ARSENICPLATFORM

HUSRB/1002/121/075

Mađarska-Srbija
IPA prekogranični program

Značaj različitih oblika arsena u strategiji upravljanja vodosnabdevanjem

Arzén különböző formáinak jelentősége a vízellátás igazgatásának stratégiájában

The significance of different arsenic species for water supply management strategy

Mr Malcolm Watson

Projekat sufinansira Evropska unija

Novi Sad, 04.09.2012.

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Overview

- Arsenic in drinking water sources - a global problem
- Different arsenic species and their relative toxicities
- Human metabolism of arsenic
- Drinking water legislation
- Water supply strategy
- Regional situation, arsenic presence and health issues
- Arsenic removal technologies
- Recommendations for moving forwards to deal with the arsenic issue

Sadržaj prezentacije

- Arsen u izvorištima vode za piće kao globalan problem
- Različiti oblici arsena i njihova toksičnost
- Metabolizam arsena kod ljudi
- Voda za piće - regulativa
- Strategija vodosnabdevanja
- Regionalno stanje, prisustvo arsena i posledice na zdravlje
- Tehnologije za uklanjanja arsena
- Preporuke za dalji rad vezane za problematiku arsena

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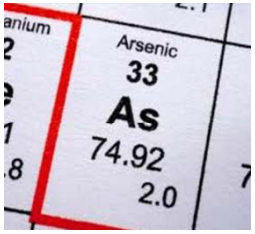
Arsenic in the environment
Arsen u životnoj sredini



- Arsenic the element
  - Atomic mass: 75
  - Metalloid, mono-isotopic
  - Natural abundance in the Earth's crust: 2.1 ppm

- Arsenic compounds
  - Oxidation states: +3 and +5 are most common,
  - -3 and 0 also possible
  - Most common arsenic containing minerals:
    - arsenopyrite (FeAsS)
    - orpiment (As_2S_3)
    - realgar (α-As_4S_4)

- Arsen kao element
  - Atomska masa: 75
  - Metaloid, mono-izotopski
  - Prirodni fon (sadržaj) u zemljinoj kori: 2,1 ppm

- Jedinjenja arsena
  - Oksidaciona stanja: +3 i +5 su najzastuplenija;
  - -3 i 0 su takođe moguća
  - Najzastupleniji minerali koji sadrže arsen:
    - arsenopirit (FeAsS)
    - orpiment (As_2S_3)
    - realgar (α-As_4S_4)

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As in drinking water – a global issue

As u vodi za piće – globalni problem

- Estimates suggest more than 130 million people worldwide drink water which is naturally contaminated by As.
- Istraživanja su procenili da 130 miliona ljudi u svetu pije vodu koja je prirodno kontaminirana arsenom.

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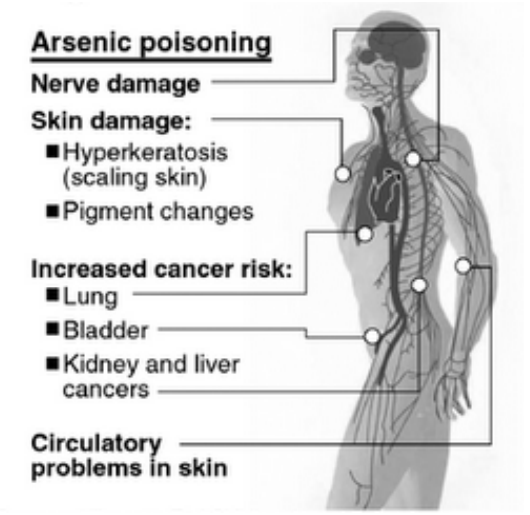
Chronic arsenic exposure
Hronična izloženost arsenu



- The skin lesions occurring most frequently in arsenic-exposed humans are hyperkeratosis, hyperpigmentation, and skin cancer.
  - Arsenical hyperkeratosis occurs most frequently on the palms and soles.
  - Confounding factors for arsenic-induced skin cancer may include exposure to sunlight, chronic liver disease, and nutritional status.

- Kod ljudi izloženih arsenu najčešće se javljaju promene na koži, hiperkeratosis, hiperpigmentacija i rak kože.
  - Arsensom indukovan hiperkeratosis se najčešće javlja na dlanovima i tabanima.
  - Arsenom-indukovan rak kože je teško razlikovati od raka kože prouzrokovanog sunčevom svetlošću, hroničnim oboljenjima jetre i ishranom.

