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A Comparison of Various Chelating Agents in the Diagnosis of Chronic Metal Exposure- An Update

E. Blaurock-Busch[1,*], Yvette M. Busch[2]

[1]Research director at Micro Trace Minerals Laboratory, Hersbruck, Germany
[2]CEO at Micro Trace Minerals Laboratory, Hersbruck, Germany
*Corresponding author: ebb@microtrace.de

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Abstract When selecting a chelating agent for treatment purposes, making an informed choice is critical and the question frequently asked is: which chelating agent is best? We thus evaluated and compared the binding ability of DMPS, DMSA, EDTA, Na-thiosulfate, Alpha-Lipoic Acid. Our aim was to identify which of the chelators would be most useful for the chelation treatment of which metal. While our previous study indicated that for single or multiple metal exposures, the intravenous application of DMPS seemed best suited as a provocation test, this follow-up study demonstrates that other chelating agents may be equally or more suitable. Our data also demonstrates that oral application can be an alternative to the use of intravenous chelation.

Keywords: *DMPS, DMSA, EDTA, Sodium-Thiosulfate, Lipoic Acid, Chronic Metal Intoxication, Chelation, Urine provocation, Metal Intoxication*

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1. Introduction

Since World War I, the role of chelating agents has been well documented. Internationally, poison center list the use and indication of specific chelating substances for cases of acute intoxications.

With the increase in environmental pollution, environmentally-caused ailments are on the rise. Patients afflicted typically suffer from diffuse symptoms associated with multiple metal overexposure and chelation therapy is becoming a recognized treatment option.

Metals circulate in blood before they are excreted, mostly via urine, feces, and sweat. Remaining metals are stored in various organ tissues. Diagnostically, tests utilized to diagnose a chronic metal intoxication involve blood and urine testing. To determine the degree of long-term exposure, environmental physicians are utilizing provocation tests, which involves a urine test taken after the application of a chelating agent. The metal-chelates found in this specimen serve as a treatment guide.

Until recently, the chelating agents mostly used for metal provocation were DMSA, DMPS and EDTA. With the availability of Sodium-Thiosulfate and Alpha-Lipoic Acid environmental physicians broadened their medical spectrum and Poison Centers around the world guide physicians by providing information about the use of chelating agents.

CaNa2EDTA (Calciumdisodium-EDTA) is listed internationally at Poison Centers in the Asian-Pacific Region, in Europe, Canada and the US, primarily for the treatment of lead poisoning. These centers provide 24/7 telephone consultation services.

DMSA (Dimercaptosuccinic acid) also referred to as Succimer has been FDA-approved as an oral antidote for lead poisoning in children in 1991. Poison center also list DMSA as an antidote for mercury, or as a general antidote for heavy metal exposure. [1]

DMPS (2,3-Dimercapto-1-propanesulfonic acid) is registered under the name of Dimaval in Germany. It is recommended for intravenous use in acute mercury poisoning (metallic, vapour, inorganic or organic compounds), if oral application or treatment by means of a gastric tube are not possible. The German Poison Center GIZ Nord list it as an antidote for lead, organic and inorganic mercury, and a number of unspecified heavy metals.[2]

Sodium thiosulfate is part of the World Health Organization's list of essential medicines, recommended as a chelator in the treatment of arsenic, lead, mercury and bismuth poisoning.

Alpha-Lipoic Acid (ALA), a fatty acid and promising chelating agent has been noted to have chelation properties, particularly for mercury and the treatment of neurological ailments. [3,4]

A number of potentially toxic metals are of concern to environmental physicians, we therefore evaluated the chelating ability of the chelating agents mentioned above. The summary of results is shown in the diagrams below.

Sample collection

For this study, urine samples were received from

physicians, practicing chelation therapy. Prior to sampling, patients were informed not to eat fish for 3 days prior to sampling as fish can contain arsenic or mercury. Patients were also instructed not to take supplements or algae products, which can be a potential source of toxic metals.

