaspar *et al.*, 2019). Sawdust and wood shavings are other byproducts of wood-based industrial operations. The forestry operations like thinning, pruning, logging and harvesting are likewise a rich source of forest residual biomass (Domingo *et al.*, 2019).

Globally, about 2.5 billion people still depend on traditional energy sources such as wood, forest and agricultural waste or dung (Röder & Welfle, 2019). Average fuelwood consumption in the Himalayan temperate region of Pakistan is estimated to be 3.10 kg day⁻¹ (Shaheen *et al.*, 2017). More than half of biomass is supplied from forests and forest-based industries all over the world (Anon., 2019). Energy security is vital to economic growth of a country (Pfau *et al.*, 2014; Naik *et al.*, 2010). All fuels on burning produce greenhouse gases and biomass as well, but their net quantity is far less than that of produced by fossil fuel burning. It is due to the fact that the biomass releases the same amount of CO₂ that was fixed during its photosynthesis. That is why, vegetation is considered both as a carbon sink and carbon source (Röder & Welfle, 2019).

The analyses of physical and chemical properties of forest residues are critical for their value-addition and optimum and efficient utilization. Material characterization is traditionally focused on proximate and ultimate analyses for assessing the fuel attributes. Identifying the renewable and alternate energy sources is need of the hour. It is an important practice for the feedstock production of selected biomass and to improve their efficiency. In considering the economic feasibility, production of biofuels is a profitable and eco-friendly energy resource (Czekała *et al.*, 2020).

The biomass densification is an advance technique that helps in improving combustion properties and per unit amount of energy (Yang *et al.*, 2022). This processing of raw material into uniform sized solid biofuels improves energy efficiency, lessen the handling and transporting costs and enhance stability (Nunes *et al.*, 2019; Moran *et al.*, 2009). Upgrading the conventional energy resources, such as fuelwood and using energy-efficient appliances can benefit the environment and promote sustainable economic growth (Moeen *et al.*, 2016). Forest residues and industrial wood waste can be best utilized by converting it into pellets and briquettes. They are a rich source of renewable energy across the world (Tyurin *et al.*, 2020). Material characterization helps in the optimization of biomass densification process as well as ascertains their suitability as a fuel. Solid biofuel production and optimized densification are best achieved by understanding the characteristics of biomass.

The sustainability and economic feasibility of biofuel industry is greatly dependent on availability of raw material. Therefore, the quantity of accumulated residues in a forest ecosystem is important to know. This quantification provides information about the availability of residues in a specific forest type over a certain period of time. These assessments also help in developing strategies and policies for green economy by fulfilling the energy demands and reducing the pressure on natural resources.

The primary objectives of this study were to enumerate residue accumulation by dominated tree-species in the moist temperate forests of Pakistan and material characterization of these residues as a biofuel. Additional, the physical and chemical characteristics of forest residues were compared by species to identify the most suitable raw material for producing value-added solid biofuels.

Material and Method

Description of the study area: This study was conducted in the moist temperate forests of Pakistan found between 33.907° to 34.909°N and 73.3943° to 73.6507°E. These forests are extended up to the dry temperate forests and to a small extent to the sub-alpine forests. Their elevation ranges from 1700 to 3350 m above sea level (Champion *et al.*, 1965). These forests are renowned for their tall, evergreen conifers, enchanting, lush green valleys and mesmerizing scenic beauty. Dominant tree species of moist temperate forests of Pakistan are *Cedrus deodara* [Deodar], *Pinus wallichiana* [Blue pine], *Picea smithiana* [Spruce] and *Abies pindrow* [Fir] (Malik *et al.*, 2023). Moist temperate forests in Pakistan remained under high exploitation pressure for wood supplies in the past. It is encouraging that the ban on green felling has helped improve their conditions over the past three decades. Geographic location of the study area is shown in (Fig. 1).

Methods: The species selected for the investigation were Fir (*Abies pindrow*), Deodar (*Cedrus deodara*) and Blue pine (*Pinus wallichiana*). Selected species constitute more than 70% of this forest type. Four locations named: Dunga Gali (District Abbottabad), Jhika Gali (District Murree), Thandiani (District Abbottabad) and Kaghan (District Mansehra) were chosen for sample collection.

