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Review

Antimony toxicity in soils and plants, and mechanisms of its alleviation

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ABSTRACT

Antimony (Sb) is a non-essential element for plants, animals, and humans. With increased anthropogenic inputs from mining and industrial activities, ore processing, vehicle emissions, and shooting activities, elevated Sb levels in the environment have become a growing concern. Despite of its non-essentiality, some plants can take up and accumulate Sb in relatively high concentrations in their organs. At increased concentration in edible plant parts or medicinal herbs it may pose health risks to humans and livestock. Although most of Sb is stored in root tissues, a smaller quantity of this metalloid can be translocated to the shoot depending on the plant species, where it exerts a variety of deleterious effects. Its chemical speciation has an influence on its behavior in the environment and its ecotoxicity. Inhibition of photosynthesis, modified root and leaf anatomy, activation of plant antioxidant system, or disruption of plant membrane system are some of the negative effects of Sb on plant growth and development. Studies on mitigation methods are quite important in order to produce food crops in a safe way. Application of silicon, selenium, biochar, nanoparticles, and microorganisms are proven to be emerging strategies for reducing the Sb toxicity.

1. Introduction

Antimony (Sb) belongs to group 15 of Periodic Table along with arsenic (As) and has no biological role in living organisms (Filella et al., 2009). The term “antimony” is widely believed to be originated from two Greek words, i.e., “anti” plus “monos”, gives a meaning of “a metal not found alone” (Multani et al., 2016) even though the origin is uncertain. It has properties of both metals and non-metals so referred to as a “metalloid”. Despite of its non-essentiality, Sb is ninth in terms of most-exploited metal(loid)s (Roper et al., 2012). Most of the Sb is used to produce diantimony trioxide (Sb_2O_3), which is used as a catalyst in the production of polyethylene terephthalate (PET) (a popular material used on food and beverage packaging) (Filella, 2020), as a flame retardant in the production of plastic, textiles, rubber, pigments, paints, coatings, ceramics, and crystal glass. Antimony is a critical component of many alloys, particularly those used in the manufacture of ammunition and batteries. Antimony is a common contaminant on shooting ranges and military bases (Filella et al., 2020). The element is also used in the treatment of leishmaniasis and bilharziasis (Bullón et al., 2021).

Studies on the Mont Blanc glaciers suggested elevated levels of antimony in the Roman era because of early mining and smelting activities in ancient Europe (Preunkert et al., 2019). Antimony research has been largely neglected in decades when it comes to the environment because of its insolubility and scarcity (Filella et al., 2002), but it has recently gained attention due to its environmental impact (Zhang et al., 2021; Bolan et al., 2022). Antimony is listed as a potential carcinogen (Gebel, 1997) and a priority pollutant by USEPA and EU.

Elevated exposure of Sb to human through many ways including drinking water has caused cancer (Gebel, 1997). Studies have shown that pollution by Sb has become a global concern (Amarasiriwardena and Wu, 2011). Studies conducted in southwest China claim that uncontrolled mining activities affect human health and found relatively higher amount of Sb from human hair ($\geq 3\ \mu g\ g^{-1}$) (Liu et al., 2011). Antimony enrichment and accumulation have been reported in peat bogs in Europe and in polar ice caps in the Canadian arctic during the past few decades (Shotyk et al., 1996; Krachler et al., 2005).

Plants primarily absorb Sb from the soil, which is influenced by a variety of parameters including phytoavailability, soil type, Sb speciation, plant species, etc. (Filella et al., 2002; Cidu et al., 2014). Antimony can be mainly found in two oxidation states: antimonite (SbIII) and

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antimonate (SbV), with the former being more harmful to plants (Filella et al., 2002; Reimann et al., 2010; Multani et al., 2016). Antimony has been linked to negative impacts on plants such as stunted growth (Chai et al., 2017), reduced biomass and photosynthesis (Tschan et al., 2008; Pan et al., 2011), generation of reactive oxygen species (Chai et al., 2016; Ortega et al., 2017), and lipid peroxidation (Vaculíková et al., 2014). As a result, it is necessary to consider mitigation strategies to protect plants and organisms associated with them. Very few studies have been conducted on remediation strategies on Sb which includes application of Si and Se (Feng et al., 2013; Vaculíková et al., 2014, 2016), biochar, arbuscular mycorrhiza, salicylic acid, growth regulators, and nano particles (Cui et al., 2017; Tandy et al., 2017; Wang et al., 2018; Gu et al., 2019, 2020; Silvani et al., 2019; Wu et al., 2019).

Given the environmental importance of Sb, research on its behavior in plants is critical because plants are the primary producers and have the potential to transport Sb to higher trophic levels. Through this review, we attempted to connect the knowledge about the fate of Sb in soils and plants, its mobility, bioavailability, and toxicity at higher concentrations. Additionally, we also investigated possible mitigation strategies of Sb in soils and plants, including phytoremediation, the use of various amendments, microorganisms, nanoparticles, and more.

2. Antimony in soils and environment

More than 100 primary and secondary Sb minerals are found in the nature (Anderson, 2012). Most of the primary Sb minerals are insoluble sulphides. At oxygen rich conditions primary minerals are weathered to form secondary minerals releasing Sb into the soil (Multani et al., 2016; Roper et al., 2012). The most important natural source of Sb, stibnite (Sb_2S_3), is associated with its principal weathering products senarmontite (cubic Sb_2O_3), valentinite (orthorhombic Sb_2O_3), and stibiconite (Sb_3O_6OH) (Filella et al., 2009; Hu et al., 2016).

Concentration of Sb in Earth’s crust is approximately 0.2–0.3 mg Sb kg^{-1} (Schulz et al., 2017). According to studies, the natural concentration of Sb varies by region due to the extent of mineralisation, differences in parent material, and different sampling methods (Gregori et al., 2003; Wilson et al., 2010). For example, concentration of Sb in Florida soils ranged from 0.06 to 0.79 mg Sb kg^{-1} across seven orders (Chen et al., 1999), whereas soils and sediments in the Netherlands contain about 3 mg Sb kg^{-1} (Crommentuijn et al., 2000). Total concentrations of Sb in unpolluted freshwaters, ocean and sediments are well below 1 µg Sb L^{-1} (Filella et al., 2009).

Elevated levels of Sb in the environment are mainly due to anthropogenic activities such as waste incineration, mining, smelting, combustion of fossil fuels and the road traffic (Guéguen et al., 2012; Sun et al., 2019). Concentration of Sb in topsoil near mining areas (up to 4400 mg Sb kg^{-1}) have been found to be greater than that in bed rock (up to 500 mg Sb kg^{-1}) (Cidu et al., 2014). Mining activities have reported to introduce metalloids like Sb into the rivers and drinking water in China, the world top Sb producer now (Sun et al., 2019). Studies conducted in Japan revealed that brake abrasion dust and waste fly ash were the dominant sources of Sb in the air particulate matter (Iijima et al., 2009).

2.1. Speciation of antimony in soil

Majority of Sb released from different sources ends up in soil which then enters different trophic levels and may cause harmful effects. Antimony exists in various oxidation states (−III, 0, III, V) but in natural systems it is found in III, V, and methylated forms (Filella et al., 2009; Reimann et al., 2010; Wilson et al., 2010; Multani et al., 2016). Toxicity of Sb to both soil microorganisms and plants depends on its species and increase in the order of methylated Sb $<$ Sb(V) $<$ Sb(III) (Gebel, 1997; Filella et al., 2002).

According to reports, Sb is lethal to soil microorganisms because it inhibits microbial community growth and reduces several enzyme activities such as urease, dehydrogenase, acid phosphatase, and it is also known to affect microbial respiration in soils (Guillamot et al., 2014; Wang et al., 2011, 2021). However, microbes that use Sb-containing minerals as energy sources can survive in highly contaminated Sb environments (Deng et al., 2021). These microorganisms can influence the fate of Sb in soil by participating in its oxidation and transformation. Soil bacteria from the genera *Pseudomonas*, *Comamonas*, *Agrobacterium*, and *Acinetobacter*, for example, are known to oxidize Sb(III) to Sb(V) (Li et al., 2016, 2017).

Biomethylation of Sb is a process where inorganic Sb can act as methyl acceptor and can be converted to mono-, di-, and tri-methyl Sb in the environment by bacteria and fungi (Filella et al., 2002; Filella, 2010; Ji et al., 2018). Trimethyl Sb is common in soil rhizosphere as reported by Wei et al. (2015). Very low concentrations of methylated or organic Sb forms are detected compared to the inorganic ones (Wilson et al., 2010).

Speciation of soil Sb mostly depends on pH and redox potential of soil (Fig. 1). In aqueous solutions, Sb(V) exists in a wide range of pH (2.7–10.4), whereas Sb(III) readily forms insoluble $Sb(OH)_3$ irrespective of pH (Herath et al., 2017). In aerobic conditions Sb(V) is more common, while Sb(III) exists in reducing or anaerobic conditions (Karimian et al., 2019; Mitsunobu et al., 2006). However, contrary results were recorded by Fawcett et al. (2015), where Sb(V) and Sb(III) were spotted in reduced and oxidised conditions, respectively. In waterlogged conditions, soil or sediment becomes anoxic and thus Sb(V) reduces to Sb(III). At very lower reduction potential Sb(III) can even be reduced to elemental Sb or stibine (Wilson et al., 2010). In anaerobic conditions, the biomethylation process is enhanced, and Sb(III) compounds are more likely to biomethylate than Sb(V) compounds (Filella et al., 2009). Low redox potential is known to promote the mobility of methylated Sb (Frohne et al., 2011). The dominant form of Sb(V) in almost all relevant pH values is the octahedral antimonate ion ($Sb(OH)_6^-$) in contrast to tetrahedral arsenate and phosphate ions (Pauling, 1933). The most abundant Sb(III) species found in the form of uncharged antimonous acid indicates the higher mobility than Sb(V) (Wilson et al., 2010).

2.2. Mobility and bioavailability of Sb in soil

Only a small portion of Sb is available to plants despite of its higher total concentration in soil (Lintschinger et al., 1998). Mobility, bioavailability, and toxicological effects of Sb depend on its chemical speciation, total Sb content, soil properties, redox potential, and the presence of other cations (Filella et al., 2002; Wilson et al., 2010; Levresse et al., 2012).

