



## PROPORTIONS OF ARSENIC AND ANTIMONY IN BIOGEOCHEMICAL PROVINCES AS HEALTH RISK FACTORS

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*To perform efficient activities aimed at managing population health risks, it is necessary to examine regularities related to distribution of chemical elements in the biosphere; especially in so called biogeochemical provinces of natural or technogenic origin. We used semimetals of arsenic and antimony as an example to show that similarity of their physical and chemical properties is accompanied with similar effects they produce on living organisms. However, amphoteric character of arsenic and antimony determines wide range of possible interactions between these elements and biological molecules in a body. As a result, combined influence exerted by these substances on living organisms leads to both antagonistic relations and competition between them and to synergy as well. Basing on reviewed literature data, we showed that animals selectively limited accumulation of arsenic in their bodies and consumed less toxic antimony in greater quantities in case of biochemical anomalies while plants were much less selective and accumulated toxic arsenic easily. Accordingly, any activities aimed at population health risk reduction that are to be performed on territories of biogeochemical provinces should take into account peculiarities related to accumulation of these elements in bodies of warm-blooded animals and people. These peculiarities should also be taken into account when hygienic research programs and hygienic inspections are drawn up. When such research is performed experts should do the following: to analyze ways and chemical forms of elements migration in the environment; to determine molecular mechanisms of elements penetration into a cell and conditions of various scenarios of their metabolism and biological efficiency.*

**Key words:** arsenic and antimony compounds, biogeochemical provinces, biogeoecosis, population health, medical and prevention activities.

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Live organisms existing within a specific biogeochemical province have to rearrange their life processes. It leads to a specific elements imbalance in a body that is to be eliminated with special medical and preventive technologies [1–4].

Absorption of chemical elements from the environment by warm-blooded organisms depends on chemical properties and aggregate state of an element, its quantity, accompanying elements, and properties of body tissues that contact it as well [5]. The conditions are multiple and it makes absorption of chemical elements by a body a truly situational process. Therefore, given specific con-

ditions existing in this or that biogeochemical province, it is necessary to determine regularities of introduction, accumulation, and excretion out of a body for elements that are specific for it. Given this multiple dependency of toxic effects produced by chemicals, I.M. Trakhtenberg noted [6] that it was necessary to perform an obligatory examination of overall quantitative contamination of the environment in biogeochemical provinces in order to work out efficient preventive measures.

Biogeochemical anomalies, both natural and technogenic ones, have been studied for more than 50 years and it allowed to determine what

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substances occurred in them, as well as reasons and conditions for formation of biogeochemical provinces; to understand and formulate regularities related to their formation; to work out strategy and tactics of activities aimed at providing prevention of population health disorders [7]. Growth in industrial outputs requires new resource provision based on new technological solutions both in production and in exploration of new mineral deposits and in recultivation of previously used ones [8].

Arsenic is a widely spread element which is contained in a lot of minerals, especially metals-containing ones. Arsenic is a metalloid, that is, it is a substance that is between metals and non-metals; so, if we want to get a better insight into regularities of its dispersion, it is interesting to analyze biogeochemical properties of other metalloids. We can take antimony as an example as it is another element that has been used by people from the earliest times and, what is also important, also accompanies a lot of other metals [9]. Antimony (Sb) and its compounds were enlisted among toxic or hazardous substances that require immediate attention on the 43th session of the World Health Assembly held in Geneva in 1990 [10, 11].

Metals and metalloids have a common capability to interact with sulfhydryl groups of biological molecules that participate in nervous impulses conduction, tissue respiration, muscle contraction, cellular membranes penetrability etc. A reaction between ions of metals and metalloids with SH-groups results in occurrence of insoluble compounds, so called mercaptides, that leads to disorders in certain biochemical processes underlying development of intoxication [12].

**Our research goal** was to perform a comparative analysis of toxicometry and toxicokinetics parameters of arsenic and antimony under conditions of their biogeochemical anomaly.

Arsenic is unique due to its occurrence everywhere, in minerals, rocks, soils, and water, in plants and animals. Average arsenic concentration in rivers is equal to 3 µg/l; in surface waters, about

10 µg/l; in seas and oceans, just about 1 µg/l<sup>1</sup>. Arsenic concentration in soils usually varies from 0.1 to 40 mg/kg. Much greater quantities of arsenic, up to 8 g/kg, can be found in areas where arsenic ores occur, as well as in volcanic regions [13].