Site selection at each location was based on spatial diversity, species composition, and topographic conditions. To achieve the study objectives, residue samples i.e., freshly fallen leaves (needles) were collected from the selected sites. The samples were collected from the pure stands of each species under investigation with the help of circular quadrat of one square meter size (radius 0.56 m). The collected samples were oven-dried

and then weighed for dry weight estimation. This dry weight of residue samples provided the residue accumulation of focused species at certain locations. These samples were then minced in the grinder for physical, proximate and ultimate analyses. Proximate analysis included calorific value, ash content and volatile matter while ultimate analysis comprised of finding the carbon and sulfur content.

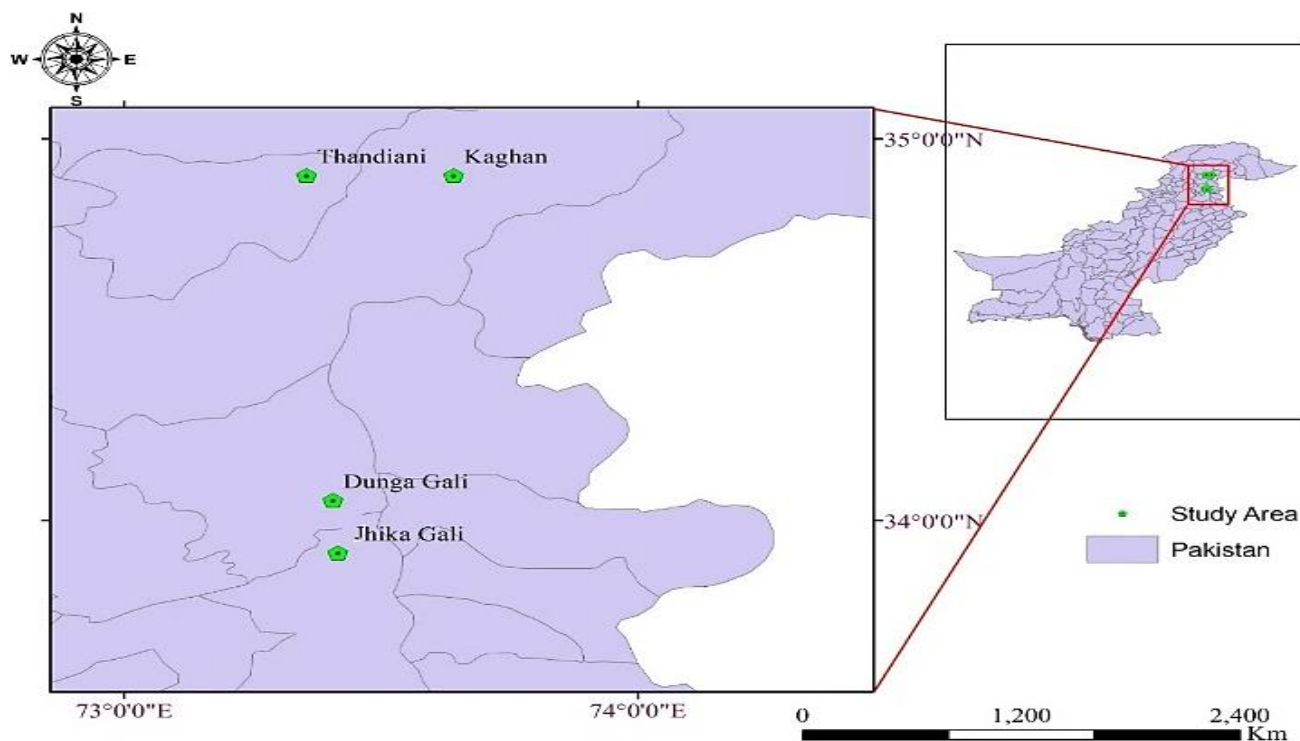


Fig. 1. Location of the study area in Pakistan.