Protocol instructions, including sampling instructions were provided to physicians. The samples included in our study are from chronically exposed patients, male and female adults. To avoid external contamination, samples were collected into metal-free tubes, provided by the laboratory. Provocation urine samples were taken after the application of either oral or intravenously applied chelating agents. The urine collection time reflected the specific chelator's half-life.

Sample Analysis

At the laboratory, samples were acid-digested with certified metal-free nitric acid involving closed vessel microwave digestion. For sample dilution ultrapure water was used.

Urine metal analysis was performed using the Agilent ICP-MS spectrophotometer with Octopole Reaction System (ORS), a relatively new and improved type of mass spectrometers. With five times the sensitivity of its predecessor and increased matrix tolerance, the ORS system replaces both GFAA and ICP-OES instruments in addition to older generation ICP-MS systems.

Certified urine standards and in-house standards were used for quality control and for validation processes. To avoid the potentially great margin of error that can result from patients' fluid intake, or from in-correctly provided sample volume, results are reported in mcg/g creatinine for all elements. Patient age and sex was used to determine urine creatinine levels.

Statistics

All urine results reflect a 95 Percentile. We compared the statistically derived urine metal concentration of provocation tests involving oral and intravenously applied chelating agents.

2. Specifics about Chelating Agents

DMPS (2,3-Dimercapto-1-propanesulfonic acid)

DMPS belongs to the thiol group, binding metals to sulfhydryl groups. DMPS is registered in Germany since 1997 under the name Dimaval (Heyl, Berlin) and is available as a prescription item in various countries. [5] It is available in capsule form for oral treatment (1 capsule DMPS-Heyl contains 100mg), and in 5ml ampules, containing 250mg for intravenous application. DMPS ampules are also available as Unithiol.

DMPS is is a water-soluble analog of BAL (British Anti Lewisite), but unlike BAL there is no potential risk of redistributing metals to the central nervous system (CNS).[6] Most importantly, DMPS causes fewer side effects than BAL. Successful clinical trials have been conducted in Germany and other countries, such as Malaysia. [7]

The bioavailability of oral DMPS is approximately 40%, which means more than half of the orally provided substance is not absorbed, [8] remaining in the digestive tract until excreted. [9]

The half-life of DMPS is about 20 minutes, and the distribution is not dose-dependant. [10] DMPS is not able to cross the blood brain barrier (BBB) [11]. Excretion of DMPS and its metabolites is relatively fast following intravenous application. The highest concentration of DMPS-metal chelates is seen in urine within two hours. [12]

After oral application, the highest concentration of DMPS is seen after 3 hours. About 80% is excreted in the urine within 5-6hrs. [13] After the application of oral DMPS, a 3hr urine collection time is recommended.

DMSA (Dimercaptosuccinic acid), also called Succimer

Like DMPS, Succimer is a water-soluble analog of dimercaprol (BAL). It has a wide therapeutic index and causes few side effects. [14] Like DMPS, this chelating agent belongs to the thiol compounds, binding metals with sulfhydryl groups. In the USA, DMSA was registered under the trade name of Chemet. It has a history of being used for the detoxification of lead and mercury, particularly in children and sensitive adults. The recommended treatment dose is 10-30mg/kg body weight.

DMSA has been found to be primarily albumin-bound in plasma through a disulfide bond with cysteine with very little remaining unbound. [15] DMSA is primarily distributed in extravascular space. [16] Like oral DMPS, the bioavailability for oral DMSA is at best 40%. [17] In adult human volunteers, the peak concentration occurred in 3.0+ 0.45 hours after 10 mg/kg dosing orally and the majority of the metal chelate elimination occurs within 24 hours. Most (>90%) conjugates as DMSA-cysteine disulfide. [18] Renal clearance is greater in healthy adults than in children. [19]

The EDTAs

The chelating agent EDTA (Ethylene Diamine Tetraacetic Acid) belongs to the group of Polyaminopolycarboxyic acids, which are known as aminopolycarboxylates or APCAs. EDTA is an organic compound containing one or more amino groups and two or more carboxyl groups, which act as strong chelating agents that form stable, water-soluble complexes with divalent and trivalent metal ions.

