# Anderson Acceleration. Applications to equilibrium chemistry

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# Plan

1. Study on a 1D case of reactive transport
   - Mathematical formulation of the chemical system
   - Positive Continued Fractions (PCF)
   - Anderson acceleration (AA(m))
   - Chemical test
   - Results

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# Chemical reactions at thermodynamic equilibrium

## Reactions at equilibrium

$$\sum_{j=1}^{n_p} \mu_{ij} \chi_j \rightleftharpoons \mathcal{C}_i \qquad i = 1, ..., n_s$$

$\chi_j$ : Concentration of components (of cardinality $n_p$)
$\mathcal{C}_i$ : Concentration of secondary species (of cardinality $n_s$)
$\mu$ : The stoichiometric matrix

# Mathematical model

## Law of mass action

$$\mathcal{C}_i = K_i \prod_{k=1}^{n_p} \chi_k^{\mu_{ik}}$$

$K$ : Equilibrium constant

## Mass conservation

$$T_j = \sum_{i=1}^{n_s} \mu_{ij} \mathcal{C}_i$$

# Mathematical model

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$K$ : Equilibrium constant

## Mass conservation

$$T_j = \sum_{i=1}^{n_s} \mu_{ij} \mathcal{C}_i$$

## Nonlinear system of equations

We seek $\chi \in \mathbb{R}^{n_p}$ such that

$$T_j = \sum_{i=1}^{n_s} \mu_{ij} (K_i \prod_{k=1}^{n_p} \chi_k^{\mu_{ik}})$$

pour tout $j \in \{1, \ldots, n_p\}$

# Matrix mathematical model

## Nonlinear problem

We seek $\chi \in \mathbb{R}^{n_p}$ such that

$$F(\chi) = 0,$$

with $F : \mathbb{R}^{n_p} \to \mathbb{R}^{n_p}$ (Nonlinear)

## Anderson Acceleration AA

Anderson acceleration is a method of accelerating the convergence rate of iterations of the fixed point ; This technique is used to find the solution of fixed point equations.

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# Positive Continued Fractions (PCF) method

**Problem reformulation :**

$$T_j = \sum_{i=1}^{n_s} \mu_{ij}\,\mathcal{C}_i = \sum_{\mu_{ij}>0} \mu_{i,j}\,\mathcal{C}_i + \sum_{\mu_{ij}<0} \mu_{i,j}\,\mathcal{C}_i$$

Sum of reactants : $SR_j = \begin{cases} \sum_{\mu_{i,j}>0} \mu_{i,j}.\mathcal{C}_i & T_j \geq 0 \\ |T_j| + \sum_{\mu_{i,j}>0} \mu_{i,j}.\mathcal{C}_i & T_j < 0 \end{cases}$

Sum of products : $SP_j = \begin{cases} T_j + \sum_{\mu_{i,j}<0} |\mu_{i,j}|.\mathcal{C}_i & T_j \geq 0 \\ \sum_{\mu_{i,j}<0} |\mu_{i,j}|.\mathcal{C}_i & T_j < 0 \end{cases}$

$$Equilibrium \Leftrightarrow SR_j(\chi) = SP_j(\chi)$$

# Fixed-point problem

## PCF (Fixed-Point Method)

We construct a sequence $\chi_j^n$ such that

$$\chi_j^{n+1} = \chi_j^n \left( \frac{SP_{j,n}(\chi^n)}{SR_{j,n}(\chi^n)} \right)$$

# Fixed-point problem

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$$\chi_j^{n+1} = \chi_j^n \left( \frac{SP_{j,n}(\chi^n)}{SR_{j,n}(\chi^n)} \right)$$

## Change of variable :

$$\omega = log_{10}(\chi)$$

# Fixed-point problem

## PCF (Fixed-Point Method)

We construct a sequence $\chi_j^n$ such that

$$\chi_j^{n+1} = \chi_j^n \left( \frac{SP_{j,n}(\chi^n)}{SR_{j,n}(\chi^n)} \right)$$

