

# Ecological phytochemistry of Cerrado (Brazilian savanna) plants

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**Abstract** The Cerrado (the Brazilian savanna) is one of the vegetation formations of great biodiversity in Brazil and it has experienced strong deforestation and fragmentation. The Cerrado must contain at least 12,000 higher plant species. We discuss the ecological relevance of phytochemical studies carried out on plants from the Cerrado, including examples of phytotoxicity, antifungal, insecticidal and antibacterial activities. The results have been classified according to activity and plant family. The most active compounds have been highlighted and other activities are discussed. A large number of complex biochemical interactions occur in this system. However, only a small fraction of the species has been studied from the phytochemical viewpoint to identify the metabolites responsible for these interactions.

**Keywords** Ecological phytochemistry · Semiochemical · Biocommunication

## Introduction

Cerrado (the Brazilian savanna) is one of the vegetation formations of great biodiversity in Brazil and it has experienced strong deforestation and fragmentation. Cerrado is a phytogeographic domain in which the savanna physiognomies predominates, but this domain also contains riparian forest, seasonal semi-deciduous forest, seasonal deciduous forest, *campo úmido* (wet grassland), amongst others (Batalha 2011; Forzza et al. 2010; Coutinho 2002). In the Cerrado, the herbaceous and shrubby constituents predominates, both of which are heliophilic and alternate in importance between different physiognomies (Coutinho 2002). As described by Sano et al. (2008), the Cerrado is the most fragmented environment of Brazil (Fig. 1). The original total area was 205 million hectares (Instituto Brasileiro de Geografia e Estatística 2004) and this is the second largest Brazilian domain, after the Amazonian (Klink and Machado 2005; Forzza et al. 2010). Cerrado extends over the States of Bahia, Goiás, Maranhão, Mato Grosso, Mato Grosso do Sul, Minas Gerais, Paraná, Piauí, Tocantins and São Paulo.

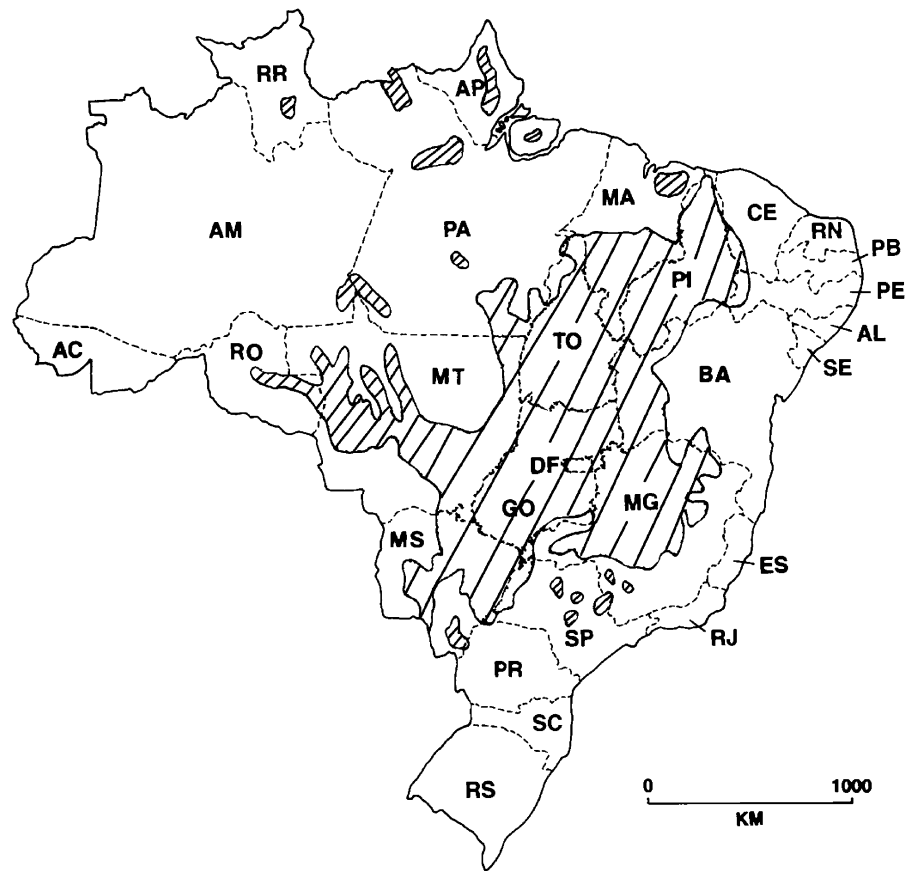
Apart from being a world hotspot (Mittermeier et al. 2004), Cerrado is the richest tropical savanna on the planet, with more than 12,000 plant species, in which at least 30 % are endemic (Klink and Machado 2005; Forzza et al. 2010). As described by Forzza et al. (2010) the richest angiosperms families of the Cerrado are Asteraceae, Fabaceae, Orchidaceae, Poaceae, Eriocaulaceae, Melastomataceae and Rubiaceae. Among

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**Fig. 1** Extent of the domain Cerrado domain in Brazil



the savanna physiognomies, the richest families are Fabaceae, Asteraceae, Poaceae, Rubiaceae, Bignoniaceae, Myrtaceae, Malpighiaceae, Malvaceae and Apocynaceae (Goodland 1979; Ratter 1980; Batalha and Mantovani 2001).

The climate is seasonal with dry winters. The average annual temperatures are around 23 °C and the annual precipitation is between 1,000 and 1,800 mm (Instituto Nacional de Meteorologia 2013). The winters are very dry and the minimum temperatures can reach negative values (Coutinho 2002). The Cerrado is developed on old soils that are hydrated by aquifers and are deep, acidic and oligotrophic, but also with a higher amount of aluminum (Haridasan 2001). Another important factor in Cerrado biodiversity is fire, which naturally occur in short periods in rainy seasons due to lightning storms (Moreira 2000). The flora of the savanna physiognomies shows adaptations to fire, such as thick cork on bark, xylopodium, leaves with dense cuticles and deep roots. Fire during the rainy season facilitates the germination of some seeds,

accelerates the nutrient cycle, induces the sprouting of lateral and subterranean buds and also enables the synchronicity of flowering in some species, thus increasing cross pollination (Coutinho 2002).

The most important threats to the Cerrado conservation are its deforestation and fragmentation, the soil degradation and the dispersion of exotic species of crops and pastures, like soy and African grasses (Carmo et al. 2011; Klink and Machado 2005).

These grasses are the most important invasive alien plants in the Cerrado fragments and agricultural fields placed over Cerrado, especially the grasses *Urochloa decumbens* (Stapf) R.D. Webster and *Melinis minutiflora* P. Beauv. Both species are negatively correlated with native species, suggesting strong competitive behavior (Pivello et al. 1999). African grasses were introduced in Brazil as cattle pasture, but they spread all over the country (Filgueiras 1990; Williams and Baruch 2000). They are present in at least 30 % of Cerrado fragments studied and their presence are strongly favored by the presence of roads and urban

areas (Durigan et al. 2007). The African grasses produce large quantities of biomass that dehydrate during the dry seasons and act as a ‘fuel’ to very fierce fires, that are more frequent, stronger and longer than Cerrado plants can resist (Klink and Machado 2005; Carmo et al. 2011). Fierce fires increase the mortality of plants, especially young individuals and seeds, and result in more open physiognomies, with different patterns of water use and carbon flux efficiencies (Medeiros and Miranda 2005; Miranda et al. 2009a).