For chelation purposes, Poison Centers list two types of EDTA, namely NaEDTA (Ethylenediaminetetraacetic acid, also refered to as Disodiumedetat) and CaNaEDTA (calcium-disodium ethylenediamine tetraacetic acid) The difference between CaNaEDTA and NaEDTA needs attention. NaEDTA easily binds with calcium and is approved for the treatment of hypercalcaemic conditions. Because of this calcium-binding ability, NaEDTA should not be used in children. [20] Calcium DiNatrium EDTA is a complex in which four carboxylic acid groups and two amine groups from EDTA combine with calcium ions (Ca^{2+}) in strong coordinate covalent bonds. Calcium ions are locked in this structure and are prevented from participating in any other reactions. Because CaNaEDTA is already complexed with calcium, no additional calcium binding can be expected during the chelation process.

NaEDTA can be complexed with magnesium, resulting in NaMgEDTA (EDTA magnesium disodium). NaMgEDTA easily binds calcium and therefore, it is not recommended for use in children. Since Magnesium increases artery dilation, it is used in patients with vascular diseases. [21]

Sodium-Thiosulfate

Sodium thiosulfate is used to treat cyanide poisoning.

[22] Other uses include treatment of hemodialysis and chemotherapy. In September 2022, the U.S. Food and Drug Administration (FDA) approved sodium thiosulfate under the trade name Pedmark to lessen the risk of ototoxicity and hearing loss in infant, child, and adolescent cancer patients receiving the chemotherapy medication cisplatin. [23] Environmental physicians have used sodium thiosulfate for chelation purposes.

Alpha Lipoic Acid (ALA)

Alpha-lipoic acid, also known as thioctic acid, is a naturally occurring dithiol compound. Both in vivo and in vitro studies demonstrate that ALA exhibits the ability to scavenge free radicals, chelate redox-active transition metals and regulate the detoxification of heavy metals. It has been suggested that ALA may be useful to protect against and reverse arsenic-induced cell toxicity. [24]

Comparison of Metal-binding

The following provides information on metals and chelating agents used for chelation practices. The urinary values are listed in the respective diagrams as mcg/g Creatinine, representing a 95Percentile.

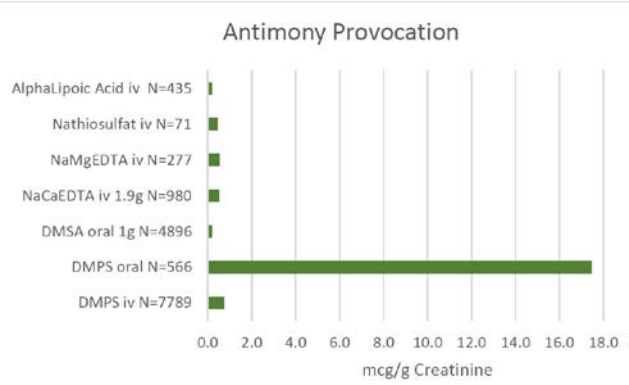
N= Amount of data sets statistically evaluated

Antimony (Sb)

Antimony's toxicity is relatively well documented. Most will enter the environment due to mining and industrial activities [25] and its toxicity is derived by its binding to thiol-containing enzymes. According to the Agency for Toxic Substances and Disease Registry (ATSDR) certain compounds of Antimony have beneficial effects when used for medical reasons, particularly those for certain types of parasites. Some side effects have been reported, including heart problems, nausea and vomiting, and muscle and joint pain. [26]

Diagram 1 indicates that intravenous DMPS is the choice of chelator used in the treatment of an Antimony overexposure.

