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Allelopathic Activity and Characterization of Allelopathic Substances from *Elaeocarpus floribundus* Blume Leaves for the Development of Bioherbicides

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Abstract: To help protect the environment as well as increase agricultural production, the use of synthetic herbicides must be reduced and replaced with plant-based bioherbicides. *Elaeocarpus floribundus* is a perennial, evergreen, and medium-sized plant grown in different areas of the world. The pharmaceutical properties and various uses of *Elaeocarpus floribundus* have been reported, but its allelopathic potential has not yet been explored. Thus, we carried out the present study to identify allelopathic compounds from *Elaeocarpus floribundus*. Aqueous MeOH extracts of *Elaeocarpus floribundus* significantly suppressed the growth of the tested species (cress and barnyard grass) in a dose- and species-dependent way. The three most active allelopathic substances were isolated via *cis*-chromatographic steps and characterized as (3*R*)-3-hydroxy-β-ionone, *cis*-3-hydroxy-α-ionone, and loliolide. All three substances significantly limited the seedling growth of cress, and the compound (3*R*)-3-hydroxy-β-ionone had stronger allelopathic effects than *cis*-3-hydroxy-α-ionone and loliolide. The concentrations of the compounds required for 50% growth inhibition (I_{50} value) of the cress seedlings were in the range of 0.0001–0.0005 M. The findings of this study indicate that all three phytotoxic substances contribute to the phytotoxicity of *Elaeocarpus floribundus*.

Keywords: *Elaeocarpus floribundus*; bioherbicides; weed control; (3*R*)-3-hydroxy-β-ionone; *cis*-3-hydroxy-α-ionone; loliolide

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1. Introduction

The plant *Elaeocarpus floribundus* Blume (local name: Jalpai in Bangladesh) is an evergreen, moderate-sized tree belonging to the family Elaeocarpaceae [1]. It has a 12.0–16.0 m high bole with a spreading-type crown. The leaves are simple, thin, and leathery with a large pointed tip, margin toothed, and green, but frequently some leaves are orange or red. The flowers bloom from April to May, and green fruit mature during August to October. The fruit is edible and usually used to prepare pickles and chutney. The tree is very familiar in Bangladesh but is also found in other countries such as India, Australia, Madagascar, Mauritius, Fiji, Malaysia, Hawaii, Japan, and China [1,2]. In Bangladesh, this tree generally grows on the ridges and slopes of hills in a clay and sandy soil and is found in the natural forests of the Sylhet, Chittagong, Cox's Bazar, Chittagong Hill Tracts, Mymensingh, and Gazipur districts. It is also planted in homestead areas with minimum cultural practices.

The bark and leaves are used for mouthwash and the fruit contains antiseptic properties [3]. *Elaeocarpus floribundus* wood is used to make paper pulp, plywood, fiberboard, and light construction materials [4]. The different parts of the tree such as bark, leaf, root, and fruit are commonly used to treat various diseases [5]. It is applied as folk medicine to treat diabetes, rheumatoid arthritis, dysentery, and high blood pressure [6]. Furthermore, this tree is reputed to possess biological properties such as antibacterial, antiseptic, anti-aging, antioxidant, antitumor, and anticancer [6–9]. The plant has also been shown to possess stabilizing and reducing activities in nanoparticles of silver biosynthesis with growth suppression potential against gram-negative and -positive bacteria [10]. Although research has been carried out on the biological properties and various functions of *Elaeocarpus floribundus*, the allelopathic potential of this tree has not yet been explored. Therefore, this research aimed to determine the phytotoxicity of *Elaeocarpus floribundus* leaves and to detect allelopathic compounds that might be useful for developing natural herbicides or bioherbicides.

Weed plants in crop fields reduce the quantity and quality of agricultural output, causing huge financial losses for farmers [11]. Weeds are unwanted, destructive plant species that inhibit the growth and development of beneficial plant species and lower their output potential. They contend with agricultural crop plants for various resources like space, light, water, and nutrients, resulting in lower crop yields [12]. Weeds are ubiquitous in crop fields, reducing crop output and raising production expenses, making the production of crops less cost-effective [13]. They reduce crop production by disrupting crop growth through competition or allelopathy or both [14]. Weeds attack different crop growth components, resulting in production losses of 15–66% for direct-sowing paddies, 18–65% of corn, 50–76% of soybean, and 45–71% of groundnut crops [15]. They are the most damaging invasive plants, capable of causing yield reductions of up to 100% [16], and are expected to cost the world economy more than $40,000 million per year [17]. To control weeds in crop fields, significant quantities of herbicides are applied, presenting a substantial risk. The effects of herbicides on soil, surface water, and groundwater are directly associated with less biodegradability, percolation, and persistence. In recent times, herbicides used to manage weed species have caused serious problems for crop plants, the environment, and people. Herbicide-resistant weeds have become common since the first record of herbicide application during the 1950s due to the development of fundamental evolutionary procedures [18]. Currently, herbicide resistance is a severe problem for farmers in managing weeds in their crop fields. For instance, the triazine group of herbicides has been commonly used largely for their effects on the photosynthesis of various weeds [19–21]. Farmers, the general public, and legislators are aware of the high cost and unsustainable nature of present weed-control practices. Recent discussions have focused on banning commonly applied herbicides like glyphosate and the rising necessity for organic products [22].

