Functional Predictor Variables for Leaching Potential of Arsenic and Selenium from Coal Fly Ash

Award #DE-FE0031748

FY22 FECM Spring R&D Project Review Meeting

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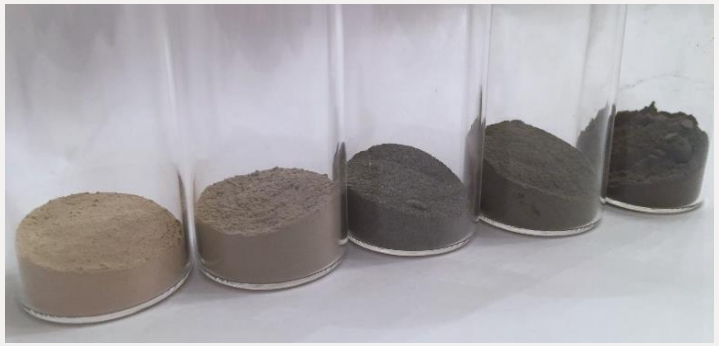
1. Motivation and Significance

What are the risks of coal ash disposal sites?

Dan River Steam Station, Feb. 2014

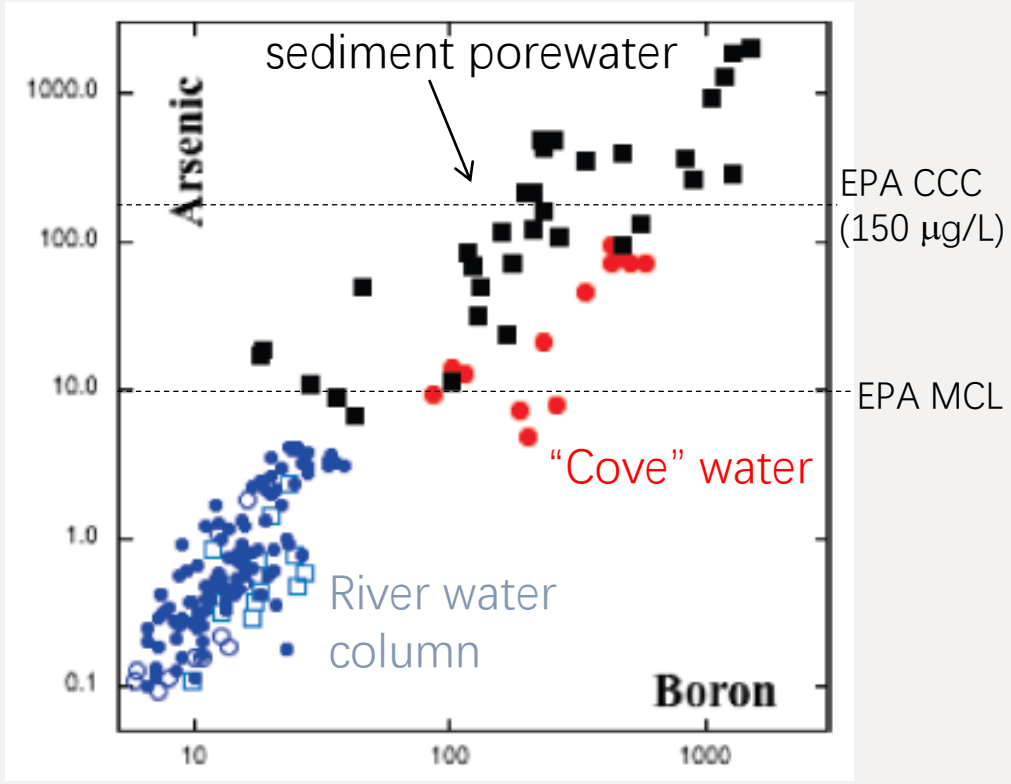


Sutton Plant at Sutton Lake, Sept. 2018

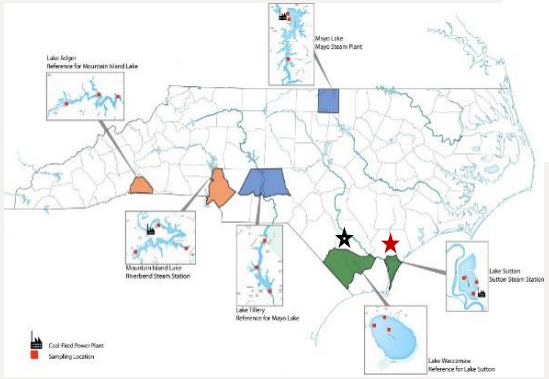
1. Motivation and Significance

Arsenic and selenium are coal ash constituents that can pose problems near disposal sites