Diagram 1: Urinary Antimony Concentration after Provocation



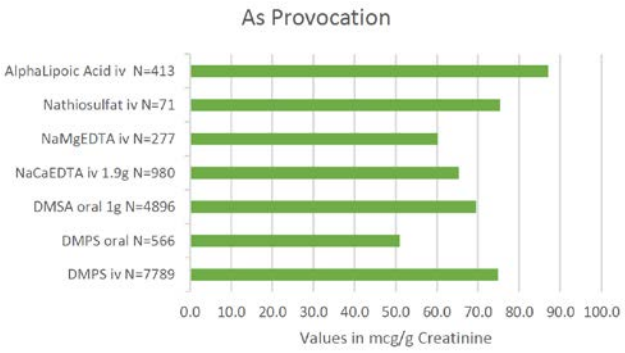
Arsenic (As)

Arsenic intoxication, also known as arsenicosis, is a medical condition primarily caused by exposure to contaminated drinking water, soil, or industrial sources. It affects over 200 million people globally, with high prevalence in regions like Bangladesh and West Bengal, Argentina, Chile, China, Mexico, Pakistan, and the United States.

Symptoms of acute poisoning include severe nausea, vomiting, abdominal pain, and watery diarrhea (often described as "rice-water" stools). Severe cases can lead to hypovolemic shock, encephalopathy, seizures, and multi-organ failure. Chronic of long-term exposure causes skin thickening, hyperpigmentation, Mees' lines (nail bands), peripheral neuropathy (numbness/tingling), and increased risk of cancer (skin, lung, bladder

For treatment purposes, DMPS is generally recommended. As Diagram 2 indicates, the use of oral ALA (Alpha Lipoic Acid), oral DMSA or Sodium-thiosulfate present an alternative.

Diagram 2: Urinary Arsenic Concentration after Provocation



Beryllium (Be)

Exposure to beryllium in the workplace can lead to a sensitized immune response. People who have a particular genetic characteristic called HLA-DPB1-Glu polymorphism are more likely to become sensitized to beryllium. This sensitivity stimulates a specific type of immune cell known as CD4+ T cells. The various signals from the immune system result in granuloma formation, small areas of inflammation in the body.

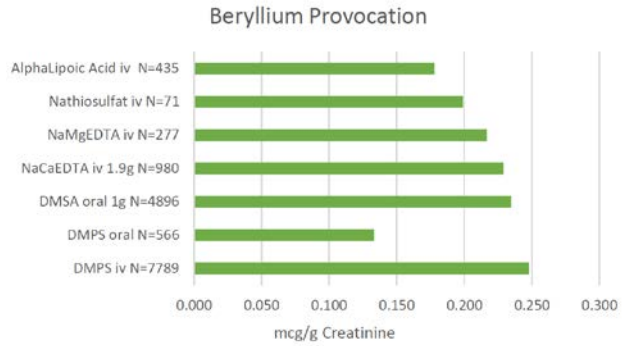
The commercial use of beryllium requires the use of appropriate dust control equipment and industrial controls at all times, because inhaled beryllium-containing dusts is toxic, and can cause berylliosis, a chronic life-threatening allergic disease. [27] Berylliosis is typically manifested by chronic pulmonary fibrosis and, in severe cases, right-sided heart failure and death. [28] Chronic berylliosis resembles sarcoidosis in many respects, and the differential diagnosis is often difficult.

Chronic exposure to beryllium was noticed in people who work near nuclear and beryllium production industry. According to studies in the 1970s and 1980s, the number of employees exposed to beryllium in the U.S. went from 21,200 to 800,000. Other data suggests that berylliosis clusters may be connected to exposure to concrete dust.

After beryllium exposure, the first and most important step is removal from the source of exposure. The US researchers Cash R, Shapiro R et al found the use of EDTA chelation in the treatment of beryllium pneumonoconiosis effective. [29]

Our statistical evaluation of provocation urines involving various chelating agents indicate that all the chelating agents included in this study provide a means of detoxing Beryllium; oral DMPS seemed the least effective.