Soil properties such as pH, cation exchange capacity (CEC), soil texture, soil organic matter (SOM) content, oxides and hydroxide content, ageing time, etc., have an impact on the bioavailability of Sb in soil (Wilson et al., 2010; Bagherifam et al., 2021). As previously discussed, the oxidation states of Sb change under different environmental conditions of pH and redox potential (Fig. 1), which eventually affects their bioavailability (Mitsunobu et al., 2006; Fawcett et al., 2015; Karimian et al., 2019). A rise in pH has been reported to increase the amount of soluble Sb that can be easily sorbed onto organic matter (Nakamaru and Martín Peinado, 2017). During the transformation of metal(loid) species in soil, organic matter can act as an electron donor, and a competing ligand. Binding of Sb to SOM can reduce Sb bioavailability, most likely through the formation of stable Sb-humic acid complexes (Steely et al., 2007). However, contrary observations were recorded by Nakamaru and Martín Peinado (2017) where application of organic amendments to soil increased the bioavailability of Sb which attributed to increased pH, CEC, and other factors. The amount of sand, silt, and clay in a soil also influences its bioavailability of Sb. High sand content, for example, increases mobility and bioavailability, whereas high clay and silt content has an inhibitory effect (Chang et al., 2022).

Majority of Sb is bound to iron (Fe) and aluminium (Al)-bearing minerals making it unavailable to plants (Hiller et al., 2012; Leuz et al.,

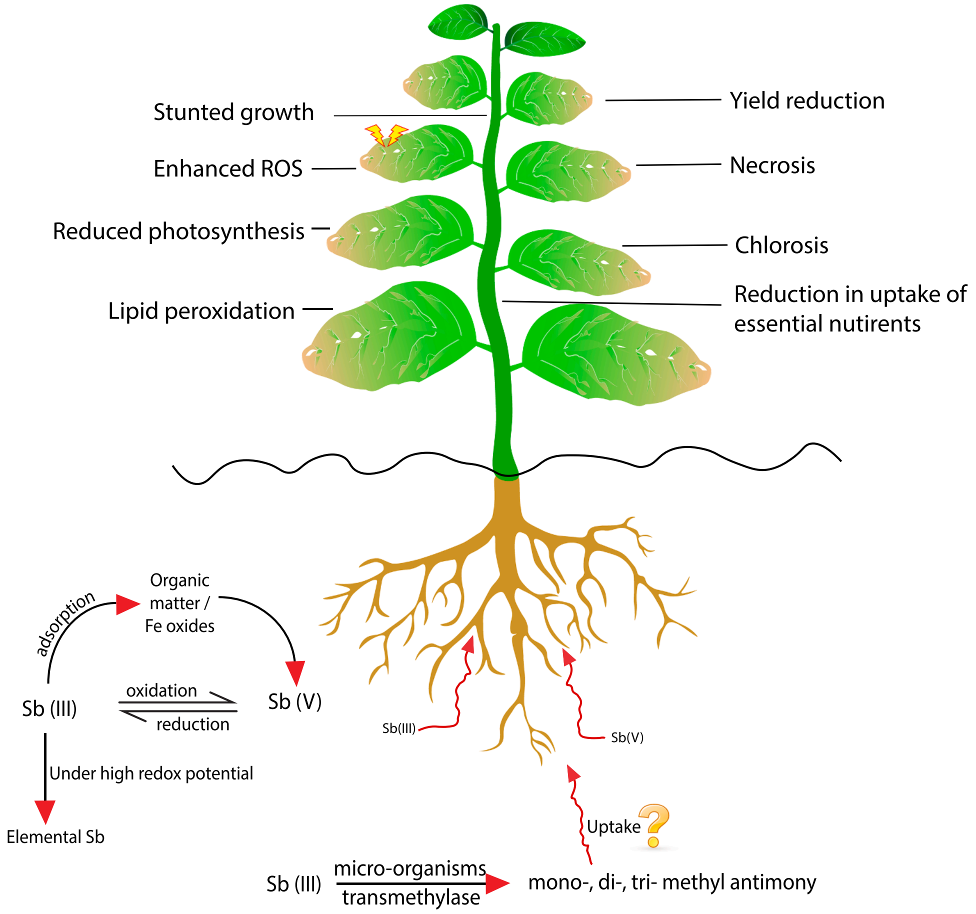

Fig. 1. Schematic diagram depicting bioavailability of Sb in soil and its toxic effects on plants under Sb stress.

2006; Lintschinger et al., 1998; Wilson et al., 2010). Supporting this, Karimian et al. (2018) found that dissolution of jarosite, a Fe-bearing mineral at pH 5.5 enhanced the mobility of Sb. Antimony III is adsorbed to goethite over wide range of pH than Sb(V) (Ackermann et al., 2008). Pentavalent Sb is commonly found in soil water extract over wide redox range. It might be due to preferential sorption of Sb(III) to soil particles/ Fe oxides and rapid oxidation of Sb(III) by Fe oxides (Okkenhaug et al., 2011). Interaction of Sb(III) with Fe(III) might have major role in speciation of Sb; wherein, photocatalytic oxidation of Sb (III) by different Fe species under sunlight might influence fate of Sb species in surface soils and waters (Kong et al., 2016). The bio-accessibility of Sb is reported to be less than 30% of total for most of the tested soils due to immobilisation of Sb by Fe hydroxides and sulphides present in soils (Herath et al., 2017).

Mining sites either active or abandoned, that prone waterlogging are at high risk of Sb(III) mobilization and toxicity (Hockmann et al., 2014). One of the reason might be enhanced reduction of Sb(V) mediated by microbes and subsequent release of Sb(III) by reduction and dissolution of Fe hydroxides. Studies on phytoavailable Sb (using chemical extraction methods) on highly contaminated mining soil in southern China found only 0.4–3.5% of easily phytoavailable Sb, 1.6–8.3% of moderately phytoavailable Sb, and 88–98% of non-phytoavailable Sb. This variation in phytoavailability of Sb in soil was mainly due to its binding with different soil constituents (He, 2007). The effect of soil ageing on Sb availability is not properly considered in most of the soil studies (Lin et al., 2020). There have been reports of decreased availability of Sb(III) and Sb(V) with ageing of soils previously spiked with Sb, and phytotoxicity of Sb was not even found in the majority of Sb(V)-treated soils (Lin et al., 2020; Diquattro et al., 2021).

3. Antimony absorption mechanisms at the cellular level: what we know so far

Data regarding uptake mechanisms of Sb are considerably less available when compared with similar metalloids such as As. Uptake of As is well known and has been thoroughly studied than Sb. While both As and Sb belong to the same group in the Periodic Table along with phosphorus (P), there was an assumption in previous years that they share the same uptake mechanisms. Arsenate [As(V)] behaves as a chemical analogue of inorganic phosphate [P(V)] and it was found that uptake of As(V) occurs through the phosphate pathway (Woolson et al., 1973; Asher and Reay, 1979). A study conducted on *Pteris vittata* L., an As hyperaccumulator, examining P and As movement in their rhizoids bysynchrotron X-ray microprobe showed that As(V) and P were co-transported till endodermis, where As(V) reduced to As(III). The absence of P accelerated the movement of As, therefore Lei et al. (2012) concluded that As(V) and P may share the same transporter.

However, to our understanding the entrance route of Sb(V) into the cells has not yet been identified (Fig. 2). One of the notable hypotheses about its uptake, proposed by Tschan et al. (2009), offered two Sb(V) routes into cells. One is the uptake of Sb(V) into the root symplasm via low-selectivity transporters, in which Sb anions replace important nutrient anions such as Cl^- and NO_3^-. Another route is via apoplasm and endodermis without a fully established Casparian strip (Fig. 2). There is evidence that plants do not use the P(V) pathway to take up Sb(V). For example, Tschan et al. (2008) reported that the addition of P(V) in to the medium where *Zea mays* L. and *Helianthus annuus* L. were growing, did not decrease the uptake of Sb(V), and in a study using *Leishmania*, Brochu et al. (2003) found that the addition of As(V) did not inhibit the accumulation of Sb(V). This could be due to the fact that Sb(V) is a stable