To reduce the dependence on synthetic herbicides, researchers are looking for alternative natural sources such as various compounds (secondary metabolites) that could be used to control weeds [23]. Allelopathy can aid in the biological control of weeds, plant diseases, and pests, which can help to enhance plant and ecosystem production. Allelopathy may be defined as the procedure or process through which a component or organism affects another component or organism, by producing allelopathic compounds [24]. It is an ecofriendly strategy for controlling weeds, increasing crop productivity, reducing the use of synthetic chemicals in the agriculture sector, and recovering biological microorganisms [25,26]. Allelopathy is also considered to be a biological process encompassing the liberation of substances that might have a stimulative influence, but commonly show an inhibitory influence, on the survival, growth, emergence, and reproduction of other plants. Bioherbicides that are prepared from allelochemicals provide minimal risk to the agro-ecosystem and the health of people [27]. Some allelopathic compounds are readily soluble in water, which makes them easy to use because there is no need to add surfactants [28]. Compared with chemical herbicides, allelochemicals have more eco-friendly structures. Allelopathic bioherbicides are well known for having less toxicity and a short

life in the environment, as well as having numerous modes of action, which decreases the chances of weeds developing herbicide resistance [29]. Hence, allelochemicals are a potential candidate for bioherbicide development.

Bioherbicides are developed from plant extracts and phytotoxins of various microbes (mycoherbicides) and are considered an important tool for controlling weeds [30]. They do not usually have persistent properties, which means they do not stay active in the environment of crops for long periods of time, do not pollute the soil and water, and do not harm non-target components. Extracts of different plants, which have traditionally been used for nutritional or medicinal purposes, could be employed to develop more environmentally friendly bioherbicides for weed control. Bioherbicides made from plant extracts have displayed promising effects against various weeds. Different plant extract substances contain very specific suppressing potential against the growth of weeds without any detrimental effect on crop plants [31]. This might be described through the variations in the susceptibility of the enzymes or persistence of the specific acceptors in the weed plants that intuit and respond with phytotoxic substances [32]. Plants release various secondary metabolites or compounds (called allelochemicals) such as phenolics, alcohols, flavonoids, fatty acids, steroids, and terpenoids, which inhibit the growth, development, and reproduction of surrounding vegetation, including weed species [27]. Therefore, allelopathic compounds from plants could be applied to manage weeds, which would be helpful for the environment.

2. Materials and Methods

2.1. Plant Materials

Fresh and mature leaves of *Elaeocarpus floribundus* Blume were gathered from various parts of the Noakhali Science and Technology University, Noakhali, (22°47′31″ N and 91°06′07″ E), Bangladesh, during April–May 2019. The leaves were washed with distilled water to remove dirt, debris, and other contaminants. The washed samples were kept in a shady area until dry and then ground into a grainy leaf powder using a blender. The leaf powder was sealed in a plastic bag and kept at 2 °C for later analysis. Two plant species were chosen for the allelopathic growth activity assay, one monocot species, barnyard grass (*Echinochloa crus-galli* (L.) P. Beauv.), and one dicot species, cress (*Lepidium sativum* L.).

2.2. Extraction and Growth Bioassay

A preliminary extraction experiment was performed to determine the phytotoxic potential of *Elaeocarpus floribundus* and to develop an accurate isolation procedure: 80 g of *Elaeocarpus floribundus* leaf powder was extracted by saturating in 400 mL of 70% aqueous MeOH (methanol) for two days in the dark and filtered using a single layer of filter paper (No. 2, 125 mm; Toyo Ltd., Tokyo, Japan). The remaining solid portions were immersed again in the same amount of MeOH for one day and filtered once again. These filtrates were combined and evaporated using a Rotavapor at 40 °C. The leaf extracts were immersed in 150 mL of MeOH to obtain the desired bioassay concentrations (0.001, 0.003, 0.01, 0.03, 0.1, and 0.3 g DW (dry weight) equivalent extract/mL), and these concentrations were applied to the filter paper (No. 2, 28 mm; Toyo Ltd.) in 28 mm Petri dishes. After drying the extracts, the Petri dishes were soaked with 0.6 mL of 0.05% aqueous (H_2O) solution of polyoxyethylene sorbitan monolaurate (Tween 20; Nacalai Tesque, Inc., Kyoto, Japan). Ten uniform seeds of cress and ten germinated seeds of barnyard grass were placed in each Petri dish. In addition, Petri dishes were treated with H_2O (aqueous) Tween 20 solution without the extracts of *Elaeocarpus floribundus* as a control treatment. Finally, the Petri dishes were kept in a growth chamber for 48 h in the dark at 25 °C, and after 2 days of incubation the seedlings were measured.