Diagram 3: Urinary Beryllium Concentration after Provocation

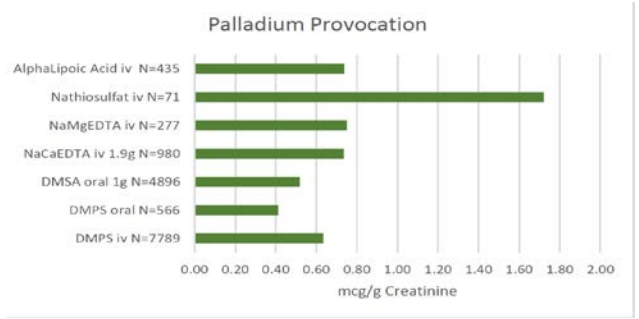


Palladium (Pd)

Palladium is used in dentistry, electrical appliances and jewellery, but the greatest increase in Pd demand has been in automotive emission control catalysts. This increase has been shown in air and dust samples. Pd2+ is regarded as having low toxicity but is a potent enzyme inhibitor.

Of the chelators used in this study, Diagram 4 indicates that intravenously applied Na-Thiosulfate showed the best binding ability.

Diagram 4: Urinary Palladium Concentration after Provocation



Lead (Pb)

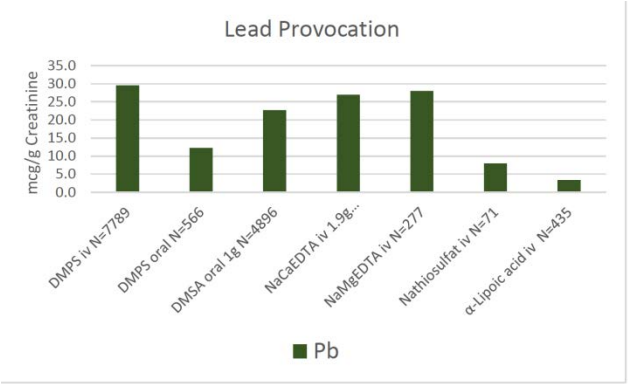
Acute lead intoxication is no longer as common as it was in prior centuries. In many countries, gasoline and paint no longer contain lead; however chronic lead overexposure remains a health problem. Lead continues to be present in dirt, dust, certain toys, and old house paint. Although new building codes require lead-free solder, lead is still found in the drinking water of old homes. Lead bullets, fishing sinkers, and soil contaminated by decades of car exhaust or years of house paint scrapings contain lead. Hobbies involving soldering, stained glass, jewelry making, pottery glazing, and miniature lead figures place people at risk for lead overexposure. Small children are at a greater risk of being affected by lead exposure.

The EDTAs have been considered the lead chelators of choice, Diagram 5 indicates that the intravenous use of DMPS or the EDTAs seem suitable chelator for lead. Oral DMSA provides an alternative.

Diagram 5: Lead (Pb) concentration after chelation (95%ile)

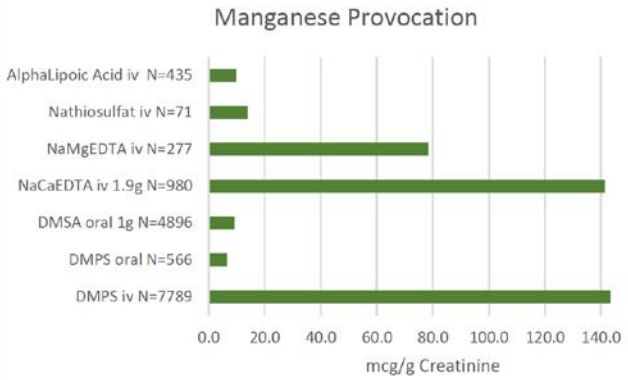
Manganese (Mn)

Manganese is a trace element necessary for good health. In excess, manganese is toxic. Inhaling Mn-dust or fumes may cause irritation of the lungs, possibly leading to pneumonia. Workers exposed to high levels of manganese experienced behavioural changes and other nervous system effects, resulting in slow and clumsy movements. This combination of symptoms when sufficiently severe is called "manganism", which closely resembles symptoms of Parkinson syndrome.