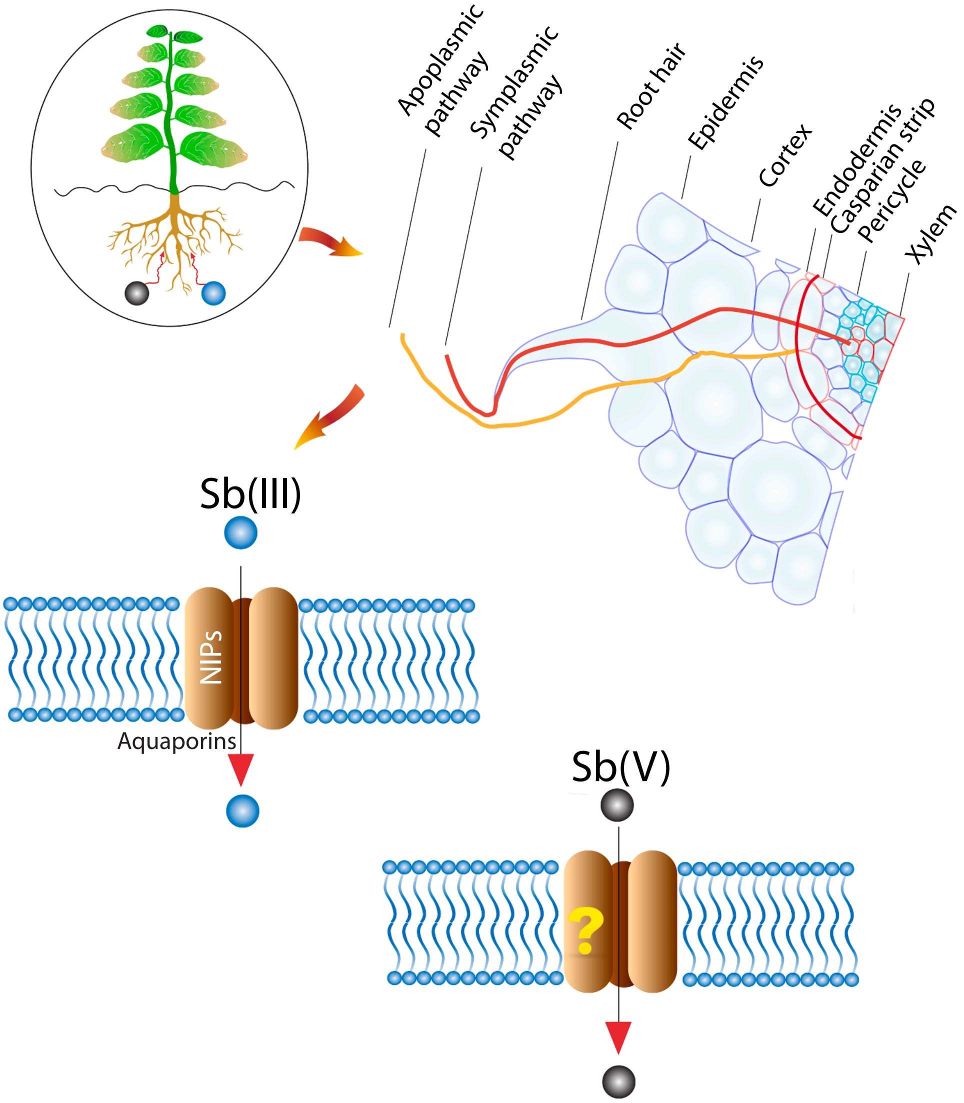

Fig. 2. Mechanisms of Sb uptake by plants. Sb can be taken up and transported through root tissues via apoplasmic or symplasmic pathway. Sb(III) is taken up through cell membranes via aquaporins (mostly NIPs), while membrane transport of Sb(V) is still unclear.

form in aqueous solutions; $Sb(OH)_6^-$ does not compete with phosphate uptake. Structural variations may also play a role. Sb(V) has an octahedral structure, whereas phosphate P(V) and As(V) have a tetrahedral structure (Pauling, 1933; Ngo et al., 2016). However, there have been instances of As(V) enhancing Sb accumulation in plant tissues, particularly in the roots of As-hyperaccumulating ferns *Pteris cretica* L. and *P. vittata* (Feng et al., 2011a; Müller et al., 2013). According to Müller et al. (2013), As(V) alters the cell membrane's integrity and permeability, resulting in an increase in uptake of Sb.

Unlike Sb(V), Sb(III) is a neutral molecule which resembles arsenite [As(III)] in its structure and size. As(III) and Sb(III) have similar transport properties and can enter and exit the cell via a passive or active pathway. Competition between Sb(III) and As(III) was observed in *Leishmania* (Brochu et al., 2003) and rice (*Oryza sativa* L.) (Meharg and Jardine, 2003), indicating similar absorption mechanism. In aqueous solution, As(III) and Sb(III) are mostly found as trihydroxylated uncharged molecules, i.e. $As(OH)_3$ and $Sb(OH)_3$, which are structurally similar to glycerol at neutral pH (Ramírez-Solís et al., 2004; Porquet and Filella, 2007). Therefore, As(III) and Sb(III) are easily transported by the aquaglyceroporins and membrane proteins permeable for water and glycerol (Bhattacharjee et al., 2008) (Fig. 2). The first Sb(III) transporter, GlpF, was identified in *Escherichia coli* (Sanders et al., 1997). GlpF belongs to the aquaglyceroporin subfamily of the major intrinsic proteins (MIP) which are selective for water and other uncharged solutes, such as glycerol and urea (Bienert et al., 2008b). Following that, Wysocki et al. (2001) found that the glycerol channel Fps1p mediates the uptake of As(III) and Sb(III) in *Saccharomyces cerevisiae*.

Plant aquaporins responsible for metalloid uptake are classified into four subfamilies corresponding to distinct subcellular localizations: 1) small basic intrinsic proteins (SIP) located in the endoplasmic reticulum, 2) plasma membrane intrinsic proteins (PIP), 3) tonoplast intrinsic proteins (TIP) responsible for water transport, and 4) Nodulin26-like intrinsic membrane proteins (NIP), which are localized to plasma and intracellular membranes to mediate transport of NH_3, $B(OH)_3$ or $Si(OH)_3$ (Ma and Yamaji, 2006; Takano et al., 2006). It is presumable that these NIPs are also responsible for uptake of $As(OH)_3$ and $Sb(OH)_3$. Bienert et al. (2008a) have shown that *O. sativa* OsNIP2;1 and *Arabidopsis thaliana* L. (Heynh.) AtNIP5;1 are permeable for the metalloids As(III) and Sb(III). AtNIP5;1 and OsNIP2;1 are localized to the distal plasma membrane domain of the endodermis in *Arabidopsis* roots (Takano et al., 2006), and endodermis and exodermis of rice roots (Ma and Yamaji, 2006). These cell layers represent the border for apoplasmic transport. Ions taken up into the shoot via the vascular system must be transported through plasma membrane to reach the symplasm. Kamiya and Fujiwara

(2009) found that NIP1;1 is the determinant of Sb(III) tolerance, and that Sb(III) transport is mediated by NIP1;1 in *A. thaliana*. Study conducted on both crop plants and non-crop plants showed that majority of Sb transporters in plants belonged to NIPs and are expressed in the roots (Azad et al., 2018) (Fig. 2).

The passive diffusion of As(III) and Sb(III) through the cell membrane is not the only transport route that exists. Active transporters for As were also discovered in *S. cerevisiae*. This mechanism, which is conserved from bacteria to plants, has been shown to involve a cluster of three genes: *ACR1*, *ACR2*, and *ACR3*. Acr3p, a protein encoded by *ACR3*, is evolved specifically to extrude As(III) out of the cell. In addition, the transport of Sb(III) via yeast Acr3p has also been demonstrated. Maciaszczyk-Dziubinska et al. (2012) showed that the yeast Acr3p is a plasma membrane antiporter that catalyzes As(III)/H^+ and Sb(III)/H^+ exchange in *S. cerevisiae*. Acr3p displays similar low affinity for both As (III) and Sb(III) but transport of As(III) is three times faster than Sb(III).

4. Plant-to-plant differences in Sb absorption

Antimony uptake varies widely among plant species, growing sites, and underlying substrate conditions (Table 1). Most commonly, the highest plant Sb concentrations occur in mining areas where the concentration of Sb in the soil is appreciably high (sometimes more than 1000 mg Sb kg^{-1} in soil). For example, Baroni et al. (2000) found that *Achillea ageratum* L. growing in an abandoned Sb-mining area where total soil Sb was more than 9000 mg Sb kg^{-1} with extractable concentration of 793 mg Sb kg^{-1}, accumulated 367 mg Sb kg^{-1} in the basal leaves and 1105 mg Sb kg^{-1} in inflorescences. The content of Sb was 2–3 times higher in roots than in leaves of *Plantago lanceolata* L., while *Silene vulgaris* (Moench) Garcke showed a high resistance to Sb, as well as the ability to collect and translocate Sb to the shoots. Vaculík et al. (2013) investigated concentration of Sb in some medicinal plants growing in old mining sites where the soil concentration of Sb was between 146 and 4463 mg Sb kg^{-1}. They discovered that plants including *Fragaria vesca* L., *Taraxacum officinale* L., and *Plantago medium* L. accumulated high levels of Sb in their roots and shoots, implying that traditional medicine made from them might be detrimental to humans. It is crucial to keep in mind that dust contamination of the stomatal chambers of leaves obtained at such mining sites may cause the root-to-shoot transfer to be overestimated.

In contrast to these investigations, there are some reports of low Sb uptake in plants growing on contaminated sites. Concentrations of Sb in tree stems and herbaceous plants were reported to be less than 5 mg Sb kg^{-1} in the soils with 663 mg Sb kg^{-1} in Portugal (Pratas et al., 2005). Cidu et al. (2014) found that a heavily contaminated soil (up to 4400 mg Sb kg^{-1}) in an abandoned mine in Sardinia had a relatively low available Sb (100 mg Sb kg^{-1}), resulting in low Sb translocation into aboveground plant parts in *Pistacia lentiscus* L. and *Asparagus* (maximum 9.3 mg Sb kg^{-1}). These reports indicate that high level of Sb in growth media does not necessarily result in significant accumulation of Sb in plant tissues (Table 2).

Factors affecting the transfer of Sb from soil to plant are as follows:

- *Bioavailability of Sb*

One of the factors governing the uptake of Sb from soil into plants is bioavailability of Sb in the soil solution, although some portion of Sb can be taken up by leaves from air pollution. The concentration of Sb in plant tissues is positively correlated with its solubility in the medium. Plants take up Sb mostly from soils in natural conditions. The potential bioavailability and mobility of Sb varies depending on the solvent. The percentage for the extractable fraction of Sb showed the following order: Ammonium nitrate NH_4NO_3 (0.57%) Sampled diamine tetraacetic acid (EDTA) (0.50%) > Acetic acid (HAc) (0.22%), Water (H_2O) (0.19%)

Table 1
Concentration of Sb in various plant species under different studies.