Physical analysis: Bulk density (BD) is the weight of any material in a given volume. The sample was first placed in the known volume container. Then the weight of sample filled in the container was measured with electronic balance of 0.01g sensitivity. Bulk density was calculated by using Equation 1 (Bhagwanrao & Singaravelu, 2014).

$$BD = \frac{W_2 - W_1}{V} \quad \text{Equation 1}$$

where

- BD = Bulk density of the sample (g cm⁻³)
- w₂ = Weight of the container and sample (g)
- w₁ = Weight of the container (g)
- v = Volume of the container (cm³)

Proximate analysis: Proximate analysis consists of detecting calorific value, ash content and volatile matter. Calorific value is the key indicator of fuel quality and is the heat produced during burning. It was estimated by adopting ASTM D4809-18 method in the static bomb calorimeter. This analysis was performed at Hydrocarbon Development Institute of Pakistan (HDIP) Islamabad laboratory. An amount of 0.1 mg was placed in the sealed cup and

weighed. One ml of water was added to the bomb and charged with oxygen. The sample was placed in the bomb calorimeter at an initial temperature of 25°C. Temperature changes were noted after fixed intervals and at the end gross calorific value was determined accordingly. Equation 2 was used to estimate calorific value of the given samples.

$$Q_g = (tW - e_1 - e_2 - e_3 - e_4) \times \left(\frac{100}{m}\right) \quad \text{Equation 2}$$

where

- Q_g = Gross calorific value expressed in MJ kg⁻¹,
- t = Corrected temperature rise,
- W = Energy equivalent to calorimeter (MJ/°C),
- e₁, e₂, e₃, e₄ = Corrections,
- m = Mass of sample

Ash content of the samples was calculated by using ASTM E1755-01 method. A muffle furnace was used to perform the test. Five grams finely milled sample was placed in crucible and completely burnt at a temperature of 575±25°C for six hours. The samples were taken out and weighed on a digital balance. Ash content was calculated by using Equation 3.

$$\text{Ash (\%)} = \frac{[(\text{Crucible weight} + \text{Ash}) - \text{Crucible weight}]}{\text{Sample ODW}} * 100 \quad \text{Equation 3}$$

*ODW: Oven dry weight

Volatile matter is an unstable material present in a fuel that is lost abruptly on heating. ASTM -E872-82-1998 standard was adopted for volatile matter estimation. Initially, one gram of milled sample was placed in the

$$\text{Volatile matter (\%)} = \text{Weight loss (\%)} - \text{Moisture (\%)}$$

Equation 4

Ultimate analysis: Ultimate analyses were carried out in the LECO Analyzer CS 244/ CS 300 in HDIP laboratory. This analyzer uses infrared absorption and thermal conductivity to measure flammable gases within the sample. This approximates carbon and sulfur concentrations in the sample by combustion method. An amount of 30-50 mg sample was poured in crucibles and analyzed. After 100 seconds carbon and sulfur percentages were displayed on the screen. Relevant oxidants were used for the analyses.

The collected data was subjected to statistical analysis. Analysis of Variance (ANOVA) was carried out to compare characteristics of species-specific residues.

Results

Physical characteristics: The results for BD are presented in (Fig. 2). Residue samples consisting of fallen leaves (needles) of Deodar were found to have the highest BD of 0.61 g cm^{-3} and Blue Pine showed the lowest BD and its value was 0.40 g cm^{-3} . Further, the BD of Fir residue was 0.51 g cm^{-3} .

ANOVA results for BD of species-specific residues are presented in (Table 1). The results indicated that the values are significantly different from each other with a p -value of 0.0052.

Proximate analysis: The calorific values of the residues associated with tree species are presented in (Fig. 3). The highest calorific value was shown by Blue Pine residue with a value of 18.6 MJ kg^{-1} followed by Fir and Deodar having 17.6 MJ kg^{-1} and 17.5 MJ kg^{-1} , respectively.

Table 2 indicates the results of ANOVA for residues derived from three species in the study area. A p -value of <0.05 indicated a statistically significant difference for calorific value of residues originated from three different species.

The results of ash content analysis of the residues related to Blue Pine, Fir and Deodar are shown in (Fig. 4). Fir showed the highest (11.40%) and Deodar showed the lowest ash content (3.75%).