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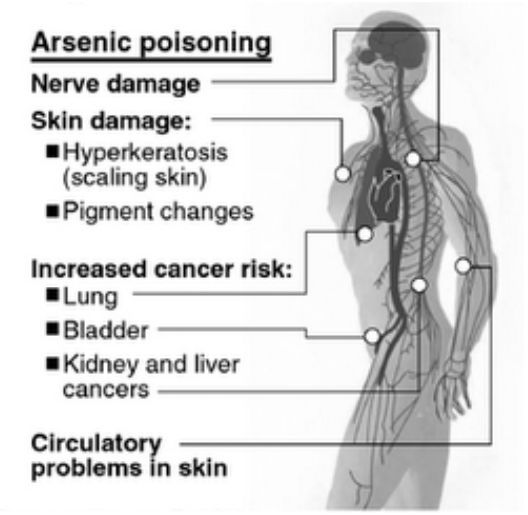
Chronic arsenic exposure
Hronična izloženost arsenu



- Chronic arsenic ingestion may also cause cancers of the bladder, kidney, liver, lung and prostate.
- Both acute and chronic exposure may result in a wide range of adverse cardiovascular effects.
  - Association between chronic arsenic exposure and peripheral vascular disease, hypertension, and cardiovascular disease.

- Hroničan unos arsena takođe može da prouzrokuje rak bešike, bubrega, jetre, pluća i prostate.
- Akutna i hronična izloženost može izazvati neželjene kardiovaskularne efekte.
  - Veza između hronične ekspozicije arsenu i perifernih vaskularnih bolesti, hipertenzije i kardiovaskularnih oboljenja.

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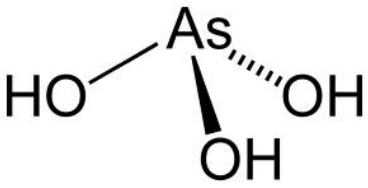
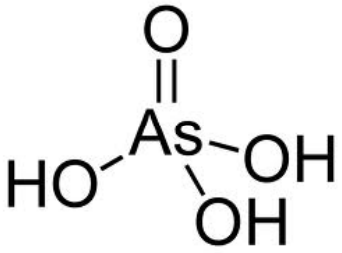
Arsenic species and their relative toxicities
Oblici arsena i njihova relativna toksičnost

As(III)
arsenite
arsenit



As(V)
arsenate
arsenat



- **Inorganic arsenic – iAs – Neorganski arsen**
  - iAs^{III} eg. Arsenous acid
  - iAs^{III} eg. Arsenitna kiselina
  - iAs^{V} eg. Arsenic acid
  - iAs^{V} eg. Arsenatna kiselina

- iAs^{III} generally considered as much as 60 x more toxic than iAs^{V}.
- iAs^{III} se generalno smatra oko 60 puta toksičnijim u odnosu na iAs^{V}.

- 70-80 mg iAs^{III} reported to be fatal for humans.
- Utvrđeno je da je 70-80 mg iAs^{III} fatalno za čoveka.

- Organic arsenic – different metabolites of arsenite and arsenate, historically considered much less toxic than inorganic arsenic
- Organski arsen – različiti metaboliti arsenita i arsenata, ranije su smatrani manje toksičnim od neorganskih oblika arsena.

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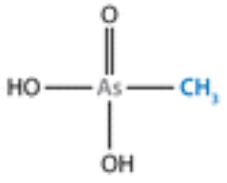
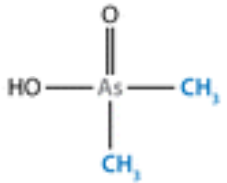
Organic arsenic species
Organski oblici arsena

- **Methylated arsenic species**
  - mono and dimethylated species
- **Metilovani oblici arsena**
  - mono i dimetilovani oblici

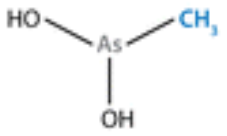


Monomethylarsonic acid
MMA

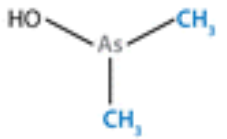


Dimethylarsinic acid
DMA

  - In humans (and other higher organisms), created in the liver
  - Kod ljudi (i drugih viših organizama), najstaju u jetri.