2.3. Isolation and Purification of the Substances

To isolate and to identify the bioactive substances, a substantial extraction was performed with 2.8 kg of *Elaeocarpus floribundus* leaf powder by following the above extraction

procedure. The resulting extracts were desiccated using a Rotavapor (40 °C) to obtain aqueous (H_2O) crude extracts. The crude extracts were corrected to pH 7.0 using a 1 M solution of phosphate buffer and partitioned six times with the same amount of ethyl acetate to get aqueous and ethyl acetate portions. The biological effect of both portions was determined using a cress bioassay. The ethyl acetate portion had a stronger effect and this portion was chosen to isolate the phytotoxic substances. Accordingly, the ethyl acetate portion was evaporated to dryness after removing water using anhydrous Na_2SO_4, and then chromatographed with a column of silica gel (60 g of silica gel 60, spherical, 70–230 mesh; Nacalai Tesque, Inc.). The column was then eluted with 150 mL of ethyl acetate in n-hexane, increased by 10% per step (*v*/*v*), and finally two times with methanol (300 mL). From the results of the phytotoxic bioassay experiment, the highest potential was found with 80% ethyl acetate in n-hexane, which was then evaporated and applied to a Sephadex LH-20 column (GE Healthcare Bio-Sciences AB, SE-751 84 Uppsala, Sweden). The column was eluted with 150 mL of H_2O methanol (methanol 20–80% (*v*/*v*), increased by 20% each step, and 300 mL methanol), and the maximum potential was observed with 40% H_2O methanol (another active fraction with 50% H_2O methanol for compound **3**), which was applied to a reverse-phase C_{18} cartridge. The cartridge was loaded with 15 mL of H_2O methanol (methanol 20–80% (*v*/*v*), and methanol 30 mL). The strongest biological activity was found with 40% H_2O methanol (another active fraction with 30% H_2O methanol for compound **3**), which was purified using reverse-phase HPLC (500 × 10 mm I.D. ODS AQ-325; YMC Ltd., Kyoto, Japan) at a flow rate of 1.5 mL/min with 50% H_2O methanol (40% H_2O methanol for compound **3**), and the chromatogram was recorded at 220 nm wavelength and 40 °C oven temperature. The two most active phytotoxic compounds, compound **1** and **2**, were detected at the retention times of 95–101 and 140–146 min (78–83 min for compound **3**), respectively, which were finally purified using reverse-phase HPLC (4.6 × 250 mm I.D., S-5 µm, Inertsil® ODS-3; GL Science Inc., Tokyo, Japan) at a flow rate of 0.8 mL/min with 40% H_2O methanol (20% for compound **3**) and detected at the retention times of 65–69 and 78–92 min (67–76 min for compound **3**), respectively. Last, all the compounds (compounds **1**, **2**, and **3**) were characterized by HRESIMS, ^{1}H NMR (500 MHz, $CDCl_3$), and specific rotation.

2.4. Bioassay of the Identified Compounds

The identified compounds, (3*R*)-3-hydroxy-β-ionone, *cis*-3-hydroxy-α-ionone, and loliolide, from *Elaeocarpus floribundus* were immersed in cold methanol to make six assay concentrations (0.00001, 0.00003, 0.0001, 0.0003, 0.001, and 0.0015 M). The bioactivity of the compounds was tested with cress as previously described.

2.5. Analysis

The assay experiments were carried out by following a CRBD (completely randomized block design) replicated three times, and the entire assay test was duplicated twice. The recorded data were presented as mean ± standard error. ANOVA (analysis of variance) was measured using SPSS statistical package version 20.0 (SPSS Inc., Chicago, IL, USA), and meaningful variations among the treatments and control were determined using Tukey's HSD test at the 0.05 level of provability. The concentrations required for 50% inhibition of the growth (I_{50} value) of the tested plants in the bioassay experiments were calculated using GraphPad Prism 6.0 (GraphPad Software, Inc., La Jolla, CA, USA).

3. Results

3.1. Phytotoxic Activity of the Elaeocarpus floribundus Extracts

The *Elaeocarpus floribundus* extracts (aqueous methanol) were shown to have a considerable phytotoxic effect on the cress and barnyard grass. Growth inhibition increased with increasing extract concentration and also differed between the two species (Figure 1). No statistically significant growth effects were found with the concentration of 0.001 g DW equivalent extract/mL of *Elaeocarpus floribundus* extracts, but growth inhibition was observed at higher concentrations. At 0.01 g DW equivalent extract/mL, the seedlings

were suppressed more than 50%, except barnyard grass shoots (30%), compared with the control treatment. Notably, at 0.1 g DW equivalent extract/mL, the growth of the cress shoots (0.38%) and roots (1.49%), and barnyard grass roots (4.16%) were less than 5% of the control treatment, whereas the barnyard grass shoot growth was 17.89% of control. However, at 0.3 g DW equivalent extract/mL, the *Elaeocarpus floribundus* extracts completely inhibited the seedling growth of cress and barnyard grass roots but not barnyard grass shoots. The concentration required for 50% growth inhibition (I_{50} value) of the seedlings was 0.00553–0.0289 g DW equivalent extract/mL (Table 1).

