Supporting information cover page

# Adsorption of arsenic on polyaluminum granulate

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## Tables

**Table S1**: Chemical composition of synthetic water (SW) and groundwater from the Pannonian Basin (Hungary and Romania) with geogenic As contamination.

| | SW | | Pannonian Basin groundwater[a] | |
|---|---|---|---|---|
| pH | 7.5 ± 0.1 | | 8.2 ± 0.2 | |
| Alkalinity (mM) | 8.0 | | 9.2 ± 5.3 | |
| | *mg/L* | *mmol/L* | *Average mg/L* | *Range mg/L* |
| Chloride | 21.3 | 0.6 | 21 | 2-159 |
| Sodium | 184.0 | 8.0 | 198 | 92-299 |
| Calcium | 40.0 | 1.0 | 16 | 4.2-33.7 |
| Magnesium | 15.0 | 0.6 | 4.6 | 0.7-13.8 |
| Phosphate | - | - | 0.513 | 0.1-1.4 |
| Iron | - | - | 0.258 | 0.01-0.78 |
| As(tot) | - | - | 0.123 | 0.02-0.21 |

[a] Data from Rowland et al. (2011), group 1 general groundwater

**Table S2:** Physicochemical properties of polyaluminum granulate (PAG)

| | |
|---|---|
| Size | 1-3 mm |
| Skeleton density | 1.93 $g/cm^3$ |
| BET surface area | 35 ± 1 $m^2/g$ |
| Total Porosity* | 219 $mm^3/g$ |
| Macropores (radius >50 nm) | 79.4 $mm^3/g$ |
| Mesopores (radius 2-50 nm) | 140 $mm^3/g$ |

* Approximately 50% of the pores are in the 25-75 nm radius range (comp. Figure 1).

**Table S3:** Linkage of As(V) removal (in %) to Ca:As ratio based on initial aqueous concentrations. $[Ca]_{initial}$ = 1 mmol/L. Conditions for Ca:As ratio = 1 is in bold. Shaded areas depict Ca:As ratio < 1, and low As(V) removal efficiencies in synthetic water (SW), whereas removal efficiencies in DW are similar for all As(V) concentrations.

| $[As(V)]_{initial}$ | | Ca:As ratio | $[As]_{removed}$ (in %) | |
|---|---|---|---|---|
| mg/L | mmol/L | | in SW | in DW |
| 0.02 | 0.000 | 3256.52 | 94.52 | 93.12 |
| 0.22 | 0.003 | 335.87 | 86.02 | 94.03 |
| 2.25 | 0.030 | 33.29 | 96.26 | 91.73 |
| 10.00 | 0.134 | 7.49 | 98.82 | 98.36 |
| 22.30 | 0.298 | 3.36 | 99.82 | 95.45 |
| **71.00** | **0.948** | **1.05** | **99.62** | **n.a.** |
| 200.00 | 2.670 | 0.37 | 69.56 | 99.33 |
| 240.00 | 3.204 | 0.31 | 45.42 | 97.91 |
| 289.00 | 3.858 | 0.26 | 53.29 | 97.24 |