Plant species	Plant part	Conc. of Sb (mg kg^{-1})	Conc. of Sb in media (mg kg^{-1})	Source of Sb	Type of experiment	References
Zea mays	Shoots	41	18	Potassium hexahydroxy antimonate (SbV)	Hydroponics	(Tschan et al., 2008)
Helianthus annuus	Shoots	77	30	Potassium hexahydroxy antimonate (SbV)	Hydroponics	(Tschan et al., 2008)
Pteris cretica	Roots Fronds	358 28.7	20	Potassium antimony tartrate hemihydrate (SbIII)	Hydroponics	(Feng et al., 2009)
Microlepia hancei	Root Fronds	123 53.3	20	Potassium antimony tartrate hemihydrate (SbIII)	Hydroponics	(Feng et al., 2009)
Cyrtomium fortunei	Roots Fronds	224 10.2	20	Potassium antimony tartrate hemihydrate (SbIII)	Hydroponics	(Feng et al., 2009)
Cyclosorus dentatus	Roots Fronds	124 27.2	20	Potassium antimony tartrate hemihydrate (SbIII)	Hydroponics	(Feng et al., 2009)
Triticum aestivum	Roots Leaves	5 0.16	150	Antimony (V) nitrate (SbV)	Hydroponics	(Shtangeeva et al., 2011)
Zea mays	Roots Shoots	22.01 45	500	Potassium antimony tartrate (SbIII)	Pot experiment (spiked)	(Pan et al., 2011)
Secale cereale	Roots Leaves	42 8	75	Antimony trichloride (SbIII)	Hydroponics	(Shtangeeva et al., 2012)
Triticum aestivum	Roots Leaves	69 3	75	Antimony trichloride (SbIII)	Hydroponics	(Shtangeeva et al., 2012)
Zea mays	Roots	5700	50	Antimony tartrate (SbIII)	Hydroponics	(Vaculíková et al., 2014)
Helianthus annuus	Roots Shoots	7700 50	5	Potassium antimony tartrate hemihydrate (SbIII)	Hydroponics	(Vaculík et al., 2015a, b)
Solanum lycopersicum	Roots Shoots	308 7798	50	Antimony potassium tartrate (SbIII)	Hydroponics	(Peško et al., 2016)
Acorus calamus	Roots Shoots	73.27 19.34	1000	L-Antimony Potassium Tartrate (SbIII)	Pot experiment (spiked)	(Zhou et al., 2018)
Acorus calamus	Roots Shoots	71.54 17.25	1000	Potassium acid pyroantimonate (SbV)	Pot experiment (spiked)	(Zhou et al., 2018)
Triticum aestivum	Roots	162	400	Antimony potassium tartrate (SbIII)	Hydroponics	(Ma et al., 2019)
Oryza sativa	Roots	660	20	Antimony potassium tartrate (SbIII)	Hydroponics	(Feng et al., 2020)
Ipomoea aquatica	Shoots	14	570	Potassium hexahydroxoantimonate (SbV)	Pot experiment (spiked)	(Egodawatta et al., 2020)
Amaranthus tricolor	Roots Shoots	803 348	1400	Antimony potassium tartrate (SbIII)	Pot experiment (spiked)	(Zhong et al., 2020)

Table 2
Antimony levels in different plant species grown in soils at Sb-contaminated sites around the world.

Plant species	Plant part	Conc. of Sb (mg kg^{-1})	Sampled area	Conc. of Sb in substrate (mg kg^{-1})	References
Achillea ageratum	Basal leaves Inflorescence	1367 1105	Abandoned mining area Southern Tuscany	9197	(Baroni et al., 2000)
Plantago lanceolata	Root Basal leaves	1150 569	Abandoned mining area Southern Tuscany	6529	(Baroni et al., 2000)
Silene vulgaris	Root Shoot	249 1164	Abandoned mining area Southern Tuscany	6530	(Baroni et al., 2000)
Cistus ladanifer	Leaves	20.2	San Antonio mine, Italy	2444	(Murciego et al., 2007)
Dittrichia viscosa	Leaves	266	San Antonio mine, Italy	2444	(Murciego et al., 2007)
Hippochcaete ramosissima	Shoots	144	Xikuangshan Sb deposit area, Hunan, China	5949	(Qi et al., 2011)
Boehmeria nivea	Shoot	4029	Active mining area Xikuangshan, China	5579	(Okkenhaug et al., 2011)
Agrostis capillaris	Root Shoot	402 69	Former antimony mine, ribes valley, Eastern Pyrenees	2904	(Bech et al., 2012)
Primula elatior	Root Shoot	128.7 12.9	Abandoned mining site Dúbrava, Dechtárka, Slovakia	523	(Vaculík et al., 2013)
Fragaria vesca	Root Shoot	703.6 269.6	Abandoned mining site Poproč, Agnes, Slovakia	523	(Vaculík et al., 2013)
Taraxacum officinale	Root Shoot	436.9 241.0	Abandoned mining site Poproč, Agnes, Slovakia	4463	(Vaculík et al., 2013)
Hedysarum pallidum	Roots Shoots	90.02 72.20	Abandoned antimony mining area of Djebel Hamimat (Algeria)	19436	(Benhamdi et al., 2014)
Lygeum spartum	Roots Shoots	61.04 18.58	Abandoned antimony mining area of Djebel Hamimat (Algeria)	19436	(Benhamdi et al., 2014)
Pistacia lentiscus	Roots Leaves	3.39 1.51	Abandoned mining area Sardinia Italy	3700	(Cidu et al., 2014)
Sedum emarginatum	Roots	1052	Mining area, Southwest China	1732	(Ning et al., 2015)
Chenopodium album	Roots	1219	Mining area, Southwest China	1732	(Ning et al., 2015)
Sedum lineare	Roots Shoots	23522 3092	Mining area, Southwest China	1732	(Ning et al., 2015)
Raphanus sativus	Roots shoots	0.56 1.31	Sb processing facility, Australia	110	(Ngo et al., 2016)
Rumex obtusifolius	Shoots	0.3	Shooting range soil, Horwin central Switzerland	50	(Hockmann et al., 2014)
Oryza sativa	Grains	0.027	Contaminated agricultural soil, Zijiang river basin, Hunan Province, China	18.1	(Zhang et al., 2020, 2019)

$>$ Ammonium acetate (NH_4OAc) ($<$0.09%) (He, 2007). In soil rich in Sb but with only a small proportion of Sb extractable with NH_4NO_3, Hammel et al. (2000) discovered that the possible dose of Sb for plants is just 0.06–0.59% of the total Sb content. When the plants grown in soil spiked with KSbO-tartrate (mobility of Sb was higher), concentration of Sb in the tissues was much higher.

The sequential leaching procedure showed three categories of Sb potential phytoavailability in soils:

☐ Easily phytoavailable Sb (water soluble plus cation exchangeable fraction)
☐ Moderately phytoavailable Sb (carbonate fraction, Fe/Mn hydrous oxide fraction and sulphidic/organic bound Sb)
☐ Not phytoavailable (residual fraction) (He, 2007).

- *Speciation of Sb*

  Another factor affecting the uptake of Sb in plants is Sb speciation. Older wheat seedlings (from 4 to 9 days) growing in a water spiked with Sb(V) accumulated more Sb than seedlings growing in a water spiked with Sb(III), but for younger (4 day-old) seedlings, the opposite trend was observed (Shtangeeva et al., 2012). Ren et al. (2014) observed a higher translocation factor of Sb(V) than Sb(III) in rice indicating high mobility of Sb(V) than Sb(III). Accumulation of Sb was higher on Sb(V) spiked soil than on Sb(III) spiked soil in sunflower plants, but no difference was found for maize (Tschan et al., 2010). Results of Wan et al. (2013) indicate that Sb might primarily be taken up as Sb(III) by *Lolium perenne* L. and as Sb(V) by *Holcus lanatus* L. Antimony uptake was much higher in all investigated species (*S. vulgaris*, *Achillea wilhelmsii* C. Koch, and *Thlaspi arvense* L.) when Sb was supplied as Sb(III) compared to Sb(V), and root and shoot concentrations were significantly higher in the Sb(III) treatment than that of the Sb(V) treatment (Jamali Hajiani et al., 2017). These findings clearly show that Sb uptake differs between plant and Sb species.
- *Concentration of coexisting ions in soil*

  Elements such as P and calcium (Ca) can also influence the bioavailability of Sb. Phosphorus can induce the release of Sb into the soil pore water via competitive adsorption to soil particles as they both are anions and Sb becomes available for plants. Oppositely, Ca can decrease Sb concentration in soil pore water because of Ca and Sb co-precipitation in soils (Xi et al., 2011; Okkenhaug et al., 2012).
- *pH of the medium*

Contrary result was later described by Evangelou et al. (2012) who found that plants on calcareous soil (pH 8.5) accumulated 1.5–2.6-fold higher concentrations of Sb than on the acidic soil. The highest concentrations of Sb were accumulated in shoots of *Rumex acetosa* L. and *Polygonum aviculare* L. These findings could be attributed to the higher Sb solubility in soils with high pH value, although other factors like relationships between soil pH, sorption of Sb to Fe/Mn oxy(hydr)oxides, clays, etc. need to be considered as well.

5. Organ, tissue, and subcellular distribution of Sb in plants

Antimony is mostly taken up by plant roots, with only a minor quantity being translocated to the aerial plant parts (Table 2). As previously stated, this property is found in plants that naturally grow near old mine sites, smelters, and roadways where Sb levels are expected to be elevated. However, at these sites there is always a risk of aerial contamination of plant samples by air dust, therefore not always the data from contaminated sites might be 100% reliable. Similarly, laboratory experiments also suggest that the root is the most important plant organ for Sb accumulation (Shetty et al., 2021a). The root appears to be an ideal environment for Sb deposition, which may be related to the strategy of non-accumulating plants to prevent element transport into reproductive tissues. Despite this, some plant species can accumulate a significant amount of Sb in their aerial parts. As a result, Sb translocation and bioaccumulation can differ significantly between plant species and sites (Table 3 and Table 4). Probably one of the best recognized Sb-accumulating plant species is *Boehmeria nivea* (L.) Gaudich with more than 4000 mg Sb kg^{-1} in shoots that was found in the biggest actively mining Sb region in the world in Xikuangshan, China (Okkenhaug et al.,

Table 3
Translocation factor for Sb in different plant species.

Plant species	Plant part	Translocation factor from root	Type of study	References
Achillea ageratum	Shoot	0.98	Field study	(Baroni et al., 2000)
Plantago lanceolata	Basal leaves	0.49	Field study	(Baroni et al., 2000)
Silene vulgaris	Shoots	5.38	Field study	(Baroni et al., 2000)
Pueratia phaseoloides	Shoots	6.65	Field study	(Qi et al., 2011)
Dendranthema indicum	Shoots	5.47	Field study	(Qi et al., 2011)
Zea mays	Shoots	2.05	Pot study	(Pan et al., 2011)
Lolium perenne	Shoots	0.15	Pot study	(Wan et al., 2013)
Holcus lanatus	Shoots	0.005	Pot study	(Wan et al., 2013)
Pistacia lentiscus	leaves	0.445	Field study	(Cidu et al., 2014)
Hedysarum pallidum	Shoots	1.21	Field study	(Benhamdi et al., 2014)
Lygeum spartum	Shoots	0.34	Field study	(Benhamdi et al., 2014)
Chenopodium ambrosioides	Shoots	1.3	Field study	(Ning et al., 2015)
Polygonum capitatum	Shoots	1.3	Field study	(Ning et al., 2015)
Raphanus sativus	Shoots	2.7	Field study	(Ngo et al., 2016)
Solanum lycopersicum	Shoots	0.04	Hydroponics	(Peško et al., 2016)
Helianthus annuus	Leaves	0.12	Hydroponics	(Ortega et al., 2017)
Acorus calamus	Shoots	0.23	Pot study	(Zhou et al., 2018)
Amaranthus tricolor	Shoots	0.23	Pot study	(Zhong et al., 2020)
Vetiveria zizanioides	Shoots	0.03	Hydroponics	(Yu et al., 2020)

Table 4
Bioaccumulation factor of Sb in different plant species.