Strong increases in the distribution of invasive plants can occur because of lack of natural predators (Callaway et al. 1999) or because they can alter the soil properties, the rate of decomposition and nutrient cycles (Weidenhamer and Callaway 2010). Another way of better establishment of invasive species is by allelopathy. This novel hypothesis predicts that these plants could have secondary metabolites that are new to the invaded community and act as allelochemicals (Callaway and Aschehoug 2000; Callaway and Ride-nour 2004). Therefore, allelochemicals of invasive plants in the Cerrado are an important area of phytochemistry to be investigated.

### Plant families

All species studied in this review had their names and authors revised using the Brazilian list of plant species (Lista de Espécies da Flora do Brasil 2013). The vast majority were angiosperms. A total of 71 species had their bioactivity studied and this represents only 0.60 % of the 12,000 plant species noted by Forzza et al. (2010) in the Cerrado. This value is very small considering that this environment is a world hotspot (Mittermeier et al. 2004). Only 32 families studied in this review have been reported to show bioactivity. The families associated with the highest number of published papers in this respect are Myrtaceae (18 occurrences), Fabaceae (11 occurrences), Bignonia-ceae (3 occurrences) and Melastomataceae (3 occurrences) and these correspond to the families with the highest abundance in the savanna physiognomies (Goodland 1979; Ratter 1980; Batalha and Mantovani 2001).

The Myrtaceae family represents 211 species in 14 genera only in the savanna physiognomies (Cardoso and Sajo 2006), which represents 5 % of the plant

biodiversity in this area (Batalha and Mantovani 2001). Bioactivity studies on Myrtaceae showed phytotoxic and molluscicidal activity of some species. As observed by Keszei et al. (2008), this family is known for the elevated concentration of terpenes in their leaves and for the strong qualitative and quantitative variation of such compounds among the *taxon*, populations and individuals. The results published by Imatomi et al. (2013a) do not indicate grouping of bioactivity for the 15 studied species by a taxonomic approach. The most studied Myrtaceae genera were *Eugenia* L. (6 occurrences), *Myrcia* D.C. (5 occurrences), *Psidium* L. (4 occurrences) and *Stryphnodendron* Mart. (3 occurrences).

The Fabaceae family represents 14 % of the species of the savanna physiognomies (Batalha and Mantovani 2001). Fabaceae have higher concentrations of nitrogen in their tissues than other plants because their roots show symbiosis with *Rhizobium* bacteria, which are able to fix atmospheric nitrogen (Vitousek et al. 2010). Therefore, this family has an advantage on the oligotrophic soils of the Cerrado (Bustamante et al. 2006), which could be one of the reasons why it is widely distributed in this domain. The genus associated with the most studies concerning bioactivity is *Stryphnodendron* (3 occurrences). The results showed phytotoxic, molluscicidal, fungicidal and antimicrobial activity for some species.

The Bignoniaceae family represents 5 % of the plants of the savanna physiognomies (Batalha and Mantovani 2001). Many Bignoniaceae species are used in medicine because of their bactericidal, fungicidal and antiophidic properties (Hiruma-Lima and Di Stasi 2002; Portillo et al. 2001; Núñez et al. 2004; Park et al. 2005). There was no predominance of genus in the bioactivity studies. The publications concerning Bignoniaceae describe phytotoxic, fungicidal and antimicrobial activities of some species. Iridoids and flavonoids are compounds that are normally present in this family (Blatt et al. 1998; Watson and Dallwitz 2007).

Melastomataceae also appears in many publications on bioactivity. This is the sixth most abundant family in the savanna physiognomies (Batalha and Mantovani 2001). The genus *Miconia* Ruiz & Pav. was the most commonly described (2 occurrences). The publications covered phytotoxic and molluscicidal activity.

## Plant parts

The most widely used plant parts in the studied species in the present review were leaves but secondary metabolites can be found in all plant tissue, including stems, flowers, fruits, seeds, roots and rhizomes (Putnan and Tang 1986). As described by Friedman (1995), all plant organs have the potential to store these compounds, but the quantities and the manner in which they are produced differ depending on the species. However, leaves and roots are normally the main sources of these substances (Wu et al. 2009). Nutrients are costly to Cerrado plant species because these plants are exposed to oligotrophic soils and fire (Moreira 2000; Haridasan 2001). To protect their foliage from herbivores, leaves usually use defence mechanisms such as coriaceous texture, trichomes and elevated amounts of phenolic compounds and tannins (Marquis et al. 2002; Corrêa et al. 2008; DaSilva and Batalha 2011). Leaves extracts and leaves isolated compounds of Cerrado plants showed all studied bioactivities, phytotoxic (Tables 1, 2, 3), molluscicidal (Table 4), insecticide (Table 5), fungicide (Table 6) and antibacterial (Table 7).

Barks and roots were the most used plant parts after leaves. Not only thick cork on bark can be present in Cerrado plants to prevent from herbivore, but also secondary metabolites can play a role on this function. This is the case of *Stryphnodendron adstringens* (Mart.) Coville and *S. polyphyllum* Mart. which shows condensed tannins on bark (Bezerra et al. 2002). The revised publications showed barks and stems of Cerrado plants with phytotoxic (Tables 1, 2, 3), molluscicidal (Table 4), insecticide (Table 5), fungicide (Table 6) and antibacterial (Table 7) activities.

On the other hand many secondary metabolites can be synthesized and stored in the roots (Flores et al. 1987). Some of them can even be exudate, transforming the environment of the donor plant, as mediators of mineral acquisitions (Dakota and Phillips 2002), reducing pathogens or inhibiting other plants by allelopathy (Bertin et al. 2003; Bais et al. 2006). Roots of Cerrado plants showed phytotoxic (Tables 1, 2, 3), insecticide (Table 5), and antibacterial (Table 7) activities.

The occurrence of fire in the Cerrado is an important factor to be considered when choosing plant parts to be phytochemically studied because, in the

case of a fire, a high percentage of the plant community will present young leaves after a while. Young leaves commonly contain higher amounts of tannins and phenols than mature leaves (Coley 1983; Langenheim et al. 1986) and they therefore show higher allelopathic activity (Marchi et al. 2008). On the other hand, roots are normally not affected by fire, especially those that happen during rainy season.