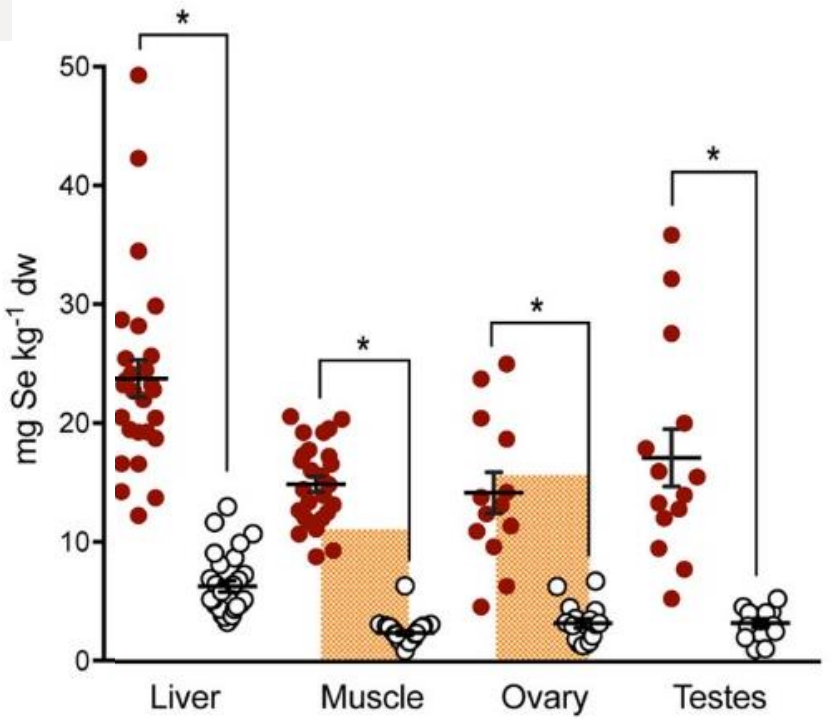
TVA-Kingston coal ash spill disaster:
River water and sediments



Ruhl *et al.*, ES&T 2010



Sutton Lake, NC (near coal ash pond) vs. **Lake Waccamaw, NC** (reference site)