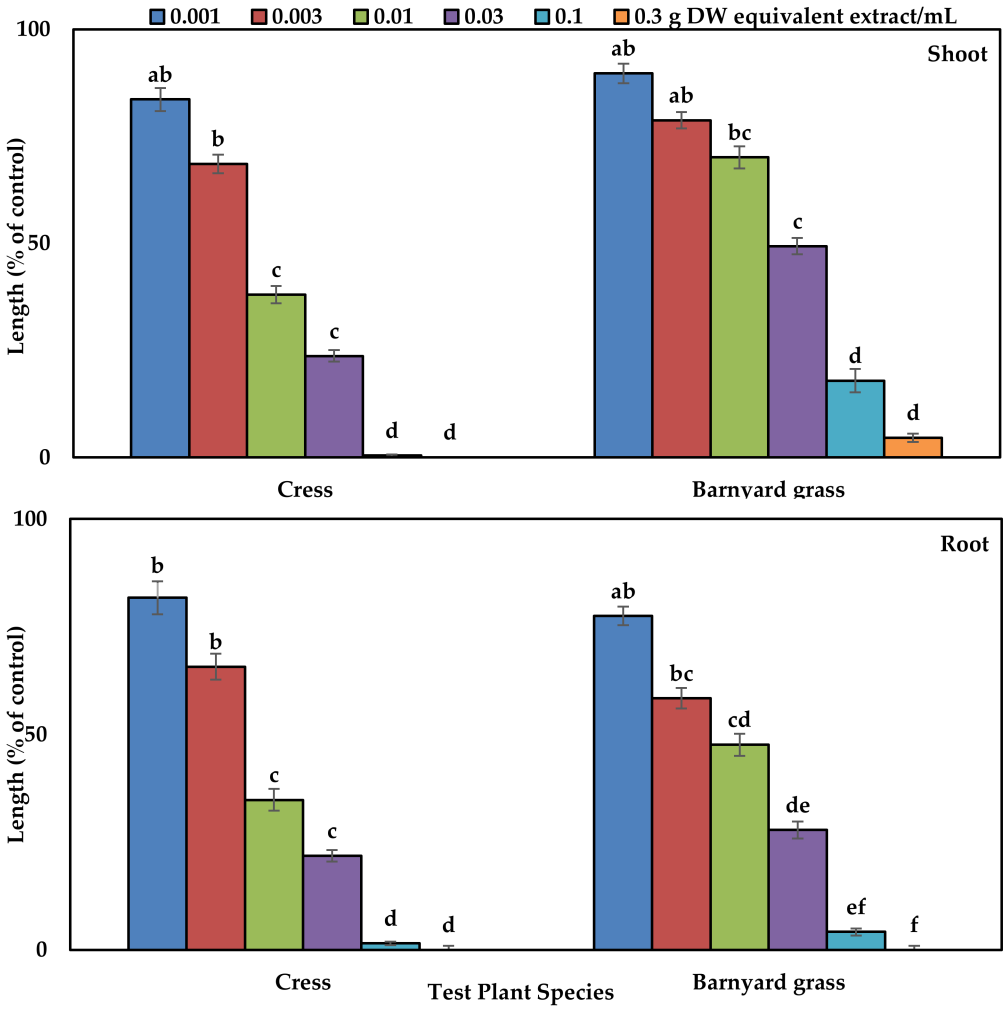

Figure 1. The phytotoxic potential of *Elaeocarpus floribundus* leaf (aqueous methanol) extracts against the seedling growth of cress and barnyard grass at various concentrations. The mean ± standard error was determined from two different experiments (replicated thrice; seedlings per treatment were 10, and total $n = 60$). Various letters signify significant differences according to Tukey's HSD test at the probability level of 0.05.

Table 1. The concentrations required for 50% growth inhibition (I_{50} value) of the cress and barnyard grass seedlings by the *Elaeocarpus floribundus* (aqueous methanol) leaf extracts.

Test Plant Species		I_{50} Value (g DW Equivalent Extract/mL)	
		Shoot	Root
Dicot	Cress	0.00622	0.00553
Monocot	Barnyard grass	0.0289	0.0076

3.2. Characterization of the Allelopathic Compounds

The molecular formula of compound **1** (yielding 2.8 mg) was determined as $C_{13}H_{20}O_2$ using HRESIMS. The ^{1}H NMR spectrum of compound **1** was determined in $CDCl_3$, resulting in four methyl proton signals at δ_H 2.30 (3H, s), 1.77 (3H, s), 1.12 (3H, s), and 1.11 (3H, s); two olefinic proton signals at δ_H 7.21 (1H, d, J = 16.4) and 6.11 (1H, d, J = 16.4); four methylene proton signals at δ_H 2.43 (1H, dd, J = 17.5, 5.7), 2.09 (1H, dd, J = 17.5, 9.5), 1.79 (1H, dd, J = 12.1, 3.7, 2.2), and 1.49 (1H, t, J = 12.1); and a methine proton signal at δ_H 4.01 (1H, m). By comparing this ^{1}H NMR spectrum of compound **1** with earlier presented data, this substance was identified as (3*R*)-3-hydroxy-β-ionone (Figure 2) [33].

