

# (S, S)-Ethylenediaminedisuccinic Acid (free acid)

## Crop Production

### Identification of Petitioned Substance

<b>Chemical Name:</b>	16	<b>Trade Names:</b>
Ethylenediamine-N,N'-disuccinic acid	17	
<b>IUPAC name:</b>		<b>CAS Number :</b>
2-[2-(1,2-dicarboxyethylamino)ethylamino]butanedioic acid.	18	20846-91-7
	19	
<b>Other Names:</b>		<b>Other Codes:</b>
EDDS, EDSS, Ethylenediamine-N,N'-disuccinic acid,	20	
N,N'-ethylenedi-L-aspartic acid, N,N'-Ethylenedi-L-	21	
aspartic acid, N,N'-Ethylenediamine disuccinic acid,	22	
N,N'-Ethylenediaspartic acid	23	
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### Characterization of Petitioned Substance

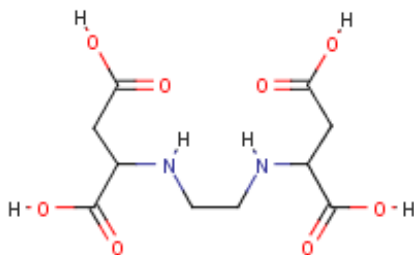
Note: Chelation is a process in which free metal ions combine with ligands (chelators, chelating agents) to form metal complexes. With respect to free metal ions, metal ions in complexes are less reactive, less subject to precipitation processes, and remain water-soluble for a longer time. Nutrient metals stay water-soluble for a longer time so that plants/animals assimilate more. Toxic metals also stay water-soluble for a longer period of time and cause more damage such as suppressing plant growth. Previously precipitated/adsorbed metal ions form complexes with available chelating agents, are released back to water-soluble, and cause different effects (such as being transported to underground water or to different geographical locations). Some basic concepts and issues related to chelation such as complex stability, ligand stability, reversible processes, competition processes, etc, are presented in Appendix A: Chelation and related issues.

#### Composition of the Substance:

Ethylenediamine-N,N'-disuccinic acid (EDDS) is one of the aminopolycarboxylic acids (APCAs). One commercial product is tri-sodium salt (Na<sub>3</sub>-EDDS) with a CAS number of 178949-82-1.

There are two chiral centers in the structure of EDDS (Fig. 1) and consequently there are two enantiomeric isomers: (R,R')-EDDS, and (S,S')-EDDS, and one meso isomer (R,S)-EDDS (Neal and Rose, 1968; Schowanek et al., 1997). These isomers have about the same efficiency, in terms of complex stability constants, in forming complexes with metal ions (Orama et al., 2002). As given below in the section of "Biodegradability," (R,R)-EDDS and (R,S)-EDDS are partially or wholly un-biodegradable. Most literature is focused on (S,S')-EDDS. (S,S')-EDDS is denoted as EDDS hereafter, unless otherwise specifically noted.

Fig. 1: Structure of EDDS



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## Properties of the Substance:

### Basic Properties

The basic properties of EDDS are listed in Table 1: Physical and chemical properties of EDDS (P&G Environmental Science data; US EPA 40 CFR Part 180.920-Documents 0002; Sigma MSDS).

Table 1: Physical and chemical properties of EDDS acid

Molecular formula	C <sub>10</sub> H <sub>16</sub> N <sub>2</sub> O <sub>8</sub>	Molecular weight	292.24
Density	1.59 g/cm <sup>3</sup>	Melting point	220-222°C
Solubility in water	slightly soluble	Acidity (pKa)	2.4, 3.9, 6.8, 9.8

EDDS is a white granular solid without characteristic odor. One commercial product of tri-sodium salt is prepared in solution form. The flammability of EDDS is not high. The solubility of EDDS acid is 0.015 g/100 g at 20°C, but the solubility of tri-sodium salt (Na<sub>3</sub>-EDDS) is > 1000 g/L at 20°C. These properties are very similar to the chelating agent EDTA (ethylenediaminetetraacetic acid). No other unique or special properties are available.

### Biodegradability

Biodegradation is a biotic process in which a substance is decomposed to components by microorganisms (as a reference, for example, a photodegradation is an abiotic process in which a substance is decomposed under light exposure). EDDS is a chelating agent and specifically exists as a ligand (i.e. EDDS acid) or as a complex (i.e. EDDS complex). The stabilities of EDDS acid and EDDS complex are different and should be investigated respectively (Metsarinne et al., 2001; Vandevivere et al., 2001; Bucheli-Witschel & Egli, 2001; and Nowack, 2002).

Based on the result of Schowanek et al. (1997), out of the three isomers of EDDS, (S,S)-EDDS was biodegradable, while (R,R)-EDDS and (R,S)-EDDS were partially or wholly un-biodegradable. In that research, those three types of EDDS compounds were labeled with radioactive <sup>14</sup>C isotope and added to a simulated sewage system. After these compounds were degraded, organic carbon was decomposed to inorganic carbon (i.e. CO<sub>2</sub> gas). By collecting CO<sub>2</sub> gas above the sewage system, measuring its <sup>14</sup>C radioactivity, and comparing the measured activity with the originally added <sup>14</sup>C activity, the percentages of degraded EDDS compounds were calculated. Close to 96% of (S,S)-EDDS was found mineralized (degraded) within one month. Similar conclusion was also made by Takahashi et al. (1997).

Vandevivere et al. (2001) investigated the stability of EDDS complex in a sewage/sludge treatment simulation test. It was found that the biodegradability (or the stability) of EDDS complex was metal ion dependent. For example, the concentrations of some EDDS complex (Cr, Cd, Mg, and Pb) decreased to less than 20% of original concentrations within 5-10 days in the test, while the concentrations of other EDDS complex (Cu, Co and Ni) remained virtually unchanged for more than 15 days.

The following is directly quoted from US EPA 40 CFR Part 180.920-Documents 0002:

#### IV. Environmental Fate and Drinking Water Considerations

(S,S)-EDDS is a chelating agent. Several studies of (S,S)-EDDS describe it as a naturally occurring compound from strains of bacteria (Bucheli-Witschel, 2001; Goodfellow, 1997; Witschel, 1998). (S,S)-EDDS was “isolated from culture filtrate of the actinomycete *Amycolatopsis orientalis*” during an antibiotic screening program (Bucheli-Witschel, 2001). In examining environmental fate and biodegradation, studies describe (S,S)-EDDS as degrading rapidly (Witschel, 1998; Bucheli-Witschel, 2001). The rapid biodegradation properties of (S,S)-EDDS will greatly reduce the amount that could occur in run-off into drinking water. Therefore, the Agency has determined that contributions of concern to drinking water are not expected from the use of this chemical as an inert ingredient in pesticide formulations applied to growing crops.

The paper of Bucheli-Witschel (2001), cited above, actually is Bucheli-Witschel and Egli (2001). This paper is a 38-page long review article titled “Environmental fate and microbial degradation of aminopolycarboxylic acids,” extensively focused on NTA (nitrilotriacetate) and EDTA. Merely ¼ page on page 84 of the paper was devoted for

111 “Biodegradation of EDDS.” The discussion was totally based on the works of Takahashi et al. (1997) and Schowanek  
 112 et al. (1997). The paper of Witschel (1998) actually is Witschel and Egli (1998). This paper is titled “Purification  
 113 and characterization of a lyase from the EDTA-degrading bacterial strain DSM 9103 that catalyzes the splitting of  
 114 [S,S]-ethylenediaminedisuccinate, a structural isomer of EDTA,” and is not specifically about the rate at which EDDS  
 115 is biodegraded.

116  
 117 Bucheli-Witschel and Egli (2001) and Nowack (2002) indicated that the literature about the environmental chemistry  
 118 of several aminopolycarboxylates including EDDS was very sparse. Vandevivere et al. (2001) stated that the  
 119 biodegradability of metal-EDDS complex was not well established. The research by Takahashi et al. (1997) and  
 120 Schowanek et al. (1997) were mainly about the biodegradation of EDDS acid in simulated sewage systems. The  
 121 biodegradability of EDDS in soil was investigated (Hauser et al., 2005; Meers et al., 2005; Tandy et al., 2006; and  
 122 Meers et al., 2008). A general conclusion was that EDDS was biodegradable in soil. However, more research might  
 123 still be needed, as discussed in Appendix B: Biodegradation of EDDS.

124  
 125 Chelating Capacity

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 127 Aminopolycarboxylic acids (APCAs) form strong and water-soluble complexes with metal ions (Bucheli-Witschel  
 128 and Egli, 2001; Nowack, 2002). EDDS, one of the APCAs, forms stable hexadentate (six binding sites, see Appendix  
 129 A) chelates with metals such as copper, zinc and lead. The stability constants are metal dependent. For example, the  
 130 constants are, expressed as log (k) where k is a stability constant, 22.0, 18.4, 13.4 and 8.57, respectively for Fe(III),  
 131 Cu(II), Zn(II) and Mn(II) (Orama et al., 2002). Stability constants are not always available from experimental data  
 132 and frequently estimated from some basic thermodynamic properties. The constants of some metals and radionuclides  
 133 are listed in Jones and Williams (2001), Vandevivere et al. (2001), and Bucheli-Witschel and Egli (2001).