Mean Loading to Sutton Lake (O4) = 0.85 kg Se day^{-1}

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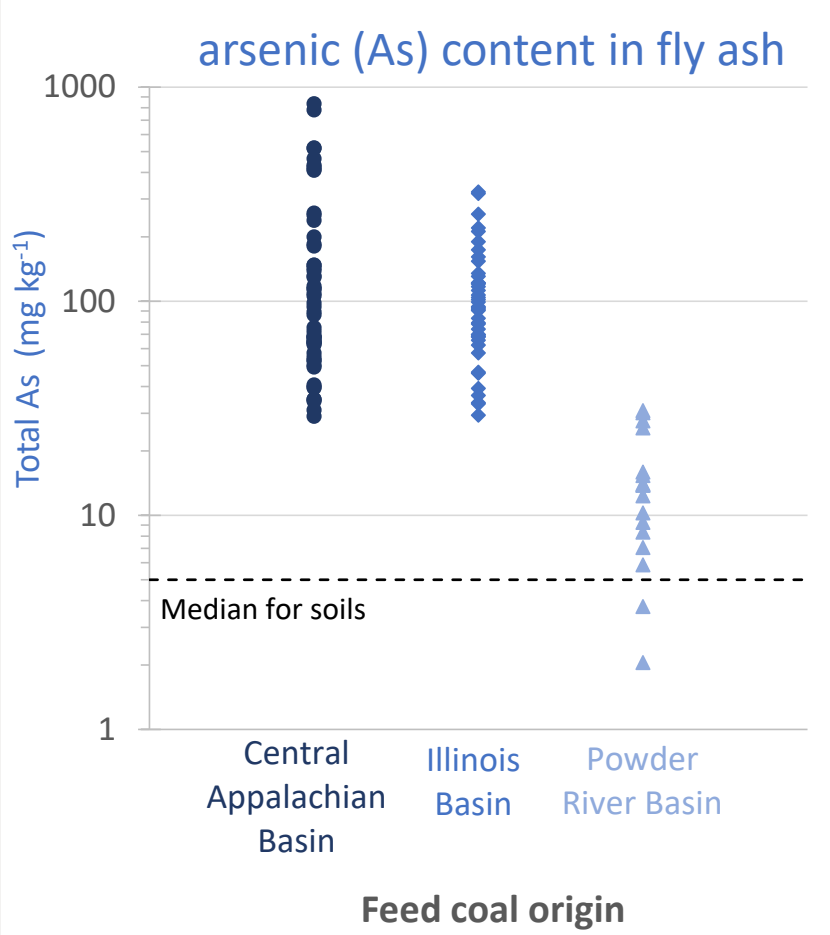
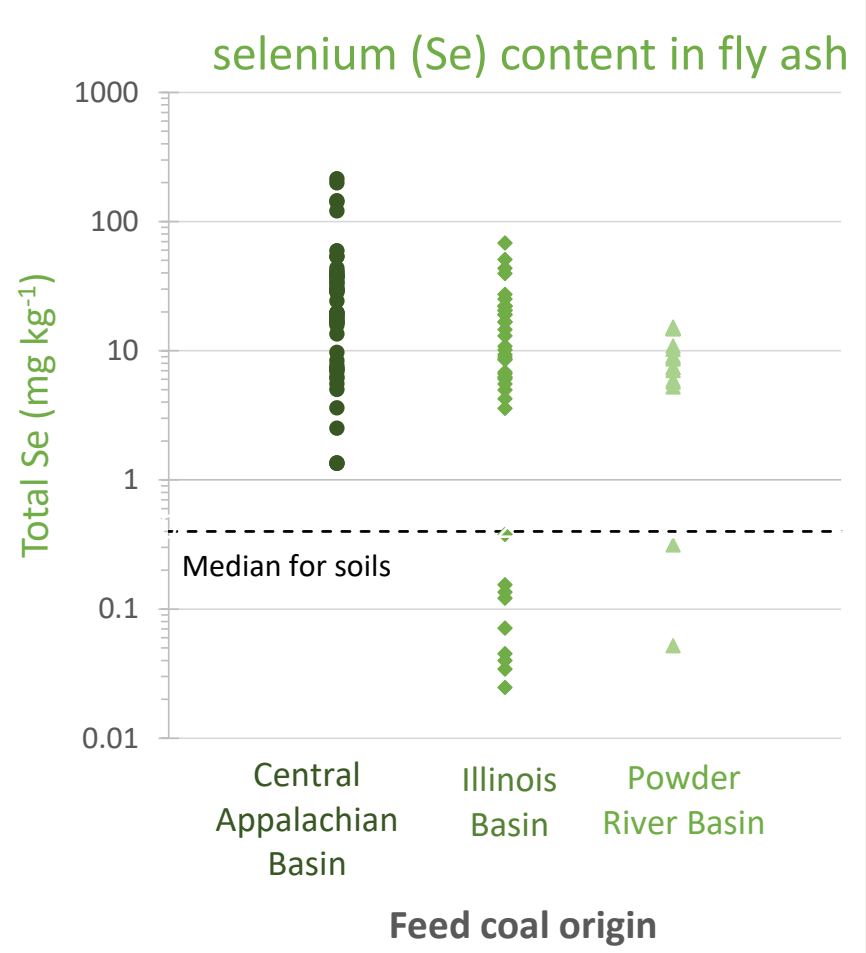
1. Motivation and Significance

- Widely variable amounts of As and Se in coal fly ash
- Depends partly on the type of feed coal





Taggart et al. 2016 ES&T

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2. Project Objectives and Methods

Easy and efficient predicting protocol for leachability of arsenic and selenium based on practical leaching data

- Explore functional predictor variables
  - based on analysis of leaching experiment of arsenic and selenium in coal fly ash

- Build and training predicting regression model
  - Multivariate linear regression
  - Lasso Regression

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2. Project Objectives and Methods

Easy and efficient predicting protocol for leachability of arsenic and selenium based on practical leaching data

Arsenic and Selenium analysis methods

Coal Fly Ash
- Coal feedstock
- Air pollution controls
- Operations (startup, steady state)
- 52 fly ashes involved

Deterministic
- ICP-MS
- HPLC
- Synchrotron XAS (bulk and microprobe)

As/Se content and speciation
Major and minor elements conc.

As, Se mobilization potential
TCLP, LEAF

Leachable As/Se
Leaching pH

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2. Project Objectives and Methods

Easy and efficient predicting protocol for leachability of arsenic and selenium based on practical leaching data

- **Multivariate linear regression**
- **Variable Selection**
  - Bivariate association between predictor variables and leachable As/Se concentrations and speciation
- **Linear regression model**
  - Variable with associations of $P<0.20$ selected for a multivariate linear regression model
- **Lasso Regression**
  - Variable selection & model training
  - L1 regularization