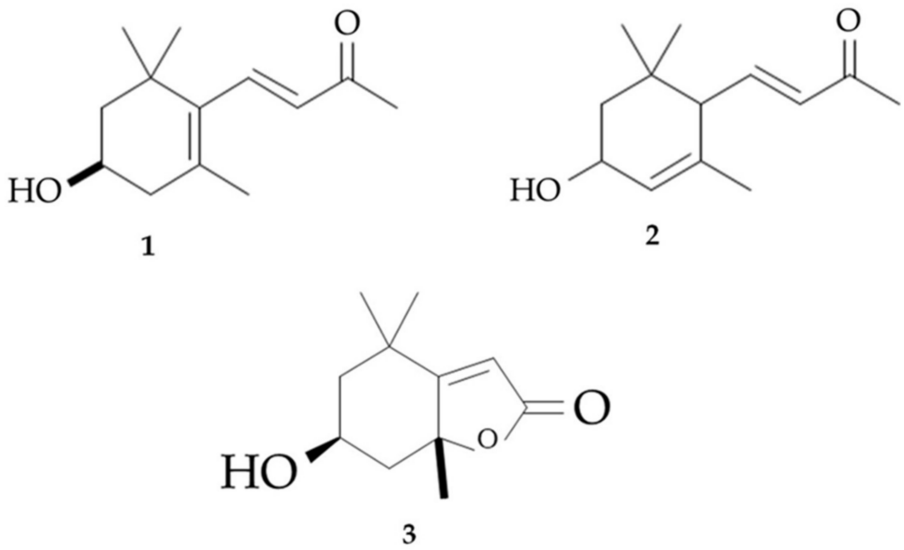

Figure 2. The molecular structure of the characterized allelopathic compounds from *Elaeocarpus floribundus* leaf extracts: **1**. (3*R*)-3-hydroxy-β-ionone, **2**. *cis*-3-hydroxy-α-ionone, **3**. loliolide.

The molecular formula of compound **2** (yielding 3.7 mg) was determined as $C_{13}H_{20}O_2$ using HRESIMS. The ^{1}H NMR spectrum of compound **2** was determined in $CDCl_3$, resulting in four methyl proton signals at δ_H 2.26 (3H, s), 1.63 (3H, t, J = 1.6), 0.97 (3H, s), and 0.89 (3H, s); three olefinic proton signals at δ_H 6.63 (1H, dd, J = 15.8, 9.6), 6.07 (1H, d, J = 15.8), and 5.59 (1H, brs); two methylene proton signals at δ_H 1.70 (1H, dd, J = 13.1, 6.6) and 1.39 (1H, dd, J = 13.4, 9.8); and two methine proton signals at δ_H 4.25 (1H, m) and 2.27 (1H, d, J = 10.0). By comparing this ^{1}H NMR spectrum of compound **2** with earlier presented data, this substance was identified as *cis*-3-hydroxy-α-ionone (Figure 2) [34].

The molecular formula of compound **3** (yielding 5.5 mg) was determined as $C_{11}H_{16}O_3$ using HRESIMS. The ^{1}H NMR spectrum of compound **3** was determined in CD_3OD, resulting in three methyl proton signals at δ_H 1.76 (3H, s), 1.47 (3H, s), and 1.28 (3H, s); an olefinic proton signal at δ_H 5.75 (1H, s); four methylene proton signals at δ_H 2.42 (1H, dt, J = 13.8, 2.4), δ_H 1.97 (1H, dt, J = 14.4, 2.3), δ_H 1.75 (1H, dd, J = 13.8, 4.0), and 1.53 (1H, dd, J = 14.4, 3.8); and one methine proton signal at δ_H 4.21 (1H, m). By comparing this ^{1}H NMR spectrum of compound **3** with earlier presented data, this substance was identified as loliolide (Figure 2) [35].

3.3. Biological Potential of the Identified Compounds

The three identified compounds were assayed to determine their biological potential against cress at the various concentrations. The assay results showed that the biological potential of the identified compounds against the seedling growth of cress varied significantly, and the phytotoxic activity increased with the increasing concentration of the compounds (Figures 3–5). Significant variation in the seedling growth of the cress occurred at the concentrations of 0.0001 M or more with compound (3*R*)-3-hydroxy-β-ionone, and

0.00003 M or more with compound *cis*-3-hydroxy-α-ionone and compound loliolide. At the concentration of 0.0003 M, the shoot and root growth of the cress seedlings were restricted to 40.24 and 33.28% of control, respectively, by compound (3*R*)-3-hydroxy-β-ionone; 57.44 and 44.53% by compound *cis*-3-hydroxy-α-ionone; and 55.18 and 45.20% by compound loliolide. At the highest concentration (0.0015 M), compound (3*R*)-3-hydroxy-β-ionone inhibited the seedling growth to 11.53 and 8.37% of control, respectively; compound *cis*-3-hydroxy-α-ionone to 20.80 and 17.19%; and compound loliolide to 12.14 and 9.50%. The compound concentrations required for 50% growth inhibition (I_{50} value) of the cress seedlings were in the range of 0.0001–0.0005 M (Table 2). From Table 2, it is clear that the inhibitory potential of (3*R*)-3-hydroxy-β-ionone was higher than that of *cis*-3-hydroxy-α-ionone and loliolide.