## Allelopathic activity

From the viewpoint of chemical ecology, the most frequent publications have concerned allelopathy (Tables 1, 2), although most of these studies were performed on extracts and their phytotoxic activity was tested on the germination and growth of cultivated species, under laboratory conditions. Many authors as Inderjit and Callaway (2003), Kaur et al. (2009), Souza Filho et al. (2010) and Meiners et al. (2012), discuss that these assays do not represent the natural conditions present in the ecosystems. For example, some publications showed that the microbiota can change allelochemical activity (Inderjit et al. 2008; Kaur et al. 2009). However, phytotoxicity assays can be used to biodirect the isolation and identification of the compounds responsible for phytotoxicity (Inderjit and Weston 2000; Macías et al. 2000). After the identification of these substances, they can be used on field studies, as in the evaluation of their concentration on soil, the interaction with other substances and microorganisms, their mode of action and others (Blum 1999; Inderjit and Weston 2000; Inderjit and Duke 2003). These are important aspects that should be studied in Cerrado plants to confirm their allelopathic activity.

Even phytotoxic studies have only been developed very recently and it is worth highlighting the screening conducted by Gorla and Perez (1997) on the species *Miconia albicans* (Sw.) Steud., *Lantana camara* L., *Leucaena leucocephala* (Lam.) de Wit and *Drimys winteri* J.R. Forst & G. Forst.; Silva et al. (2006) on 15 native tree species; Maraschin-Silva and Áquila (2006) on *Erythroxylum argentinum* O.E. Schulz, *Luehea divaricate* Mart., *Myrsine guianensis* (Aubl.) Kuntze and *Ocotea puberula* Rich Nees.; Gatti et al. (2007) on 12 different species of a variety of genera and families, and the recent contribution of Imatomi

**Table 1** Species with phytotoxic activity in extracts

Species	Family	Plant part	References
<i>Xylopia aromatica</i> (Lam.) Mart.	Annonaceae	Leaves	Gatti et al. (2007)
<i>Davilla elliptica</i> A.St.-Hil.	Dilleniaceae	Leaves	Gatti et al. (2007)
<i>Erythroxylum argentinum</i> O.E. Schulz	Erythroxylaceae	Leaves	Maraschin-Silva and Áquüila (2006)
<i>Senna occidentalis</i> L. Link	Fabaceae, Caesalpinioideae	Leaves, stem	Candido et al. (2010)
<i>Hymenaea stigonocarpa</i> Mart. Ex. Hayne	Fabaceae: Caesalpinioideae	Leaves, fruits	Oliveira et al. (2002)
<i>Machaerium acutifolium</i> Vogel	Fabaceae: Faboideae	Leaves	Povh et al. (2007)
<i>Anadenanthera falcata</i> (Benth.) Speg.	Fabaceae: Mimosoideae	Leaves	Gatti et al. (2007)
<i>Stryphnodendron adstringens</i> Mart. Coville	Fabaceae: Mimosoideae	Leaves	Silva et al. (2006)
<i>Leucaena leucocephala</i> (Lam.) de Wit	Fabaceae: Mimosoideae	Leaves	Gorla and Perez (1997)
<i>Andira humilis</i> Mart. ex Benth	Fabaceae: Papilionoideae	Leaves, stem	Periotto et al. (2004)
<i>Ocotea puberula</i> Rich Nees.	Lauraceae	Leaves	Maraschin-Silva and Áquüila (2006)
<i>Ocotea odorifera</i> Rohwer	Lauraceae	Leaves, stem, roots	Gatti et al. (2008)
<i>Luehea divaricata</i> Mart.	Malvaceae	Leaves	Maraschin-Silva and Áquüila (2006)
<i>Miconia albicans</i> (Sw.) Steud.	Melastomataceae	Leaves	Gorla and Perez (1997), Gatti et al. (2007)
<i>Trembleya parviflora</i> (D. Don) Cogn.	Melastomataceae	Leaves	Borghetti et al. (2005)
<i>Eugenia dysenterica</i> D.C.	Myrtaceae	Leaves	Borghetti et al. (2005), Pina et al. (2009), Giotto et al. (2007)
<i>Campomanesia adamantium</i> (Cambess. O. Berg.)	Myrtaceae	Leaves	Borghetti et al. (2005)
<i>Blepharocalyx salicifolius</i> Kuth O. Berg	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Campomanesia pubescens</i> O. Berg	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Psidium australe</i> Cambess.	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Psidium cinereum</i> Mart.	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Psidium laruotteanum</i> Cambess.	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Psidium rufum</i> Mart. Ex DC.	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Eugenia bimarginata</i> O. Berg	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Eugenia klotzschiana</i> O. Berg	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Eugenia myrcianthes</i> Nied.	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Eugenia puniceifolia</i> (Kunth) DC.	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Myrcia bella</i> Cambess.	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Myrcia lingua</i> (O. Berg) Mattos	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Myrcia multiflora</i> DC.	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Myrcia splendens</i> DC.	Myrtaceae	Leaves	Imatomi et al. (2013a)
<i>Myrcia tomentosa</i> (Aubl.) DC.	Myrtaceae	Leaves	Imatomi et al. (2013b)
<i>Ouratea spectabilis</i> (Mart. ex Engl.) Engl.	Ochnaceae	Leaves	Silva et al. (2006)
<i>Sapindus saponaria</i> L.	Sapindaceae	Leaves	Grisi et al. (2012)
<i>Pouteria ramiflora</i> (Mart.) Radlk.	Sapotaceae	Leaves	Silva et al. (2006)
<i>Pouteria torta</i> (Mart.) Radlk.	Sapotaceae	Leaves	Nascimento et al. (2007)
<i>Siparuna guianensis</i> Aubl.	Siparunaceae	Leaves	Gatti et al. (2007)
<i>Solanum lycocarpum</i> A. St.-Hil.	Solanaceae	Leaves	Oliveira et al. (2004), Aires et al. (2005)

**Table 1** continued

Species	Family	Plant part	References
<i>Solanum palinacanthum</i> Dunal	Solanaceae	Leaves	Oliveira and Campos (2006)
<i>Lantana camara</i> L.	Verbenaceae	Leaves	Gorla and Perez (1997)
<i>Qualea grandiflora</i> Mart.	Vochysiaceae	Leaves	Silva et al. (2006)
<i>Qualea parviflora</i> Mart.	Vochysiaceae	Leaves	Borghetti et al. (2005)
<i>Drimys winteri</i> J.R. Forst and G. Forst.	Winteraceae	Leaves	Gorla and Perez (1997)