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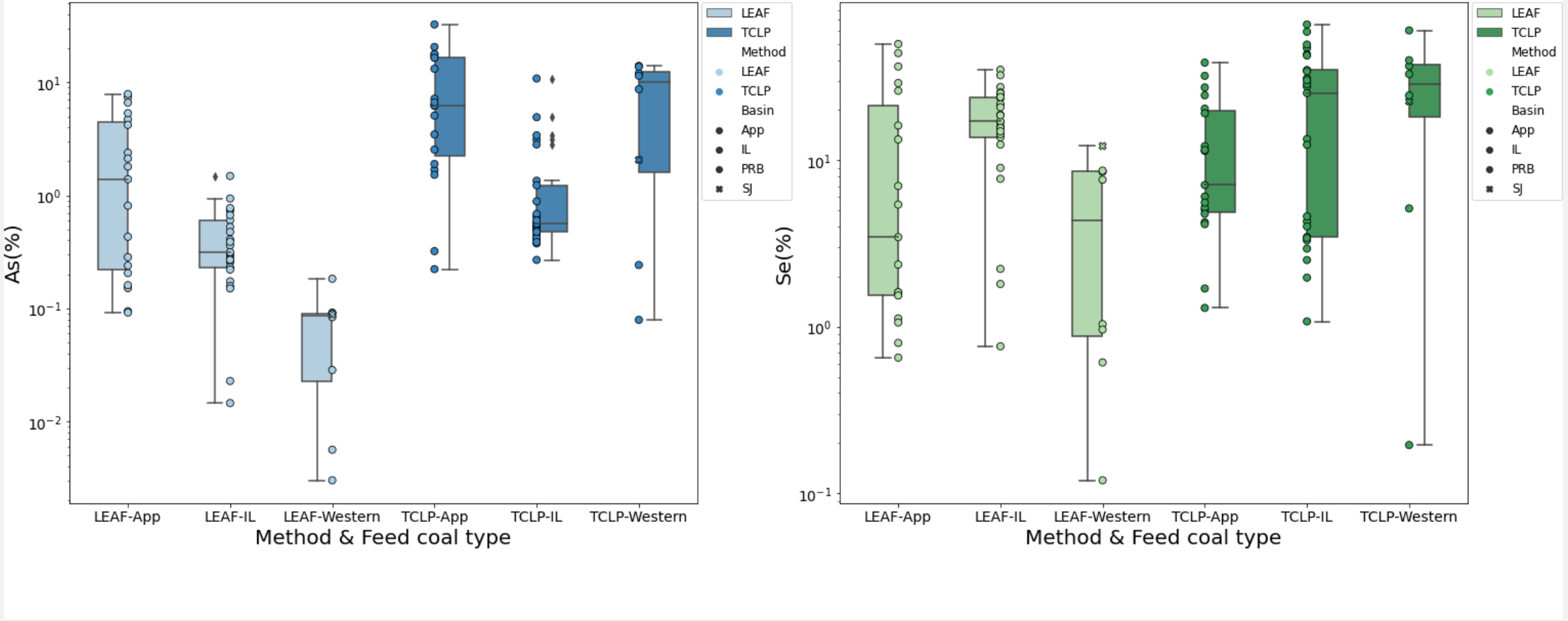
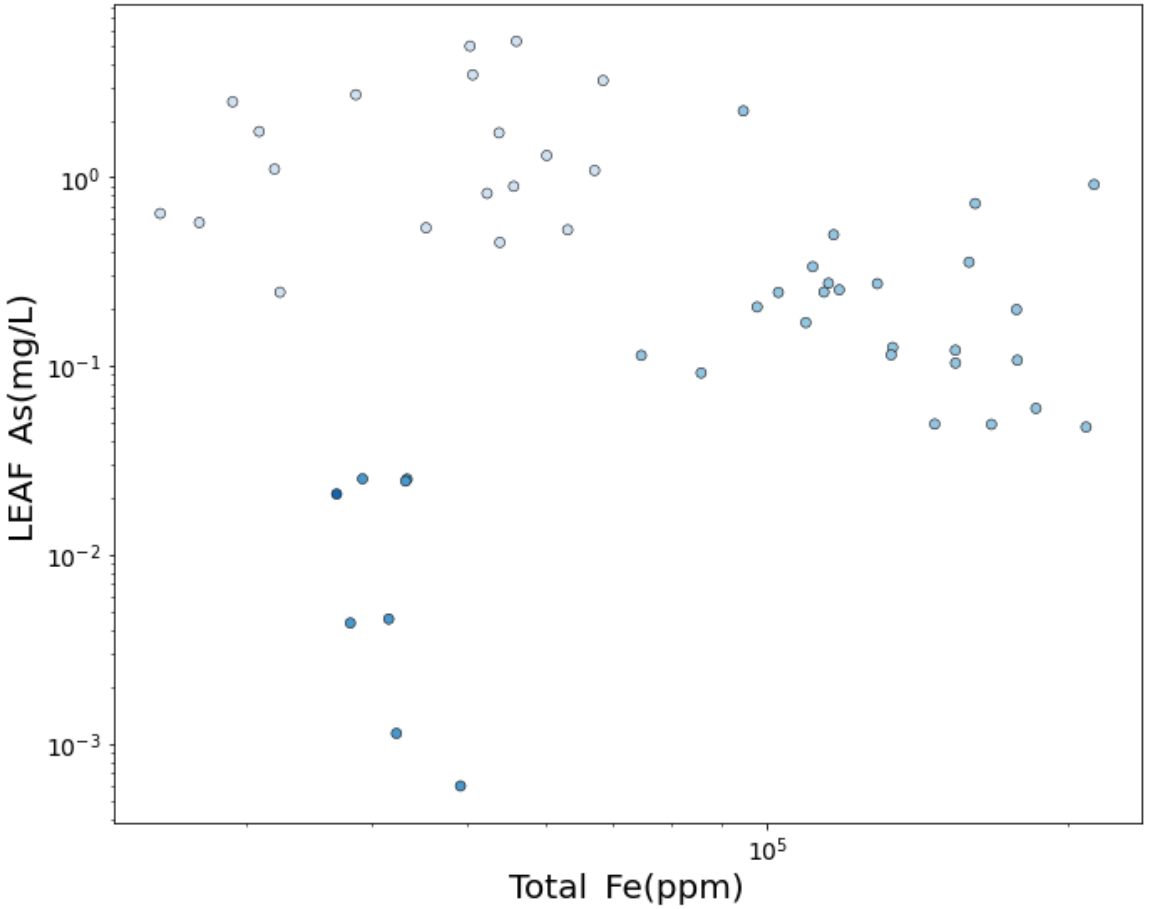
3. Functional Predictor Variables