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 135 Both of EDDS and EDTA are aminopolycarboxylic acids with similar structures (Fig. 2 Structure of EDTA). The six-  
 136 member rings of EDDS function effectively as chelating agents (Fig. 3. Metal-EDDS complex).

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 138 Fig. 2 Structure of EDTA

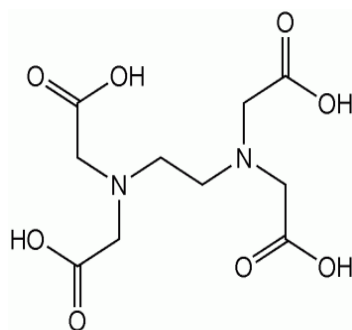
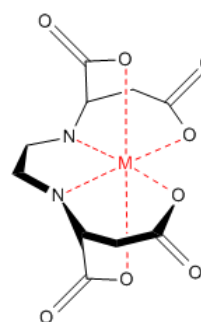


Fig. 3 Metal-EDDS complex



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 142 The chelating capacity of EDDS is compared to that of EDTA, in terms of complex stability constant. One example is  
 143 shown below.

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Formation Reaction	Formation Constant
$[\text{Fe}(\text{H}_2\text{O})_6]^{3+} + (\text{S,S})\text{-EDDS}^{4-} \rightarrow \text{Fe}[(\text{S,S})\text{-EDDS}]^- + 6 \text{H}_2\text{O}$	$K_{\text{EDDS}} = 10^{20.6}$
$[\text{Fe}(\text{H}_2\text{O})_6]^{3+} + \text{EDTA}^{4-} \rightarrow \text{Fe}(\text{EDTA})^- + 6 \text{H}_2\text{O}$	$K_{\text{EDTA}} = 10^{25.1}$

145  
 146 In the work of Jones & Williams (2001), stability constants of the EDDS and EDTA complexes were estimated for  
 147 these ions: Sb, Co, Mn, Ce, Ru, Eu, Pu, Am, UO<sub>2</sub>, NpO<sub>2</sub>, Fe, Cr, Ni, Mg, Zn, Mo, Nd, and Gd, respectively. The ratio  
 148 of (log K<sub>EDDS</sub>)/(log K<sub>EDTA</sub>) was 0.76±0.13. Based on this, the ETDA complex is generally more stable than EDDS  
 149 complex.

150  
 151 The estimated formation and stability of a complex, based on the stability constants, is a “thermodynamic approach”  
 152 and is a “possibility.” The actual formation and stability of a complex is determined by the kinetics in which the  
 153 complex is formed (Nortemann, 1999; Nowack, 2002; and Bucheli-Witschel and Egli, 2001). In a specific case, the  
 154 stability constant alone might be insufficient to predict the biodegradability or stability of a metal complex

155 (Vandevivere et al., 2001). Chelating agents are used in soil washing and phytoextraction. There was sufficient  
 156 evidence to suggest that copper and zinc were more stable with EDDS than with EDTA, while lead and cadmium  
 157 were less stable with EDDS than with EDTA (Kos & Lestan, 2003; Luo et al., 2005; Meers et al., 2005; and Ko et al.,  
 158 2010).

159  
 160 Toxicity

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 162 The following toxicity values are directly quoted from Table 2 of US EPA 40 CFR Part 180.920-Documents 0002.

163  
 164 “Table 2. Acute Toxicity”

166 Parameter	166 Toxicity Value	166 MRID
167 Oral LD50 - rats	> 2,700 mg/kg	46323201
168 Dermal LD50 – rat	> 2,000 mg/kg	46309104
169 Inhalation LC50 - rat	> 1.49 mg/L	46323203
170 Eye irritation - rabbit	Mild irritant	46309107
171 Dermal irritation - rabbit	Not an irritant	46323205
		46309109
173 Dermal sensitization – guinea pig	Not a sensitizer	46309110
		46309111

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 177 The following statements are directly quoted from US EPA 40 CFR Part 180.920-Documents 0002.

178  
 179 “The available studies show (S,S)-EDDS is poorly absorbed and rapidly excreted from the body,  
 180 and it has low toxicity in acute, repeat dose, and developmental studies. The results of the (S,S)-  
 181 EDDS studies indicate developmental toxicity is present only at high dosage levels (limit dose)  
 182 and only in the presence of maternal toxicity (limit dose). Based on this information, there is low  
 183 concern, at this time, for increased sensitivity to infants and children to (S,S)-EDDS when used as  
 184 an inert ingredient in pesticide formulations. For the same reason, a safety factor analysis has not  
 185 been used to assess risk and, therefore, the additional ten-fold safety factor for the protection of  
 186 infants and children is also unnecessary.”

187  
 188 “Considering the low toxicity, poor absorption, and rapid biodegradation properties of (S,S)-  
 189 EDDS, residues of concern are not anticipated from dietary exposures (food and drinking water)  
 190 or from residential exposures (inhalation and dermal). Utilizing a highly conservative aggregate  
 191 exposure assessment, EPA has concluded that aggregate exposures to (S,S)-EDDS are more than  
 192 three orders of magnitude less than the dose at which no adverse effects were seen in the most  
 193 sensitive animal study and are therefore below the level of concern. In addition, this highly  
 194 conservative exposure assessment is protective of any possible non-occupational exposures to  
 195 (S,S)-EDDS as it results in exposure estimates orders of magnitude greater than the high-end  
 196 exposure estimates for residential uses of pesticides routinely used by the Office of Pesticide  
 197 Programs.”

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 199 “Taking into consideration all available information on (S,S)-EDDS, it has been determined that  
 200 there is a reasonable certainty that no harm to any population subgroup, including infants and  
 201 children, will result from aggregate exposure to this chemical when used as an inert ingredient in  
 202 pesticide products when considering dietary exposure and all other nonoccupational sources of  
 203 pesticide exposure for which there is reliable information. Therefore, the exemption from the  
 204 requirement of a tolerance for residues of (S,S)-EDDS under 40 CFR 180.920 can be considered  
 205 safe under section 408(q) of the FFDCA.”  
 206



207  
 208 **Specific Uses of the Substance:**

209  
 210 EDDS is mainly used as a chelating agent (e.g. Grzman et al., 2003; Tandy et al., 2004; Hauser et al., 2005; Duquene  
 211 et al., 2009; and Ko et al., 2010). Metal ions such as iron, manganese, copper, cadmium and lead cause various kinds

212 of interferences and problems in different application areas such as industry, agriculture, consumer products, etc.  
213 Chelating agents combine with metal ions to form complexes and keep these ions water-soluble.

214

215 The following is directly quoted from the petition (page 4 of the petition).

216

217 “The proposed substance ([S,S]-EDDS free acid, CAS # 20846-91-7, trade name Enviomet C265),  
218 is intended to be used as an inert ingredient in multiple pesticide formulations. It is intended to be  
219 a substitute for other, less readily biodegradable synthetic chelating agents currently used in these  
220 pesticides. The pesticide formulations containing EDDS are intended to be sold to kill pests of  
221 many types. These pesticides are registered for use on and around nearly all crop types (fruits,  
222 vegetables, fruit trees, outdoor ornamentals, lawns, greenhouses, field crops, nurseries, etc.) in  
223 both the homeowner and commercial agricultural markets.”

224

225 Based on the majority of literature, EDDS is used as a chelating agent but not as other specific uses.

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### 227 **Approved Legal Uses of the Substance:**

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229 U.S. Environmental Protection Agency:

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231 The U.S. Environmental Protection Agency (EPA) established the exemption from the requirement of a tolerance for  
232 residues of (S,S)-EDDS when used as an inert ingredient sequestrant or chelating agent in pesticide formulations  
233 applied to growing crops only under 40 CFR Part 180.920 (EPA-HQ-OPP-2008-0250; FRL-8362-4; effective  
234 November 14, 2008).

235

236 The following is directly quoted from US EPA’s Document 0001 (US EPA 40 CFR Part 180.920).

237

238 **“SUMMARY:** This regulation establishes an exemption from the requirement of a tolerance for  
239 residues of (S,S)-Ethylenediaminedisuccinic acid (CAS Reg. No. 20846-91-7) ((S,S)-EDDS)  
240 when used as an inert ingredient sequestrant or chelating agent in pesticide formulations applied to  
241 growing crops only under 40 CFR 180.920. Associated Octel Company, Limited, submitted a  
242 petition to EPA under the Federal Food, Drug, and Cosmetic Act (FFDCA), as amended by the  
243 Food Quality Protection Act of 1996 (FQPA), requesting an exemption from the requirement of a  
244 tolerance. This regulation eliminates the need to establish a maximum permissible level for  
245 residues of (S,S)-Ethylenediaminedisuccinic acid.

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247 **DATES:** This regulation is effective November 14, 2008.”