**Table 2** Species tested with no phytotoxic activity

Species	Family	Plant part	References
<i>Aspidosperma tomentosum</i> Mart.	Apocynaceae	Leaves	Silva et al. (2006), Gatti et al. (2007)
<i>Schefflera vinosa</i> (Cham. and Schldtl.) Frodin and Fiaschi	Araliaceae	Leaves	Gatti et al. (2007)
<i>Didymopanax vilosum</i> E. March	Araliaceae	Leaves	Silva et al. (2006)
<i>Kielmeyera variabilis</i> Mart. and Zucc.	Calophyllaceae	Leaves	Silva et al. (2006)
<i>Kielmeyera coriacea</i> Mart. and Zucc.	Calophyllaceae	Leaves	Gatti et al. (2007)
<i>Diospyros hispida</i> A. DC.	Ebenaceae	Leaves	Gatti et al. (2007)
<i>Anadenanthera falcata</i> (Benth.) Spig.	Fabaceae: Mimosoideae	Leaves	Silva et al. (2006)
<i>Machaerium villosum</i> Vogel	Fabaceae: Faboideae	Leaves	Silva et al. (2006)
<i>Senna rugosa</i> (G. Don) H. S. Irwin and Barneby	Fabaceae: Caesalpinaceae	Leaves	Gatti et al. (2007)
<i>Stryphnodendron polyphyllum</i> Mart.	Fabaceae: Mimosaceae	Leaves	Gatti et al. (2007)
<i>Byrsonima coccolobifolia</i> Kunth	Malpighiaceae	Leaves	Silva et al. (2006)
<i>Byrsonima verbascifolia</i> (L.) DC.	Malpighiaceae	Leaves	Silva et al. (2006)
<i>Rapanea guianensis</i> Aubl.	Myrsinaceae	Leaves	Silva et al. (2006)
<i>Styrax ferrugineus</i> Nees and Mart.	Styracaceae	Leaves	Silva et al. (2006)
<i>Vochysia tucanorum</i> Mart.	Vochysinaceae	Leaves	Silva et al. (2006)

et al. (2013a) on 15 species of the Myrtaceae family. However, as discussed above, the number of species in question is very small compared to the biodiversity present in the Cerrado. Although this number may seem too small to be representative, most studies have uncovered significant effects, suggesting that allelopathy plays an important role in the ecology of Cerrado.

As mentioned previously, the Myrtaceae family has been the most widely studied. In all cases, tests were carried out on leaf extracts. Of the 15 species studied by Imatomi et al. (2013a), 12 showed activity. *Blepharocalyx salicifolius* Kuth O. Berg, *Psidium australe* Cambess., *Eugenia puniceifolia* (Kunth) DC., *Myrcia multiflora* DC. and *Myrcia splendens* DC. showed higher levels of activity against lettuce, onion and tomato seedlings. *Eugenia dysenterica* DC. (Borghetti et al., 2005; Pina et al., 2009; Giotto et al., 2007) and *Campomanesia adamantium* (Cambess. O. Berg.) (Borghetti et al., 2005) have also been studied from this

family. Extracts from *E. dysenterica* showed a high level of activity on sesame and radish (*Raphanus sativus* L.) seedlings. The activity was expressed on root development, morphology and root hairs. *E. dysenterica* also showed similar effects when tested against lettuce. This family has been studied from the phytochemical viewpoint and monoterpenes, sesquiterpenes, triterpenes, flavonoids, tannins and alkaloids have all been identified. However, there is only one precedent of an allelopathy study in which bio-guided isolation was carried out and this involved *M. tomentosa* (Imatomi et al., 2013b). Two major flavonoids were isolated from the most active fractions of this plant: juglanin (1) and aviculin (2). The presence of these compounds could be related to the activity observed in extracts (Fig. 2). The latter compound showed activity in other bioassays and it was active on DPPH radical scavenging, 15-lipoxygenase and xanthine oxidase inhibition (Le et al. 2012).

**Table 3** Species with phytotoxic activity and identified compounds

Species	Family	Plant part	Compounds identified	References
<i>Aristolochia esperanzae</i> Kuntze	Aristolochiaceae	Leaves, stem, roots	Fargesin	Gatti et al. (2004, 2010)
<i>Memora peregrina</i> (Miers) Sandwith	Bignoniaceae	Roots, leaves	Allantoin, (+),6 $\beta$ -hydroxyipolamiide, hyperin, 3'- <i>O</i> -methylhyperin, 4-hydroxy- <i>N</i> -methylproline, $\beta$ -sitosterol, $\alpha$ -amirin, $\beta$ -amirin, lupeol and others	Grassi et al. (2005)
<i>Caryocar brasiliense</i> Cambess.	Caryocaceae	Fruits	Ethyl gallate, 5-hydroxyfurfural, gallic acid, methyl shikimate, and mixtures of $\beta$ -D-fructopyranose and $\beta$ -D-fructofuranose, $\alpha$ - and $\beta$ -D-glucose, lupeol and oleic acid and $\beta$ -sitosterol, stigmasterol and oleic acid	Ascari et al. (2010)
<i>Byrsonima crassa</i> Nied.	Malpighiaceae		Amenthoflavone	Almeida et al. (2007)
<i>Myrcia tomentosa</i> (Aubl.) DC.	Myrtaceae	Leaves	Avicularin and juglanin	Imatomi et al. (2013b)
<i>Rapanea umbellata</i> (Mart.) Mez	Primulaceae	Leaves, Stem, roots	Lutein, (–)-catechin, 3-geranyl-4-hydroxy-5-(3'',3''-dimethylallyl)-benzoic acid, 5-carboxy-7-(3'',3''-dimethylallyl)-2-(1'-hydroxy-1',5'-dimethylhex-4'-enyl)-2,3-dihydro-benzofuran and 6-carboxy-8-(3'',3''-dimethylallyl)-3 $\alpha$ -hydroxy-2a-methyl-2-(4'-ethylpent-3'-enyl)-3,4-dihydrobenzopyran	Novaes et al. (2013a,b), Januário et al. (1991, 1992)
<i>Rapanea ferruginea</i> (Ruiz and Pav.) Mez	Primulaceae	Leaves	3-Geranyl-4-hydroxy-5-(3'',3''-dimethylallyl)-benzoic acid, 5-carboxy-7-(3'',3''-dimethylallyl)-2-(1'-hydroxy-1',5'-dimethylhex-4'-enyl)-2,3-dihydrobenzofuran	Novaes (2011), Januário et al. (1991)
<i>Myrsine guianensis</i> (Aubl.) Kuntze	Primulaceae	Leaves	5-Carboxy-7-(3'',3''-dimethylallyl)-2-(1'-hydroxy-1',5'-dimethylhex-4'-enyl)-2,3-dihydro-benzofuran, 6-carboxy-8-(3'',3''-dimethylallyl)-3 $\alpha$ -hydroxy-2a-methyl-2-(4'-methylpent-3'-enyl)-3,4-dihydrobenzopyran, 24( <i>E</i> )-3-oxo-dammara-20,24-dien-26-al, 24( <i>E</i> )-3 $\alpha$ -hydroxycycloart-24-en-26-al	Maraschin-Silva and Áquila (2006), Januário et al. (1992)

The Fabaceae family is also among the most studied and 7 of 11 species have shown phytotoxic potential in extracts: *Hymenaea stigonocarpa* Mart. Ex. Hayne (Oliveira et al., 2002), *Machaerium acutifolium* Vogel (Povh et al., 2007), *Anadenanthera falcata* (Benth.) (Gatti et al., 2007), *S. adstringens* Mart. Coville (Silva et al., 2006) and *L. leucocephala* (Lam.) de Wit (Gorla and Perez, 1997). Most of the assays were conducted on lettuce seeds but seeds from other species such as tomato, onion, radish, wheat and cucumber were also investigated. The most markedly affected parameters were the germination percentage (*S. adstringens*), the germination rate (*A. falcata*, *Andira humilis* Mart. ex Benth, *Senna occidentalis* L. Link) or both (*H. stigonocarpa*, *M. acutifolium*), and the root growth (*S. occidentalis*, *A. humilis*). These studies did not

encompass the isolation of the products responsible for the activity, although the types of metabolites present in the active fractions were assigned in some publications. For example, *S. adstringens* was the most active arboreal species of those studied by Silva et al. (2006), with its extracts showing activity at a concentration of 2 % (w/v). This activity was attributed to the presence of terpenoids. Another case is *L. leucocephala*, which showed stimulatory activity on cucumber germination and tomato root growth. This effect was attributed to the presence of alkaloids and catechinic tannins (Gorla and Perez 1997). The study of *H. stigonocarpa* (Oliveira et al., 2002) is also worth highlighting as the inhibitory effect remained in the soil for at least 90 days, with the highest activity produced by the fruit extract.