Table 3 showing results of ANOVA for ash content. A p -value of 0.0026 indicated a highly significant difference among species for ash content. A material producing low ash content is considered good for use as a fuel.

Figure 5 presents the volatile matter percentage in the residues of selected tree species in the study area. The volatile matter of Deodar, Fir and Blue Pine was 84.4%, 81.57% and 78.91%, respectively.

Ultimate analysis: Carbon content of residues associated with tree species is shown in (Fig. 6). Fir showed the highest carbon content of 48.40% and Blue Pine showed the lowest carbon content of 46.39%.

The results of ANOVA for carbon content are presented in (Table 5). A p -value of 0.5397 indicated a statistically significant difference among species for carbon content.

It is worth mentioning that none of the residue from the investigated three species i.e., Blue Pine, Fir and Deodar indicated sulfur content.

muffle furnace at $950 \pm 20^\circ\text{C}$ for seven minutes. Later, samples were weighed again and the weight loss was the volatile matter in the sample following Equation 4.

Table 1. Analysis of variance (ANOVA) of bulk density among species in the study area.

Source of variation	SS	df	MS	F	p-value	F crit
Between groups	0.068	2	0.03418	14.41544	0.0052	5.1433
Within groups	0.0142	6	0.0024			
Total	0.0824	8				

* Level of significance $\alpha=0.05$

Table 2. Analysis of variance (ANOVA) among calorific value of residues associated with three species in the study area.

Source of variation	SS	df	MS	F	p-value	F crit
Between groups	1566003	2	783001.4	3.177	0.049	3.285
Within groups	8132057	33	246426			
Total	9698060	35				

* Level of significance $\alpha=0.05$

Table 3. Analysis of variance (ANOVA) among ash content of the residues associated with three species in the study area.

Source of variation	SS	df	MS	F	p-value	F crit
Between groups	131.4768	2	65.738	8.4325	0.0026	3.554
Within groups	140.3245	18	7.796			
Total	271.8013	20				

Table 4. Analysis of variance among volatile matter of the residues associated with three species in the study area.

Source of variation	SS	df	MS	F	p-value	F crit
Between groups	45.1703	2	22.585	0.902	0.4544	5.143
Within groups	150.1947	6	25.0324			
Total	195.365	8				

A p -value of 0.4544 indicated a significant difference among species for volatile matter (Table 4)

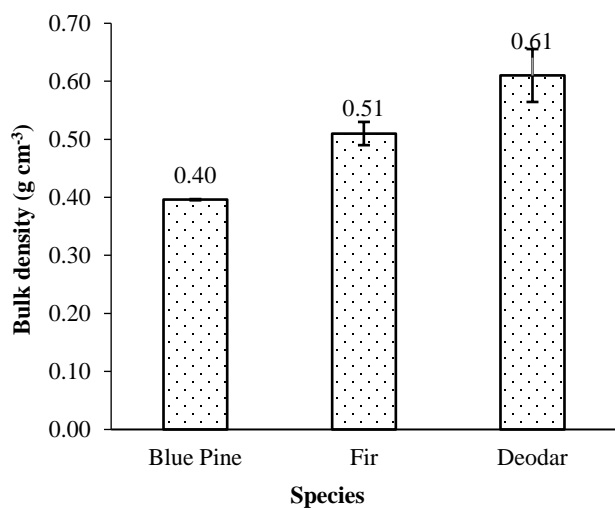
Table 5. Analysis of variance (ANOVA) among carbon content of the residues associated with three species in the study area.

Source of variation	SS	df	MS	F	p-value	F crit
Between groups	24.218	2	12.109	0.628	0.5397	3.285
Within groups	635.8946	33	19.269			
Total	660.1131	35				