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250 U.S. Food and Drug Administration:

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252 The U.S. Food and Drug Administration (FDA) approved Food Contact Notification (US FDA FCN 000799). In this  
253 FCN, Innospec requested that the tri-sodium salt of EDDS (a close chemical relative of EDDS) be allowed as a  
254 chelating agent in the manufacture of food-contact paper and paperboard. FDA approved that request on February 6,  
255 2008 (FDA FCN 000799).

256

257 The following is directly quoted from FDA Memo “FCN No. 799 – [S,S]-ethylenediaminedisuccinic acid, trisodium  
258 salt, as a chelating agent in the manufacture of paper and paperboard” (US FDA FCN 000799).

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260 **“Finding of No Significant Impact:**

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262 A food contact notification (FCN No. 799), submitted by Innospec Limited, to provide for the safe  
263 use of (S,S)-ethylenediaminedisuccinic acid, trisodium salt, as a chelating agent in the  
264 manufacture of paper and paperboard.

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266 The Environmental Review Team has determined that allowing this notification to become  
267 effective will not significantly affect the quality of the human environment and, therefore, will not  
268 require the preparation of an environmental impact statement. This finding is based on  
269 information, submitted by the notifier, in the notification, which includes an environmental  
270 assessment, dated December 24, 2007.”

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#### Others

No other major approved uses of EDDS were found.

#### **Action of the Substance:**

EDDS combines with metal ions to form metal complexes. The process and the product depends on the metal ions and other parameters such as pH (e.g. Nowack, 2002; Orama et al., 2002; Wu et al., 2007; Yip et al., 2009).

For example, the stability constants of several metal-EDDS complexes, as summarized in Tandy et al. (2006), are  $10^{6.34}$ ,  $10^{7.77}$ ,  $10^{12.7}$ ,  $10^{14.46}$ ,  $10^{20.46}$  and  $10^{23.68}$  for  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Cd}^{2+}$ ,  $\text{Pb}^{2+}$ ,  $\text{Cu}^{2+}$  and  $\text{Fe}^{3+}$ -EDDS complexes, respectively. EDDS preferentially combines with transition metals and/or heavy metals such as Cd, Pb, Cu and Fe over alkaline elements such as Ca and Mg. By using this differential action, EDDS was used for the preferential removal of heavy metals from contaminated soil (Grcman et al., 2003; Tandy et al., 2004).

#### **Status**

#### **U.S. Environmental Protection Agency:**

See above in "Approved legal uses of the substance."

#### **U.S. Food and Drug Administration:**

See above in "Approved legal uses of the substance."

#### **Association of American Feed Control Officials, Inc.**

No specific items were found.

#### **International:**

No specific items were found.

#### **Evaluation Questions for Substances to be used in Organic Crop or Livestock Production**

#### **Evaluation Question #1: Is the petitioned substance formulated or manufactured by a chemical process?** **(From 7 U.S.C. § 6502 (21).)**

As proposed in the petition, EDDS is produced by mixing two major chemicals and several other supporting chemicals (calcium hydroxide, sodium hydroxide, and hydrochloric acid) in a heated and pressurized reactor to react chemically under pre-specified conditions for a duration of about half a day. The reaction product is crystallized after acidification and then is separated, as solid material, from other byproducts with physical processes.

As a reference, (S,S)-EDDS was produced chemically starting from the chemicals of L-aspartic acid, sodium hydroxide, and 1,2-dibromoethane at a laboratory experimental scale (Neal and Rose, 1968). L-aspartic acid was reacted with sodium hydroxide at low temperature (ice-bath) to form sodium L-aspartate salt. Other chemicals (sodium carbonate and ethanol) were added and the mixture was heated to reflux. The other major chemical, 1,2-dibromoethane, was added slowly to the heated mixture. The reaction was given about one day to finish. The reaction product was cooled, acidified, and separated physically from other by-products.

328 (S,S)-EDDS was produced when L-aspartic acid was reacted to 1,2-dibromoethane, (R,R)-EDDS was produced when  
329 D-aspartic acid was reacted to 1,2-dibromoethane, and the mixture of 25% (S,S)-EDDS, 50% (R,S)/(S,R)-EDDS and  
330 25% (R,R)-EDDS was produced when maleic anhydride was reacted to ethylene diamine (Neal and Rose, 1968; .  
331 Schowanek et al., 1997).

332  
333 **Evaluation Question #2: Is the petitioned substance formulated or manufactured by a process that chemically**  
334 **changes the substance extracted from naturally occurring plant, animal, or mineral sources? (From 7 U.S.C. §**  
335 **6502 (21).)**

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337 EDDS was reported to be produced naturally by a number of microorganisms (Nishikiori et al., 1994; Zwicker et al.,  
338 1997; and Takahashi et al., 1999; Bucheli-Witschel and Egli, 2001). EDDS is a pure chemical compound with a  
339 relatively simple molecular structure but not a mixture of compounds or a substance with great structure complexity  
340 and/or component variations. No evidence is available to indicate that the manufactured EDDS would be different  
341 chemically from the naturally occurring EDDS.

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343 **Evaluation Question #3: Is the petitioned substance created by naturally occurring biological processes? (From**  
344 **7 U.S.C. § 6502 (21))**

345  
346 EDDS was found to be produced by microorganisms in soil (Nishikiori et al., 1984).

347  
348 In a laboratory fermentation experiment, EDDS was produced by bacteria *Amycolatopsis orientalis* (Zwicker et al.,  
349 1997). With feeding solutions of glycerol (carbon source), glutamic acid (phosphorus source) and urea (nitrogen  
350 source), the concentration of EDDS in fermentation medium reached to 20 g/L after about 42 days of fermentation  
351 time. The fermentation medium must be low in zinc content. In fact, the fermentation was carried out in glass  
352 containers only and the scale-up in larger (metal) tanks was not realized. The product EDDS was purified using a  
353 three-step process consisting of an acid precipitation, an ethanol washing, and a final crystallization step.

354  
355 EDDS was produced by bacteria isolated from soil and sludge (Takahashi et al., 1999). In a reaction mixture  
356 composed of ethylenediamine (200 mmol/L) and fumaric acid (200 mmol/L) in 50 mmol/L phosphate buffer (pH7.5)  
357 at 30°C, the bacterium *Acidovorax* sp. (TNT149) produced 71 mmol/L (21 g/L) of EDDS in 24 hours. Other bacteria,  
358 *Sphingomonas*, *Brevundimonas*, or *Pseudomonas*, produced less than 1 g/L of EDDS in 24 hours.

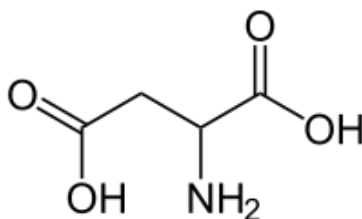
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360 **Evaluation Question #4: Is there environmental contamination during the petitioned substance's manufacture,**  
361 **use, misuse, or disposal? (From 7 U.S.C. § 6518 (m) (3).)**

362  
363 Publicly available data are scarce about the environmental contamination during the EDDS' manufacture, use,  
364 misuse or disposal. From the MSDS (material safety data sheet), EDDS is not considered as hazardous. EDDS is  
365 considered to be low-toxicity by US EPA and allowed to be used in food-contacting paper and paper board by US  
366 FDA. EDDS chemically functions like EDTA and provides strong chelating capabilities.

367  
368 Two major chemicals are used for manufacturing the product of EDDS, as proposed in the petition. One chemical is  
369 L-aspartic acid with CAS # 56-84-8, and the other chemical is dibromoethane with CAS # 106-93-4.

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Aspartic acid CAS # 56-84-8

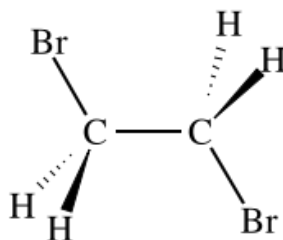


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378 Aspartic acid (L-aspartic acid, asparagic acid, or S-aminobutanedioic acid), one of the nonessential amino acids,  
379 occurs in animals and plants, especially in young sugar cane and in sugar beet molasses. Aspartic acid is allowed in  
380 the compounds for use in foods for infants and children, as listed in Codex Alimentarius (Codex – L aspartic acid).

381  
382 Aspartic acid is colorless crystals with a low to medium solubility (0.45 g/100 mL). Dust explosion is possible if in  
383 powder or granular form, mixed with air. If dry, aspartic acid can be charged electrostatically by swirling, pneumatic  
384 transport, pouring, etc. Aspartic decomposes on burning producing toxic gases including nitrogen oxides. Aspartic  
385 acid reacts violently with oxidants. The substance can be absorbed into the body by ingestion. Other relevant risk  
386 evaluations are listed in “International chemical safety cards” (ICSC-aspartic acid) and in “National Institute for  
387 Occupational Safety and Health” (NIOSH-aspartic acid).