**Figures**

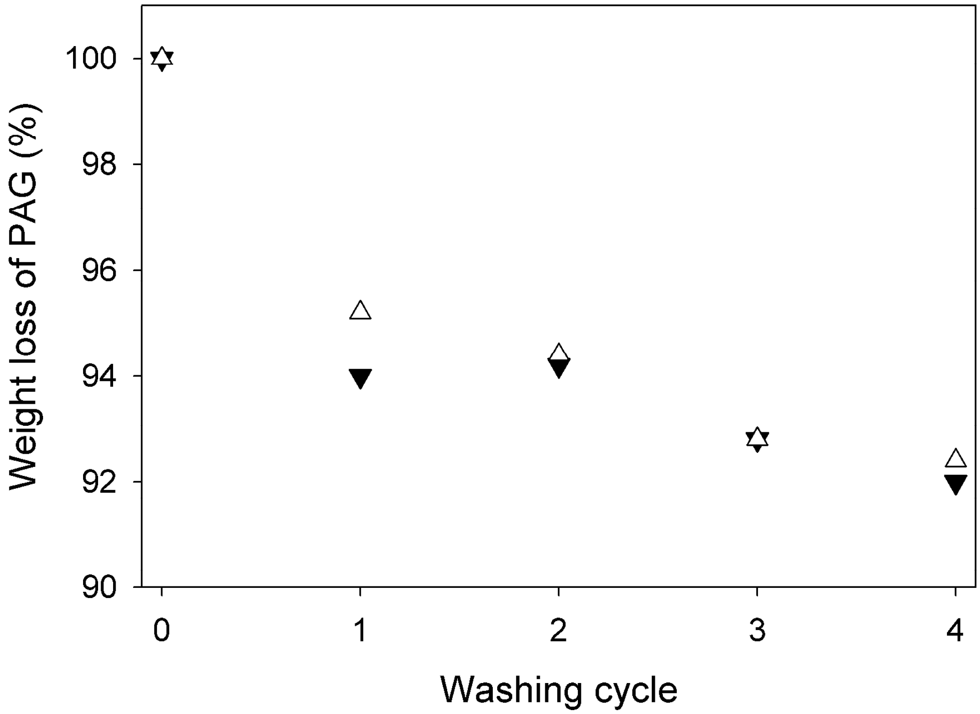


**Figure S1:** Mechanical stability of PAG during washing procedures. White and black triangles represent duplicate measurements. Washing cycle 1: rinsed 4 times with deionized water, washing cycle 2: shaken for 1 hour at 150 rpm, cycle 3: shaken for 16 hours at 150 rpm, cycle 4: shaken for 72 hours at 150 rpm; After each procedure, supernatant water was decanted, and granulates were dried for 6 hrs at 40 °C.