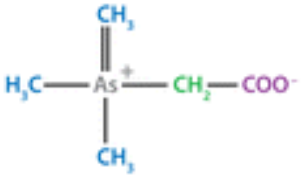


Monomethylarsonous acid
MMA(III)

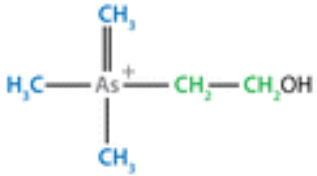


Dimethylarsinous acid
DMA(III)

- **Arsenobetaine and arsenocholine**
  - Known as 'fish arsenic' – marine animals
  - Not generally considered toxic
- **Arsenobetain i arsenoholin**
  - Ne smatraju se toksičnim
  - Poznat kao "riblji arsen" – morske vrste



Arsenobetaine



Arsenocholine

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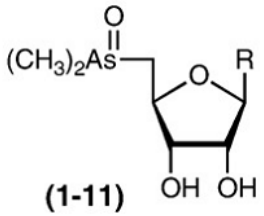
- **Arsenosugars Arsenošećeri**
- Found in algea
  (bottom of food chain)
- Pronađeni kod algi (najjednostavnija karika lanca ishrane)
- not considered toxic
- ne smatraju se toksičnim
- could potentially be broken down by during cooking into MMA^V and DMA^V
- Mogu se delimično razložiti tokom kuvanja u MMA^V and DMA^V



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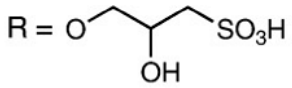

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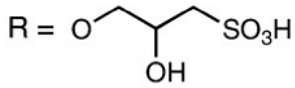

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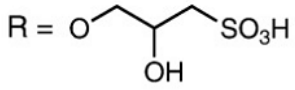

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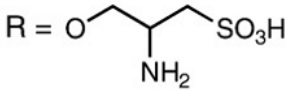

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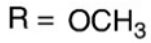

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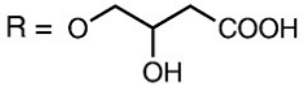

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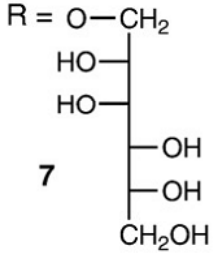

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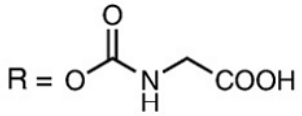

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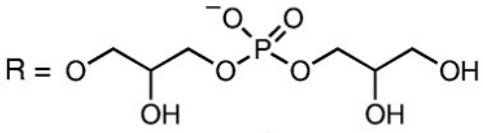

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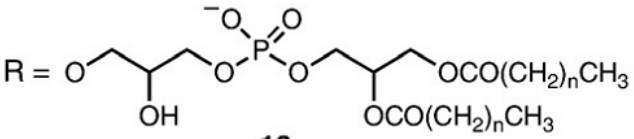

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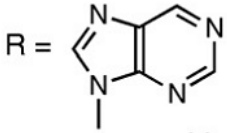

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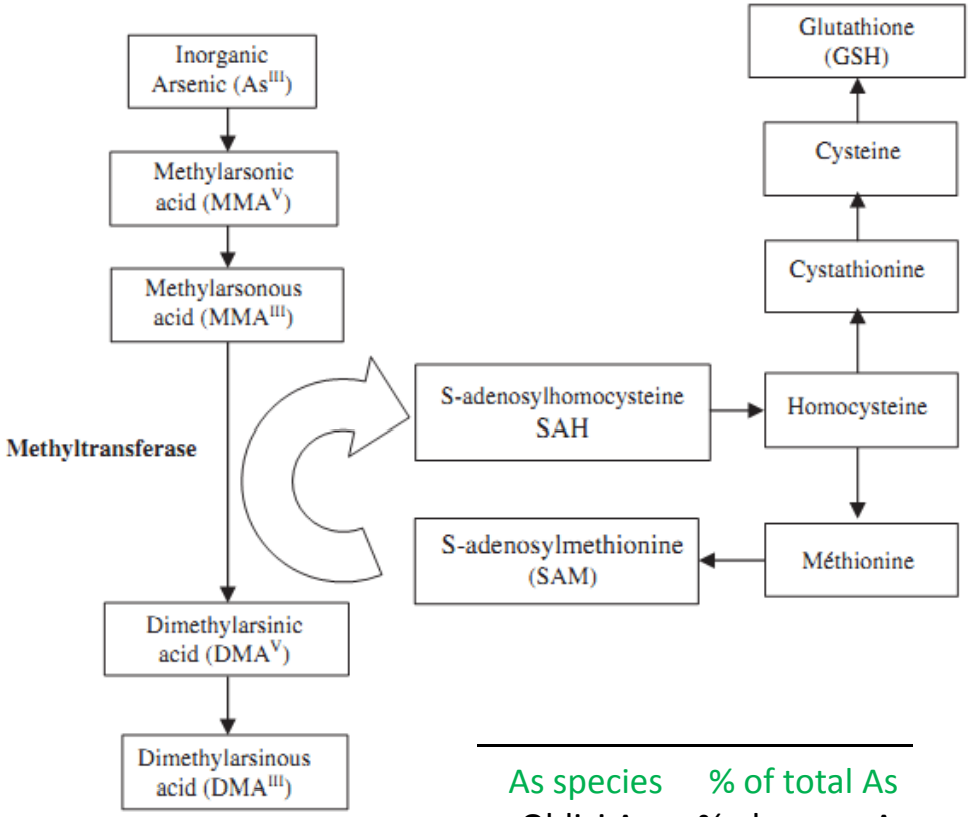
Human arsenic metabolism
Metabolizam arsena kod ljudi