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Dibromoethane CAS # 106-93-4



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393  
394 The other chemical is dibromoethane (or 1,2-dibromoethane, ethylene dibromide, ethylene bromide, EDB, and glycol  
395 bromide). The trade names are “Bromofume” and “Dowfume.” It is a colorless liquid with a melting point of 9-10°C.  
396  
397 Dibromoethane is reasonably anticipated to be a human carcinogen and has been banned by US EPA for most kinds of  
398 uses since 1984.  
399  
400 The following potential danger is directly quoted from “Agency for toxic substances and disease registry” (ATSDR-  
401 dibromoethan)

402  
403 “SUMMARY:

404  
405 Exposure to 1,2-dibromoethane can result from drinking groundwater or breathing air that is  
406 contaminated. This is most likely to occur in the workplace or from living near a hazardous waste  
407 site. 1,2-dibromoethane can affect the brain, damage skin, damage sperm in males, and even cause  
408 death if exposure is very high. This chemical has been found in at least 27 of 1,416 National  
409 Priorities List sites identified by the Environmental Protection Agency.”

410  
411 The following environmental consequence is directly quoted from “Agency for toxic substances and disease registry”  
412 (ATSDR-dibromoethan)

413  
**“What happens to 1,2-dibromoethane when it enters the environment?”**

It moves into the environment from manufacturing use and leaks at waste sites.

When released, it quickly moves to air and will evaporate from surface water and soil to the air.

It dissolves in water and will move through soil into the groundwater.

Small amounts remain attached to soil particles.

It breaks down slowly in air (over 4-5 months), more quickly in surface water (2 months), and hardly at all in groundwater.

It is not expected to build up in plants or animals.”

414  
415



416 The following regulations are directly quoted from the “11<sup>th</sup> Report on Carcinogens” by US Department of Health and  
417 Human Services (11<sup>th</sup> ROC)

418

419

**“Regulations**

420

421

**DOT**

422

1,2-Dibromoethane is considered a hazardous material and special requirements have been set  
423 for marking, labeling, and transporting this material.

424

**EPA**

425

Clean Air Act

426

NESHAP: Listed as a Hazardous Air Pollutant (HAP).

427

NSPS: Manufacture of substance is subject to certain provisions for the control of Volatile  
428 Organic Compound (VOC) emissions.

429

Urban Air Toxics Strategy: Identified as one of 33 HAPs that present the greatest threat to  
430 public health in urban areas.

431

Comprehensive Environmental Response, Compensation, and Liability Act

432

Reportable Quantity (RQ) = 1 lb.

433

Emergency Planning and Community Right-To-Know Act

434

Toxics Release Inventory: Listed substance subject to reporting requirements.

435

Federal Insecticide, Fungicide, and Rodenticide Act

436

Most registrations have been cancelled.

437

Resource Conservation and Recovery Act

438

Listed Hazardous Waste: Waste codes in which listing is based wholly or partly on substance -  
439 U067, K117, K118, K136.

440

Listed as a Hazardous Constituent of Waste.

441

Safe Drinking Water Act

442

Maximum Contaminant Level (MCL) = 0.00005 mg/L.

443

**FDA**

444

Action levels for 1,2-dibromoethane in food and in animal feed range from 0.01-150 ppb.

445

Maximum permissible level in bottled water = 0.00005 mg/L

446

**OSHA**

447

Acceptable Peak Exposure = 50 ppm (maximum duration = 5-minutes).

448

Ceiling Concentration = 30 ppm.

449

Permissible Exposure Limit (PEL) = 20 ppm.

450

451

**Guidelines**

452

453

**NIOSH**

454

Ceiling Recommended Exposure Limit = 0.13 ppm (15 minute exposure).

455

Immediately Dangerous to Life and Health (IDLH) = 100 ppm.

456

Recommended Exposure Limit (time-weighted-average workday) = 0.045 ppm.

457

Listed as a potential occupational carcinogen.”

458

459 The following use of dibromoethane is directly quoted from the “11<sup>th</sup> Report on Carcinogens” by US DHHS (11<sup>th</sup>  
460 ROC)

461

462

**“Use**

463

464

Historically, the primary use of 1,2-dibromoethane was as a lead scavenger in antiknock mixtures  
465 added to gasolines. Lead scavenging agents transform the combustion products of lead alkyls to  
466 forms that are more likely to be vaporized from engine surfaces. In 1978, 90% of the 1,2-  
467 dibromoethane produced was used for this purpose. Annual consumption of 1,2-dibromoethane in  
468 the United States has decreased due to EPA regulations banning the use of lead in gasolines  
469 (IARC 1977, ATSDR 1992).

470

471

Another major use of 1,2-dibromoethane in the past was as a pesticide and ingredient of soil and  
472 grain fumigant formulations. It was used for post-harvest application to a variety of vegetable,  
473 fruit, and grain crops. It was also used to kill fruit flies on citrus fruits and in the soil to protect

474 grasses in environments such as golf courses. By 1984, EPA regulations had eliminated most of  
475 the use of 1,2-dibromoethane as a pesticide in the United States (ATSDR 1992).

476  
477 Currently, 1,2-dibromoethane is used as a chemical intermediate in synthesis and as a  
478 nonflammable solvent for resins, gums, and waxes. The major chemical made from 1,2-  
479 dibromoethane is vinyl bromide, which is used as a flame retardant in modacrylic fibers. It also  
480 has been used as an intermediate in the preparation of dyes and pharmaceuticals (ATSDR 1992).”

481  
482 In the petition, no information was given whether dibromoethane, one of the two major chemicals for manufacturing  
483 (S,S)EDDS, would be completely converted to the end-product of (S,S)EDDS. If the conversion is not 100%, no  
484 information was given whether the un-reacted dibromoethane would be mixed with the end-product of (S,S)EDDS or  
485 mixed with by-products.

486  
487 **Evaluation Question #5: Is the petitioned substance harmful to the environment? (From 7 U.S.C. § 6517 (c) (1)**  
488 **(A) (i) and 7 U.S.C. § 6517 (c) (2) (A) (i).)**

489 US EPA exempted EDDS from the requirement of a tolerance when used as an inert ingredient sequestrant or  
490 chelating agent in pesticide formulations applied to growing crops (US EPA 40 CFR Part 180.920-Document 0001).  
491 In this document, US EPA did not specify conditions for use where concentrations are limited.

492  
493 US FDA approved the use of (S,S')-EDDS in food-contacting paper or paperboard (US FDA FCN 000799).

494  
495 As given in “Appendix B: Biodegradation of EDDS,” the use of and research on EDDS are recent events relatively, in  
496 the range of 5-10 years. Information about potential harmfulness of EDDS to the environment is still limited, relative  
497 to the information about EDTA’s effects to the environment (Nortemann, 1999; Vandevivere et al., 2001; Bucheli-  
498 Witschel and Egli, 2001; and Nowack, 2002). EDTA, as a chelator, has been used for 40-50 years. In a review article  
499 by Bucheli-Witschel and Egli (2001), 256 papers were cited to provide the base for the discussion of “environmental  
500 fate and microbial degradation of aminopolycarboxylic acids.” NTA and EDTA were the major APCAs discussed in  
501 the paper.

502  
503 Quantity is one index relevant to a potential harmfulness of a substance to the environment. For example, EDTA was  
504 found in natural waters close to places where substantial use and discharge of EDTA occurred (Sillanpaa & Oikari,  
505 1996; Bucheli-Witschel and Egli, 2001). EDDS is used in domestic products such as detergents, but the usage of  
506 EDDS, in quantity and in scale, is far less than that of EDTA. In 1981, the estimated world-wide usage of EDTA was  
507  $56 \times 10^3$  metric tons, while the usage of other APCAs was  $5 \times 10^3$  metric tons (Bucheli-Witschel and Egli, 2001).  
508 Joanna et al. (1999) carried out environmental risk assessment on the use of EDDS in detergent applications. Based  
509 on the assessment, a “no immediate concern” at the anticipated usage level was proposed. Additional and other major  
510 researches about the effect of EDDS on environment are still very limited.

511  
512 In Bucheli-Witschel and Egli (2001), four types of potential environmental risks caused by APCAs were listed: “(1)  
513 adverse effects on the operation of wastewater treatment plants, (2) toxic effects of APCAs on aquatic and mammalian  
514 organisms, (3) the contribution of nitrogen from APCAs to eutrophication, and (4) the potential to mobilise metals.”  
515 However, no words were mentioned about EDDS.

516  
517  
518 **Evaluation Question #6: Is there potential for the petitioned substance to cause detrimental chemical**  
519 **interaction with other substances used in organic crop or livestock production? (From 7 U.S.C. § 6518 (m) (1).)**

520  
521 The petitioned substance EDDS works as a strong chelating agent, similar to another extensively used chelating agent  
522 EDTA. EDTA is allowed in NOP as “inert ingredients.” EDDS is expected to cause similar chemical interactions  
523 with other substances used in organic crop or livestock production. However, direct evidence to support this  
524 expectation is still very limited.

525  
526 Based on the germination and seedling growth of the water cress *Rorippa* sp., Temara et al. (2006) indicated that the  
527 germination was not significantly affected by EDDS. On the other hand, in the phytoextraction experiments, the  
528 application of EDDS in soil released adsorbed heavy metals to water-soluble. The excess heavy metals inhibited the  
529 “normal” growth of plants (e.g. Epelde et al., 2008; Duquene et al., 2009; Wu et al., 2007).