Increased arsenic concentrations in soils can exert negative influence on agricultural crops due to arsenic becoming a food chain component [14]. Geochemical background of arsenic (As) which is detected in landscape components around tailings dams of mining enterprises is characterized with high arsenic pollution, namely 57-300 mg/kg [15]. Arsenic concentration in plants that grow on a territory of natural-technogenic landscapes varies considerably, from such low values as 0.001 to 847.29 mg/kg. Average arsenic contents in plants growing in this landscape are 2.7 times higher than in plants growing in quarry-dump landscapes and almost 28 times higher than in plants growing in natural landscapes [15]. Arsenic also migrates in the environment in a form of volatile arsenic-organic compounds [13].

Arsenic concentration in a living organism usually amounts to about 6 µg/kg. Daily introduction of arsenic into a human body is insignificant and varies from 50 to 100 µg, and its semi-excretion period amounts to 30-60 hours<sup>2</sup>. When arsenic enters a body, it then concentrates in the thyroid gland, liver, kidneys, spleen, lungs, bones, hair, brain tissues, and the muscles. There are some data on arsenic accumulation in the thyroid gland making for endemic goiter development [16].

Chronic negative effects produced by arsenic are damage to skin, neurotoxicity, cardiovascular diseases, diabetes, and cancer. The International Agency for Research on Cancer (IARC) rank arsenic and its non-organic compounds as "Carcinogenic to Humans" (group I) (IARC, 1980[17]). EFSA Food Contamination Panel detected that provisional tolerable weekly intake of arsenic shouldn't exceed 15 µg/kg\*day (EFSA contam PANEL, 2009)<sup>3</sup> [18].

Geometric average arsenic concentrations in

<sup>1</sup>Turbinskiy V.V. Hygienic grounds for sanitary protection of cross-border and border sources of drinking water supply for the RF population: dissertation ...for Doctor Of Medical Sciences. Novosibirsk, 2012. 376 p. Available at: <http://www.dissercat.com/content/gigienicheskie-osnovy-sanitarnoi-okhrany-transgranichnykh-i-pogranichnykh-istochnikov-pitevo> (date of visit February 13, 2018).

<sup>2</sup>Chemical structure of Russian food products: reference book. In: I.M. Skurikhin ed. Moscow, Deli print Publ., 2002, 236 p.

<sup>3</sup>P 2.1.10.1920-04. Guidelines on assessment of population health risk under exposure to chemicals that pollute environment. Moscow, Federal center for Sanitary and Epidemiologic Surveillance of The RF Public healthcare Ministry Publ., 2004,143 p.

umbilical blood of parturient women are equal to  $0.92 \pm 1.01$  ng/ml, at  $0.43 \pm 0.88$  ng/ml ( $n = 296$ ) in healthy newborns [19]. An inhibition of arsenic elimination out of a body was detected under single oral exposure to arsenous anhydride in a dose equal to 30 mg/kg which coincided with a threshold of acute action produced by arsenous anhydride on changes in SH-group contents in blood<sup>4</sup>.

Arsenic concentration in chicken that consumed it with forage in a dose equal to 0.5-5.0 mg/kg amounted to 0.11-0.2 mg/kg in muscle tissues; 0.09-0.12 mg/kg in liver; 0.09-0.34 mg/kg in kidneys; and 0.12-0.21 mg/kg in eggs<sup>5</sup>.

Oral introduction of arsenic oxide ( $As_2O_3$ ) into sheep bodies in a dose equal to 0.5 mg/kg of an animal body weight didn't cause any clinical intoxication symptoms during 3 months after exposure. But arsenic accumulated in a dose equal to 0.2-0.3 mg/kg in the kidneys, skin, liver, and spleen; about 0.12 mg/kg, in muscle tissues and lungs; about 0.25-0.3 mg/kg, in the abomasum, duodenum, jejunum, and ileum [20].

High arsenic concentrations in drinking water, 200-500  $\mu\text{g/l}$ , are toxic for the human endocrine system [21]. Cumulative exposures to arsenic with food and drinking water are higher in people suffering from the II type diabetes than in healthy people [22, 23] and it proves it is necessary to conduct further research on a role played by moderate and low arsenic concentrations (500-200  $\mu\text{g/l}$ ) in water [24].

Experts used models of animal cells cultures to show that arsenic acted as endocrine destroyer. They detected disorders in gene expression of steroid receptor (SR) in cells exposed to non-organic arsenic (arsenite,  $iAs(+3)$ ). Low  $iAs(+3)$  concentrations (0.1–0.7  $\mu\text{M}$ ) stimulate hormone-induced transcription, but higher non-cytotoxic arsenic concentrations (1-3  $\mu\text{M}$ ) inhibit transcription [25].