## Fixed-point problem

We are looking for $\xi \in \text{R}^{n_p}$ such that

$$\omega_{n+1} = G(\omega_n)$$

avec

$$\begin{aligned} G : \mathbb{R}^{n_p} &\longrightarrow \mathbb{R}^{n_p} \\ \xi &\longmapsto G(\omega) = \omega + [log_{10}(SP(\omega)) - log_{10}(SR(\omega)] \end{aligned}$$

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# Anderson acceleration (AA(m))

## algorithm (AA(m)) :

**Input :** $\omega_0, G, m \geq 1$
$\omega_1 = G(\omega_0)$;
$\mathrm{f}_0 = G(\omega_0) - \omega_0$
**for k = 1,...**
$m_k = min(m, k)$
$f_k = G(\omega_k) - \omega_k$
$F_k = (f_{k-m_k}, ..., f_k)$
Determine : $min_\alpha ||F_k \alpha||_2$, avec
$\sum_{i=0}^{m_k} \alpha_i = 1$
$\omega_{k+1} = \sum_{i=0}^{m_k} \alpha_i{}^{(k)} G(\omega_k - m_k + i)$
**end**

# Anderson acceleration (AA(m))

**algorithm (AA(m)) :**

**Input :** $\omega_0, G, m \geq 1$
$\omega_1 = G(\omega_0)$;
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$F_k = (f_{k-m_k}, ..., f_k)$
Determine : $min_\alpha ||F_k \alpha||_2$, avec $\sum_{i=0}^{m_k} \alpha_i = 1$
$\omega_{k+1} = \sum_{i=0}^{m_k} \alpha_i{}^{(k)} G(\omega_k - m_k + i)$
**end**

**Algo :**

**cas** $m = 0$ **:**
$\omega_1 = G(\omega_0)$
$f_0 = G(\omega_0) - \omega_0$
**k = 1**
$m_1 = min(0, 1) = 0$
$f_1 = G(\omega_1) - \omega_1$
$F_1 = f_1$
$min_\alpha ||F_1 \alpha||_2 = min||f_1 \alpha_0||_2$,
avec $\alpha_0 = 1$
$\omega_2 = \alpha_0 G(\omega_1) = G(\omega_1)$

# Anderson acceleration (AA(m))

## algorithm (AA(m)) :

**Input :** $\omega_0, G, m \geq 1$

$\omega_1 = G(\omega_0);$

$f_0 = G(\omega_0) - \omega_0$

**for k = 1,...**

$m_k = min(m, k)$

$f_k = G(\omega_k) - \omega_k$

$F_k = (f_{k-m_k}, ..., f_k)$

Determine : $min_\alpha ||F_k \alpha||_2$, avec $\sum_{i=0}^{m_k} \alpha_i = 1$

$\omega_{k+1} = \sum_{i=0}^{m_k} \alpha_i{}^{(k)} G(\omega_k - m_k + i)$

**end**

## Algo :

**cas** $m = 1$ **:**

$\omega_1 = G(\omega_0)$

$f_0 = G(\omega_0) - \omega_0$

$\mathbf{k = 1}$

$m_1 = min(1, 1) = 1$

$f_1 = G(\omega_1) - \omega_1$

$F_1 = (f_0, f_1)$

$min_\alpha ||F_1 \alpha||_2 = min||f_0 \alpha_0 + f_1 \alpha_1||_2$, avec

$\alpha_0 + \alpha_1 = 1$

$\omega_2 = \alpha_0 G(\omega_0) + \alpha_1 G(\omega_1)$

# Anderson acceleration

**Problème d'optimisation sous contrainte :**

$$min_{\alpha}||F_k\alpha||_2 \quad avec \sum_{i=0}^{m_k}\alpha_i = 1$$

**Changement de variable :**

$$\alpha_0 = \gamma_0 \quad \alpha_j = \gamma_j - \gamma_{j-1} \quad \alpha_{mk} = 1 - \gamma_{mk-1}$$

$$F_k = \left(f_{k-mk}, f_{k-mk+1}, ..., f_k\right) ; \alpha = \left(\alpha_0, \alpha_1, ..., \alpha_{mk}\right)$$