530

531 As given in the answer to question #5, EDDS was approved to be used in pesticides and in food-contacting paper or  
532 paperboard. US EPA concluded that (S,S)-EDDS is a low-toxicity materials (see above “Toxicity” in the  
533 “Properties”).

534  
535 The following is directly quoted from the petition:

536  
537 a) Chemical Interactions with other substances

538  
539 EDDS is a chelating agent therefore can form multiple chemical bonds with certain single metal ions.  
540 The result is a complex but soluble molecule. Once EDDS reacts with a given metal ion, that ion is  
541 inactivated so that it cannot react normally with other elements or ions to produce precipitates or scale.

542  
543 EDDS (and other chelating agents) play a major role in soil remediation projects because its action  
544 solubilizes heavy metals from polluted soils. Other chelating agents (such as  
545 Ethylenediaminetetraacetic acid) were used in such remediation projects but EDDS is replacing other  
546 chelating agents due to its biodegradable nature.

547  
548 **Evaluation Question #7: Are there adverse biological or chemical interactions in the agro-ecosystem by using**  
549 **the petitioned substance? (From 7 U.S.C. § 6518 (m) (5).)**

550  
551 As given in “Appendix B: Biodegradation of EDDS,” the use of and research on EDDS are relatively recent events.  
552 The data relevant to this question are limited. Jaworska et al. (1999) assessed the environmental risk of EDDS used in  
553 detergent application. By using mathematical models and making numerous assumptions of relevant parameters, a  
554 “no immediate concern” conclusion was generated.

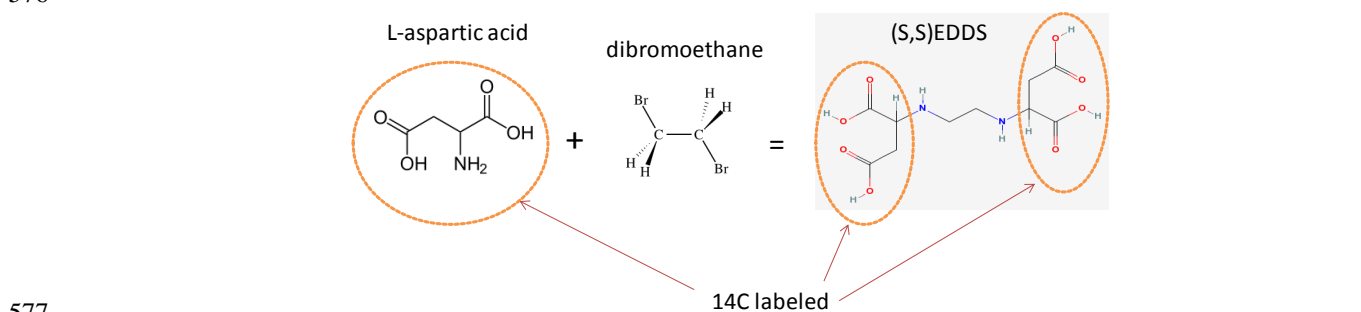
555  
556 **Evaluation Question #8: Are there detrimental physiological effects on soil, organisms, crops, or**  
557 **livestock by using the petitioned substance? (From 7 U.S.C. § 6518 (m) (5).)**

558  
559 The petitioned substance EDDS works as a strong chelating agent, similar to another extensively used chelating agent  
560 EDTA. EDTA is allowed in NOP as “inert ingredients.” EDDS is expected to cause similar physiological effects on  
561 soil, organisms, crops, or livestock. However, direct evidence to support this expectation is still very limited, since  
562 the use of and research on EDDs are still relatively recent events.

563  
564 **Evaluation Question #9: Is there a toxic or other adverse action of the petitioned substance or its breakdown**  
565 **products? (From 7 U.S.C. § 6518 (m) (2).)**

566  
567 As given above in “Toxicity” of “Properties,” the petitioned substance EDDS itself is considered to be of low toxicity  
568 by US EPA.

569  
570 The petitioned EDDS is made from 1,2-dibromoethane and L-aspartic acid. In Schowanek et al. (1997) experiment,  
571 EDDS was synthesized from 1,2-dibromoethane and L-aspartic acid. Exactly, <sup>14</sup>C labeled L-aspartic acid was used  
572 and the material EDDS was labeled on the succinate part. This <sup>14</sup>C labeled EDDS was added to a simulated sewage  
573 system, and the <sup>14</sup>C activity in evolved CO<sub>2</sub> gas was measured. By comparing the <sup>14</sup>C activity in CO<sub>2</sub> gas with the <sup>14</sup>C  
574 activity originally added to the simulated sewage system, Schowanek et al. (1997) concluded that 96% of EDDS  
575 added to the sewage system degraded to inorganic carbon in about two months.



579 Although the measured <sup>14</sup>C activity in CO<sub>2</sub> gas (i.e. inorganic carbon) was about 96% of the <sup>14</sup>C activity which was  
580 added originally to the simulated sewage system as EDDS (i.e. organic carbon), that does not necessarily mean that all

581 of EDDS had decomposed to inorganic carbon already. As shown in the above figure, EDDS was actually labeled  
582 with <sup>14</sup>C partially. The labeled part of EDDS did decompose to CO<sub>2</sub> gas, but that did not necessarily assure that the  
583 unlabeled part also decomposed to CO<sub>2</sub> gas, since that part was not directly measured. In other words, EDDS as a  
584 whole compound did decompose in about two months, but the breakdown products might not be totally inorganic.  
585 The breakdown products of the unlabeled part of EDDS may still need to be clarified.

586  
587 The potentially unbroken part is originated from 1,2-dibromoethane, a substance banned by US EPA in 1984 for most  
588 kinds of uses (See above in Question 4).

589  
590 **Evaluation Question #10: Is there undesirable persistence or concentration of the petitioned substance or its**  
591 **breakdown products in the environment? (From 7 U.S.C. § 6518 (m) (2).)**

592  
593 The petitioned substance EDDS was claimed to be biodegraded quickly. As given in “Biodegradability” of  
594 “Properties,” and as discussed in Appendix B: Biodegradation of EDDS, EDDS might degrade in natural  
595 environments, but the evidence to support the claim is still weak,

596  
597 As given in Question #9, the breakdown products of (S,S)EDDS might not be totally inorganic but no further  
598 information is available.

599  
600 **Evaluation Question #11: Is there any harmful effect on human health by using the petitioned substance?**  
601 **(From 7 U.S.C. § 6517 (c) (1) (A) (i), 7 U.S.C. § 6517 (c) (2) (A) (i) and, 7 U.S.C. § 6518 (m) (4).)**

602  
603 As given in “Toxicity” of “Properties,” (S,S)-EDDS is considered to be of low toxicity by US EPA. US FDA  
604 approved the use of (S,S)-EDDS in food-contacting paper or paperboards.

605  
606 **Evaluation Question #12: Is there a wholly natural product that could be substituted for the petitioned**  
607 **substance? (From 7 U.S.C. § 6517 (c) (1) (A) (ii).)**

608  
609 EDDS is petitioned to be used as a chelator in organic pesticides. Chelators are a class of chemical compounds which  
610 combine with metal ions to form water-soluble complexes. Some chelators such as EDTA are synthetically  
611 manufactured, and some chelators such as rhizobactin are found as trace or minor components in natural products  
612 (Bucheli-Witschel and Egli, 2001). No chelators are found as wholly natural products or as major components in  
613 natural products.

614  
615 EDDS, a simple compound, is found to be produced by microorganisms in soil (Nishikiori et al., 1984; Schowanek et  
616 al., 1997; Zwicker et al., 1997; Takahashi et al., 1999). EDDS can be synthetically manufactured but EDDS has not  
617 been found as a wholly natural product.

618  
619 **Evaluation Question #13: Are there other already allowed substances that could be substituted for the**  
620 **petitioned substance? (From 7 U.S.C. § 6517 (m) (6).)**

621  
622 EDTA is already listed in NOP as “inert ingredients” in pesticides. Generally, EDTA, used as a chelating reagent, is  
623 sufficiently available, provides better complex capability than the petitioned substance EDDS, and has been used in  
624 domestic, industrial and agricultural applications for 40-50 years.

625  
626 EDTA, a synthetic substance, was found in natural waters at 10-60 µg/L (Barber et al., 1995; Sillanpaa & Oikari,  
627 1996). The existence of a synthetic substance in natural environment has a variety of effects, as quoted below, and  
628 prompted searches for alternative chelators to replace EDTA (Zwicker et al., 1997; Takahashi et al., 1999).

629  
630 Chelating agents have the potential to perturb the natural speciation of metals and to influence  
631 metal bioavailability, and their presence may lead to the remobilization of metals from sediments  
632 and aquifers, consequently posing a risk to groundwater and drinking water (Nowack, 2002).

633  
634 EDTA is used in domestic, industrial and agricultural applications for a long time and has received substantial  
635 researches (Nortemann, 1999; Nowack, 2002). For example, the research on its biodegradation at least started in 1967  
636 (Bunch and Ettinger, 1967).