Antimony is a considerably rare element in the earth's crust,  $4 \cdot 10^{-5}$  %, although it, like arsenic, can occur in high concentrations in some regions [26]. In nature antimony usually has valence +3, more rarely +5 [27]. Compounds of trivalent posi-

tive antimony (sulfides, thiosalts, antimonites, trioxide) are most widely spread; the next place belongs to compounds of trivalent negative antimony (antimonides). Compounds of pentavalent antimony are rarely met in nature. Antimonial glitter  $Sb_2S_3$  (stibuite, antimonite) is the most widely spread antimony-containing mineral; it occurs in hydrothermal deposits as antimonial ores seams and tabular deposits [28].

Background antimony concentrations in upper soils layers amount to 0.76 mg/kg in sod-podzol soils; 0.99 mg/kg, in black earth; 0.28 mg/kg, in peaty soils. Antimony concentrations in Siberian rivers (Irtys, Ob', Tom' and Amur) amounts to 0.0007–0.002 mg/dm<sup>3</sup> <sup>5</sup>.

Antimony concentrations in tissues of trees and bushes that grow in areas with ores mineralization reach 7-50 mg/kg of dry solid matter; its average concentrations in plants parts above ground level amounts to 0.06 mg/kg of dry solid matter. Antimony concentrations in edible plants vary within 0.02-4.3  $\mu\text{g/kg}$  of raw matter. Antimony concentration in corn grains and potato tubers doesn't exceed 2  $\mu\text{g/kg}$  of dry solid matter; it reaches 29  $\mu\text{g/kg}$  in grass. Antimony concentration in barley and flax rootage amounts to 122 and 167  $\mu\text{g/kg}$  of solid dry matter respectively, and it is considerably higher than in leaves where it amounts to 10 and 27  $\mu\text{g/kg}$  of solid dry matter [28]. As concentrations of heavy metals in soils become very high, their concentrations in various parts of plants also increase. But a ratio between heavy metals concentrations in roots, culms, leaves, and reproductive organs is preserved.

Complex research performed on Dyukov Log waste storage where sulfide-containing wastes from Salairskiy ore mining and processing enterprise were kept helped to outline migration routes for drainage flows that contained increased antimony concentrations (96 MPC) and arsenic concentrations (6 MPC). It was detected that polluted drainage water penetrated water-bearing horizons that, among other things, were used as water supply sources for communal needs and drinking [29]. Plants growing on pasture lands and land-

<sup>4</sup>Zhuruli M.O. Studies on toxicokinetics of arsenous anhydride under various exposure in order to fix hygienic standards: dissertation...Candidate of Medical Sciences. Moscow, 1984, 179 p. Available at: <http://www.dissercat.com/content/izuchenie-toksikokinetiki-myshyakovistogo-angidrida-pri-razlichnykh-rezhimakh-ego-vozdeistviya#ixzz5CNZZqPjy> (date of visit January 19, 2018).

<sup>5</sup>Chupakhina O.K. Toxicity of arsenic for chicken and its distribution in poultry organs and tissues: thesis of dissertation...Candidate of Veterinary Sciences. Moscow, 1983, 21 p.

scape plants in Kadamjaiskaya biogeochemical antimony province accumulate substantial antimony concentrations, 1.2 – 16 MPC.

As per data obtained in some research, antimony can be found in a human body: in blood, 0.0033 mg/l; bone tissue,  $(0.01-0.6) \cdot 10^{-4}$  %; muscle tissues,  $(0.42-19.1) \cdot 10^{-6}$  %; a toxic dose amounts to 100 mg. Average daily introduction of antimony into a human body with food and water amounts to approximately 50  $\mu$ m. Antimony is rather slowly excreted from a body [30, 31].

Antimony in concentrations equal to 0.41-0.55 mg/kg and arsenic in concentrations equal to 0.79-0.82 mg/kg were detected in bone fragments belonging to a gray rat *Rattus Norvegicus* found in pellets of a long-eared owl that usually spends a winter in Tashkent and adjacent territories [32].

Elimination of antimony is equally related to its valence in a specific compound. Thus, when antimony trioxide was added to rats forage, 80-100  $\mu$ m of this metal were daily excreted with urine, and up to 100 mg, with stool. Pentavalent antimony is primarily excreted with urine even when it is introduced intragastrically [33].

Antimony (Sb) is similar to arsenic as per its properties; it was detected that antimony exerted inhibiting influence on enzymes that participate in carbohydrates, fats, and lipids metabolism. Just as arsenic, antimony reacts with sulfhydryl groups, has toxic properties, can probably cause immune deficiency [34], and causes functional disorders in

various organs (heart, kidneys, CNS, liver, lungs, intestines, lymphatic system and others) [35, 36].