$$
\begin{aligned}
{\color{red}F_k\alpha} &= f_{k-m_k}\alpha_0 + f_{k-mk+1}\alpha_1 + \dots + f_k\alpha_{mk} \\
&= f_{k-m_k}\gamma_0 + f_{k-mk+1}(\gamma_1-\gamma_0) + \dots + f_k(1-\gamma_{mk-1}) \\
&= f_{k-mk}\gamma_0 + f_{k-mk+1}\gamma_1 - f_{k-mk+1}\gamma_0 + \dots + f_k - f_k\gamma_{mk-1} \\
&= f_k - \gamma_0(f_{k-mk} - f_{k-mk-1}) - \dots - \gamma_{mk-1}(f_{mk} - f_{mk-1}) \\
&= f_k - \sum_{i=k-mk}^{k-1} \gamma_i(f_{i+1} - f_i) \\
&= f_k - \sum_{i=k-mk}^{k-1} \gamma_i \Delta f_i
\end{aligned}
$$

$$
\begin{aligned}
\omega_{k+1} &= \sum_{i=0}^{m_k} \alpha_i G(\omega_k - m_k + i) \\
&= \alpha_0 G(\omega_k - m_k) + \dots + \alpha_{mk} G(\omega_k) \\
&= G(\omega_k) - \sum_{i=k-mk}^{k-1} \gamma_i \Delta G_i
\end{aligned}
$$

## algorithm (AA(m)) :

**Input :** $\omega_0, G, m \geq 1$
$\omega_1 = G(\omega_0);$
$f_0 = G(\omega_0) - \omega_0$
**for k = 1,...**
$m_k = min(m, k)$
$f_k = G(\omega_k) - \omega_k$
$F_k = (f_{k-m_k}, ..., f_k)$
Determine$\alpha$ : $min_\alpha ||F_k \alpha||_2$, avec $\sum_{i=0}^{m_k} \alpha_i = 1$
$\omega_{k+1} = \sum_{i=0}^{m_k} \alpha_i^{(k)} G(\omega_k - m_k + i)$
**end**

## New Algorithm AA(m) :

**Input :** $\omega_0, G, m \geq 1$
$\omega_1 = G(\omega_0);$
$f_0 = G(\omega_0) - \omega_0$
**for k = 1,...**
$m_k = min(m, k)$
$f_k = G(\omega_k) - \omega_k$
$\mathcal{F}_k = (\Delta f_{k-m_k}, ..., \Delta f_{k-1})$
$\mathcal{G}_k = (\Delta G_{k-mk}, ..., \Delta G_{k-1})$
Determine $\gamma$ : $min_\gamma ||f_k - \mathcal{F}_k \gamma||_2$
$\omega_{k+1} = G(\omega_k) - \sum_{i=k-mk}^{k-1} \gamma_i \mathcal{G}_k$
**end**

### New Algorithm AA(m) :

**Input :** $\omega_0, G, m \geq 1$
$\omega_1 = G(\omega_0)$;
$f_0 = G(\omega_0) - \omega_0$
**for k = 1,...**
$m_k = min(m, k)$
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$\mathcal{F}_k = (\Delta f_{k-m_k}, ..., \Delta f_{k-1})$
$\mathcal{G}_k = (\Delta G_{k-mk}, ..., \Delta G_{k-1})$
Determine $\gamma$ : $min_\gamma ||f_k - \mathcal{F}_k \gamma||_2$
$\omega_{k+1} = G(\omega_k) - \sum_{i=k-mk}^{k-1} \gamma_i \mathcal{G}_k$
**end**

### Algo :

**cas** $m = 1$ **:**
$\omega_1 = G(\omega_0)$
$f_0 = G(\omega_0) - \omega_0$
$\mathbf{k = 1}$
$m_1 = min(1, 1) = 1$
$f_1 = G(\omega_1) - \omega_1$
$\mathcal{F}_1 = (\Delta f_0) = (f_1 - f_0) =$
$((G(\omega_1) - \omega_1) - (G(\omega_0) - \omega_0))$
$\mathcal{G}_1 = (\Delta G_0) = (G(\omega_1) - G(\omega_0))$
Determine $\gamma$ : $min_\gamma ||f_1 - \mathcal{F}_1 \gamma||_2$
$\omega_2 = G(\omega_1) - \gamma_0 \mathcal{G}_1 =$
$G(\omega_1) - \gamma_0(G(\omega_1) - G(\omega_0)) =$
$(1 - \gamma_0) G(\omega_1) + \gamma_0 G(\omega_0)$