Plant species	Bioaccumulation factor	Type of study	References
Zea mays	0.93 (Shoots)	Hydroponics	(Tschan et al., 2008)
Helianthus annuus	1.32 (Shoots)	Hydroponics	(Tschan et al., 2008)
Pteris vittata	0.08 (Shoots)	Soil study	(Qi et al., 2011)
Miscanthus floridulus	2.74 (Shoots)	Soil study	(Okkenhaug et al., 2011)
Boehmeria nivea	9.13 (Shoots)	Soil study	(Okkenhaug et al., 2011)
Pistacia lentiscus	(Roots)	Soil study	(Cidu et al., 2014)
Asparagus	0.015 (Shoot)	Soil study	(Cidu et al., 2014)
Sedum emarginatum	3.9 (Shoots)	Soil study	(Ning et al., 2015)
Plantago asiatica	1.0 (Shoots)	Soil study	(Ning et al., 2015)
Sedum lineare	6.7 (Shoots)	Soil study	(Ning et al., 2015)
Gnaphalium affine	1.7 (Shoots)	Soil study	(Ning et al., 2015)
Dichrocephala auriculata	1.1 (Shoots)	Soil study	(Ning et al., 2015)
Arundo donax	73.6 (Shoots)	Soil study	(Delplace et al., 2020)
Phragmites australis	96.5 (Shoots)	Soil study	(Delplace et al., 2020)
Amaranthus tricolor	0.18 (Roots) 0.04 (Shoots)	Pot study	(Zhong et al., 2020)
Vetiveria zizanioides	0.57 (Roots)	Hydroponics	(Yu et al., 2020)
Dittricha viscosa	3.98 (leaves)	Natural site	(Garrido et al., 2021)

2011). Although *Sedum lineare* Thunb. showed significantly higher Sb accumulation in roots than leaves, shoot Sb concentration was recorded over 3000 mg Sb kg^{-1} indicating its high shoot bioaccumulation potential (Ning et al., 2015). However, information on the precise locations of Sb tissue deposition and the mechanisms that cause is still limited.

Plants are thought to have a substantial tolerating and accumulating mechanism based on element compartmentation in their cytoplasm. For Sb, the largest concentration was found in the cytosol in the aboveground sections of the Cretan brake fern (*P. cretica*), relative to the cell wall and organelle fractions, even though the majority of Sb was stored in the cell wall in the roots (Feng et al., 2011a). However, when Wang et al. (2017b); Wang et al. (2017a) studied the subcellular distribution of Sb in different cell compartments of *Ficus tikoua* Bur, they found the largest Sb deposition (about 34% of Sb observed) in the cell walls of both above and below ground parts. In contrast, Yu et al. (2020) recently discovered the greatest Sb peaks in precipitates in the cytosol of roots of *Vetiveria zizanioides* L. Nash plants treated with Sb using transmission electron microscopy and X-ray analysis. They further suggest that both Sb(III) and Sb(V) could bind together with other elements like sulphur and form solid complexes that would also explain higher concentration of Sb in roots than in shoots.

Wu et al. (2020) compared the apoplasmic and symplasmic distribution of trivalent and pentavalent forms of Sb as well as their combinations in *Brassica parachinensis* L. roots. They found that both Sb(III) and Sb(V) forms are in higher concentration in symplasmic than apoplasmic flow. Moreover, the symplasmic concentration of Sb(III) was significantly higher than Sb(V), while no difference between apoplasmic Sb concentration was found between Sb(III) and Sb(V). In the cell walls lower Sb exposure (1 mg Sb L^{-1}) showed no differences between concentration of Sb(III) and Sb(V), while at higher Sb exposure (5 mg Sb L^{-1}) a higher cell wall concentration of Sb(III) than Sb(V) was found (Wu et al., 2020). They later found that the highest proportion of Sb(III) and Sb(V) is found in cell walls, followed by symplamic and apoplasmic flow, respectively. Furthermore, Wu et al. (2020) discovered that Sb(V) forms are more abundant in xylem than Sb(III) forms. Antimony recognition in tissue and cellular distribution and localization is still in its early stages, so more research is needed to better understand how Sb functions in plants.

6. Impact of Sb on plant growth and performance

Antimony in excess is reported to have toxic effects on plants (Feng et al., 2013; Natasha et al., 2019) as shown in Fig. 1. The intensity of toxicity mostly depends on speciation of Sb and certain plant species (Baek et al., 2014), however other factors, like dose, exposure time, soil type, etc., play another important feature. Inorganic Sb is comparatively more toxic than organic one. Among the inorganic forms, trivalent Sb is more toxic than pentavalent Sb (Filella et al., 2002; Gebel, 1997). It was suggested that Sb concentration of 5–10 mg Sb kg^{-1} in plants is potentially toxic and can disturb normal functioning of plants (Herath et al., 2017; Natasha et al., 2019).

6.1. Changes in growth and development induced by Sb

Antimony exhibits different levels of toxicity to different plant species as explained by Liang et al. (2018), where authors found a dose effect relationship between radish and rapeseed plants with Sb. Antimony had severe effects on germination and root elongation of rapeseed than radish. Antimony can cause leaf chlorosis and necrosis as observed by Vaculík et al. (2015a) in sunflower seedlings at 2.5 and 5 mg Sb kg^{-1}. Roots are mostly affected by Sb toxicity than shoots (Baek et al., 2014). Contrary results were observed by Chai et al. (2017), where growth of *F. tikoua* leaves were significantly inhibited when Sb concentration was higher than 30 μmol Sb L^{-1}, and no inhibitory effect of Sb was observed on stems and roots.

Exposure of Sb to plants can decrease the biomass significantly (Bech et al., 2012; Chai et al., 2017). The root and shoot biomass of paddy cultivar Nanjing-45(N) was reported to be decreased by 42% and 27% respectively at low exposure levels to Sb(V) as compared to the control treatment (Cai et al., 2016), Although the extent of decrease varied between cultivars. The leaf and root biomass of *Triticum aestivum* L. seedlings grown for 7 days in Sb enriched media was lower compared to the biomass of the seedlings germinated in Sb-free media. This decrease in biomass was observed to be correlated with increased Sb concentration (Shtangeeva et al., 2011). Similarly, Pan et al. (2011) found that the amount of Sb accumulated in maize seedling increased with increasing soil Sb concentration, and high concentration of Sb (up to 1000 mg Sb kg^{-1}) in soil clearly reduced plant growth and biomass. On the other hand, it was reported that growth of maize and sunflower seedlings in water containing up to 24 mg Sb L^{-1} for a week did not produce toxicity symptoms in the plants, and there was no significant decrease in the biomass when treated with Sb compared to the untreated control plants (Tschan et al., 2008).

6.2. Anatomical changes of root and leaf tissues caused by Sb

Although there is relatively a broad knowledge about the effects of various metals, like cadmium (Cd), lead (Pb), zinc (Zn), Al, and also metalloids like As on plant tissue structure, very little is known about Sb. Vaculík et al. (2015a) discovered that Sb reduced the thickness and size of intercellular spaces in sunflower leaves, making the leaf tissues more compact and denser. Similar effects were observed in maize roots, where reduced aerenchyma size was detected in the presence of Sb (Vaculíkova et al., 2016). As a result, roots were denser, and tissues were more compact. They also noticed that Sb promotes the development of endodermis suberization closer to the root apex. The similar phenomenon has previously been observed with other metals such as Cd or Zn, (Martinka and Lux, 2004; Zelko and Lux, 2004; Stoláriková et al., 2012; Vaculík et al., 2009, 2012) and it is thought to be a reaction of roots to protect the central cylinder from the uptake of harmful contaminants (Martinka et al., 2014). Recently, Shetty et al. (2021b) reported that Sb is slightly enhancing root lignification in giant reed (*Arundo donax* L.), although much intensive lignification occurred when roots were treated with both Sb and another metalloid Silicon (Si).

6.3. Effect of Sb on reactive oxygen species (ROS) production and plant antioxidant system

Plants subjected to any abiotic stress including metal(loid) toxicity show enhanced accumulation of reactive oxygen species (ROS) which are harmful to plants at higher concentration. Oxidative stress occurs when there is a serious imbalance in any cell compartment between the production of ROS and antioxidant defence. Excess production of ROS such as superoxide (O_2^{-}), hydrogen peroxide (H_2O_2), hydroxyl radicals (HO^{-}) and singlet oxygen ($^1O_2$) cause damage to proteins, lipids, carbohydrate, DNA and ultimately leads to cell death (Ahmad et al., 2009; Shahid et al., 2014). Not many studies have been done on free radical formation in plants upon Sb exposure. Ortega et al. (2017) observed a strong and significant increase in the production of O_2^{-} in both roots and leaves of *H. annuus* upon Sb addition. This increase was found to be dose dependent in case of roots. The authors also found that O_2^{-} affected both the vascular cylinder and epidermal cells in the root elongation zone. Similarly, H_2O_2 content was reported to be increased when Sb was treated at the rate of 50 μM to *A. thaliana* (Wu et al., 2019).

Antioxidant enzymes are used by plants to eliminate excess of ROS which include superoxide dismutase (SOD), catalase (CAT), ascorbate peroxidase (APX), monodehydroascorbate reductase (MDHAR), dehydroascorbate reductase (DHAR), glutathione reductase (GR), and guaiacol peroxidase (GPX) (Ahmad et al., 2009). In these enzymes SOD functions as a quencher to eliminate the radical of the superoxide anion (O_2^{-}) that forms H_2O_2. However, peroxidase (POD), CAT and APX are important intrinsic antioxidative enzymes in plant cells. They help in degradation of harmful H_2O_2 to harmless H_2O (Pereira et al., 2002).