**Table 4** Species with molluscicidal activity and identified compounds

Species	Family	Plant part	Compounds identified	References
<i>Schefflera vinosa</i> (Cham. and Schtdl.) Frodin and Fiaschi	Araliaceae	Aerial	Betulin, quercetin 3- <i>O</i> - $\beta$ -D-rhamnoside	Cunha et al. (2012)
<i>Kielmeyera variabilis</i> Mart. and Zucc.	Calophyllaceae	Stem	Assiguxanthone-B, kielcorin and 1,3,5,6-tetrahydroxy-2-prenylxanthone), 2,5-dihydroxy benzoic acid	Pinheiro and Cortez (2003)
<i>Caryocar brasiliense</i> Cambess	Caryocaceae	Leaves, fruits	Ethyl gallate, 5-hydroxyfurfural, gallic acid, methyl shikimate, and mixtures of $\beta$ -D-fructopyranose and $\beta$ -D-fructofuranose, $\alpha$ - and $\beta$ -D-glucose, lupeol and oleic acid and $\beta$ -sitosterol, stigmaterol and oleic acid	Bezerra et al. (2002), Ascari et al. (2010)
<i>Stryphnodendron adstringens</i> (Mart.) Coville	Fabaceae: Mimosoideae	Leaves, bark	Condensed tannins	Bezerra et al. (2002)
<i>Stryphnodendron polyphyllum</i> Mart.	Fabaceae: Mimosoideae	Leaves, bark	Condensed tannins	Bezerra et al. (2002)
<i>Miconia langsdorffii</i> Cogn.	Melastomataceae	Aerial	Oleanolic, ursolic acids	Cunha et al. (2012)
<i>Eugenia dysenterica</i> D.C.	Myrtaceae	Leaves	Condensed tannin, flavonoids, coumarins, phenolic acids and condensed tannin	Bezerra et al. (2002)
<i>Eugenia malaccensis</i> L.	Myrtaceae	Leaves, stem	Miricetine, mearnsetine, gallic acid, 3 $\beta$ - <i>O</i> -acetyl-urs-12-en-28-oic acid	Oliveira et al. (2006)
<i>Roupala montana</i> Aubl.	Proteaceae	Aerial	Quercetin 3- <i>O</i> - $\beta$ -D-glucoside, quercetin 3- <i>O</i> - $\beta$ -D-glucopyranosyl-(1-2)- $\alpha$ -l-rhamnopyranoside, isorhamnetin, 3- <i>O</i> - $\beta$ -D-glucopyranosyl-(1 $\rightarrow$ 2)- $\alpha$ -l-rhamnopyranoside	Cunha et al. (2012)

**Table 5** Species with insecticide activity and identified compounds

Species	Family	Plant part	Compounds identified	References
<i>Annona coriacea</i> Mart.	Annonaceae	Seeds, leaves	Anonaine, annoretine, romucosine and xylopine	Costa et al. (2012), Egydio et al. (2013)
<i>Memora peregrina</i> (Miers) Sandwith	Bignoniaceae	Roots, leaves	Allantoin, (+),6 $\beta$ -hydroxyipolamiide, hyperin, 3'- <i>O</i> -methylhyperin, 4-hydroxy- <i>N</i> -methylproline, $\beta$ -sitosterol, $\alpha$ -amirin, $\beta$ -amirin, lupeol and others	Grassi et al. (2005)
<i>Ocotea velloziana</i> (Meisn.) Mez	Lauraceae	Bark	(+)-Dicentrine	Garcez et al. (2009)
<i>Eugenia malaccensis</i> L.	Myrtaceae	Leaves, stem	Miricetine, mearnsetine, gallic acid, 3 $\beta$ - <i>O</i> -acetyl-urs-12-en-28-oic acid	Oliveira et al. (2006)
<i>Magonia pubescens</i> A. St. Hil.	Sapindaceae	Bark	Catechinic tannin	Silva et al. (2004)

Further 37 species have been studied, of which 26 were active. The screening of 12 species from Cerrado carried out by Gatti et al. (2007) highlighted activities for the species *Davilla elliptica* A.St.-Hil., *M. albicans* (Sw.) Steud. and, in particular, *Siparuna guianensis* Aubl. on the germination rates of lettuce and sesame.

The aqueous extracts of other species showed significant activity on the growth of lettuce [*M. guianensis* and *O. puberula* (Maraschin-Silva et al., 2006)] and on the growth of sesame [*Qualea parviflora* Mart. and *Trembleya parviflora* (D. Don) Cogn. (Borghetti et al., 2005)]. A detailed fractionation of extracts from



**Table 6** Species with fungicidal activity and identified compounds

Species	Family	Plant part	Compounds identified	References
<i>Arrabidaea brachypoda</i> Bureau	Bignoniaceae	Leaves	3',4'-Dihydroxy-5,6,7-trimethoxyflavone, cirsiolol, cirsimaritin and hispidulin	Alcerito et al. (2002)
<i>Tabebuia caraiba</i> (Mart.) Bureau		Leaves		Silva et al. (2009)
<i>Kielmeyera coriacea</i> Mart. & Zucc	Clusiaceae	Leaves		Silva et al. (2009)
<i>Stryphnodendron obovatum</i> Benth.	Fabaceae: mimosoideae	Bark	Epigallocatechin, gallocatechin and epigallocatechin-(4 $\beta$ $\rightarrow$ 8)-gallocatechin	Sanches et al. (2005)
<i>Stryphnodendron adstringens</i> (Mart.) Coville	Fabaceae: mimosoideae	Leaves		Silva et al. (2009)
<i>Hyptis cretana</i> Pohl. Ex Benth.	Labitae	Leaves		Violante et al. (2012)
<i>Ocotea corymbosa</i> (Meisn.) Mez	Lauraceae	Leaves	(2'S)-2-(propan-2'-ol)-5-hydroxy-benzopyran-4-one, 2,3-dihydro-2-methyl-benzopyran-4,5-diol, 2-methyl-5-methoxy-benzopyran-4-one, (2R)-2,3-dihydro-2-methyl-5-methoxy-benzopyran-4-one	Teles et al. (2005)
<i>Eugenia dysenterica</i> D.C.	Myrtaceae	Leaves	$\beta$ -Caryophyllene, $\alpha$ -humulene, limonene, $\alpha$ -thujene, $\alpha$ -terpineol, $\beta$ -caryophyllene oxide	Costa et al. (2000)
<i>Tocoyena formosa</i> (Cham. and Schltdl.) K. Shum	Rubiaceae	Leaves	$\alpha$ - and $\beta$ -Gardiol, and the new iridoids, mollugoside methyl ester and formosinoside	Bolzani et al. (1997)
<i>Renealmia alpinia</i> Rottb. Maas	Zingiberaceae	Leaves		Silva et al. (2009)

*Solanum lycocarpum* A. St. Hil. was carried out and the fractions of intermediate polarity showed the highest activity (Oliveira et al. 2004; Aires et al. 2005).