637  
638 The petitioner claimed that “EDDS occurs naturally in the environment and has a better environmental fate and  
639 degradation profile than the chelating agents currently allowed in organic pesticides;” and “EDDS degrades rapidly

640 and is completely mineralized.” However, the use of and research on EDDS are relatively recent events, as discussed  
641 in “Appendix B: Biodegradation of EDDS.” The biodegradation of EDDS in soil has been investigated but an  
642 unambiguous conclusion might still be too soon to make (Appendix B). EDDS might be biodegraded faster than  
643 EDTA, but the environmental consequence of EDDS might be less understood than that of EDTA currently.

644  
645 **Evaluation Question #14: Are there alternative practices that would make the use of the petitioned substance**  
646 **unnecessary? (From 7 U.S.C. § 6517 (m) (6).)**

647  
648 Chelation is a process in which chelators (chelating reagents) combine with metal ions to form complex and to keep  
649 metal ions water-soluble without changing other significant properties such as solution pH and metal ion  
650 concentrations. In this sense of keeping other significant properties such as pH and metal ion concentrations  
651 unchanged, the use of chelator can not be replaced by other alternative practices.

652  
653 The substance EDDS is petitioned to be used as “inert ingredients” in organic pesticides. As a chelator, the petitioned  
654 substance EDDS can be replaced with other kinds of chelators, if available. In fact, EDTA, a chelator, is currently list  
655 in NOP as “inert ingredients” in pesticides.

656  
657 The use of EDTA has some environmental concerns, as given in “Evaluation Question #13.”

658  
659

## 660 Appendix A: Chelation and Related Issues

661  
662 The following is quoted from "Glossary of Terms Used in Physical Organic Chemistry (IUPAC Recommendations  
663 1994)."

664 **chelation**

665 The formation or presence of [bonds](#) (or other attractive interactions) between two or more separate  
666 [binding sites](#) within the same ligand and a single central atom. A [molecular entity](#) in which there is  
667 chelation (and the corresponding [chemical species](#)) is called a "chelate". The terms bidentate (or  
668 didentate), tridentate, tetradentate... multidentate are used to indicate the number of potential  
669 binding sites of the ligand, at least two of which must be used by the ligand in forming a "chelate".  
670 For example, the bidentate ethylenediamine forms a chelate with CuI in which both nitrogen  
671 atoms of ethylenediamine are bonded to copper. (The use of the term is often restricted to metallic  
672 central atoms.)

673 The phrase "separate binding sites" is intended to exclude cases such as [PtCl<sub>3</sub>(CH<sub>2</sub>=CH<sub>2</sub>)],  
674 ferrocene, and (benzene)tricarbonylchromium in which ethene, the cyclopentadienyl group, and  
675 benzene, respectively, are considered to present single binding sites to the respective metal atom,  
676 and which are not normally thought of as chelates (see [hapto](#)). See also [cryptand](#).

677 Analogous to wrapping medicine pills with protective coats so that medicine pills are less reactive and exert longer  
678 effects, chelation could be intuitively understood as a process in which metal ions are wrapped with chelating agents  
679 so that metal ions are less reactive. The reactivity includes precipitation, adsorption, reaction with other components,  
680 and assimilation by organisms, etc.

681  
682 **Complex stability and reversible process:** Metal ion "M" combines with ligand "L" to form complex "ML."  
683 Chelation is a reversible process:  $M + L \rightleftharpoons ML$ . Metal "M" and ligand "L" form complex "ML" at one condition, but  
684 complex "ML" decomposes to metal "M" and ligand "L" at another condition. The stability constant  $k$  of "ML" is  
685 expressed as:  $k = [ML] / ([M] \times [L])$ , where square brackets denote concentrations. The higher the  $k$  value, the stable  
686 the complex "ML" is.

687  
688 **Ligand stability:** A ligand decomposes itself to its components and loses the chelating capability. The ligand  
689 decomposing process is not reversible.

690  
691 **Competition:** Assume there are two metal ions (calcium, Ca<sup>2+</sup> and lead, Pb<sup>2+</sup>) and one ligand EDTA. The stability  
692 constant  $k$  of Pb-EDTA complex is much greater than that of Ca-EDTA complex. In this case, EDTA forms Pb-  
693 EDTA preferentially. If lead is added to a system which contains Ca-EDTA originally, Ca-EDTA will decomposes  
694 and Pb-EDTA is formed.

695  
696 Initially, Ca-EDTA complex is formed:  $Ca + EDTA \rightarrow Ca-EDTA$ .

697 After Pb is added, Ca-EDTA decomposes and Pb-EDTA is formed:  $Pb + Ca-EDTA \rightarrow Ca + Pb-EDTA$ .

698  
699 **Kinetics:** Competition based on the consideration of complex stability is just one side of a story. A real complex  
700 process is controlled by the complex kinetics.

701  
702 With these concepts of "complex stability," "ligand stability," "reversible process," "competition," and "kinetics,"  
703 several processes are described here.

704  
705 **Basic application:** Example 1: Hard water contains high concentrations of calcium and magnesium. These ions form  
706 "soap scum" with detergents. Chelating agents added to detergent forms complex with these metal ions. Calcium and  
707 magnesium stay dissolved and soap scum is not formed. Example 2: Chelating reagents added to pesticides modify  
708 the effects of heavy metals in pesticides.

709  
710 **Soil washing:** Soil is soaked with water containing chelating reagents. Heavy metals such as copper, lead, and zinc,  
711 initially precipitated or adsorbed to soil, are converted to complex which is water-soluble and rinsed away from soil.

712  
713 **Phytoextraction:** Phytoextraction is an enhanced accumulation of metal ions in harvestable plant. It is an alternative  
714 remediation technology for soils polluted with heavy metals. Chelating agents enhance phytoextraction by making  
715 precipitated/adsorbed metals in soil water-soluble and available for plants.

716

## 717 Appendix B: Biodegradation of EDDS

718

719 Chelation makes metal ions less reactive and water-soluble. On one hand, chelation is used, for example, in “soil  
720 washing” in which toxic metals such as copper and lead precipitated in and/or adsorbed to soil are released back to  
721 water-soluble and rinsed away from soil. On the other hand, released toxic metals are transported to undesired places  
722 such as to groundwater reservoirs which might be sources of drinking water. An ideal chelator might be strong in  
723 forming complex and quick in decomposing. “Biodegradation” is a process in which a substance is decomposed to  
724 components by microorganisms.

725

726 From several chelators, Procter & Gamble selected EDDS in order to “commercially develop a chelator that  
727 performed equally to currently available materials but with a greatly improved biodegradation potential.” (Schowanek  
728 et al., 1997).

729

730 The list below is not exhaustive but includes most major literature about EDDS in the areas of biodegradability, soil  
731 washing and phytoextraction.

732

Year	Author(s)	Subject	Country	Relation to manufacture
1968	Neal & Rose	EDDS isomers		
1996	Schowanek et al.	Bioavailability	Belgium	P&G
1997	Schowanek et al.	Degradability	Belgium	P&G
1997	Takahashi et al.	Degradability	Japan	
1997	Zwicker et al.	Production by bacteria	Germany	
1999	Jaworska et al.	Environment assessment	Belgium	P&G
1999	Nortemann	Review: EDTA	Germany	
1999	Takahashi et al.	Production by bacteria	Japan	
2001	Bucheli-Witschel & Egli	Review: APCAs	Switzerland	
2001	Jones & Williams	Stability constants, model	U.K.	
2001	Metsarinne et al.	Photodegradation	Finland	
2001	Vandevivere et al.	Degradability	Belgium	P&G
2002	Nowack	Review: APCAs	Switzerland	
2002	Orama et al.	Stability constants, experimental	Finland	
2003	Grcman et al.	Phytoextraction	Slovenia	EDDS donated by P&G
2003	Kos & Lestan	Phytoextraction	Slovenia	EDDS donated by Octel
2004	Tandy et al.	Soil washing	Switzerland	EDDS donated by Octel
2005	Hauser et al.	Degradability and soil washing	Switzerland	EDDS donated by P&G
2005	Luo et al.	Phytoextraction	Hong Kong	
2005	Meers et al.	Degradability and phytoextraction	Belgium	EDDS donated by P&G
2005	Tandy et al.	Analytical	Switzerland	EDDS donated by P&G
2006	Finzgar et al.	Soil washing	Slovenia	EDDS donated by Octel
2006	Luo et al.	Degradability and phytoextraction	Hong Kong	
2006	Tandy et al.	Degradability	Switzerland	EDDS donated by P&G
2006	Temara et al.	Water plant germination	Belgium	P&G
2007	Polettini et al.	Soil washing	Italy	
2007	Xu & Thomson	chelating agent	Canada	
2007	Wu et al.	Phytoextraction	China	
2008	Epelde et al.	Degradability and phytoextraction	Spain	
2008	Meers et al.	Degradability	Belgium	
2009	Duquene et al.	Phytoextraction	Belgium	
2009	Sun et al.	Soil washing	China	
2009	Yip et al.	Soil washing	Hong Kong	
2010	Ko et al.	Extraction of Cu, Cr and As from wood	Taiwan	EDDS donated by Octel

733

734

735 Most papers in the list did not investigate the biodegradability of EDDS, but almost all of those papers used  
736 “biodegradable” or “environmentally friendly” as an adjective to describe EDDS, and “non-biodegradable” to  
737 describe EDTA. Those papers eventually cited a limited number of papers as the basis for those claims. The papers  
738 relevant to biodegradability of EDDS were mostly contributed by the manufactures or by the people around the world  
739 who received EDDS from the manufactures.