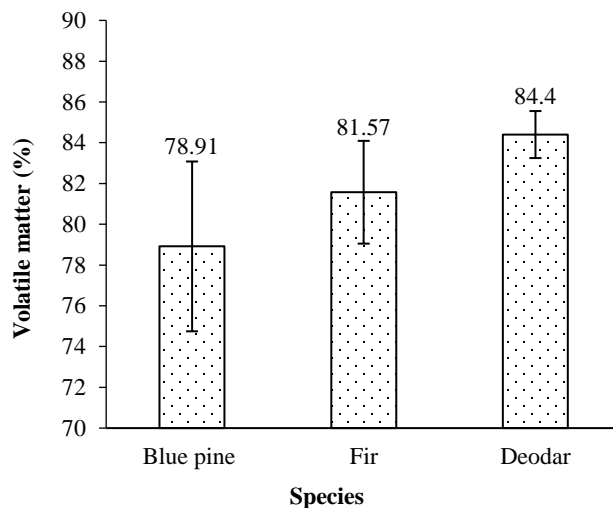
Table 6. Annual residue accumulation (kg ha⁻¹) of species by location in the study area.

Locations	Blue pine	Fir	Deodar
Kaghan	1161.5	2465.8	2040.2
Dhunga Gali	1905.3	1931.1	3026.1
Jhika Gali	2204.2	4752.1	1526.5
Thandiani	1971.7	1641.3	1776.6
Mean	1810.7	2697.6	2092.4

Quantification of residue accumulation: Figure 7 illustrates the accumulation of forest residues for Blue pine, Fir and Deodar species at four locations in moist temperate forest of Pakistan. The mean residue accumulation for Blue pine, Fir and Deodar was $1810.7 \text{ kg ha}^{-1}$, $2697.6 \text{ kg ha}^{-1}$ and $2092.4 \text{ kg ha}^{-1}$, respectively. Jhika Gali site showed the highest accumulation of Fir species and Thandiani site showed the lowest accumulation. The accumulation of Deodar was the highest at Dhunga Gali forests ($3026.1 \text{ kg ha}^{-1}$) and the lowest at Jhika Gali ($1526.5 \text{ kg ha}^{-1}$). Further Blue Pine residue accumulation was the highest in Jhika Gali sampling areas ($2204.4 \text{ kg ha}^{-1}$) and the lowest in Kaghan forest ($1161.5 \text{ kg ha}^{-1}$).



*Error bars showing standard error (SE)
Fig. 2. Bulk density of the residues by tree species in the study area.



*Error bars showing standard error (SE)
Fig. 5. Volatile matter of the residues associated with three species in the study area.

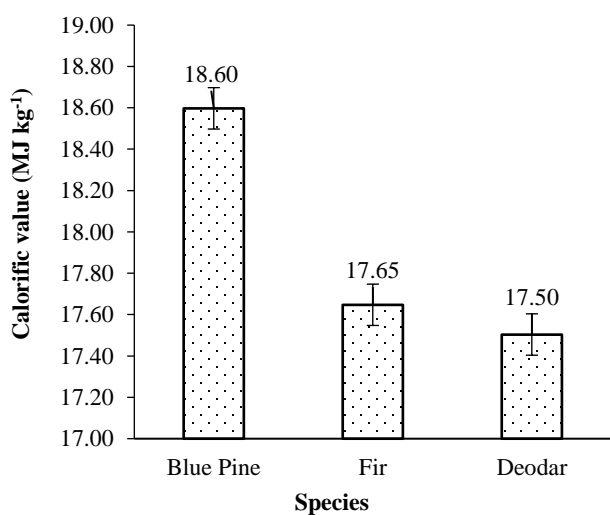
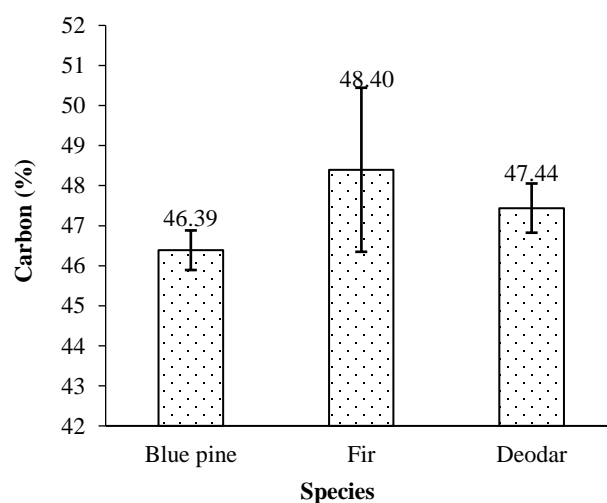
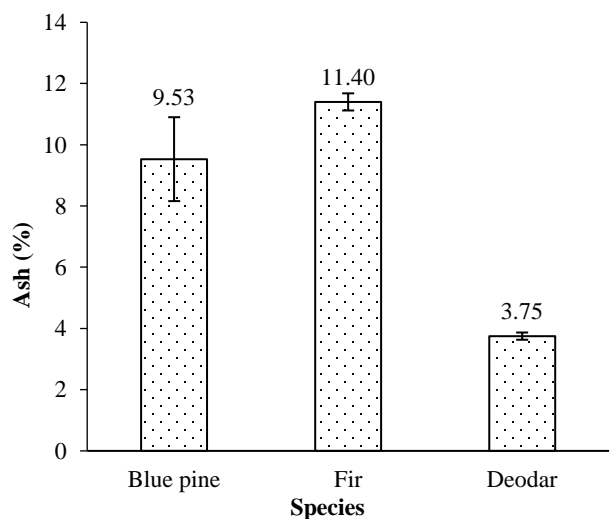