- Two main metabolic pathways for arsenic:
  - reduction of pentavalent to trivalent arsenic
  - Methylation the reaction with the methyl donor SAM (S-adenosyl –methionine) is catalysed by methyltransferases in the presence of glutathione.
- Dva metabolička puta arsena:
  - Redukcija petovalentnog u trovalentni As
  - Metilacija reakcija sa metil donorom SAM (S-adenozil-metionin) je katalizovana metiltransferazama u prisustvu glutationa.
- Process repeats until final metabolite DMA^{III} reached
- Distribution of arsenic species in urine:
- Proces se ponavlja dok ne nastane finalni metabolit DMA^{III}
- Distribucija oblika arsena u urinu:



As species Oblici As	% of total As % ukupnog As
iAs	10-30%
DMA	60-80%
MMA	10-20%

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Toksičnost metilovanih oblika
Toxicity of methylated species

- Toksičnost As(III) (neorganski i organski) usled afiniteta za sulfhidrilne grupe biomolekula kao što su glutation - formiranje As(III)-sumpor veza rezultuje inhibicijom enzimske aktivnosti.
- As(III) toxicity (inorganic and organic) due to affinity for sulfhydryl groups of biomolecules such as glutathione – formation of As(III) - sulphur bonds results in inhibition of enzyme activity.

- Letalna doza za ljudske ćelije:
- Lethal doses for human cells:
- Trovalentni metilovani oblici As su toksičniji u odnosu na petovalentne.
- Ultimately, trivalent methylated As species are more toxic than pentavalent ones

Oblici As As species	LD_{50} (µM)
iAs(III)	5.49
DMA(III)	2.16
iAs(V)	571
DMA(V)	843

Naranmandura et al, Chem. Res. Toxicol., 2007, 20 (8), pp 1120–1125

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As in drinking water legislation

As u vodi za piće - regulativa

- 1958: WHO International Standards for Drinking-water recommend MAC of 200 µg/l

  SZO Internacionalni Standardi za Vodu za piće preporučuju MDK od 200 µg/l
- 1963: WHO lower recommendation to 50 µg/l

  SZO prepuručuje smanjenje na 50 µg/l
- 1980s: Arsenic poisoning in Bangladesh

  Trovanje arsenom u Bangladešu
- 1993: WHO lower recommendation to 10 µg/l

  WHO smanjenje na 10 µg/l

Arsenic Level in Tap Water	Approximate Total Cancer Risk (2 litres consumed/day)
0.5 ppb	1 in 10,000
1 ppb	1 in 5,000
5 ppb	1 in 1,000
10 ppb	1 in 500
50 ppb	1 in 100

- Currently, WHO, EPA, EU and Serbia legislate an MAC of 10 µg/l
- concerns over carcinogenicity, lowest practical level of determination
- MAC must be measurable
- MAC must be achievable

- Trenutno SZO, EPA, EU i Srbija – MDK od 10 µg/l
- Zabrinutost usled kancrogenosti, najniži praktični nivo određivanja
- MDK moraju biti merljive
- MDK mogu biti dostignute

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Water supply strategy in the region
Strategija vodosnabdevanja u regionu

- Is there are an arsenic problem?
  - How much iAs is in the water?
  - Do we have evidence (biomarkers) of arsenic exposure?
  - Epidemiological studies – can we observe adverse health effects in the exposed population?
- How severe is the problem, how do we solve it?
  - Can we use different water sources?
  - Can we afford the technology to remove the As?
- Can we afford not to solve the problem?
  - Economic cost of treatment
  - Poor health levels increase burden on the workforce
  - Societal cost

- Da li postoji problem sa arsenom?
  - Koliki je sadržaj iAs u vodi?
  - Da li imamo dokaz (biomarkeri) za izloženosti arsenu?
  - Epidemiološke studije – da li možemo da uočimo negativne efekte na zdravlje kod populacije izložene arsenu?
- Koliko je ozbiljan ovaj problem, na koji način ćemo ga rešiti?
  - Da li možemo da koristimo druga izvorišta vode?
  - Da li možemo da priuštimo tehnologiju za uklanjanje As?
- Da li smemo da ne rešimo ovaj problem?
  - Ekonomski troškovi tretmana
  - Niski nivo zdravlja negativno utiče na radnu populaciju
  - Društveni troškovi