740

741 The biodegradation of EDDS (i.e. EDDS acid) in a simulated sewage system was contributed by Takahashi et al.  
742 (1997) and Schowanek et al. (1997). A general conclusion was that EDDS acid was biodegradable. The  
743 biodegradation of EDDS complex in a simulated sewage system was contributed by Vandevivere et al. (2001). A  
744 general conclusion was that the biodegradation of EDDS complex depended on metal ions. For example, the Cu-  
745 EDDS complex did not show significant biodegradation within an experiment period during which the EDDS  
746 complex of several other metal ions were degraded already (Vandevivere et al., 2001). Overall, the knowledge about  
747 the fate of EDDS in the environment was still limited in 2001 (Bucheli-Witschel and Egli, 2001; Vandevivere et al.,  
748 2001; and Nowack, 2002). The review article about APCAs by Bucheli-Witschel and Egli (2001) is 38 pages long,  
749 but the discussion about biodegradation of EDDS is merely ¼ page long and the discussion is totally based on the  
750 papers by Takahashi et al. (1997) and by Schowanek et al. (1997).

751  
752 Most research about the biodegradability of EDDS in soil is five years or less (e.g. Tandy et al., 2006; and Meers et  
753 al., 2008). Contrasted to the research on biodegradation of EDDS in a simulated sewage system (Takahashi et al.,  
754 1997; Schowanek et al., 1997; and Vandevivere et al., 2001), the research on biodegradation of EDDS in a simulated  
755 soil system (e.g. Tandy et al., 2006; and Meers et al., 2008) has the following differences.

- 757 • The soil system was an open and dynamic system with repeated evaporation-watering cycles, while the sewage  
758 system was a closed and steady-state system in which all of the materials were added at the beginning of the  
759 experiment and well mixed.
- 760 • The soil system was a heterogeneous system composing of soil (clay minerals, organic matters, sand, etc), water,  
761 and air. With repeated evaporation-watering cycles, the concentrations of metal ions in one physical location  
762 could be different from the concentrations of metal ions in other locations. The sewage system was a pseudo-  
763 homogeneous system.
- 764 • In the soil system, a probe-type sampler was used to collect samples for the analysis of EDDS or metal ions. The  
765 probe-type sample collected samples in the very vicinity of the probe.
- 766 • In the soil system, EDDS was applied to the top of soil, while the probe-type sampler might be placed in the  
767 middle or bottom of the soil.
- 768 • In the soil system, one gram of EDDS was added to about 1000 g of soil. Soil contains clay minerals and organic  
769 matter. EDDS could be trapped and adsorbed to soil. In the sewage system, one gram of EDDS was added to 1-3  
770 g of sludge solid. The relative percentage of EDDS trapped/adsorbed to sludge solid should be very small.
- 771 • In the soil system where the ratio of EDDS to soil was about 1 g EDDS to 1000 g soil, the amount of metal ions  
772 could be higher than the amount of added EDDs. In other words, EDDS might exist as EDDS complex rather  
773 than EDDS acid. In the sewage system, the ratio of EDDS to solid was about 1:1. EDDS could exist mainly as  
774 EDDS acid. The stabilities of EDDS acid and EDDS complex could be significantly different.
- 775 • The adsorption of chelators and metal complex to soil is greatly affected by the ratios of chelator/soil and  
776 complex/soil. In the soil system and in the sewage system, these ratios were substantially different. In other  
777 words, the adsorption of chelators and complex to soil might not be insignificant at all.

778  
779 Considering these, the research on the biodegradability of EDDS in a soil system could be more complicated than that  
780 in a sewage system. The conclusions proposed in the papers about EDDS degradation in soils might be well  
781 questioned with reasonable doubts. Two examples are provided below: paper 1 by Tandy et al. (2006) and paper 2 by  
782 Meers et al. (2008).

783  
784 Paper 1

785  
786 In “Biodegradation and speciation of residual SS-ethylenediaminedisuccinic acid (EDDS) in soil solution left after  
787 soil washing” (Tandy et al., 2006, hereafter termed as “the paper” within this section), it was stated that “This paper  
788 aims to investigate the degradation and speciation of EDDS-complexes (SS-ethylenediaminedisuccinic acid) in soil  
789 following soil washing. The changes in soil solution metal and EDDS concentrations were investigated for three  
790 polluted soils. EDDS was degraded after a lag phase of 7-11 days with a half-life of 4.18-5.60 days. .... Our results  
791 show that even in polluted soils EDDS is degraded from a level of several hundred micromoles to below 1 µM within  
792 50 days.” After a critical reading of the paper, it was found that the conclusions about the biodegradation of EDDS  
793 proposed in the paper might not be substantiated by the experimental results presented in the paper.

794  
795 The paper’s experimental set up is quoted below with some numerical marks such as “(§1-►)” inserted for later  
796 discussion convenience.

797  
798 2.3. Experimental setup



799  
800 (§1-►) Soil (12 kg DW of each) was placed in a plastic barrel with 120 l tap water and stirred with  
801 an electrical stirrer (200 rotations per minute). 20 mmol/kg Na<sub>3</sub>EDDS was added (0.24 moles) and  
802 the solution adjusted to pH 7 if necessary with 1 M HNO<sub>3</sub>. This equated to a EDDS:metal ratio of  
803 1:1 for soil 1, 4:1 for soil 2 and 2:1 for soil 3. The barrels were covered and the soils were washed in  
804 this manner for 24 h. (§2-►) The suspension was then allowed to settle for 24 h before the  
805 supernatant was removed by suction and (§3-►) the soil was rinsed for 1 h with 120 l tap water.  
806 (§4-►) After 24 h settling the supernatant was again removed. (§5-►) The soil slurry was then  
807 poured into 3 l black plant pots (4 replicates) with a disc of fine mesh (60 mm) in the bottom to  
808 prevent the soil leaking out and 2 Rhizon Flex soil moisture samplers (SMS) (Rhizosphere Research  
809 Products, Wageningen, Netherlands) were installed at a 45° angle. (§6-►) The pots were allowed to  
810 drain over night and the clear solution present on top of the soil was removed. (§7-►) The pots  
811 were then transferred to a climate chamber with a 16 h (21°C)/8 h (16°C) day/night cycle to  
812 simulate field conditions. The first soil solution was then extracted (time 0) see Section 2.4. This  
813 corresponds to day 4 after addition of EDDS. After two days no more drainage occurred and this  
814 was then taken to be 100% water holding capacity (WHC). (§8-►) Soil solution was extracted  
815 every 7 days. One day prior to this the pots were made up to 100% WHC with ultra pure water and  
816 24 h later the solution extracted. The pots were then allowed to dry until the next week.

817  
818 The measured concentrations of EDDS in the extracted soil solution samples were presented in Fig. 1 of the paper.  
819 By assigning the EDDS concentration at time zero as 100%, the EDDS concentrations in subsequent samples were 90-  
820 100% at day 7 (day 7 counted from time zero), 40-70% at day 14, 20-60% at day 21, about 15% at day 28, less 10% at  
821 day 35, and close to 0% at day 56. Based on the results, the paper concluded that EDDS was decomposed in soil  
822 solution.

823  
824 Based on the experimental setup, EDDS was added to soil (§1). After mixing, EDDS existed as EDDS acid and/or as  
825 EDDS complex (noted as M-EDDS), could be kept in soil by different mechanisms (such as adsorbed, attached, or  
826 trapped), and was distributed in water and in soil. EDDS in water was discarded (§2). The EDDS-treated soil was  
827 washed with tap water (§3), and EDDS in this washing water was discarded (§4). EDDS in water phase was further  
828 discarded (§6). Some water remained in soil as “soil solution.”

829  
830 Initially, 70 g of EDDS (as EDDS acid) was added to 12,000 g of soil (§1) (5.8 g of EDDS to 1000 g of soil). After  
831 the above preparation steps, it is not know how much EDDS remained in “soil solution,” how much EDDS was  
832 adsorbed/attached/trapped, and how much EDDS was discarded. Schowanek et al. (1997) indicated that the  
833 adsorption of EDDS to sludge/soil was insignificant. However, in Schowanek et al. (1997), the ratio of EDDS to  
834 sludge solid was about 1:1 to 1:3 and sludge solid contained less clay minerals than regular soil. In a review paper,  
835 Nowack (2002) indicated that the adsorption of chelators and complex to soil was significant: “Chelating agents have  
836 been developed to solubilize metals and keep them in solution. Therefore, it might be reasonable to assume that  
837 chelating agents decrease heavy metal adsorption by forming dissolved complexes. This, however, is only true for the  
838 very high concentrations of chelating agents used in technical applications. At low concentrations, chelating agents are  
839 able not only to decrease but also can significantly increase metal adsorption onto mineral surfaces.”