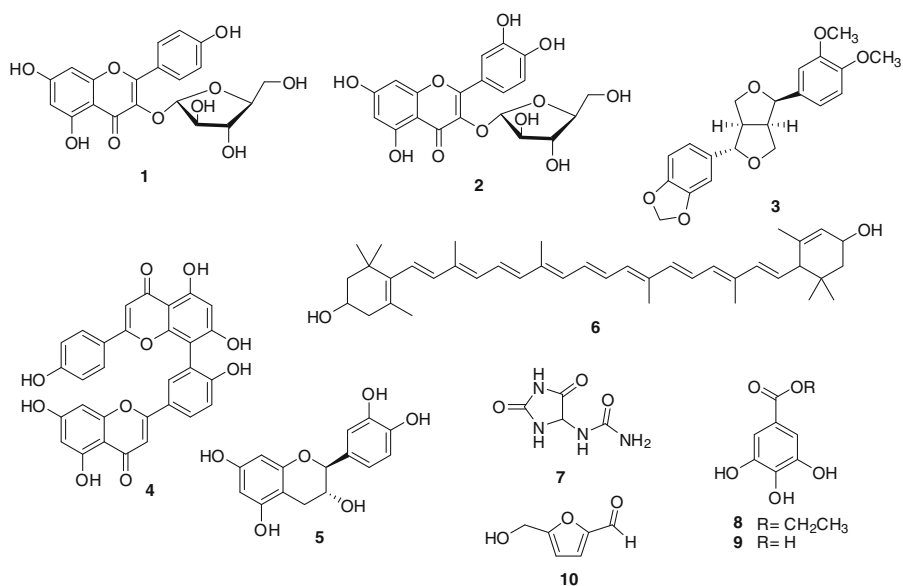
The secondary metabolite composition of these species is not well known and only a limited number of components have been identified from eight species (Fig. 2) (Table 3). The activity of the extracts of various organs of *Aristolochia esperanzae* Kuntze on lettuce and radish have been studied (Gatti et al. 2004, 2010). The most active extracts were those obtained from the roots and these affected the number and size of lateral roots of target plants. The presence of fargesin (3) has been demonstrated in these root extracts (Gatti et al. 2004, 2010) and this lignan has shown various activities, e.g. against platelet activating factor in the [3H]PAF receptor binding assay (Pan et al. 1987), histamine release in rat peritoneal mast cells (RCMP) (Shen et al. 2008), trypanomastigotes in vitro (Sartorelli et al. 2010) and it also improves dyslipidemia and hyperglycemia (Lee et al. 2012).

*Byrsonima crassa* also showed stimulatory activity on the germination and growth of tomato (Almeida et al. 2007). Amentoflavone (4), a dimeric flavone, was isolated and showed identical activity, thus explaining, at least partially, the activities of the extracts.

Three species from the family Primulaceae have shown activity: *M. guianensis*, *Rapanea ferruginea* (Ruiz & Pav.) Mez and *Rapanea umbellata* (Mart.) Mez, all three of which were studied from a phytochemical viewpoint by Januário et al. (1991, 1992). The compounds isolated from *M. guianensis* or *R. ferruginea* have not been tested for their bioactivity and therefore phytotoxic activity cannot be attributed to any. The third of these species was studied by following the standard methodology in allelopathy, i.e. bio-guided isolation (Novaes et al. 2013a, b). The most active fractions afforded (–)-catechin (5) and lutein (6). The latter compound showed phytotoxic activity whereas the first showed practically no activity. The lutein results are consistent with those

**Table 7** Species with antibacterial activity and identified compounds

Species	Family	Plant part	Compounds identified	References
<i>Hancornia speciosa</i> Gomes	Apocynaceae		Flavonoids, tannins, steroids and triterpenoids	Costa et al. (2008)
<i>Arrabidaea harley</i> A.H. Gentry	Bignoniaceae	Bark	Verbascoside and isoverbascoside	Lima et al. (2003)
<i>Austroplenckia populnea</i> (Reissek) Lundell	Celastraceae	Bark, wood, root	Friedelin, 3 $\beta$ -friedelinol, 28-hydroxyfriedelin (canofilol), populnic acid, catonic acid, epikatic acid, pristimerin, abruslactone A and $\alpha$ -amirin	Miranda et al. (2009b)
<i>Bowdichia virgilioides</i> Kunth	Fabaceae: Faboideae	Bark		Silva Junior et al. (2009)
<i>Croton urucurana</i> Baill.	Euphorbiaceae	Bark		Silva Junior et al. (2009)
<i>Hyptis cretana</i> Pohl. Ex Benth.	Labiatae	Leaves		Violante et al. (2012)
<i>Virola surinamensis</i> (Rol. Ex Rottb.) Warb.	Myristicaceae	Bark	Steroids, lignans, flavonoids and polyketide	Costa et al. (2008)
<i>Qualea grandiflora</i> Mart.	Vochysiaceae	Bark	C-methyl phenolics and flavones	Costa et al. (2008)

**Fig. 2** Allelochemicals isolated from Cerrado species

obtained previously on *Withania aristata* Pauq. (Solanaceae) (Llanos et al., 2010) as well as other activities found for this molecule, including inhibition of aflatoxin synthesis (Norton 1997). The results for (–)-catechin contradict some previous results, which indicated that this compound is responsible for the activity of *Centaurea maculosa* Lam. (Asteraceae) (Bais et al., 2003).

Various metabolites have been isolated from *Memora peregrine* (Miers) Sandwith, of which allantoin (7)

(Grassi et al. 2005) should be highlighted. This substance was found in a greater proportion in the underground organs and it showed significant in vitro stimulation of lettuce root growth.

The last of the species with phytotoxic activity, for which the composition has been studied, is *Caryocar brasiliense* Cambess. (Ascari et al., 2010). A number of metabolites have been isolated from this plant. Three of these compounds showed phytotoxic activity: ethyl gallate (8), gallic acid (9) and 5-hydroxymethylfurfural

(10). Ethyl gallate was isolated in high yield (14 % of the crude extract), whereas gallic acid showed the highest levels of bioactivity. Ethyl gallate has also been associated with cytotoxicity (Saleem et al. 2002) and antimicrobial activity (Ooshiro et al. 2009). Gallic acid has also been given a role in phytotoxic interactions of *Fagopyrum esculentum* Moench. (Polygonaceae) (Iqbal et al., 2003), along with other activities like antiaflatoxin (Prakash et al. 2012), fungicidal (Wu 2006) and molluscicidal (Lahlou 2004).