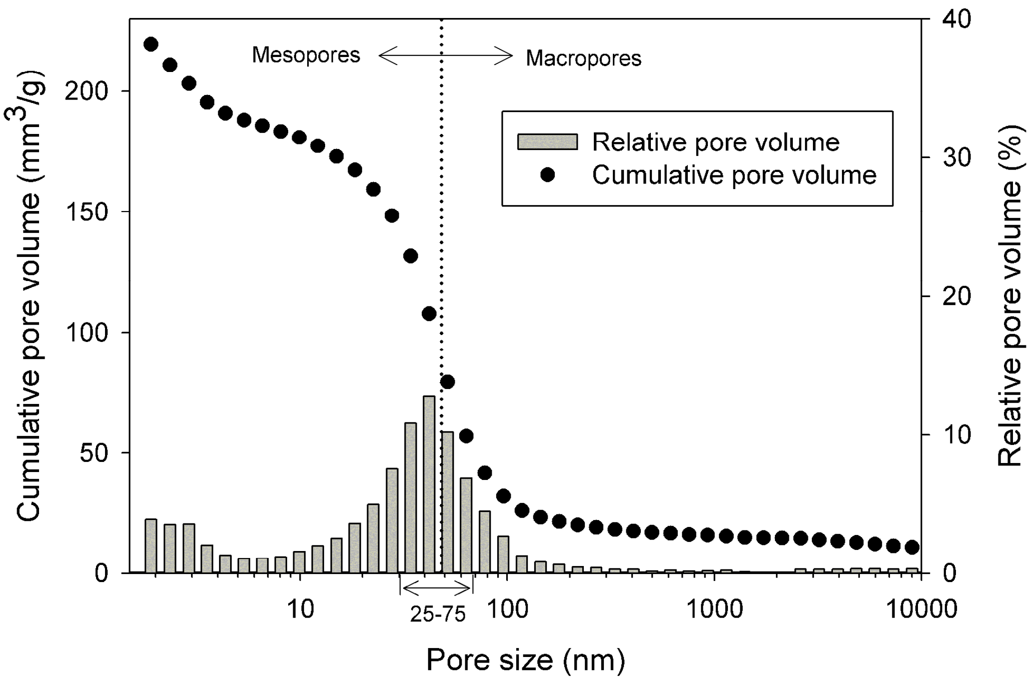


**Figure S2**: Pore size distribution in polyaluminum granulate.

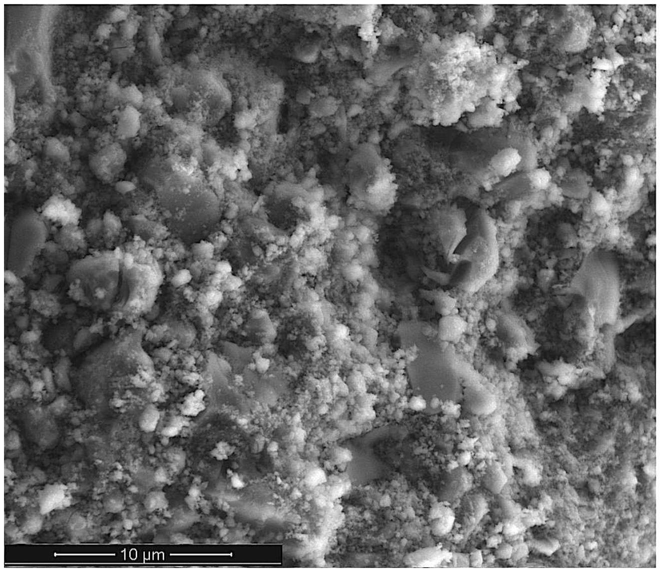

**Figure S3:** Scanning electron microscope image of the polyaluminum granulate surface.

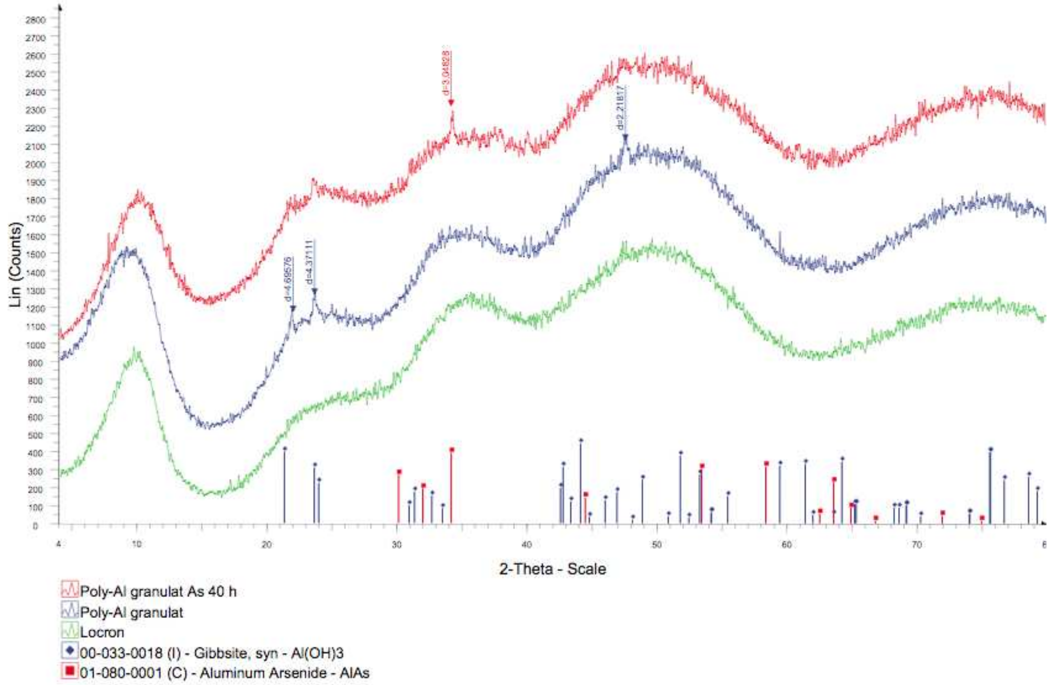


**Figure S4:** XRD pattern of dry polyaluminum granulate after production (in blue) and polyaluminum granulate that was exposed to 3.4 mM As(III) in deionized water for 40 hours and an As(III) uptake of 4.0 mmol $g^{-1}$ PAG (in red) compared to its base material, the polyaluminum chloride Locron and the XRD patterns of the Al mineral gibbsite. The measured XRD patterns exhibit the same very broad lines for both polyaluminum granulate samples as the Locron powder, indicating that the base material consists of almost X-ray amorphous material and did not change its crystallographic composition. Additionally, three small peaks in the polyaluminum granulate were identified, which might be an indication that probably some gibbsite was formed during the production process of polyaluminum granulate. The peaks were too weak, however, to clearly identify a crystalline gibbsite phase.