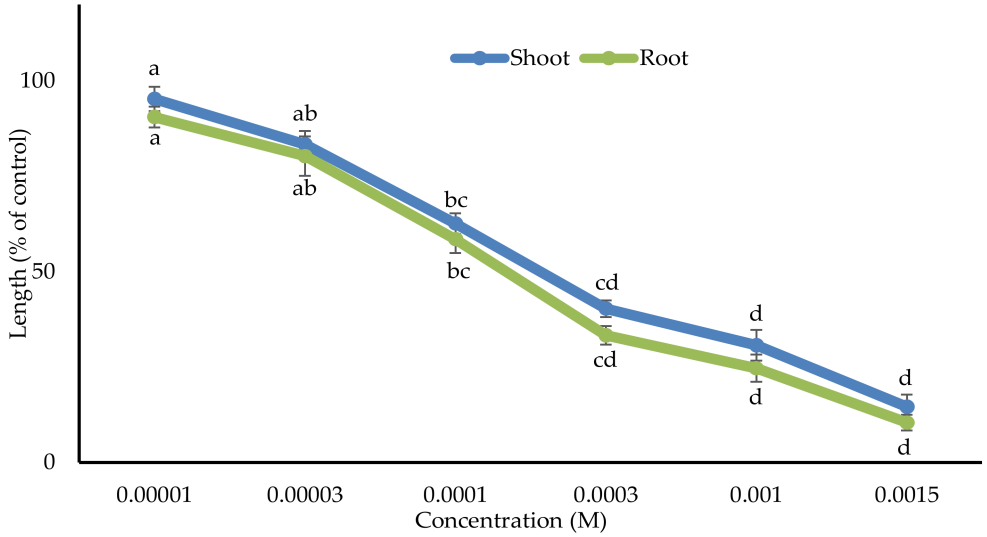

Figure 3. The phytotoxicity of compound (3*R*)-3-hydroxy-β-ionone against cress. Values indicate means ± SE from three replications (n = 30). Significant variations between control and treatment are indicated by various letters (p < 0.05–0.001).

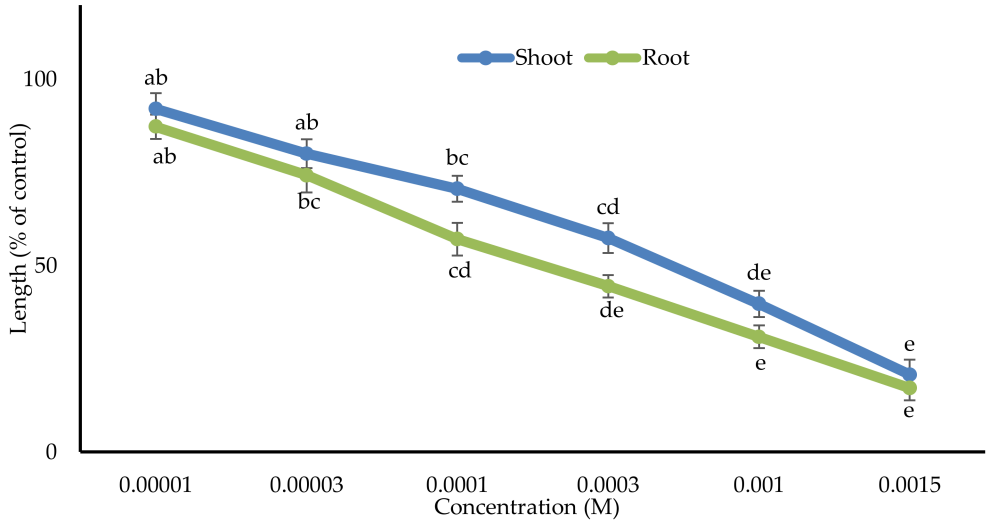

Figure 4. The phytotoxicity of compound *cis*-3-hydroxy-α-ionone against cress. Values indicate means ± SE from three replications (n = 30). Significant variations between control and treatment are indicated by various letters (p < 0.05–0.001).