Chelation therapy presents a promising approach for addressing manganese toxicity. Diagram 6 shows that intravenous NaCaEDTA and intravenous DMPS are effective manganese chelators.

Diagram 6: Manganese Provocation Results



Mercury (Hg)

Mercury intoxication is no longer a topic in occupational medicine only. In dentistry, mercury is the metal most commonly associated with health problems. In Denmark, dentists no longer use mercury amalgams, and in 2008, Sweden and Norway also banned its use. [30] Amalgam fillings may contain up to 50% of mercury. Scientists agree that dental amalgam fillings slowly release mercury vapor into the mouth. But both the amount of mercury released and the question of whether this exposure presents a significant health risk remain controversial. Diagram 7 indicates that the intravenous administration DMPS by far exceeds the mercury excretion caused by other chelating agents.

Diagram 7: Mercury (Hg) Provocation Results

Silver (Ag)

Dental fillings commonly consist of 50% mercury (50%), 22-32% silver, ~14% tin, and ~8% copper and other trace metals. Silver and tin are not considered highly toxic, nor are they carcinogenic. [31,32]

However, exposure to silver may cause silver deposits in the skin and other parts of the body. While this is not considered harmful, dentists and environmental physicians are concerned about tissue overexposure to silver.

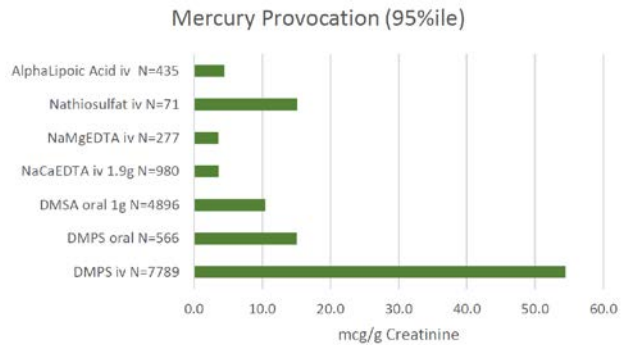
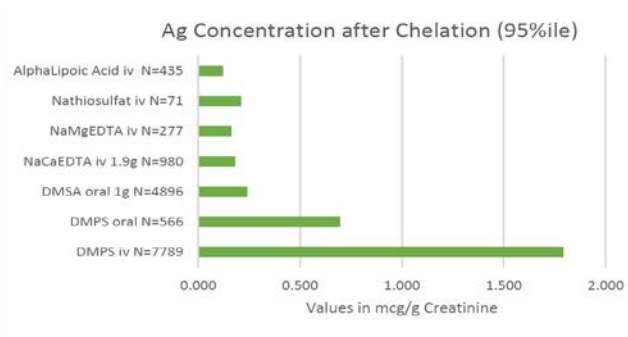


Diagram 8 indicates that silver is best bound using intravenously applied DMPS. The oral form, which has an absorption rate of 40%, seems a good alternative. In fact, Heyl Co., the supplier of Dimaval® recommends oral use.

Diagram 8: Silver (Ag) concentration after Chelation



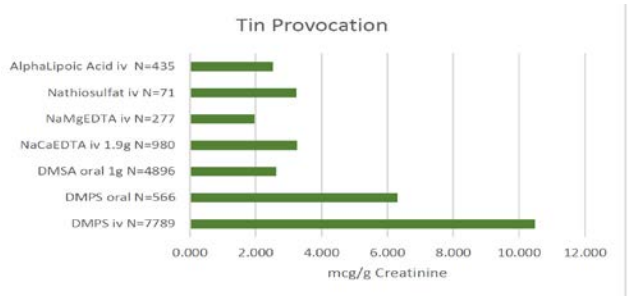
Tin (Sn)

Due to its poor gastrointestinal absorption, human and animal studies show that ingestion of large amounts of inorganic tin can cause stomachache, anemia, and liver and kidney problems. Breathing or swallowing, or skin contact with some organo-tins, such as trimethyltin and triethyltin compounds, can interfere with brain and nervous system function. [33]

The most common form of tin in commercial products is called stannous fluoride, which is found in mouth rinse or toothpaste. The most significant health risks arise from swallowing excessive amounts, which can lead to gastrointestinal distress in children.