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Yes, there is As in the water – situation in the Pannonian Basin
Da, zaista postoji As u vodi – situacija u Panonskom basenu

- Estimated million people exposed to As concentrations > 10µg/l
- Procenjeno je da je oko milion ljudi izloženo As u koncentraciji > 10µg/l

- Groundwaters taken from aquifers in sedimentary rock layers laid down during the Miocene epoch (ca 10 million years ago) by the Pannonian sea (pictured)
- Podzemna voda iz akvifera u stenovitim slojevima formirana je tokom miocena (pre 10 miliona godina) do Panonskog mora.

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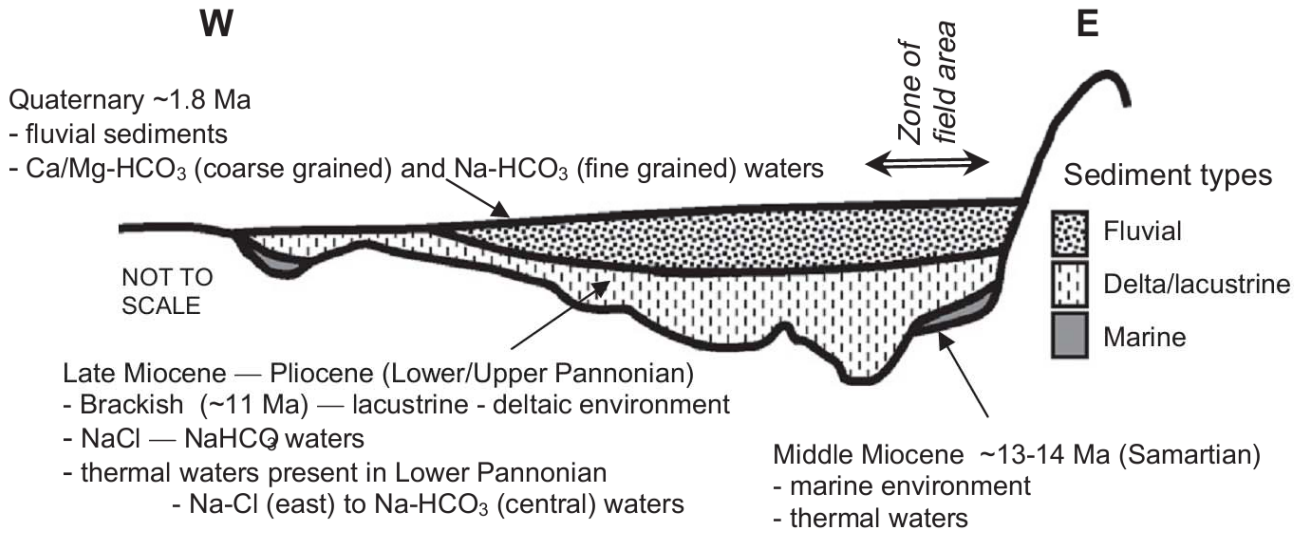
Yes, there is As in the water – situation in the Pannonian Basin
Da, zaista postoji As u vodi – situacija u Panonskom basenu

- Dissolution of As bearing Fe-minerals occurs in regions of low pH (around 7.6), high concentrations of organic ligands promote mobilisation in areas of groundwater discharge
- Rastvaranje minerala arsena i Fe se dešavalo pri pH oko 7,6, visoke koncentracije organskih liganada ubrzavaju mobilizaciju u oblasti proticanja podzemne vode.

- Despite high arsenic levels, in many cases, these aquifers represent best water available for drinking – it is microbiologically safe
- Uprkos visokom sadržaju arsena, u mnogim slučajevima ovi akviferi predstavljaju najbolju raspoloživu vodu za piće – mikrobiološki je bezbedna.