Fig. 3. Calorific value of the residues associated with three species in the study area.



*Error bars showing standard error (SE)
Fig. 6. Carbon content of the residues associated with three species in the study area.



*Error bars showing standard error (SE)
Fig. 4. Ash content of the residues associated with three species in the study area.

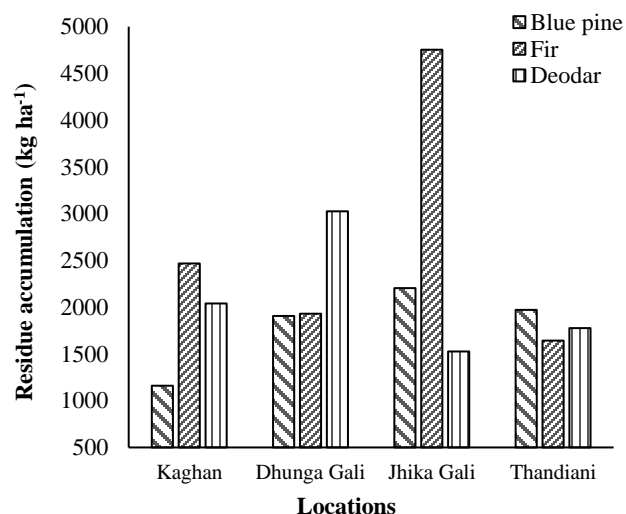


Fig. 7. Species-wise comparison of residue accumulation by location in the study area.

Discussion

Bulk density is an important feature in designing the logistic system for biomass handling and transporting. This property defines the costs of handling, transporting and storage of raw material. It influences feedstock (input) cost for making value-added product (biofuel). The BD of agriculture residue varies from Rice husk: $0.33159 \text{ g cm}^{-3}$, Rice straw: 0.380 g cm^{-3} , Sugar cane bagasse: 0.723 g cm^{-3} and cotton stalk: 0.206 g cm^{-3} (Makavan *et al.*, 2018). High BD is desirable for reducing handling, transport and storage costs. In comparison to agriculture residues, most forest residues have higher BD which is a useful character for reducing the cost of finished product.

The calorific value is the key indicator of contained energy in the material. Biomass including forest residues have comparatively less calorific value than that of fossil fuels *i.e.*, Coal: 29 MJ kg^{-1} , Oil: 42 MJ kg^{-1} , Natural Gas: 38 MJ kg^{-1} and LPG: 46 MJ kg^{-1} (Anon., 2024). Similar studies in different parts of the world have shown variable results for diverse species residues. For example, Nunez-Regueira *et al.* (2003) reported a calorific value of 7.5 MJ kg^{-1} for pine, while Pena-Vergara *et al.*, (2022) found a calorific value of $17\text{-}21.1 \text{ MJ kg}^{-1}$ for *Eucalyptus* spp. Masia *et al.*, (2007) reported lower and higher calorific values of 18.55 and 19.79 MJ kg^{-1} for Pine wood chips. Further, Silver fir biomass residues indicated calorific values in the range of 20.8 MJ kg^{-1} to 22.8 MJ kg^{-1} (Butnaru *et al.*, 2022). Our results are almost in line with the findings of Pena-Vergara *et al.*, (2022) and Butnaru *et al.*, (2022). This means that same species or species of the same genera growing under dissimilar climatic or forest condition can produce different calorific values. This helps in its selection as a fuel and specifically selection of raw material for making fuel pellets and briquettes. The blending of assorted primal matter for making value-added biofuels is principally based on calorific value of the raw material.