Rare side effects include mild tissue irritation or sloughing of the inner cheeks, and in exceptional cases, allergic contact cheilitis (swelling and blistering of the lips) triggered by the tin component. These allergic reactions are uncommon and symptoms usually disappear upon discontinuation of the product. Diagram 9 indicates that of all the chelators tested intravenously applied DMPS showed the best chelating ability.

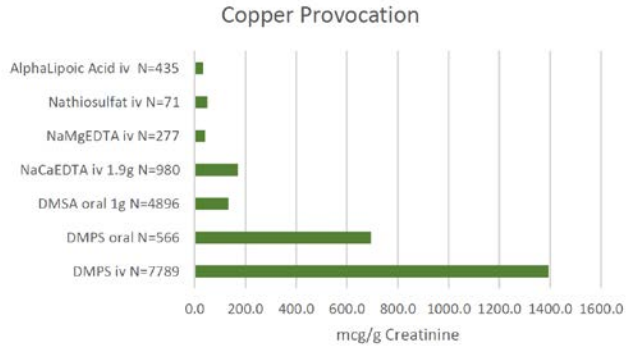
Diagram 9: Tin (Sn) concentration after chelation



Copper (Cu)

Copper (Cu) is one of the essential elements, vital for mental and physical health. Copper toxicity is a potential complication in long-term hemodialysis patients. Acquired copper toxicity can result from ingesting or absorbing excess copper. Inherited copper toxicity (as in Wilson's disease) results in accumulation of copper in the liver and other organs. Hepatic or neurologic symptoms develop. Treatment consists of a low-copper diet, zinc supplementation to prevent copper uptake and chelation. Presently, penicillamin is listed as the chelator of choice. Diagram 10 indicates that DMPS might be considered an alternative.

Diagram 10: Urinary Copper (Cu) Concentration after Provocation



Summary:

For the treatment of metal exposure, Table 1 lists the chelator of choice for the individual elements evaluated in this study. The information is based on statistics only. No consideration is given to patient symptoms and condition. To choose a provocation test for multiple metal exposures, the following list may serve as a guideline.

Table 1.

Element	DMSA oral	DMPS oral	DMPS i.v.	NaMgEDTA i.v.	CaEDTA i.v.	Na-thio-sulfate iv	a-Lipoic acid oral
Antimony		best					
Arsenic	good	good	Very good	good	good	Very good	Best*
Beryllium	good	ok	Best	good	good	good	good
Cesium			good		best		
Copper		good	best				
Chromium			best		Very good		
Lead	good		best	Very good	Very good		
Manganese			Best	Very good			
Mercury	best	good				good	
Nickel				best		Very good	

Element	DMSA oral	DMPS oral	DMPS i.v.	NaMgEDTA i.v.	CaEDTA i.v.	Na-thio-sulfate iv	a-Lipoic acid oral
Palladium						best	
Silver			best				
Thallium	ok	ok	ok	ok	ok	ok	ok
Tin		good	best				
Vanadium			Very good		best		

*The statistical evaluation of arsenic in urine after provocation with ALA came, with one exception, from a medical clinic, located in Riat, Saudi Arabia. This clinic has not submitted any other provocation urine utilizing chelating agents other than ALA. More data is needed to support our findings.

Conclusion

None of the chelating agents discussed can be considered superior to any of the others. Each has its place. However, when selecting a chelating agent for diagnostic purposes, the use of intravenously applied DMPS has a broad metal binding ability and thus seems most suitable as a provocation test for the diagnosis of a metal exposure. While the intravenous application of chelating agents provides an effective method for chelating metals, the use of oral chelators presents a reasonable alternative.

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