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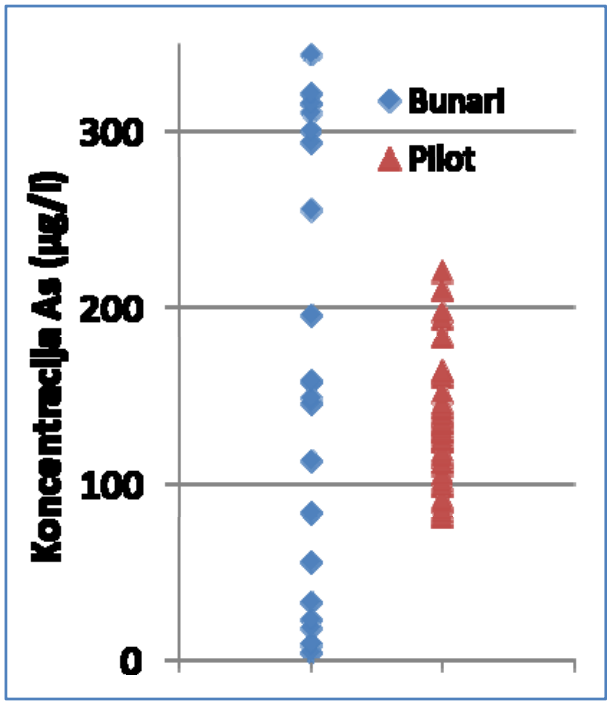
As u vodi, podaci o ekspoziciji
As in water, evidence of exposure



- Zrenjanin kao primer - Zrenjanin for example:
  - $iAs^{(III)}$: 78 % $iAs^{(V)}$: 2 % oAs: 20 %
  - Ukupni As iznosi čak do 350 µg/l As
  - Total arsenic as high as 350 µg/l As
- Bacs, Bekes , Csongrad:
  - iAs, MMA i DMA su detektovani u povišenim koncentracijama u urinu ljudi
  - iAs, MMA and DMA found in elevated levels in peoples urine
- Podaci o ekspoziciji u Srbiji i Mađarskoj su još uvek oskudni – biomarkeri koje treba ispitati podrazumevaju:
  - Oblike arsena u krvi i urinu, sadržaj As u noktima i kosi
  - Biomarkere genotoksičnosti, kao to su hromozomske aberacije, formiranje mikronukleusa
- Literature on exposure in Serbia and Hungary still scarse –biomarkers which could be investigated include:
  - As species in blood and urine, As levels in nails and hair
  - Genotoxicity biomarkers such as chromosomal aberrations, micronucleus formation

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Health consequences – Hungary
Posledice na zdravlje - Mađarska

- M. Börzsönyi, A. Bereczky, P. Rudnai, M. Csanady and A. Horvath. Epidemiological studies on human subjects exposed to arsenic in drinking water in Southeast Hungary (Epidemiološka studija o ljudima izloženim arsenu preko vode za piće u jugozapadnoj Mađarskoj). *Archives of Toxicology*, Volume 66, Number 1 (1992).

  - Found elevated levels of As in peoples hair
  - Report cases of arsenical hyperkeratosis and hyperpigmentation
  - Visok sadržaj As u ljudskoj kosi
  - Potvrđeni slučajevi arsenske hiperkeratoze i hiperpigmentacije
  - Observed statistically significant differences between frequency of spontaneous abortions and stillbirths for between two populations, one exposed to high As levels, the other not.
  - Zapažene statistički značajne razlike između učestalosti spontanih pobačaja i mrtvo rođene dece između dve populacije, jedna je bila izložena visokom sadržaju As, a druga nije.

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Health consequences: acute coronary syndrome in Zrenjanin
Posledice na zdravlje: akutni koronarni sindrom u Zrenjaninu

- Acute coronary syndrome (ACS) includes unstable angina, myocardial infarction (heart attacks) and sudden cardiac death.
- Recent study compared two populations from Zrenjanin
  - Water containing < 10 µg/l (median 1 µg/l, mean 1.6 µg/l)
  - Water containing > 10 µg/l (median 80 µg/l, mean 112 µg/l)

- Akutni koronarni sindrom (ACS) obuhvata nestabilnu anginu, infarkt miokarda (srčani napad) i iznenadnu srčanu smrt.
- Novije studije porede dve populacije u Zrenjaninu
  - Voda koja je sadržala < 10 µg/l (medijana 1 µg/l, sr. vred. 1.6 µg/l)
  - Voda koj je sadržala > 10 µg/l (medijana 80 µg/l, sr. vred. 112 µg/l)

- Vrednosti su korigovane za:
  - Starost, pol, stepen obrazovanja, gojaznost, medicinski istorijat dijabetesa, arterijske hipertenzije, dislipidemije (visok holesterol)

- Odds ratios were adjusted for potential confounders:
  - Age, gender, education level, overweight status, medical history of diabetes, arterial hypertension or dyslipidemia (high cholesterol)

D. D. Jovanović et al. Arsenic in drinking water and acute coronary syndrome in Zrenjanin municipality, Serbia. *Environmental Research*, Volume 117 (2012) 75-82.