### Molluscicidal and insecticidal activity

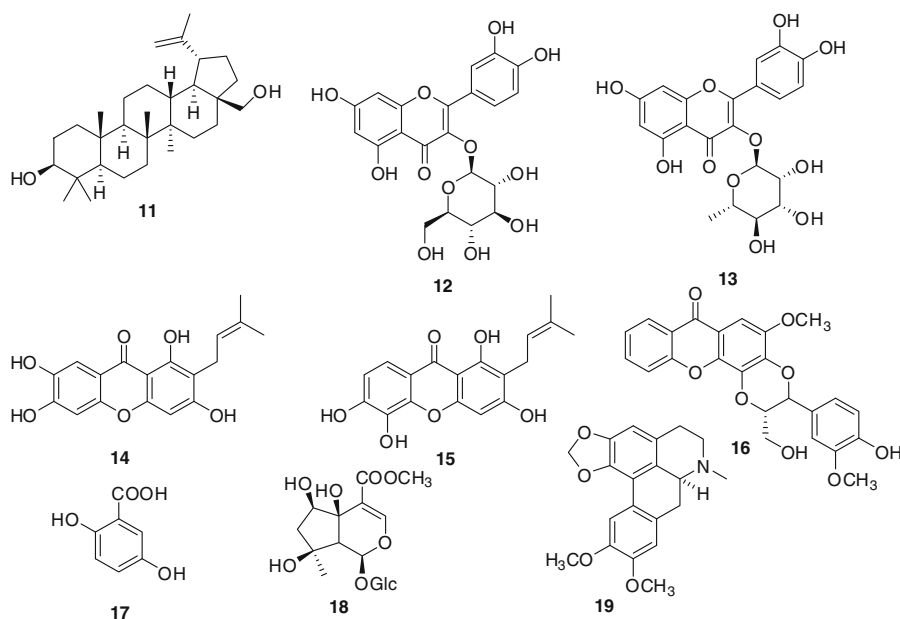
In general, the study of molluscicidal and insecticidal activity has been carried out in the context of health benefits through an ecological approach, with some exceptions that will be discussed. The main problem that can be addressed in this way is schistosomiasis. There are basically two strategies, defence against the parasite *Schistosoma mansoni* (a trematode) or against vector, mainly *Biomphalaria glabrata*—an aquatic mollusc (Table 4).

Several species have been tested against *S. mansoni*, of which three have provided active extracts: *Schefflera vinosa* (Cham. & Schltld.) Frodin & Fiaschi, *Miconia langsdorffii* Cogn. and *Roupala montana* Aubl. (Cunha et al., 2012). A phytochemical study of these species afforded several triterpenes and

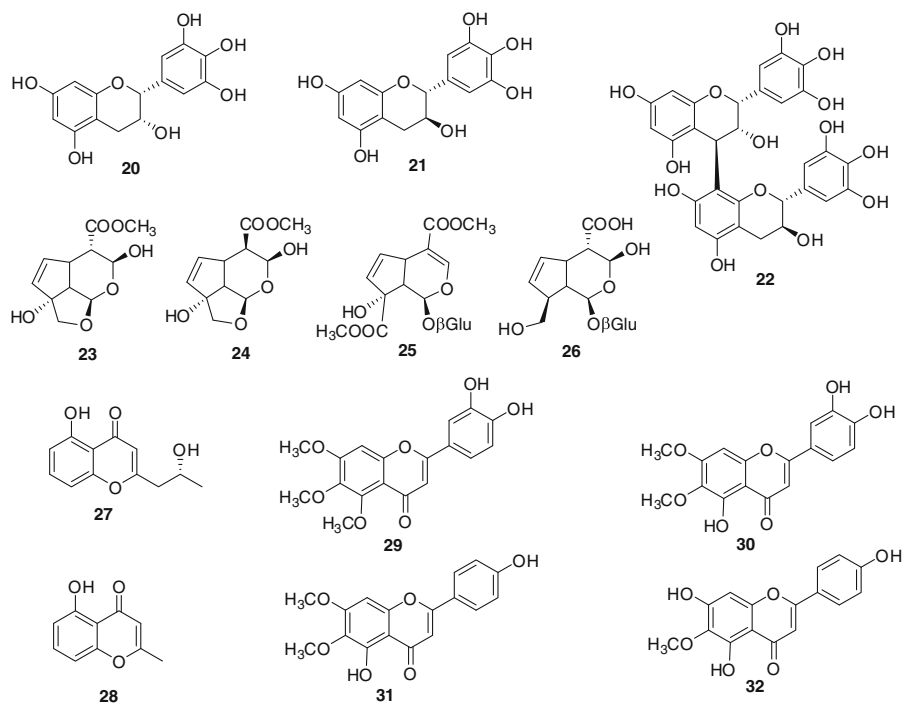
glycosylated flavonoids (Fig. 3). Three compounds induced death or reduced the motor function of the worm: the triterpene betulin (11), isolated from *S. vinosa*, and two monoglycosylated quercetin derivatives isolated from *R. montana*, namely quercetin 3-*O*- $\beta$ -D-glucoside (12) and quercetin 3-*O*- $\beta$ -D-rhamnoside (13). The triterpene 11 has a broad activity profile and the activities described include phytotoxic (Macias et al. 1994), molluscicidal (Yadav and Singh 2009), fungicidal (Jasicka-Misiak et al. 2010), insecticidal (Santos and Salvador 2007) and antimicrobial (Sinha et al. 2009). There are also precedents for the activity of the two flavonoids: antimicrobial (Metwally et al. 2010) for 12 and antioxidant (Abas et al. 2003) for 13.

The second approach is to reduce the vector population, i.e. the mollusc *B. glabrata*. A number of methanolic (*Kielmeyera variabilis* Mart. & Zucc.) (Pinheiro and Cortez, 2003) or ethanolic (*Caryocar brasiliensis*, *S. adstringens* Mart. Coville, *Stryphnodendron polyphyllum* Mart., *Eugenia malaccensis* L. and *E. dysenterica*) (Bezerra et al., 2002; Oliveira et al., 2006) extracts have shown activity against this mollusc. The composition of secondary metabolites of some species is known (*C. brasiliensis*, *S. adstringens*, *S. polyphyllum*, *E. dysenterica*) but the activity of these metabolites has not been studied. The same applies to the species *K. variabilis*, of which 3 xanthanes [assiguxanthone-B (14), 1,3,5,6-tetrahydroxy-2-prenylxanthone (15) and

**Fig. 3** Metabolites isolated from Cerrado species with molluscicidal and insecticidal activity



**Fig. 4** Metabolites isolated from Cerrado species with fungicidal activity



kielcorin (**16**) and 2,5-dihydroxybenzoic acid (**17**) were isolated. However, the activities of these compounds have not been tested (Pinheiro and Cortez 2003).