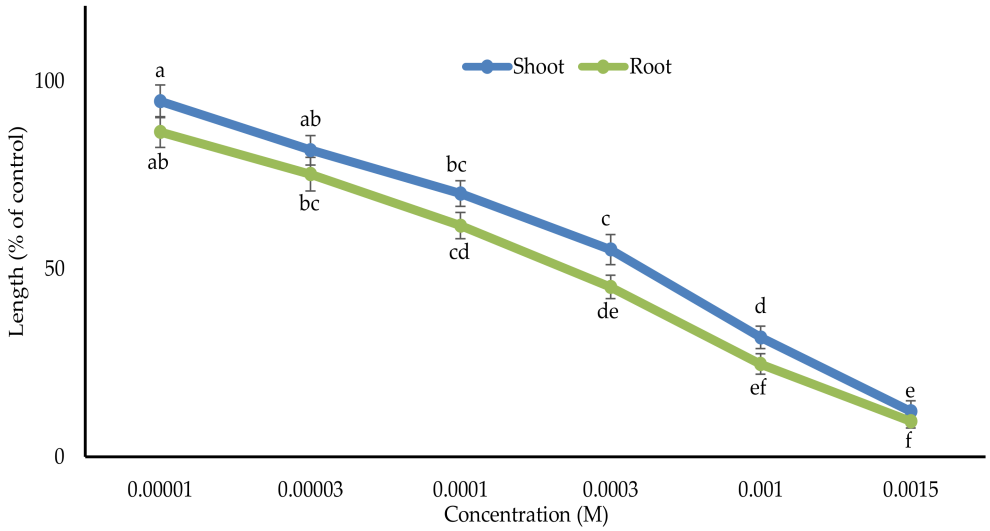

Figure 5. The phytotoxicity of compound loliolide against cress. Values indicate means ± SE from three replications (n = 30). Significant variations between control and treatment are indicated by various letters (p < 0.05–0.001).

Table 2. The concentrations required for 50% growth inhibition (I_{50} value) of the cress seedlings by the identified compounds from the *Elaeocarpus floribundus* leaf extracts.

Test Plant		(3*R*)-3-Hydroxy-β-ionone	*cis*-3-Hydroxy-α-ionone (M)	Loliolide
Cress	Shoot	0.0002	0.0005	0.0004
	Root	0.0001	0.0002	0.0002

4. Discussion

The results of the present study indicated that the *Elaeocarpus floribundus* (aqueous methanolic) leaf extracts significantly inhibited the cress and barnyard grass seedling growth in a dose-dependent way, where the severity of growth suppression was similar to the different treatment concentrations. Several studies have reported this type of dose-dependent phytotoxicity of various plant extracts against a variety of monocotyledonous and dicotyledonous test species, and our findings support those results [36–38]. The tested species in this study showed various levels of inhibition by the *Elaeocarpus floribundus* leaf extracts. The I_{50} values from Table 1 also indicate species-specific phytotoxicity of the extracts. This type of allelopathic activity of plant extracts has been reported by many researchers [39–42]. Our preceding studies with *Albizia richardiana* also show these types of dose-dependent and species-specific inhibitory activity against various tested plants [43–45].

The growth suppression potential of the *Elaeocarpus floribundus* leaf extracts might be due to secondary metabolites (growth inhibitory compounds), which may possess the ability to influence different physiological activities of targeted plants [46]. To develop environmentally friendly natural herbicides, isolating and characterizing secondary metabolites from natural sources (plants) is very important. Therefore, the present study was conducted to isolate allelopathic compounds from the *Elaeocarpus floribundus* leaf extracts, and three phytotoxic compounds were identified as (3*R*)-3-hydroxy-β-ionone, *cis*-3-hydroxy-α-ionone, and loliolide (Figure 2) using several chromatographic methods.

Allelochemicals have a number of advantages, including the ability to inhibit weed development while simultaneously being environmentally friendly. Allelochemicals (secondary metabolites) released from plant sources have been used to develop bioherbicides and to help sustain long-term agricultural production [47]. Phytochemicals have no generic specificity in the cropping system and phytotoxic activity differs from crop to crop. These variations across crop species and the phytotoxic potential of various allelochemicals

are prospects for future study. The bioherbicidal potential of different plant extracts has been demonstrated through the anatomical alteration of seedlings, such as increased lipid globules, reduced mitochondria, and degradation of the membranes of nuclei and mitochondria [48]. The findings of the current study showed that the hypocotyl growth of the examined plant species was less susceptible to the *Elaeocarpus floribundus* leaf extracts than the radical growth. Allelochemicals damage primary radical surfaces more than shoots because of a thinner cuticle layer, which permits transporting more allelopathic substances to the radical cells [49]. Thus, the cell division and cycle are disrupted along with the ultrastructure of the cellular membrane, which are responsible for radical growth inhibition [49,50]. Allelochemicals shrink metaxylem cells in the radicals, which may prevent cell enlargement due to alterations in cytokinins, ethylene, and auxin [51]. The importance of auxin for the growth and development of roots is well understood: changes in the cellular mechanisms reflect disruptions of the enzymatic functions responsible for the biosynthesis of auxin [52]. Different plant extracts influence the synthesis of proteins by causing aberrant upregulation or downregulation of the proteins. In particular, chlorophyll a or chlorophyll b chaining protein and OEEP1 (oxygen evolving enhancer protein 1) are reduced twofold or more, when exposed with plant extract. Suppression of chlorophyll a or chlorophyll b chaining protein synthesis restricted the secretion of the total chlorophyll, which influences photosynthesis [53]. OEEP1 plays an important role in discharging O_2 by water splitting, preventing the cluster of tetra manganese, ionic coverage, and also displays thioredoxin potential [54].