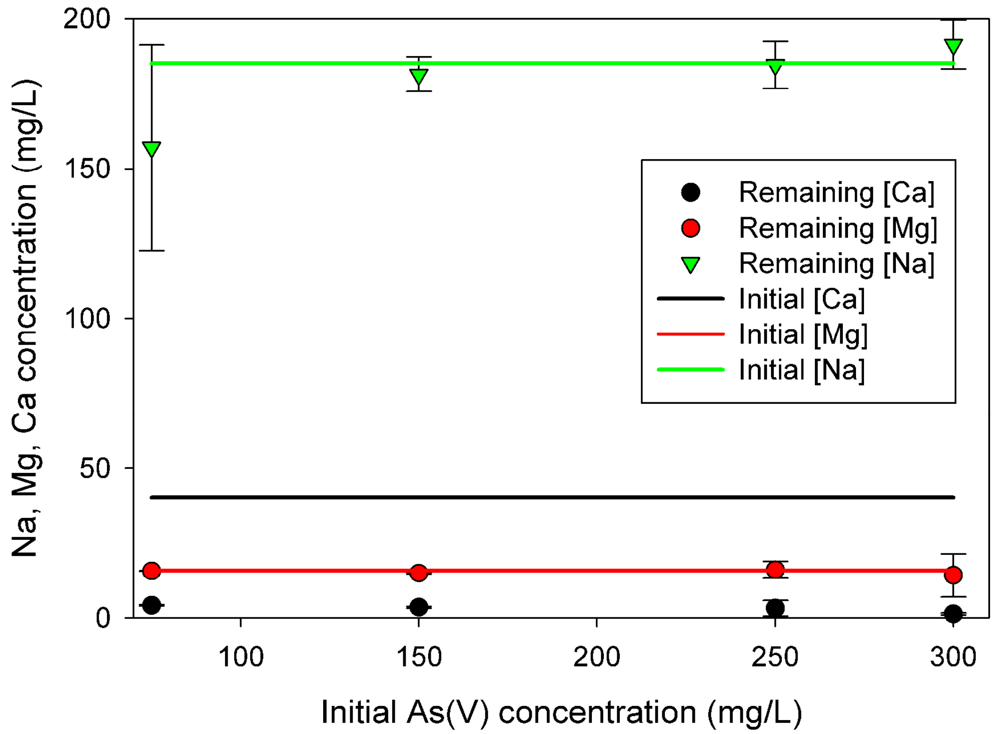


**Figure S5:** Initial and remaining concentrations of calcium (black), magnesium (red) and sodium (green) remaining in supernatant water after 20 hours contact time with PAG (results from adsorption batch experiments). Calcium was removed considerably by 90 - 97%.