**Algo :**

**cas** $m = 1$ **:**
$\omega_1 = G(\omega_0)$
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**k = 1**
$m_1 = min(1, 1) = 1$
$f_1 = G(\omega_1) - \omega_1$
$F_1 = (f_0, f_1)$
$min_\alpha ||F_1\alpha||_2$, avec $\alpha_0 + \alpha_1 = 1$
$\omega_2 = \alpha_0 G(\omega_0) + \alpha_1 G(\omega_1)$

**New Algo :**

**cas** $m = 1$ **:**
$\omega_1 = G(\omega_0)$
$f_0 = G(\omega_0) - \omega_0$
**k = 1**
$m_1 = min(1, 1) = 1$
$f_1 = G(\omega_1) - \omega_1$
$\mathcal{F}_1 = (\Delta f_0) = (f_1 - f_0) =$
$((G(\omega_1) - \omega_1) - (G(\omega_0) - \omega_0))$
$\mathcal{G}_1 = (\Delta G_0) = (G(\omega_1) - G(\omega_0))$
Determine $\gamma$ : $min_\gamma ||f_1 - \mathcal{F}_1\gamma||_2$
$\omega_2 = G(\omega_1) - \gamma_0 \mathcal{G}_1 =$
$G(\omega_1) - \gamma_0(G(\omega_1) - G(\omega_0)) =$
$(1 - \gamma_0)G(\omega_1) + \gamma_0 G(\omega_0)$

## Problème d'optimisation sans contrainte :

$$min_{\gamma}||f_k - \mathcal{F}_k\gamma||_2$$

### QR Factorisation

- $\mathcal{F}_k = Q_k R_k$
- $R_k\gamma = Q_k^T f_k$
- $\gamma = R_k \backslash Q_k^T f_k$

### QR Factorisation

$\mathcal{F}_k \in \mathbb{R}^{n\times mk}$ $R_k \in \mathbb{R}^{mk\times mk}$
$Q_k \in \mathbb{R}^{n\times mk}$
Q orthogonale et R triangulaire

# Problème d'optimisation sans contrainte :

$$min_{\gamma}||f_k - \mathcal{F}_k\gamma||_2$$

## QR Factorisation

- $\mathcal{F}_k = Q_k R_k$
- $R_k\gamma = Q_k^T f_k$
- $\gamma = R_k \backslash Q_k^T f_k$

## QR Factorisation

$\mathcal{F}_k \in \mathbb{R}^{n\times mk}$ $R_k \in \mathbb{R}^{mk\times mk}$
$Q_k \in \mathbb{R}^{n\times mk}$
Q orthogonale et R triangulaire

## Updating the QR factors

- When $k = 1$, $m_1 = 1$, $\mathcal{F}_1 = QR = \Delta f_0$ avec $Q = \frac{\Delta f_0}{||\Delta f_0||_2}$ et $R = ||\Delta f_0||_2$
- When k>1, we have $\mathcal{F}_{k-1} = QR$, where $\mathcal{F}_{k-1}$ is $n \times m_{k-1}$, and update as follows :
  - If $m_{k-1} = m$, then delete the left column of $\mathcal{F}_{k1}$, updating Q and R so that $\mathcal{F}_{k-1} \leftarrow \mathcal{F}_{k-1}(:, 2 : m) = QR$
  - Add a column on the right, updating Q and R so that $\mathcal{F}_k = [\mathcal{F}_{k-1}, \Delta f_{k-1}] = QR$

## Control of the conditioning of the matrix $\mathcal{F}_k$

The tolerance $tol > 0$ and $\mathcal{F}_k = Q_k R_k$

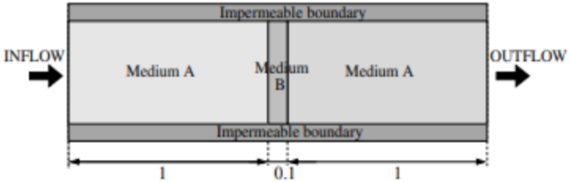
while $cond(R_k) > tol$

Remove the last column from the left of $\mathcal{F}_k$ and update it

QR factorisation : $\mathcal{F}_k = Q_k R_k$

End

## Benchmark Momas Easy




The domain is assumed to be initially at equilibrium :

- Equilibrium in A (more permeable, low porosity)
- Equilibrium in B (less permeable, high porosity)

Injection On the left side, followed by a leaching on the same side :

- Injection into A (on the left).
- Injection into B.
- Leaching.