The hormones found in plants act as signaling agents, which influence the growth and development of plants through a variety of metabolic processes. GA (gibberellin) is an important plant growth hormone that enhances hypocotyl growth [55]. Using burcucumber (seed) extracts and 2-linoleoyl glycerol (a phenolic compound of burcucumber seed extract) restricts the GA pathways and promotes the accumulation of ABA (abscisic acid), JA (jasmonic acid), and SA (salicylic acid) [53]. ABA is responsible for closing stomata, a poor rate of photosynthesis, and generating reactive oxygen species (ROS), which decrease the growth of plants and cause senescence. JA also causes the closing of stomata and initiates senescence, which both decrease the rate of photosynthesis [56]. Bioherbicides restrict the growth of weed plants by disrupting nutrient uptake, membrane accessibility, and photosynthesis. They limit the uptake of nutrients (K, Ca, Fe, and Mg) in weed plants by altering the cell membrane functions and structures [57]. The phytotoxic actions of allelochemicals against weeds increase O^{2-} (superoxide), H_2O_2 (hydrogen peroxide), and OH^- (hydroxyl) radicals, causing DNA, protein, and cell membrane damage [58]. The endonucleases, the proteases, and the death of programmed cells are induced by electrolytic shrinkage, inhibiting weed growth [59] and causing necrosis [60]. Research has suggested that allelochemicals may directly limit the antioxidant enzymatic function within cells, causing the generation of high quantities of O_2 (active oxygen), and this oxidative stress ultimately stunts seedling growth [61].

The compound (3*R*)-3-hydroxy-β-ionone is a C_{13}-norisoprenoid, the cleavage output of zeaxanthin, and is found in various stages during the development of fruit [62]. It has been reported that the compound accumulates in the seedlings of bean cultivars through irradiation by light, causing light-effected growth suppression of bean seedlings [63]. (3*R*)-3-hydroxy-β-ionone has also been isolated and identified from different plants and its growth suppression potential against several species is well documented [64–67]. It has been isolated from moss (*Rhynchostegium pallidifolium*) and reported as the main phytotoxic compound [68]. However, there is no report in the literature about identifying this compound from *Elaeocarpus floribundus*. This study is the first to document the presence of the compound and its allelopathic activity from *Elaeocarpus floribundus* leaf extracts. The compound *cis*-3-hydroxy-α-ionone is also a norisoprenoid, an important terpenoid derivative used as an attractant and aroma compound, and present in *Ducrosia anethifolia*, *Bacillus subtilis*, and raspberry [69–73]. It has also been isolated from *Anredera cordifolia* and its biological potential tested [74], but this study is the first to report on the isolation and

phytotoxicity of this compound from *Elaeocarpus floribundus* leaf extracts. Loliolide is a ubiquitous lactone [75] also found through the synthesis of C_{11}-aldehyde [76]. Loliolide has been reported in several plants and animals in different ecosystems (land and sea) [75] and has different pharmaceutical properties such as antioxidant, antifungal, antibacterial, antidiabetic, anticancer, antiviral, antituberculosis, anti-melanogenic, anti-inflammatory, and anti-aging. Previous research has isolated and determined its allelopathic activity [43,77,78], but no documents have been found in the literature about identifying and determining the allelopathic effects of loliolide from the extracts of *Elaeocarpus floribundus*.

The findings of the present study indicated that all three compounds significantly suppressed the growth of the cress seedlings (Figures 3–5). The I_{50} values indicated that (3*R*)-3-hydroxy-β-ionone possesses greater potent phytotoxicity than *cis*-3-hydroxy-α-ionone and loliolide (Table 2). Differences in the allelopathic potential of the isolated compounds may be due to the variations in their structures [79]. Thus, the phytotoxicity of (3*R*)-3-hydroxy-β-ionone, *cis*-3-hydroxy-α-ionone, and loliolide are responsible for the allelopathic effect of *Elaeocarpus floribundus*. Therefore, the allelopathic activity of *Elaeocarpus floribundus* may help to develop bioherbicides and to protect our environment from synthetic herbicide pollution. Although, the allelopathic activity of these compounds are documented in the laboratory condition. Further study needs to be done in the field condition to verify our findings.

5. Conclusions

The leaf extracts of *Elaeocarpus floribundus* showed dose-dependent allelopathic potential against the seedling growth of the examined species. Three allelopathic compounds were isolated from the *Elaeocarpus floribundus* leaf extract and identified as (3*R*)-3-hydroxy-β-ionone, *cis*-3-hydroxy-α-ionone, and loliolide via spectral analysis. These compounds significantly suppressed the growth of the cress seedlings in a dose-dependent way. The findings of this study showed that all three compounds possess allelopathic potential and may be responsible for the phytotoxic activity of *Elaeocarpus floribundus*. Therefore, *Elaeocarpus floribundus* could be used to develop bioherbicides. To confirm this result, we will set a field experiment in future.

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