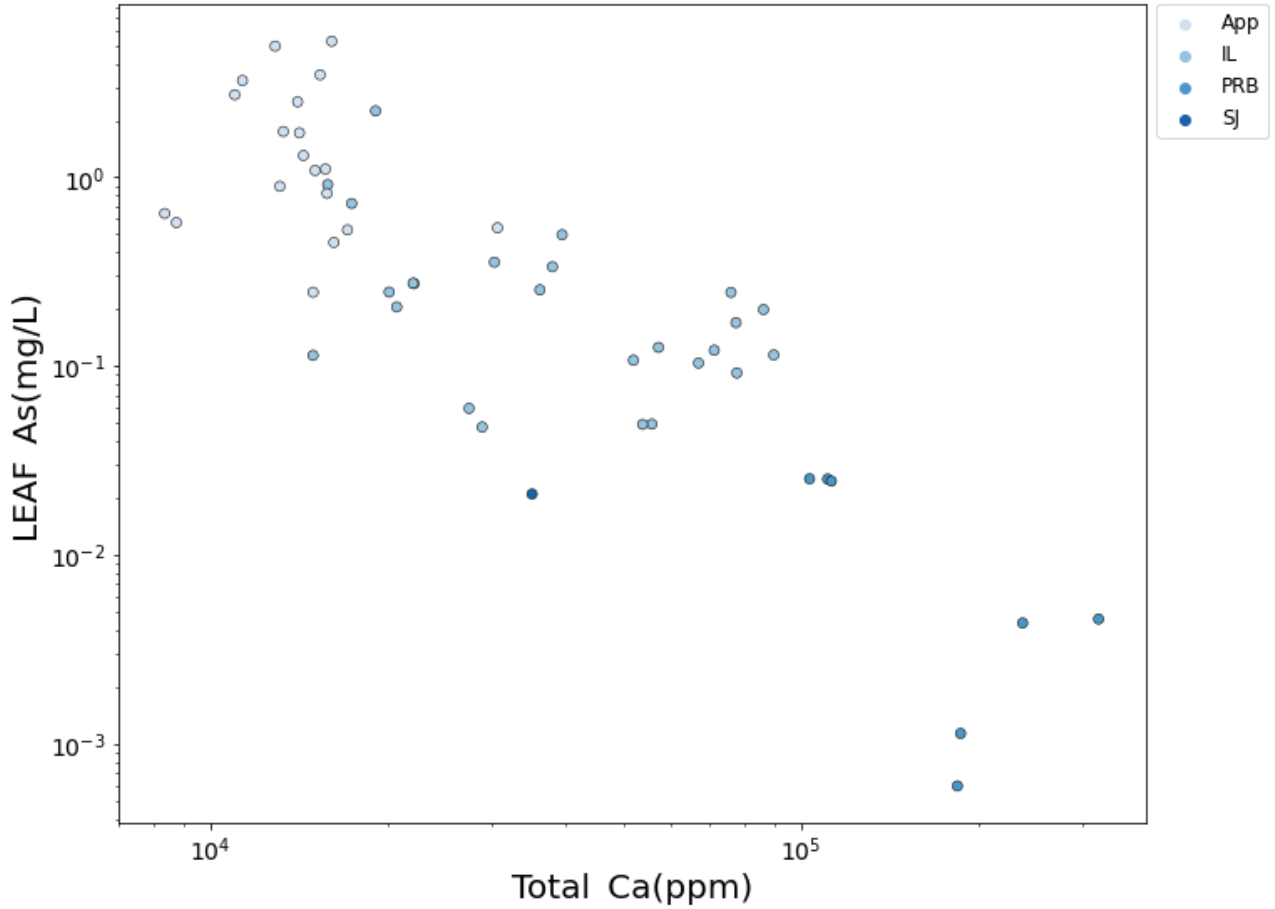
Total elemental content is not informative of leachable concentrations