Another sanitarian problem to be addressed is insecticidal activity against *Aedes aegypti* (Table 5), which is currently the mosquito that has the highest dispersion in urban areas in the world. The medical importance of this species is its vectorial capacity for the four serotypes of dengue virus and yellow fever virus. In this way, two species have been assayed against this dipteran, *E. malaccensis* (Oliveira et al., 2006) and *Magonia pubensis* (Silva et al., 2004). Only the latter species showed activity, provoking the death of the mosquito. A catequinic tannin was isolated from the active fraction and this showed a toxic effect on the intestinal epithelia of larvae.

The third approach in the study of insecticidal activity is ecological. In this context, extracts from *Memora peregrina* were tested against the moth *Anasagasta kuehniella* (Grassi et al., 2005). The glycosylated iridoid 6 $\beta$ -hydroxyipolamiide (**18**), which showed moderate activity on larval development of *A. kuehniella*, was isolated from active fractions. A biodirected study of *Ocotea velloziana* allowed the isolation of a promising larvicidal aporphine alkaloid (+)-dicentrine (**19**), due to, not only the

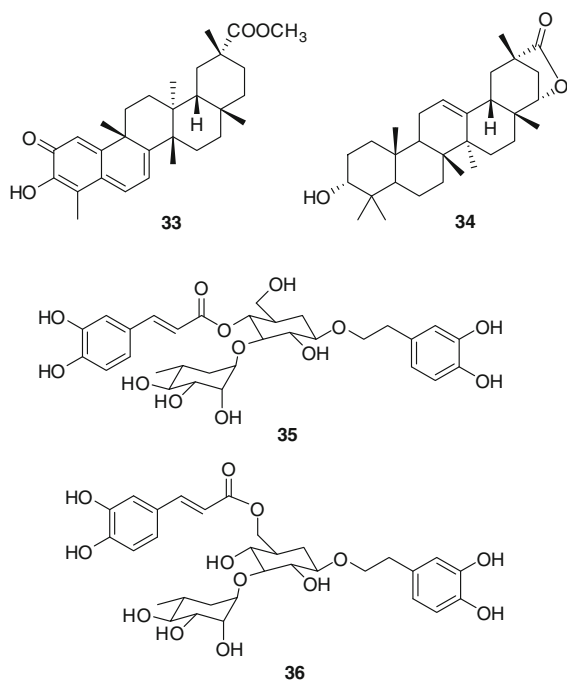
growing resistance of *A. aegypti* populations to temephos, one of the most employed commercial insecticides for their control, but also the environmental safety and human health concerns associated to their use (Garcez et al. 2009).

### Fungicidal activity

Five species have shown antifungal activity in extracts or as isolated compounds: *Stryphnodendron obovatum* Benth., *E. dysenterica*, *Tocoyena Formosa* (Cham. & Schltdl.) K. Shum, *Arrabidaea brachypoda* Bureau and *Curcularia* sp., an endophytic fungus isolated from *Ocotea corymbosa* (Meisn.) Mez (Table 6).

Some fractions from extracts of the first of these species, *S. obovatum*, have shown activity against *Candida albicans* and *Candida parapsilosis* (Sanches et al., 2005). Epigallocatechin (**20**), gallic acid (**21**) and gallic acid-(4 $\beta$  → 8)-gallic acid (**22**) have been isolated from these extracts but their activity has not been tested (Fig. 4). Essential oils obtained from *E. dysenterica* have also shown activity against *Cryptococcus neoformans* (Costa et al., 2000).

Extracts from *T. formosa* were found to be active against *Saccharomyces cerevisiae*, *Cladosporium*



**Fig. 5** Metabolites isolated from Cerrado species with antimicrobial activity

*cladosporioides* and *Cladosporium sphaerospermum* (Bolzani et al., 1997). A bioassay-guided isolation from the active fractions afforded 4 iridoids;  $\alpha$ -gardiol (23),  $\beta$ -gardiol (24), mollugosido methyl ester (25) and formosinoide (26). The mixture of  $\alpha$ - and  $\beta$ -gardiol was also tested and showed moderate antifungal activity.

Four benzopyran derivatives have been isolated from the endophytic fungus associated with *O. corymbosa* (*Curcularia* sp.). Of these, (2'*S*)-2-(propan-2'-ol)-5-hydroxy-benzopyran-4-one (27) and 2-methyl-5-methoxy-benzopyran-4-one (28) showed weak antifungal activity against *C. sphaerospermum* and *C. cladosporioides* (Teles et al., 2005).

A different case involved the study of the epicuticular wax of *A. brachypoda*, which yielded four flavonoids—3',4'-dihydroxy-5,6,7-trimethoxyflavone (29), cirsilinol (30), cirsimaritin (31) and hispidulin (32) (Alcerito et al. 2002). An assay of the flavonoids showed growth inhibition of the fungus *C. sphaerospermum*, thus reinforcing the hypothesis that this epicuticular wax is not only a physical barrier but also a chemical one, with an ecological role played by these compounds.

## Antibacterial activity

Regarding activities against bacteria, several studies were performed with extracts and isolated metabolites against bacteria. The selected plants are mainly used in traditional medicine (Table 7). It is worth highlighting the activities of the species *Qualea grandiflora* Mart., *Viola surinamensis* (Rol. Ex Rottb.) Warb. and *Hancornia speciosa* Gomes against *Staphylococcus aureus* and *S. epidermis* (Costa et al., 2008), as well as the dragon's blood from *Croton urucurana* Baill. against *S. aureus*, *S. epidermis*, *Streptococcus agalactiae*, *Streptococcus pneumoniae*, *Enterococcus faecalis*, *Salmonella typhimorium*, *Shigella flexneri* and *Proteus mirabilis* (Silva Junior et al., 2009).

A total of 12 triterpenes were isolated from *Austroplenckia populnea* extracts and, of these, pristimerin (33) showed significant activity against *Escherichia coli* and abruslactone A (34) against *S. aureus* (Miranda et al., 2009b). A mixture of the phenylpropanoid glycosides verbascoside (35) and isoverbascoside (36) was isolated from *Arrabidaea harleyi* (Lima et al., 2003) (Fig. 5). This mixture showed activity against five Gram-positive bacteria (*S. aureus*, *Micrococcus luteus*, *Bacillus subtilis*, *Bacillus mycoides* and *Enterococcus faecalis*) and two Gram-negative bacteria (*E. coli* and *Serratia marcescens*).

## Conclusions

Cerrado is a vegetation domain in which there are a large number of complex biochemical interactions. However, only a small fraction of the species has been studied from the phytochemical viewpoint with the aim of identifying metabolites responsible for these interactions. This domain is therefore a potential source of bioactive compounds that has remained almost unexplored. It is crucial to preserve the biodiversity of the Cerrado in order to deepen the study of the enormous wealth of new bioactive products that would be available.

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