The activities of these enzymatic antioxidants are reported to be higher under Sb toxicity (Feng et al., 2013, 2016; Chai et al., 2016). Vaculíková et al. (2014) found an increased activity of APX and CAT in maize roots with increasing concentration of Sb in the media, whereas GPX was reduced. Behaviour of antioxidant enzymes under Sb stress is complex and less studied. Pan et al. (2011) observed a decreased activity of SOD, and increased activity of CAT and POD under Sb stress in maize seedlings. In contrary, increased activity of SOD was observed in both leaves and roots of sunflower plants with increase in application of Sb (Ortega et al., 2017). In the same experiment, they also observed high APX and POX activity. It is known that activities of enzymatic antioxidants vary with plant species and their level of tolerance (Benhamdi et al., 2014). Feng et al. (2009) set up a hydroponic experiment using four species of fern plants (*P. cretica*, *Microlepia hancei* Prantl, *Cyrtomium fortune* J. Sm. and *Cyclosorus dentatus* (Forssk.) Ching), and observed a significant increase of POD, CAT and APX activities in all the four fern plants under Sb exposure. The unchanged biomass, accumulation of Sb in roots and lower malondialdehyde (MDA) contents in *P. cretica* compared to other ferns with elevated levels of antioxidative enzymes showed the role of antioxidative enzymes in Sb tolerance (Feng et al., 2009).

Most eukaryotic organisms produce ascorbic acid (AsA or vitamin C), a powerful, water-soluble antioxidant as scavenger of ROS to prevent or at least alleviate deleterious effects caused by ROS (Ahmad et al., 2009). Ascorbate is considered a strong antioxidant because of its ability to intervene in redox reactions, and it plays a crucial part in the system for the removal of H_2O_2 via the ascorbate-glutathione (AsA-GSH) cycle. It also plays an important role in cell division, expansion, and plant growth (Zhang et al., 2016). DHAR and GR are the enzymes related to AsA-GSH cycle. These enzymes were studied by Ortega et al. (2017) in sunflower plants under Sb stress and found that DHAR and GR increased in leaves in response to Sb stress (1 mM SbV) which indicates the increased production of AsA. Meanwhile, the content of ascorbic acid in cabbage leaf significantly decreased with increasing soil Sb concentrations but applied in the form of SbIII (Xiao et al., 2015). Recently, Espinosa-Vellarino et al. (2021) found that at 1 mM Sb, MDHAR activities reduced, DHAR increased, and GR remained unaffected in the roots of *Dittrichia viscosa* (L.) Greuter; however, in the leaves, MDHAR activity decreased while GR increased.

Glutathione (GSH) may be the most important intracellular defence against damage by ROS. Tripeptide (-GluCysGly) glutathione is one of the crucial metabolites in plants. Glutathione is a precursor of phytochelatins (PCs), which have an important role in controlling cellular heavy metal concentrations. Glutathione and its oxidized form, GSSG maintain a redox balance in the cellular compartments (Ahmad et al., 2009). Increased amounts of GSH may indicate the response of plants to Sb stress. Ji et al. (2017) observed Sb-thiol complexes in roots of *L. perenne* after treating with Sb. It seemed that Sb(III) was complexed by compounds with thiol groups, e.g. GSH and PCs in the root cells as a part of detoxification to control its entry to shoot. However, decreased amounts of total GSH + GSSG were observed in *H. annuus* under increased concentrations of Sb. An increased glutathione-S-transferase (GST) expression which may be involved in detoxification via catalyzing binding of GSH to Sb was also found (Ortega et al., 2017). The up regulation of GST under Sb exposure in *Miscanthus sinensis* Andersson indicate the role of GSH in antioxidative response to Sb stress (Xue et al., 2015). Espinosa-Vellarino et al. (2021) also suggested that Sb appears to act by binding to thiol groups, which can alter free GSH content and SOD and GST activities. However, these experiments were carried out on very young plants (3–10 days old) grown in conditions of hydroponics with Sb concentrations ranging up to 1 mM. Therefore, we may expect the different behavior of plants that grow from the beginning in natural conditions in soils, where the available concentration of Sb is usually considerably lower.

Phenolics are diverse secondary metabolites (flavonoids, tannins, hydroxycinnamate esters, and lignin) which possess antioxidant properties. Thus, polyphenols can directly eliminate ROS, and can inhibit lipid peroxidation of membranes by scavenging lipid alkoxyl radicals. Phenolic compounds, especially flavonoids, have a great capacity to modify membrane packing and fluidity. These changes affect the ability of ROS to diffuse through the membranes, and in this way decrease the peroxidation reactions (Sharma et al., 2012). Increased production of total phenols and flavonoids was observed by Ortega et al. (2017) in sunflower plants in accordance with increased application of Sb. Relative to the control there was an increase of approximately 100% in roots and 50% in leaves in response to the treatments with Sb. Feng et al. (2020) proposed that pectin in root cell walls constrains Sb outside the root cell of rice, finding a dose-dependent connection between Sb and cell wall pectin and lignin.

6.4. *Antimony and integrity of plant cell membranes*

The peroxidation of lipids (LPO) is considered as the most damaging process known to occur in every living organism. It has been recognized that during lipid peroxidation, products are formed from polyunsaturated precursors that include small hydrocarbon fragments such as ketones, MDA etc., and compounds related to them. Some of these compounds react with thiobarbituric acid (TBA) to form colored products called thiobarbituric acid reactive substances (TBARS). It takes place in both cellular and organelle membranes. The ROS generated due to any stress can react with the membrane lipids and enhance the extent of LPO which result in the loss of membrane integrity (Gill and Tuteja, 2010).

The presence of MDA and TBARS is considered as an indicator of oxidative stress and lipid peroxidation. The increase in MDA/TBARS content clearly indicates the formation of ROS under Sb stress (Benhamdi et al., 2014; Natasha et al., 2019). There are several reports indicating an increased MDA content with increasing concentration of Sb (Vaculíková et al., 2014; Cai et al., 2016; Ortega et al., 2017). An experiment conducted on lichen *Xanthoria parietina* (L.) Th.Fr showed an increase in lipid peroxidation even at low concentration of Sb (0.1 mM) (Paoli et al., 2013). Massive accumulation of TBARS in leaves of *Solanum lycopersicum* L. was observed for variants treated with two highest studied Sb concentrations (50 and 100 mg Sb L^{-1} as Sb(III)) (Peško et al., 2016). Contrasting results were obtained by Corrales et al. (2014). In their experiment with *Trifolium* sp, lower content of MDA was recorded upon Sb exposure. Amount of MDA produced in plants as a response to Sb concentration is a clear indicator of Sb tolerance. Feng et al. (2009) claimed that Sb tolerant plant species, like *P. cretica* and *Cyrtomium fortunei* J.Sm produced higher biomass with lower MDA content in their fronds at higher concentration of Sb. Seventy-eight days experiment with *F. tikoua* showed an increase in MDA content with increase in Sb concentration in early days of experiment. However, in the later stages the MDA content was found to be decreased, suggesting that *F. tikoua* could alleviate lipid peroxidation under different Sb levels over time (Chai et al., 2017).

6.5. *Effect of Sb on photosynthesis*

Antimony in excess amounts is known to affect many physiological processes in plants, mainly photosynthesis (Pan et al., 2011; Feng et al., 2013; Mirza et al., 2017). It is known to decrease chlorophylls probably by interfering with biomolecules involved in synthesis of chlorophylls (Peško et al., 2016). However, there is very little information on the effect of Sb on photosynthesis, which require development further in detail. Chlorophyll contents, net photosynthesis, stomatal conductance, and transpiration in *H. annuus* were negatively affected by Sb (Vaculík et al., 2015a). Decreased amount of chlorophylls and carotenoids were also observed by Zhou et al. (2018) when they treated *Acorus calamus* L. with Sb from 250 to 2000 mg Sb kg^{-1}. Pan et al. (2011) observed that chlorophyll content in maize shoots decreased markedly at higher concentrations of Sb (50–1000 mg Sb kg^{-1}) applied to soil. Photosystem II might be the target site of Sb toxicity as explained by Pan et al. (2011), where they found that the maximum photochemical efficiency (F_V/F_M) responded similar to chlorophyll content in maize shoot at higher concentrations of Sb. The maximum photochemical efficiency (F_V/F_M) was recorded 0.62, and the value 0.8 or above indicate a healthy photosystem apparatus. Similar results were noted in an investigation conducted on an aquatic algae *Synechocystis* sp. Zhang et al. (2010) found that exposure of Sb at 10 mg Sb L^{-1} for 24 h inhibited O_2 evolution, fluorescence yield at 684 nm, maximum quantum yield for primary photochemistry, and damaged cellular components. Site of Sb inhibition was found both on donor and acceptor side of PSII (Zhang et al., 2010). However, we can not neglect the fact that in this study an acute, strong and short-term exposure on algae was shown, therefore, long-term exposure of plants to considerably smaller doses might be different. Anyhow, the effects on photosynthesis are more pronounced when Sb interacts with other non-essential elements, like As. Shetty et al. (2021a) found that net photosynthesis rate was significantly reduced when Sb and As were applied simultaneously compared to Sb alone, which is similar to the case with naturally contaminated area. Combined with the reduction in chlorophyll content, disruption of the large subunit of RuBisCO (RuBisCO LSU) (Spot 1350) were observed in abundance under Sb stress in *M. sinensis* (Xue et al., 2015). There are reports of ultrastructural alterations in response to Sb stress. For example, plasmolysis, impairment of the thylakoid system of the algae and cytoplasmic lipid droplets were found in *Xanthoria parietina* (L.) Th.Fr. when treated with as low as 0.1 mM Sb (Paoli et al., 2013).