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Health consequences: acute coronary syndrome in Zrenjanin
Posledice na zdravlje: akutni koronarni sindrom u Zrenjaninu

- Population exposed to arsenic 3 times more likely to suffer from ACS: a statistically significant difference
- Stanovništvo izloženo arsenu ima 3 puta veću šansu da boluje od ACS: statistički značajna razlika

- Peripheral arterial disease (significantly increases risk of cardiovascular events) almost 4 times more likely in population exposed to arsenic
- Periferna arterijska bolest (značajno povećava rizik od kardiovaskularnih obolenja), skoro 4 puta više za populaciju izloženu arsenu

- Slight increase in mortality observed, not statistically significant
- Blago povećanje mortaliteta, statistički nije značajno

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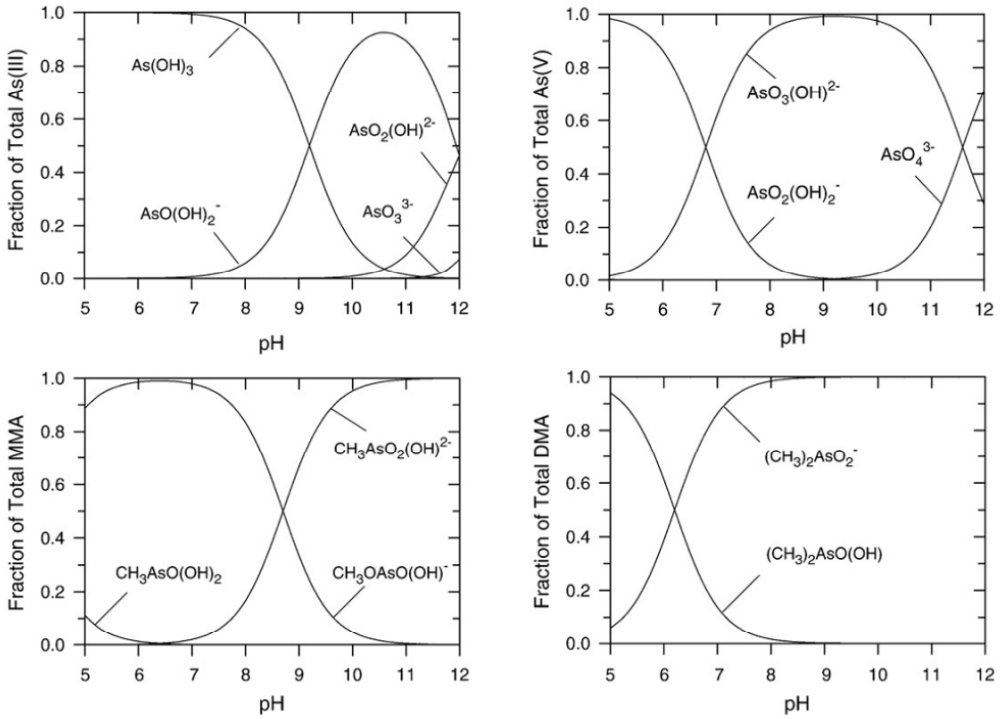
How does arsenic speciation effect the choice of removal technology?
Kako oblici arsena utiču na izbor tehnologije za njegovo uklanjanje?

- The different arsenic species have different physical-chemical characteristics, therefore technologies may be better at removing some arsenic species than others.
- Različiti oblici arsena imaju različite fizičko-hemijske karakteristike, stoga su tehnologije efikasnije za uklanjanje pojeinih oblika arsena.



- For example:
  The distribution of As(III), As(V), MMA and DMA hydroxide species as function of pH at 25°C.

- Na primer:
  Distribucija As(III), As(V), MMA , i DMA hidroksid kao funkcije pH na 25°C.

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How does arsenic speciation effect choice of removal technology?
Kako oblici arsena utiču na izbor tehnologije za uklanjanje?

- **Technologies equally effective at removing $iAs^{(III)}$ and $iAs^{(V)}$:**
  - Adsorption on zerovalent iron
  - Adsorption on iron oxide coated sand
  - Fixed-bed upflow bioreactors (uses biological oxidation of Fe and Mn ions which in turn adsorb As, less effective at higher (>80 µg/l) As concentrations
- **Technologies which are much more effective at removing $iAs^{(V)}$ than $iAs^{(III)}$**
  - Coagulation/filtration with ferric salts or alum
  - Adsorption on activated alumina
  - Some other adsorption technologies
- **Preoxidation step to turn all $iAs^{(III)}$ into $iAs^{(V)}$**
  - chlorine, permanganate and ozone all very effective