The information regarding ash content of fuels is important for improving burning efficiency through conversion techniques (Biswas *et al.*, 2022). The high ash content may cause fouling and slagging in combustion chamber. In a study, Silver fir showed the ash content of less than 5% by weight (Butnaru *et al.*, 2022). A study by Masia *et al.*, (2007) reported 5.95% ash content for Pine wood chips. However, it ranged between 1.85 and 37.79% for various bio-based residues from different sources. Although biomass is comprised of small amount of ash content yet it somehow affects combustion process. The values observed in this study was found contradictory with the results of Pinewood residues observed in the study performed by Naik *et al.*, (2010). Ash content observed in Pinewoods was $1.5 \pm 0.2\%$ which is far less than that of Blue Pine and Fir (Naik *et al.*, 2010). Generally, lignocellulosic biomass has volatile matter in the range of 70-90% which is found to be almost similar to our findings in this study (Biswas *et al.*, 2022). Fuels having high volatile content are highly reactive with superior flame during burning (Ajimotokan *et al.*, 2019).

The fuels having high carbon content produces higher amount of energy and also contributes much in greenhouse gas emissions (Anon., 2024). These residues have less carbon content compared to the coal, oil, natural gas and

LPG that have 75%, 85%, 75% and 82% carbon contents, respectively and hence produce relatively less emissions compared to fossil fuels (Anon., 2024). Generally, agriculture and forestry biomass showed the same trend with carbon content in the range of 41-49% (Naik *et al.*, 2010).

Sulfur present in fuel converts into SO_2 upon combustion and releases it as a pollutant in the environment. Biofuels have an advantage over fossil fuels due to absence of sulphur as was the findings of this study. However, some studies reported residue of Pine and Spruce contains a very little sulfur *i.e.*, 0.25% (Aniszewska *et al.*, 2017).

The highest mean annual accumulation of residue was observed to be 5269 kg ha^{-1} for a deciduous forest in Brazil (Gomes *et al.*, 2022). The residue accumulation in coniferous forests usually found to be in the range of $1420\text{-}4000 \text{ kg ha}^{-1}$ (Kavvadias *et al.*, 2001). In another study by Gairola *et al.*, (2009), Fir showed the annual residue accumulation of about 2358 kg ha^{-1} in three subalpine forests. Our findings regarding residue accumulation on forest floor are in general agreement with reported information in scientific literature.

Conclusion

Material characterization is the key step for the efficient utilization of biomass and its conversion. Blue pine residue was found to have the highest calorific value shown among studied species and considered the best for biofuel production. This species is considered more environmentally friendly as its burning produces less carbon dioxide because of low carbon content. The bulk density of blue pine was also lesser compared to other species investigated in this study. This might be due to the large needle size and its greater volume.

Blue pine needles indicated calorific value of 18.6 MJ kg^{-1} whereas Fir leaves have the lowest calorific value of 17.6 MJ kg^{-1} . Out of three species investigated, the highest ash content was produced by Fir followed by Blue Pine and Deodar. The volatile matter was the lowest in Blue Pine needles and the highest in Deodar leaves. Carbon content was found to be in the range of 46-49% in all three species. Sulfur was not detected in any of the species investigated in this study.

Apart from characterization, the data for residue accumulation indicated that Fir trees produced the highest ($2697.6 \text{ kg ha}^{-1}$) and blue pine trees produced the lowest ($1810.7 \text{ kg ha}^{-1}$) residue. The trees of Fir species accumulated the highest residue among dominated species of moist temperate forests and at some locations more than double compared to Blue Pine. Further, the highest carbon content of Fir makes it a better option for its use as a feedstock for making value-added biofuel from forest residues.

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