As discussed above, Sb can hinder uptake of several macro- and micro-elements (Feng et al., 2013; Ortega et al., 2017). The deficiency of some elements such as nitrogen (N), P and potassium (K) can inhibit photosynthesis. Low concentration of K was found to reduce total chlorophyll content up to 50% (Longstreth and Nobel, 1980). Decreased uptake of K was reported by Shtangeeva et al. (2011) at higher concentration of Sb in *T. aestivum*. The decrease in concentration of Fe and magnesium (Mg) can diminish the synthesis of photosynthetic pigments which in turn can lead to reduced photosynthesis (Ortega et al., 2017). However, enhanced Ca uptake was seen in rice plants exposed to 20 mg Sb L^{-1}, while other essential elements such as K and Cu were decreased, and this was correlated with several antioxidant system parameters, suggesting a Ca induced regulatory mechanism in plants (Zhu et al., 2020b).

7. Alleviation of Sb toxicity in soils and plants

A variety of methods have been reported to ameliorate Sb toxicity (Fig. 3). However, most of the studies are limited to laboratory conditions mostly in artificial growth substates. External application of amendments such as Si and Se to Sb-contaminated substrates have been studied extensively in the pot trials (Feng et al., 2013; Vaculíková et al., 2014, 2016). Recent approaches of amelioration include biochar amendment, utilization of mycorrhiza and other microorganisms, salicylic acid, growth regulators and use of nano particles (Cui et al., 2017; Tandy et al., 2017; Wang et al., 2018; Gu et al., 2019, 2020; Silvani et al., 2019; Wu et al., 2020; Bolan et al., 2022). Yet, more research is needed to determine the optimum strategy for reducing the toxicity of Sb in both plants and soils.

7.1. *Phytoremediation and phytomanagement*

Concept of phytoremediation and phytomanagement of metal(loid) contaminated soils has been well developed in the last decades as cost-effective, efficient, environment- and eco-friendly, in situ applicable, and solar-driven remediation strategy (Ali et al., 2013; Wang et al., 2017a; Wang et al., 2017b; Garbisu et al., 2020; Moreira et al., 2021). There are several reports documenting the uptake of Sb in vegetation

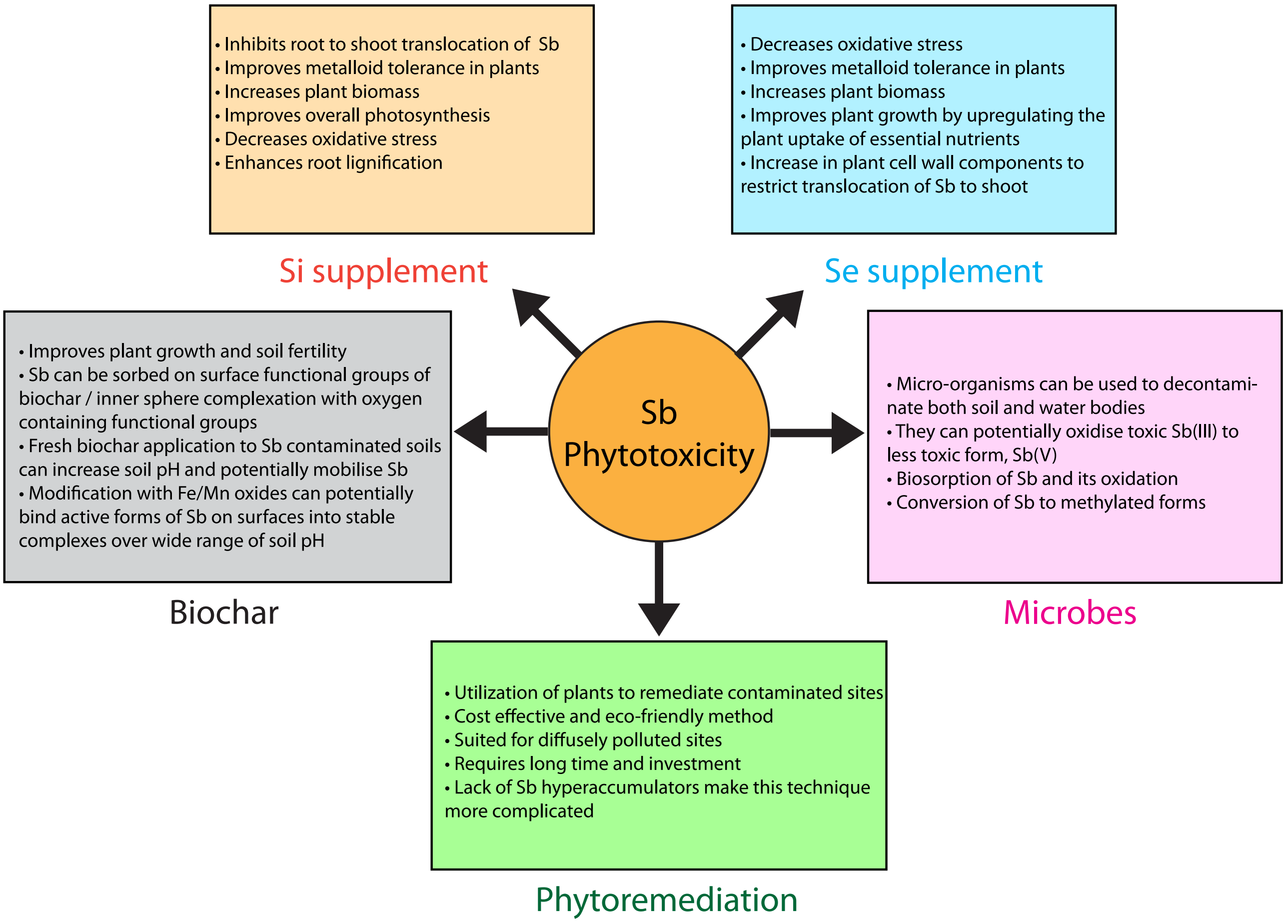

Fig. 3. Possibilities of Sb soil remediation and phytotoxicity alleviation. Sb can be remediated in contaminated waters and soils via application of biochar, iron-based particles, microbes, or by phytoremediation methods such as phytoextraction or phytostabilization. In plants, the potential of arbuscular mycorhizal fungi, and application of Si or Se could mitigate Sb phytotoxic effects.

grown in both active and abandoned mining areas (Baroni et al., 2000; Pratas et al., 2005; Murciego et al., 2007; Okkenhaug et al., 2011; He et al., 2012; Vaculík et al., 2013). Therefore, such plant species might be potentially used for cleaning of Sb-polluted substrates. However, there is always a possibility of foliar exposure of contaminants in the field conditions influencing the measured Sb concentrations in plant tissues, therefore these reports need to be evaluated with caution. Additionally, the ability of plant to take up Sb can also be improved by chemically assisted phytoremediation. For instance, Wang et al. (2018) reported the enhanced As and Sb accumulation in *Brassica juncea* (L.) Czern compared to the control with the addition of thiosulfate in a pot study using metal(loid)-polluted soil (initial soil pH - 8.38) sampled from mining site. However, phytostabilization of the contaminated sites can be a better option compared to phytoextraction, since there is no need to deal with disposal of contaminated plant material. However, phytoextraction helps to gradually decrease the concentration of contaminants in the site over the period (Ali et al., 2013). Nowadays, the concept of phytotechnologies have developed into holistic approach wherein it considers the assessment of suitable site specific phytotechnologies in combination with site management resulting in economic and environmental benefits termed as phytomanagement (Moreira et al., 2021; Mourinha et al., 2022). Even though phytomanagement techniques hold a great promise with environmentally sustainable approach, there are not so many on-site studies to persuade the stakeholders of its economic benefits and widespread employment. Therefore, extensive research employing a phytomanagement strategy on Sb-contaminated lands is highly desirable.

7.2. *Microorganisms*

Microorganisms play a vital role in geochemical cycle of elements. Especially, metal(loid) oxidizing bacteria can be very effective in decontamination of metal(loid) polluted soil and water bodies. For example, novel bacterium *Acinetobacter* sp. JH7, isolated from the mine tailings has been reported to be effective in decreasing Sb toxicity by biosorption and oxidation of Sb(III) to Sb(V) (Gu et al., 2019). Metal(loid)-oxidizing bacteria such as *Ferrovum*, *Thiomonas*, *Gallionella*, and *Leptospirillum*, were found to be highly effective in total Sb and Sb(III) removal from the contaminated water bodies (Sun et al., 2016). Similarly, Sb oxidizing bacteria *Bacillus* sp. S3, has been reported to be promising inoculant for bacteria-assisted phytoremediation on Sb-contaminated sites, which helps in reducing plant uptake of Sb (Gu et al., 2020).

Arbuscular mycorrhizal fungi (AMF) are important microorganisms which play a vital role in functioning of ecosystems including abiotic and biotic stress tolerance (Krishnamoorthy et al., 2015). *Glomus* species of AMF are distributed widely in metal(loid) contaminated soils showing an adaptation to metal(loid) stress. Inoculation of plants with *Glomus* sp. reported to have a promoting effect on shoot biomass (Rasouli-Sadaghiani et al., 2019) and showed an improvement in metal tolerance (Cui et al., 2019). However, very few studies have been carried out with respect to Sb toxicity alleviation by AMF. Wei et al. (2016) suggested that AMF inoculation can help to alleviate Sb toxicity by inhibiting the reduction process of Sb(V) to Sb(III). However, it was also observed that mycorrhizal colonization might also increase plant Sb concentrations mainly by reducing rhizosphere soil pH due to elevating acid phosphatase activity resulting in release of Sb from mineral fractions. Similarly, Pierart et al. (2018) also suggested that there is a risk in increased accumulation of Sb in aerial/edible parts of vegetable crops either by inoculation or naturally present AMF. Recently, Xi et al. (2022) found that AMF could moderate impairment of *P. cretica* fronds, exposed to Sb, by maintaining the sufficient protein levels for ribosomal functioning, photosynthesis activity and to counter ROS production. They further suggested that AMF associated with *P. cretica* might be very useful to enhance phytoremediation effectiveness of Sb contaminated sites (Xi et al., 2022).