840  
841 Except the first soil solution sample which was collected at time zero after the EDDS-treated soil was made “ready” in  
842 the plant pots (§7), the subsequent soil solution samples were collected after the EDDS-treated soil was repeatedly  
843 subject to day/night cycles (§7) and subject to additional input of ultra clean water (i.e. dry/wet cycles) (§8).

844  
845 “Soil” and “soil solution” were not two clearly separated physical entities, but interchanged and interacted closely.  
846 EDDS was kept in soil either strongly or weakly. It could be well expected that initial soil solution samples contain  
847 more EDDS than the subsequent samples, since most weakly kept EDDS would be released from soil to soil solution  
848 quickly. In other words, the result of Fig. 1 would still be obtained even if EDDS did not degrade at all.

849  
850 Even if the releasing of EDDS from “soil” to “soil solution” is not considered, it could be well expected that initial  
851 soil solution samples contain more EDDS than the subsequent samples. EDDS in soil solution was a limited source  
852 and would reach to zero content after ultra clean water was repeatedly added to the EDDS-treated soil. Each addition  
853 of clean water would deplete EDDS from the soil (or soil solution). Therefore, the result of Fig. 1 would still be  
854 obtained even if EDDS did not degrade at all.

855  
856 Specific to the experiment setup, there could be at least three scenarios, individually or combined, to explain the  
857 experimental results of Fig. 1: differential release of EDDS from soil to soil solution, limited amount of EDDS in soil

858 and/or soil solution relative to repeat depletion, and degradation of EDDS in soil solution. The paper ascribed the  
859 experimental results solely to the degradation of EDDS in soil solution without mentioning other potential  
860 mechanisms. In analog, one person can take pickles out of a bottle, wash the pickles initially, rinse the pickles  
861 repeatedly with fresh water, and measure the salt in the rinses. Not surprisingly the concentrations of salt will be  
862 higher in initial rinses and lower in subsequent rinses. It is true that the salt concentrations in these rinses decrease  
863 with increasing time, however, no one would conclude that salt is decomposed within this time period.

864

865 Paper 2

866

867 In “Degradability of ethylenediaminedisuccinic acid (EDDS) in metal contaminated soils: Implications for its use soil  
868 remediation” (Meers et al., 2008, hereafter termed as “the paper” within this section), it was stated that “This study  
869 examines heavy metal mobilization in three polluted soils varying in soil composition, with specific attention for  
870 competitive behaviour for complexation between the various metals and major elements, such as Al, Fe, Mn, Ca and  
871 Mg. .... EDDS was fully degraded within a period of 54 d in all soils regardless of initial delay.” After a critical  
872 reading of the paper, it was found that the conclusions about the biodegradation of EDDS proposed in the paper might  
873 not be substantiated by the experimental results presented in the paper.

874

875 The paper’s experimental set up is quoted below with some numerical marks such as “(§1-►)” inserted for later  
876 discussion convenience.

877

## 878 2.2. Soil experiment

879

880 (§1-►) The pot experiment was conducted under outdoor conditions to mimic behaviour of EDDS  
881 under natural conditions. The experiment was performed in open air, with collection and  
882 recirculation of percolate in case of excess rainfall to prevent leaching of the chelate and mobilized  
883 metals from the system. (§2-►) Temperatures ranged between 6-18°C (night) and 16-30°C (day)  
884 over the course of the growing season. To induce biological activity in the soil experiments, pots  
885 containing 3 kg of soil (dry weight) were planted with *Zea mays* at the start of the growing season  
886 (May 2004). After 4 months of incubation, (§3-►) the pots were treated with 7.5 mmol EDDS per  
887 pot added as Na<sub>3</sub>-EDDS (Octel Performance Chemicals, Cheshire, United Kingdom). Application  
888 was divided over three separate doses (dissolved in 3 × 200 mL deionized water), spread over a  
889 period of 1 week. (§4-►) The pots were fitted with Rhizon soil solution samplers (MOM-type;  
890 Eijkelpark Agrisearch, Giesbeek, the Netherlands). (§5-►) Soil solution samples were collected at  
891 regular intervals over a period of 54 d following treatment. (§6-►) ..... (§7-►) Dissolved organic  
892 carbon (DOC) in the soil solution was determined using a TOC-500 analyzer (Shimadzu, Duisburg,  
893 Germany). (§8-►) ..... (§9-►) DOC concentrations present more direct additional information in  
894 regards with chelate degradability.

895

896 The measured concentrations of DOC in soil solution samples were presented in Fig. 2 of the paper. Samples shown  
897 at time zero of Fig. 2 were the first samples collected following treatment (§5). DOC concentrations in the samples  
898 were 50 mg/L at day 0, 300-400 mg/L at days 2, 600-900 mg/L at day 9, 300-400 mg/L at day 30, and 50 mg/L at day  
899 45. Based on the results, the paper concluded that DOC concentration decreased with increasing time after a lag  
900 phase, and EDDS was degraded in soil solution.

901

902 Based on the experimental setup, the application of 7.5 mmol EDDS was divided over three separate doses (dissolved  
903 in 3 × 200 mL deionized water), spread over a period of 1 week (§3). The concentration of 2.5 mmol of EDDS (Na<sub>3</sub>-  
904 EDDS or C<sub>10</sub>H<sub>13</sub>N<sub>2</sub>Na<sub>3</sub>O<sub>8</sub>) in 200 mL of deionized water is 0.0125 mmol/mL or 0.0125 mol/L of EDDS. This  
905 solution contains 1.5 g/L or 1,500 mg/L of DOC.

906

907 Initially, 2.2 g of EDDS (as EDDS acid) was applied to 3000 g of soil (0.7 g of EDDS to 1000 g of soil). Soil solution  
908 samples were collected following treatment (§5). However, the concentrations of DOC in the first samples (at time  
909 zero) were 50 mg/L. After 600 mL of EDDS (1,500 mg/L DOC) was applied to 3 kg of soil in a period of one week  
910 (§2 & §3), the expected DOC concentration should be about 1,500 mg/L, even considering some dilution by water  
911 which was initially in soil. Where did the rest (actually more than 95%) of EDDS go?

912

913 DOC in soil solution then increased from 50 mg/L at time zero to a maximum of 600-900 mg/L at day 9. The paper  
914 indicated that “The initial increases in DOC and metal concentrations observed during the first 200-240 h are due to  
915 the treatment with EDDS, added in three applications spread over the duration of a week.” This statement is difficult  
916 to understand and to accept. After the application of EDDS, why did the DOC concentration kept increasing with

917 increasing time in 9 days? There seemed to be a source of DOC to the soil in these nine days. Then why was the  
918 maximum concentration of DOC only 600-900 mg/L? This accounted for 50% of added DOC. Where was the other  
919 50%? Was the other 50% decomposed (degraded) already in 9 days?  
920

921 After 9 days, the DOC concentrations kept decreasing with increasing time. The paper concluded that EDDS  
922 degraded. EDDS might really degrade. However, would there be other explanation(s)? Can the observed variations  
923 of DOC concentrations from day zero to day nine be ignored since those variations did not support the conclusion of  
924 “degradation of EDDS?” Experimental results should not be selectively used to support a conclusion. “A lag phase”  
925 did not explain the increase in DOC concentration observed from day zero to day nine.  
926

927 From the above discussion, it is very hard to accept that the conclusion about degradation of EDDS in soil was  
928 substantiated by the experimental results.  
929

930 The “Rhizon soil solution samplers” (§4) is a probe type sampler like a pH electrode, and collect soil solution at the  
931 vicinity of probe. The paper did not specify how many samplers were used and where the samplers were placed (§4).  
932 The following is just a speculation which might explain the observed results of Fig. 2.  
933

934 The sampler (one or several) was placed somewhere between the top and the bottom of soil. EDDS was applied to the  
935 top of soil. At time zero, the solution collected in the sampler contained 50 mg/L DOC since the applied EDDS was  
936 still in the top of soil and had not reached to the sampler which was placed away from where EDDS was applied.  
937 With time, the EDDS zone migrated down from the top of soil to the bottom of soil (and migrated sideways). This  
938 migration was possible due to the experiment set up “The experiment was performed in open air, with collection and  
939 recirculation of percolate in case of excess rainfall to prevent leaching of the chelate and mobilized metals from the  
940 system” (§1). When the EDDS zone migrated towards to the sampler, the DOC concentration increased with  
941 increasing time and reached to a maximum. When the EDDS zone migrated away from the sample, the DOC  
942 concentration decreased with increasing time. With migration and rainwater input, EDDS was distributed and/or  
943 retained in different places and the measured maximum concentration was substantially less than the expected  
944 maximum concentration.  
945

946 If the above speculation is reasonable, the concentration variations potentially caused by migration and other  
947 mechanisms should not be used as the evidence to support the biodegradation of EDDS.  
948  
949

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