[5] Carrayrou, J., Kern, M. and Knabner, P. Reactive transport benchmark of MoMaS. Computation Geosci., 14 (2010).

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# 1D Easy test case of the MoMas benchmark

Morel table

| Espèces | $X_1$ | $X_2$ | $X_3$ | $X_4$ | S | K |
|---|---|---|---|---|---|---|
| $C_1$ | 0 | -1 | 0 | 0 | 0 | $10^{-12}$ |
| $C_2$ | 0 | 1 | 1 | 0 | 0 | 1 |
| $C_3$ | 0 | -1 | 0 | 1 | 0 | 1 |
| $C_4$ | 0 | -4 | 1 | 3 | 0 | 0.1 |
| $C_5$ | 0 | 4 | 3 | 1 | 0 | $10^{35}$ |
| $CS_1$ | 0 | 3 | 1 | 0 | 1 | $10^{6}$ |
| $CS_2$ | 0 | -3 | 0 | 1 | 2 | $10^{-1}$ |
| Total concentration T | $T_1$ | $T_2$ | $T_3$ | $T_4$ | $T_s$ | |
| Initial conditions | | | | | | |
| Zone A | 0 | -2 | 0 | 2 | 1 | |
| Zone B | 0 | -2 | 0 | 2 | 10 | |
| Boundary conditions | | | | | | |
| Injection t $\leq$ 5000 | 0.3 | 0.3 | 0.3 | 0 | 0 | |
| Leaching t$\geq$ 5000 | 0 | -2 | 0 | 2 | 0 | |

$$C_1 = 10^{-12}.X_2^{-1}$$
$$T_3 = X_3 + C_2 + C_4 + 3.C_5 + CS_1 = 0$$

# 1D Easy test case of the MoMas benchmark

**Law of mass action**

$$C_1 = 10^{-12}.X_2^{-1}$$
$$C_2 = X_2.X_3$$
$$C_3 = X_2^{-1}.X_4$$
$$C_4 = 10^{-1}.X_2^{-4}.X_3.X_4^3$$
$$C_5 = 10^{35}.X_2^{4}.X_3^{3}.X_4$$
$$CS_1 = 10^{6}.X_2^{3}.X_3.S$$
$$CS_2 = 10^{-1}.X_2^{-3}.X_4.S^2$$

**Mass conservation**

$$T_1 = X_1 = 0$$
$$T_2 = X_2 - C_1 + C_2 - C_3 - 4.C_4 + 4.C_5 + 3.CS_1 - 3.CS_2 = -2$$
$$T_3 = X_3 + C_2 + C_4 + 3.C_5 + CS_1 = 0$$
$$T_4 = X_4 + C_3 + 3.C_4 + C_5 + CS_2 = 2$$
$$TS = S + CS_1 + 2.CS_2 = 1$$

**Nonlinear system of equation**

$$T_1 = X_1 = 0$$
$$T_2 = X_2 - 10^{-12}.X_2^{-1} + X_2.X_3 - X_2^{-1}.X_4 - 4.10^{-1}.X_2^{-4}.X_3.X_4^3 + 4.10^{35}.X_2^{4}.X_3^{3}.X_4 + 3.10^{6}.X_2^{3}.X_3.S - 3.10^{-1}.X_2^{-3}.X_4.S^2 = -2$$
$$T_3 = X_3 + X_2.X_2^{-1}.X_4 + 10^{-1}.X_2^{-4}.X_3.X_4^3 + 3.10^{35}.X_2^{4}.X_3^{3}.X_4 + 10^{6}.X_2^{3}.X_3.S = 0$$
$$T_4 = X_4 + X_2^{-1}.X_4 + 3.10^{-1}.X_2^{-4}.X_3.X_4^3 + 10^{35}.X_2^{4}.X_3^{3}.X_4 + 10^{-1}.X_2^{-3}.X_4.S^2 = 2$$
$$TS = S + 10^{6}.X_2^{3}.X_3.S + 2.10^{-1}.X_2^{-3}.X_4.S^2 = 1$$