7.3. Silicon and selenium

Several studies have shown that external application of additives such as silicon (Si) and selenium (Se) can reduce the uptake of Sb and ameliorate its toxicity in crop plants. Silicon has been proven to have beneficial effects on plants (Lux et al., 2002) despite of its non-essentiality. Soil application or root exposure to Si/Se has been reported to be beneficial for the overall plant growth and development compared to foliar application (Pilon et al., 2013), and proved to be more effective in enhancing wheat growth in Cd-contaminated soils and decreasing Cd concentrations in wheat grains (Zhou et al., 2021). Many studies have revealed that Si helps to improve their tolerance against metal(loid) stresses such as Cd (Vaculík et al., 2009, 2015b), chromium (Cr) (Ahmad et al., 2016), As (Pandey et al., 2016) etc. Huang et al. (2012) demonstrated that 1 mM Si in the form of potassium silicate significantly inhibited Sb transfer from root to shoot, and reduced Sb accumulation in shoots of rice plants. Similarly, Si addition to Sb treated roots was found to decrease oxidative stress symptoms documented by lower lipid peroxidation, proline accumulation, and decreased activity of antioxidative enzymes (ascorbate peroxidase, catalase, and guaiacol peroxidase) (Vaculíková et al., 2014). It also mitigated the toxic effects of Sb on root morphology and enhanced root aerenchyma formation (Vaculíková et al., 2016). Silicon (1 mM) in the form of sodium silicate applied to Sb-treated *A. donax* plants significantly improved photosynthesis, contents of photosynthetic pigments, growth, and biomass by reducing Sb translocation to the upper parts of the plant. Additionally, the roots of plants where Si and Sb were applied together had increased lignification which in turn might have restricted the translocation to the upper parts (Shetty et al., 2021b).

Selenium in plants is considered as an antioxidant (Hartikainen et al., 2000). Several possible ways have been suggested for Se to be involved in detoxifying metal(loid)s: 1) alleviating the oxidative stress generated from metal(loid)s; 2) preventing the metabolism of metal(loid)s; and 3) inhibiting the uptake of metal(loid)s through the antagonistic effects (Zhu et al., 2009). Very few studies have been conducted on mitigatory effect of Se on Sb. Addition of 1 mg Se L^{-1} in the form of sodium selenite with 5 mg Sb L^{-1} decreased leaf MDA content and increased biomass of rice plants, indicating an antagonistic role of Se to Sb, and suggested that Se due to its antioxidant property might have alleviated the Sb-induced oxidative stress (Feng et al., 2011b). Feng et al. (2013) proposed that Se reduces the toxicity of Sb by (1) the direct inhibition of Sb uptake; and (2) by regulating the uptake of some essential elements, such as Ca, Mg and K; however, the actual mechanism of Se on Sb toxicity is still unknown (Feng et al., 2016). Meanwhile, Liu et al. (2019) found that the reduced concentration of Sb in roots in the presence of Se could be due to changes in cell wall components, with increased formation of pectin, hemicellulose, and lignin. However, most of Sb-alleviating results using Si and Se comes from hydroponics, therefore, addition of Si and Se-based soil amendments to prove their mitigation potential in Sb-polluted soils would be essential.

7.4. Biochar

The other possible ways to reduce Sb toxicity may be through addition of biochar (Cui et al., 2017; Silvani et al., 2019). Biochar is the carbon-rich product obtained by thermal decomposition of organic material under limited supply of oxygen (O_2), and at relatively low temperatures ($< 700\ ^{\circ}C$) (Lehmann and Joseph, 2009). A pot culture study was conducted to understand the effect of commercially produced biochar application (feedstock source: not mentioned) on maize plants grown in Sb spiked soil (Zhu et al., 2020a). This study reported that addition of biochar reduced Sb toxicity symptoms by improving antioxidant enzyme activity, increasing nutrient (N, P, and K) uptake, and decreasing Sb concentration in shoots by reducing Sb translocation from root to shoot. Iron enriched biochar (Source: *Eupatorium adenophorum* (Spreng.) King & H.Rob) using iron oxide-hydroxide (FeOOH) found to have stronger effect in Sb remediation compared with untreated biochar application (feedstock source: timber waste) to military shooting range soil which was again dependent on the oxygen containing surface functional groups, including iron oxides (Silvani et al., 2019). Cui et al. (2017) reported that inner-sphere complexation with oxygen-containing functional groups and coordination with π electrons with Sb(III) might be possible sorption mechanisms on biochar. Further, they reported the catalytic oxidation of Sb(III) to Sb(V) on biochar surface. In addition, biochar application was found to influence Sb speciation in soil, via changing the activity of bacteria that can cause Sb mobilization or immobilization (Hua et al., 2021). However, application of biochar has been reported to have negative impact on remediation by increasing the mobility of Sb in soil bound to organic/ inorganic binding sites due to increase in soil pH. Increase in the application rate of wood biochar from 0% to 5% has remarkedly shown 2-fold increase in soil Sb availability in a *technosol* (Lomaglio et al., 2017). Modified biochar, on the other hand, is an emerging option for immobilizing hazardous metal(loid)s in soil. A study conducted on Fe-modified rice husk hydrochar showed its potential ability to adsorb and immobilize Sb, both in aqueous solution and in soil. The Fe oxides formed during hydrothermal process thought to have combined with Sb to form geochemically stable minerals, and thereby immobilizing it and making it unavailable to plants (Teng et al., 2020). Additionally, Fe-modified biochar was shown to increase the efficiency of Sb remediation significantly (Silvani et al., 2019).

7.5. Iron containing amendments

Iron-based amendments have been reported to have high selectivity for Sb (Mariussen et al., 2015; Okkenhaug et al., 2013, 2016; Silvani et al., 2019). Antimony retention was found to be irreversible in ferrosols (Li et al., 2018) indicating the affinity of Sb to form stable complex with Fe-(hydro)oxides. Even the oxidised forms, Sb(V) on the surface found to be tightly bound because of the highest clay content and crystalline Fe and Al oxides. Application of $FeSO_4$ was found to be very effective in lowering Sb leaching from soil by the reduction of Sb(V) to Sb(III), which binds more strongly to Fe hydroxides over a wide range of pH (Tandy et al., 2017). Amorphous iron oxides are known to form surface complexes with both cationic and anionic substances (Okkenhaug et al., 2013; Silvani et al., 2019). Combined application Fe-Mn oxides with organic amendments such as peat soil have proven to reduce the Sb uptake in *Brassica campestris* L. (Rong et al., 2021). Sekula et al. (2018) showed an efficient way how to eliminate Sb and As in mining drainage water from old mines by use of zero-valent iron. Nano-zero valent iron (nZVI) is known to adsorb inorganic contaminants and form surface mediated complexes such as As(III, V), Sb(III, V), U (VI), Cr(VI), Pb(II), Cd(II) (Kanel et al., 2005; Dorjee et al., 2014; Zhao et al., 2016; Galdames et al., 2020; Shaheen et al., 2022). Studies conducted by Dorjee et al. (2014) showed that nZVI reduces Sb(V) to Sb(III) and got adsorbed via chemisorption. Hiller et al. (2021) also reported nZVI as a very successful tool for Sb immobilisation in topsoils collected from the different industrial origin, and suggested it as suitable for application to multimetal(loid)-contaminated soils (Sb, As, Pb) at a wide pH range. Similarly, nano MgO was proven to have adsorption ability to Sb(III) from water (Xu et al., 2020). Combination of nanomaterials and plant growth promoting microorganisms can be a great prospect in removal of Sb from contaminated soil. However, detailed investigations involving large scale pilot plot or field experiments are required to know the role of these additives in mitigating the toxicity of Sb.

8. Conclusion and future prospect

Toxicity of various elements in high concentrations, such as Cd, Pb, or As has been investigated for a long time in all living systems. In this view, research interest for Sb was a bit neglected, although the level of knowledge about behaviour and toxicity of Sb for the environment, including plants, has dramatically increased during the last years, and is currently summarized also in this review. Although Sb is primarily taken up by plant roots and root – to – shoot translocation of Sb is in many plant species limited, still the risk of human and animal diet intoxication exists through consumption of food and feed made from contaminated crops, fodder or through herbal products prepared from plants collected on contaminated sites. Moreover, in this review we aimed to show the current state of the art about possible mechanisms of Sb toxicity alleviation in soil-plant level. We have also identified some missing gaps in our knowledge that need to be consider in the future:

- There are significant gaps in the mechanisms of uptake and translocation, particularly for Sb(V). Unlike other metal(loid)s, topics such as Sb transport, xylem loading, detoxification, and vacuole sequestration etc., received less attention and must be thoroughly investigated in order to broaden our understanding of Sb behavior in plants.
- Plants are the primary components of the food chain, therefore mitigation mechanisms of Sb toxicity both at substrate and plant levels cannot be ruled out. This is important for production of safe food and feed and closely relates with use of Sb contaminated water for irrigation.
- The use of specific plant species to clean metal polluted substrates (Cd, Zn, Ni, As, etc.) via phytoextraction is a well-known topic. However, not many Sb accumulators have been identified so far, nor it is explained how they achieve efficient Sb translocation. Most of the species rather showed less Sb translocation. Therefore, a deep screening of potential new Sb (hyper)accumulators on contaminated soils is important. Alternatively, gathering the mechanisms how reduced root-to-shoot transfer of Sb might be important for use of plants for phytostabilisation of highly Sb contaminated mining sites in order to set up suitable and achievable phytomanagement strategies.
- The use of elements such as Si, Se, and other soil amendments like biochar, and microorganisms are emerging options for reducing Sb toxicity. Use of nanoscale zero valent iron (nZVI) and Fe-modified biochar has been proven to be more efficient in Sb remediation than unmodified biochar; even in the multi-metal(loid) contaminated sites with Sb, As and Pb. However, a real focus on field-trial studies, and deep understanding of mechanism of their interaction with various soil types, microbiome, site conditions and wide spectra of plants should be in a view of the scientific community in the coming years.
- Iron has been shown as efficient agent to bind the soluble As and Sb from contaminated mining water. It is worth to know whether Fe nanoparticles could be offered to plants and whether this nano-phytotechnology could decrease the root exposure of soluble Sb concentrations.

CRediT authorship contribution statement

VCS and MV conceptualised the draft, VCS, RS and MV wrote the original draft, VCS prepared tables, RS prepared figures, MV revised and edited the draft, all authors have commented and approved the final manuscript.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

No data was used for the research described in the article.

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