3. Functional Predictor Variables

Major elements correlation with Arsenic leachability





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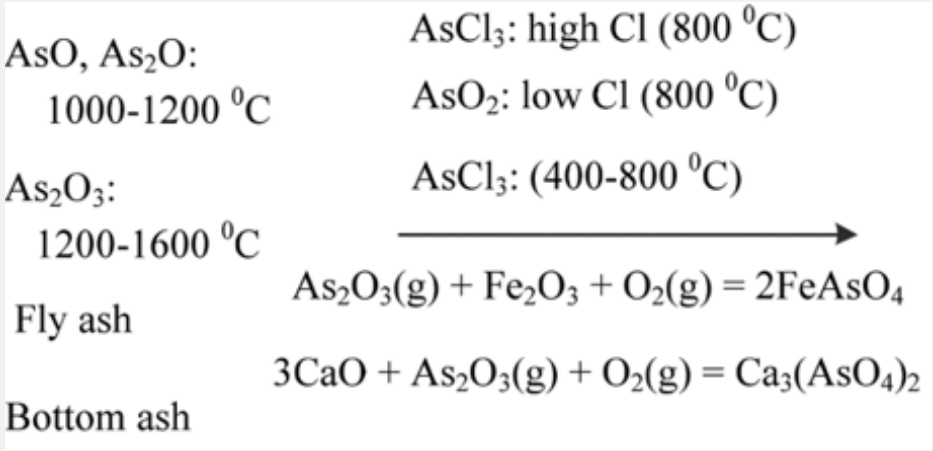
3. Functional Predictor Variables

Tentative mechanism



Zhao, S. et al, *Energy and Fuels,* 2018

Thermodynamic calculation

Component Interactions	Stable Species
	As
TEs-O_2-K	$K_3AsO_4(s)$
TEs-O_2-Ca	$Ca_3(AsO_4)_2(s)$
TEs-O_2-Al	$AlAsO_4(s)$
TEs-O_2-Fe	$FeAsO_4(s)$
TEs-O_2-Mg	$Mg_3(AsO_4)_2(s)$