Inhalation exposure to antimony aerosols that occur in working area air causes its higher concentrations in workers' bodies; they increase from 0.5 mg% to 2.1 mg% in blood; from 0.86 mg% to 1.86 mg% in urine; from 1.6 mg% to 7,8 mg% in hair [37].

Fivefold intraperitoneal introduction of metal antimony suspension in peach oil into white rats in a dose equal to 50 mg/kg of a body weight caused increased antimony concentrations in blood  $(10.46 \pm 1.22; 6.58 \pm 0.74$  mg%). Antimony accumulated in internal organs in the following concentrations: muscles,  $1.49 \pm 0.35$  mg%; lungs,  $1.38 \pm 0.2$  mg%; skin,  $1.14 \pm 0.3$  mg% [38].

Antimony concentrations in sheep bodies on a territory of antimony biogeochemical province amount to 3.66–12.7 mg/kg in the heart; 4.00–12.16 mg/kg, lungs; 2.6–10.2 mg/kg, kidneys; 3.6–10.0 mg/kg, muscles; concentration gradient is 2–2.4 [39]. Daily antimony introduction into a human body detected in Kadamdjayskaya antimony province, Republic of Kirgizstan, amounts to 8.54 mg while it is equal to only 1.22 mg per day on a reference territory [40], and a reference dose of antimony for chronic oral introduction into a body amounts to 0.0004 mg/kg. This increased antimony introduction exerts its negative influence primarily on dextrose and cholesterol contents in blood<sup>3</sup>.

Table

Arsenic/antimony ratio in geological and biological environment in biogeochemical provinces

| Environmental object                                  | Arsenic, mg/kg<br><u>min-max</u><br>average | Antimony, mg/kg<br><u>min-max</u><br>average | Average As/Sb ratio |
|---|---|--|---------------------|
| Soil  | 0,1–40,0<br>10,0                            | 0,28– 0,99<br>0,76                           | 13,1                |
| Water   | 0,0007–0,005<br>0,0029                      | 0,00005– 0,0007<br>0,00037                   | 7,8                 |
| Plants (antimony and arsenic biogeochemical province) | 57–300<br>100                               | 7–50<br>20                                   | 5,0                 |
| Background (reference territory)                      | 0–6,01<br>1,4                               | 0,02–4,3<br>0,7                              | 2,0                 |
| Animals:  | 0,79–0,82                                   | 0,41–0,55                                    | –                   |
| - wild rats (bones)                                   | 0,80  | 0,47   | 1,7                 |
| - sheep (lungs, kidneys, muscles)                     | 0,41–1,54<br>0,98                           | 6,41–8,08<br>7,25                            | 0,14                |
| - people (blood)                                      | 0,43–0,92<br>0,68                           | 3,3<br>3,3                                   | 0,21                |
| Threshold toxic does, mg/day                          | 20,0  | 100,0  | 0,2                 |
| Lethal dose for a man, mg                             | 50–340<br>180                               | 500–1000<br>750                              | 0,24                |

Research conducted in antimony biogeochemical provinces located in Fergana Valley revealed that adult population living there daily consumed approximately 0.1-0.15 mg of antimony with food and water and it was 10-15 times higher than a usual introduction on a reference territory [41].

Each pathology has its specific element structure and concentrations, including maximum ones, as well as changes in an aggregate parameter of their accumulation [42–44]. Antimony and arsenic are endocrine destroyers [45].

As/Sb ratios are obtained on the basis of collected data on arsenic and antimony concentrations in geological and biological objects. The results are shown in the Table.

The obtained ratios show that arsenic concentrations are 2-13 times higher than antimony ones in geological environmental objects, and, on the contrary, antimony concentrations are 5-20 times higher in biological media of living organisms. Arsenic contents in bones is 1.7 times higher than antimony contents, just like in geological objects.

Higher antimony concentrations than arsenic ones in a body are determined by antimony being less toxic than arsenic.

Therefore, plants are less selective and tend to accumulate toxic arsenic easily when growing in biogeochemical anomalies while animals selectively limit its introduction into their bodies and try to consume less toxic antimony instead. Accordingly, all medical and prevention activities performed in biogeochemical provinces with increased arsenic and antimony occurrence are to take into account peculiarities of these elements accumulation in bodies of warm-blooded animals and people. Programs for in-depth research within hygienic examinations, inspections, or investigations should also take these peculiarities into account. Research should include analysis of directions and chemical forms in which elements migrate in the environment, establishment of molecular mechanisms with which elements penetrate into a cell, and conditions for various scenarios of their metabolism and biological activity.

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