- **Podjednako efikasne tehnologije za uklanjanje $iAs^{(III)}$ i $iAs^{(V)}$**
  - Adsorpcija na nultovalentnom gvožđu
  - Adsorpcija na gvožđevitom pesku
  - Fiksirani bioreaktori (primenjuje bilošku oksidaciju Fe i Mn jona koji adsorbuju As, manja efikasnost pri većim (>80 µg/l) koncentracijama As
- **Tehnolgije koje su efikasnije za uklanjanje $iAs^{(V)}$ nego $iAs^{(III)}$**
  - Koagulacija/filtracija sa solima gvožđa i aluminijuma
  - Adsorpcija na aktiviranom aluminijum-oksidu
  - Druge tehnologije za adsorpciju
- **Predoksidacijioni korak za prevođenje $iAs^{(III)}$ u $iAs^{(V)}$**
  - Hlor, permanganat i ozon, svi su veoma efikasni

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Other factors influencing choice of As removal technology
Drugi faktori koji utiču na izbor tehnologije za uklanjanja As

- What other contaminants need to be removed?
- How much water do we need to treat?
- Which As species are present, do we need an oxidation step?
- Degree of As removal – are the As levels low enough after treatment?
- What sort of wastes are generated, how can they be disposed of?

- Koje još kontaminante u vodi treba ukloniti?
- Koju količinu vode treba tretirati?
- Koji oblici As su prisutni, da li je potreban oksidacioni korak?
- Stepen uklanjnaja As – da li je sadržaj As nakon tretmana dovoljno nizak?
- Koja se vrsta otpada generiše, na koji način se može odlagati?

- Koliki su investicioni troškovi?
- Koliki su troškovi održavanja?
- Koliko je proces složen, da li se može automatizovati, koliko kvalifikovanih radnika je potrebno?
- Da li se lako može implementirati na već postojeće postrojenje za tretman vode?

- How much does it cost to build?
- How much does it cost to maintain?
- How complicated is the process, can it be automated, how qualified do the maintainers need to be?
- Can it be added easily to existing treatment plants_

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Sledeći korak?

- Da bi se sagledali rizici za ljudsku populaciju usled različitih oblika arsena prisutnih u vodi, neophodni su dodatni podaci:
- Nivo izloženosti arsenu
- Detaljne epidemiološke studije u regionu
  - Literatura vezana za ovu regiju je vrlo ograničena
- Bez ovih dodatnih podataka, teško je odrediti prioritete i definisati koji se problemi moraju hitno rešavati

Going forwards

- To make informed decisions about the control of risks to human health which stem from arsenic species present in drinking water, more data is required:
- Actual As exposure levels in the effected populations.
- In depth epidemiological studies in the region
  - Literature for this region is very limited
- Without this additional data, hard to prioritise and define which are the most urgent problems we need to solve

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Going forwards

In the absence of such data, two points to consider in the meantime:

- Regional groundwater sources have relatively poor chemical quality, but in many cases, they are better than the surface water sources: its better to get skin cancer in 30 years than die from an infection next week.
- The presence of As in drinking water **will** have an economic cost. The choice is whether to pay for it early via the provision of clean drinking water, or to pay for it later, in increased healthcare requirements for a large number of unnecessarily sick people.

Sledeći korak?

U odsustvu navedenih podataka, dve stvari se razmatraju:

- Regionalna izvorišta podzemne vode su relativna lošeg hemijskog kvaliteta, ali u velikom broju slučajeva, bolja su od površinske vode: bolje je dobiti rak kože za 30 godina nego umreti od infekcije sledeće nedelje.
- Prisustvo arsena u vodi za piće podrazumeva ekonomske troškove. Izbor je ili da se plati i obezbedi zdravstveno bezbedna voda za piće, ili da se cena plaća kasnije, u vidu zdravstvenih troškova velikog broja bespotrebno bolesnih ljudi.

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Going forwards

In the absence of such data, two points to consider in the meantime:

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Sledeći korak?

U odsustvu navedenih podataka, dve stvari se razmatraju:

- Problem prisustva arsenu u vodi zaista postoji i dosta je ozbiljan. Ovaj problem potrebno je rešiti.
- Prisustvo arsena u vodi za piće podrazumeva ekonomske troškove. Izbor je ili da se plati i obezbedi zdravstveno bezbedna voda za piće, ili da se cena plaća kasnije, u vidu zdravstvenih troškova velikog broja bespotrebno bolesnih ljudi.

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Hvala na pažnji!

Köszönöm a figyelmet!

Thanks for listening!

Dobri susedi
zajedno stvaraju
budućnost

Novi Sad, 04.09.2012.

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