Combustion, P. C., Minerals, 2021

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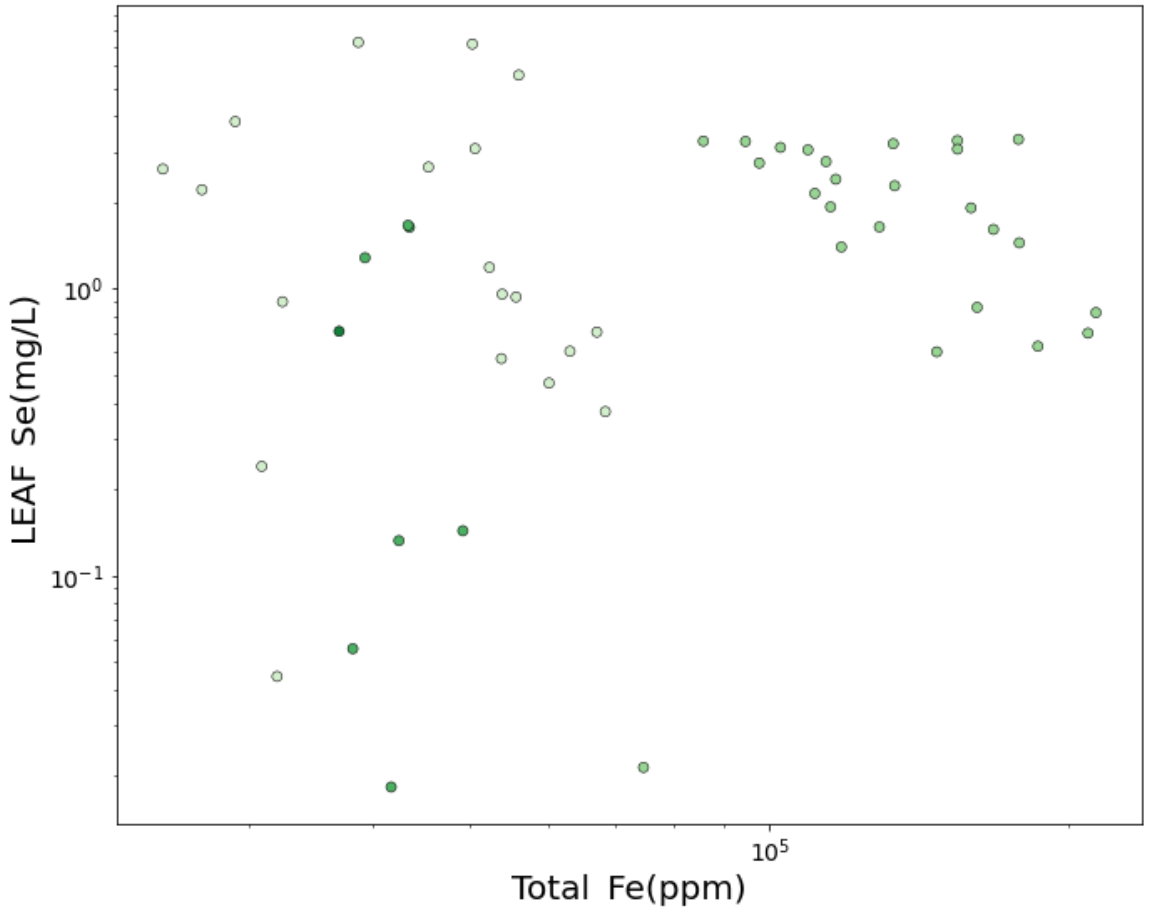
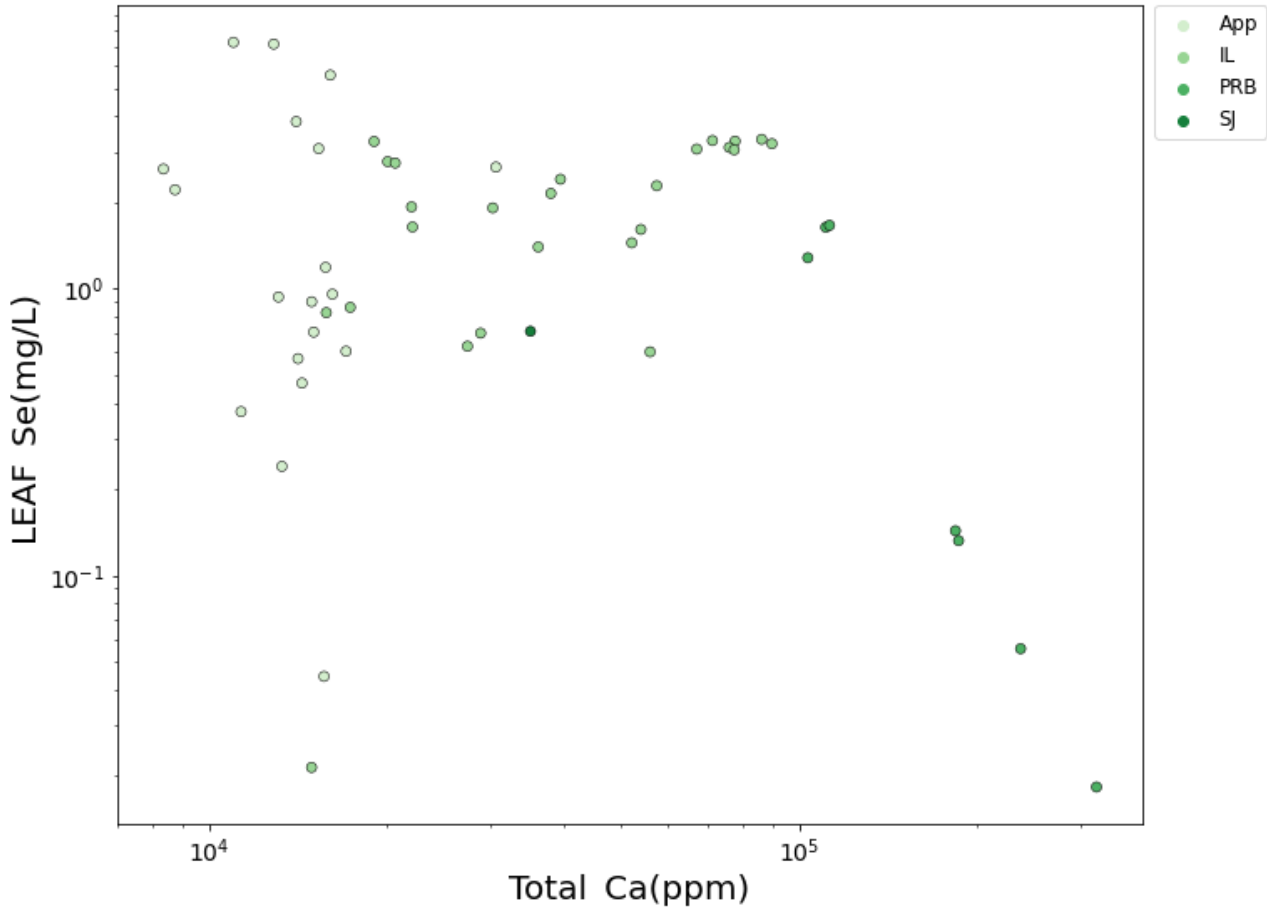
3. Functional Predictor Variables