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# Results

Donnée initiale : $x_0 = [0.3; 0.4; 10^{-11}; 0.21; 0.6]$

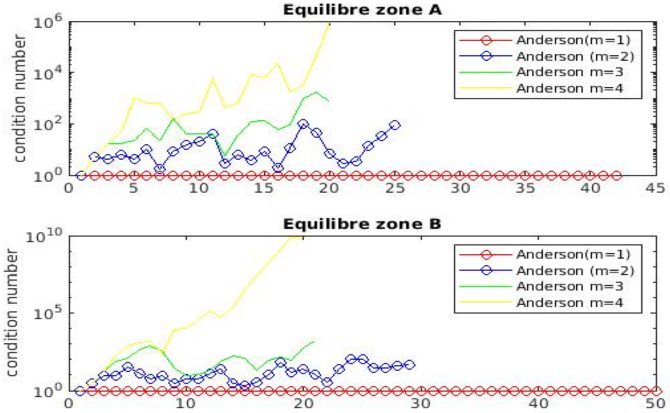
| | Picard Pcf | AA PCF |
|---|---|---|
| $TA = [10^{-30}; -2; 10^{-30}; 2; 1]$ | 0.0127 | 0.0047 |
| $TB = [10^{-30}; -2; 10^{-30}; 2; 1]$ | 0.0078 | 0.0028 |

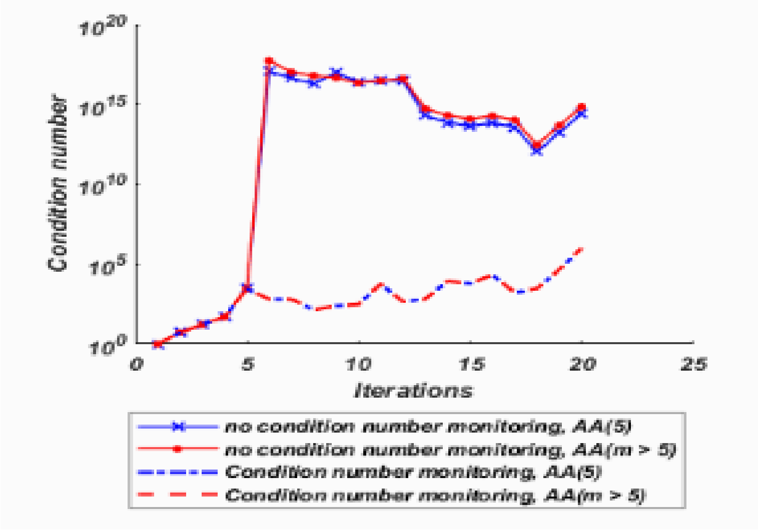
Table – Comparaison CPU AA PCF vs Picard PCF

| | Picard Pcf | AA PCF |
|---|---|---|
| $TA = [10^{-30}; -2; 10^{-30}; 2; 1]$ | 450 | 20 |
| $TB = [10^{-30}; -2; 10^{-30}; 2; 1]$ | 250 | 16 |

Table – Comparaison nb d'Itération AA PCF vs Picard PCF

# Results


Equilibre zone A
condition number
$10^0$
$10^2$
$10^4$
$10^6$
0 5 10 15 20 25 30 35 40 45
Anderson(m=1)
Anderson (m=2)
Anderson m=3
Anderson m=4
Equilibre zone B
condition number
$10^0$
$10^5$
$10^{10}$
0 10 20 30 40 50
Anderson(m=1)
Anderson (m=2)
Anderson m=3
Anderson m=4


Condition number
$10^{20}$
$10^{15}$
$10^{10}$
$10^{5}$
$10^{0}$
0
5
10
15
20
25
Iterations
no condition number monitoring, AA(5)
no condition number monitoring, AA(m > 5)
Condition number monitoring, AA(5)
Condition number monitoring, AA(m > 5)

Thank you !