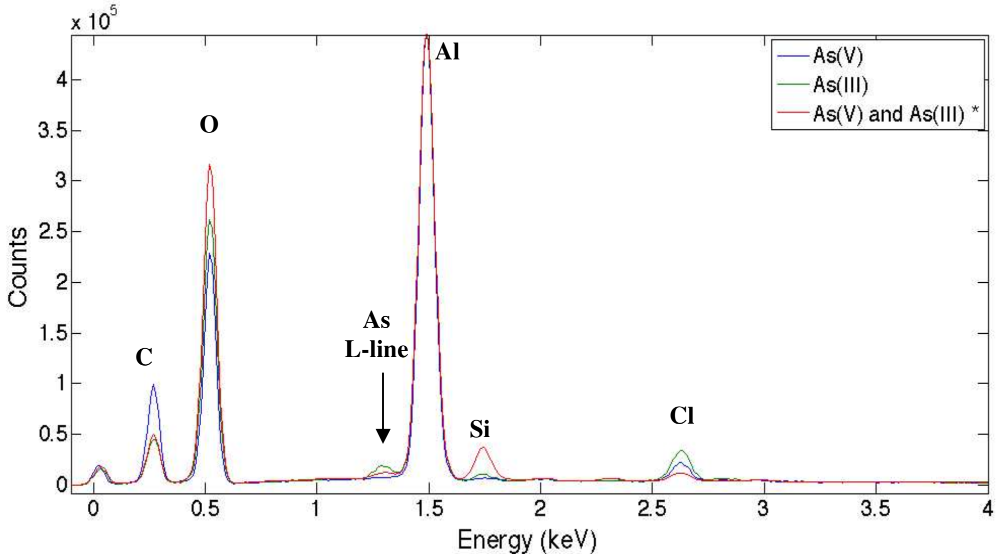


**Figure S6:** Al-normalized spectra of the centre area of three batches of PAGs loaded with As(V), As(III), and both, As(V) and As(III) during 20 hours.* As(V) was adsorbed for 20 hours, and was followed by 20 hours As(III) adsorption.

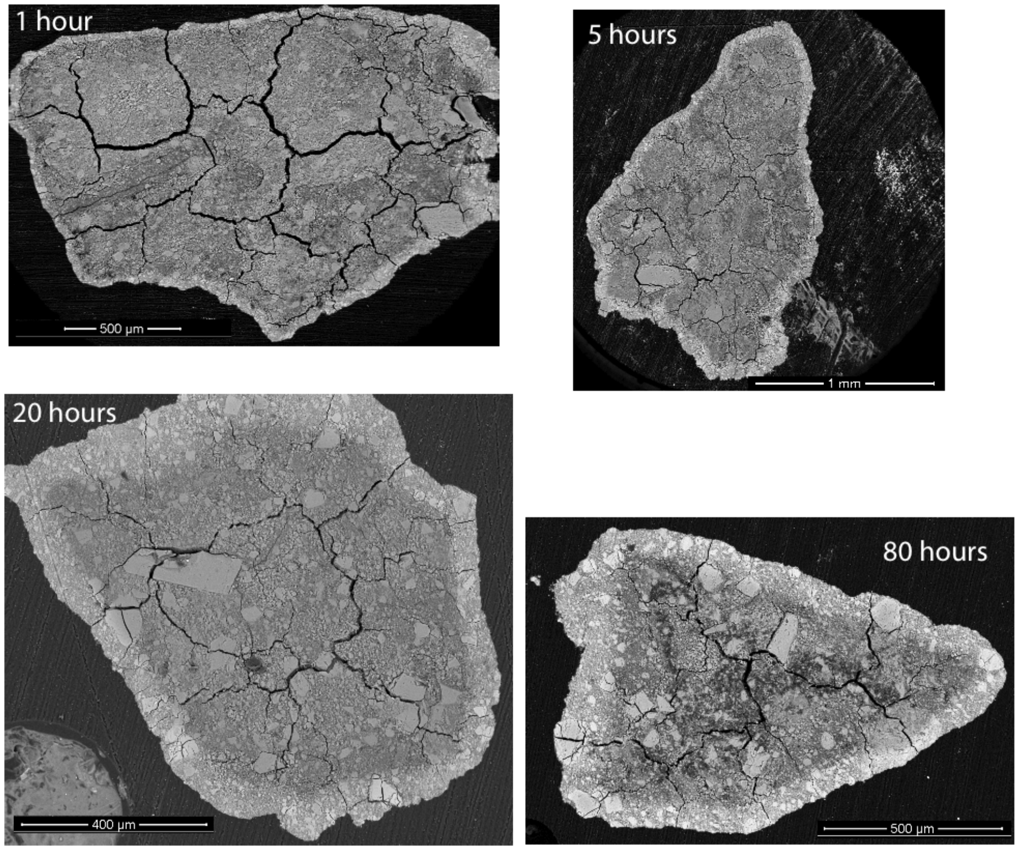


**Figure S7:** Backscattered electron images of polyaluminum granulate, which was in contact with 3.8 mmol As/L for 1, 5, 20, and 80 hours. Polished thin sections show the regularity of the As-rich rims allowing consistent rim-width measurements as presented in Table 5. The occasional broadening of the As-rich layer can be explained by a cutting effect due to non-perpendicular polishing of the grain's surface.