Major elements correlation with Selenium leachability





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3. Functional Predictor Variables

Thermodynamic calculation

Component Interactions	Stable Species
	Se
TEs-O_2-K	SeO_2(s)
TEs-O_2-Ca	SeO_2(s)
TEs-O_2-Al	SeO_2(s)
TEs-O_2-Fe	SeO_2(s)
TEs-O_2-Mg	$MgSeO_3$(s)

Combustion, P. C., Minerals, 2021

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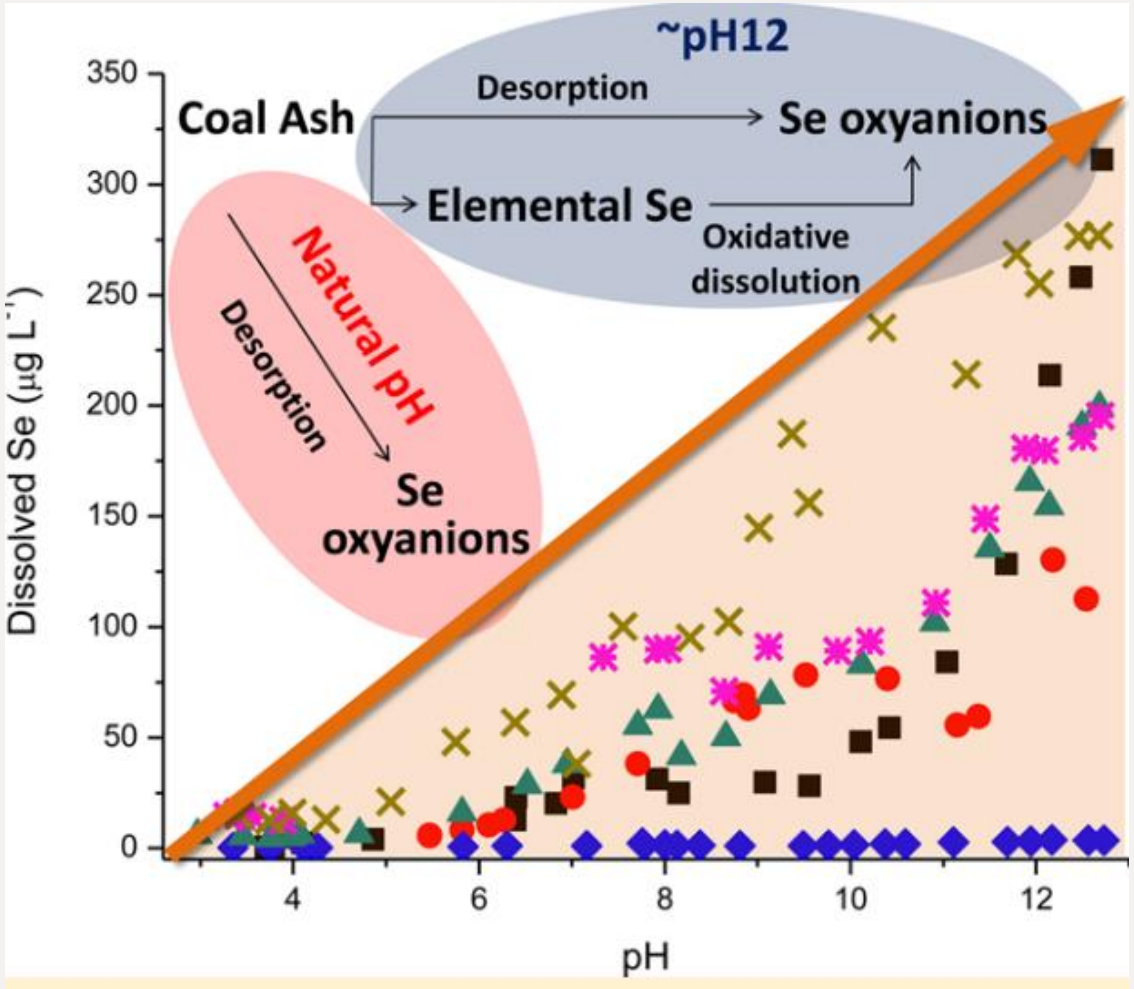
3. Functional Predictor Variables

pH has impact to leachability but insufficient



Liu, Y.T. et al, *Environmental science & technology, 2013*

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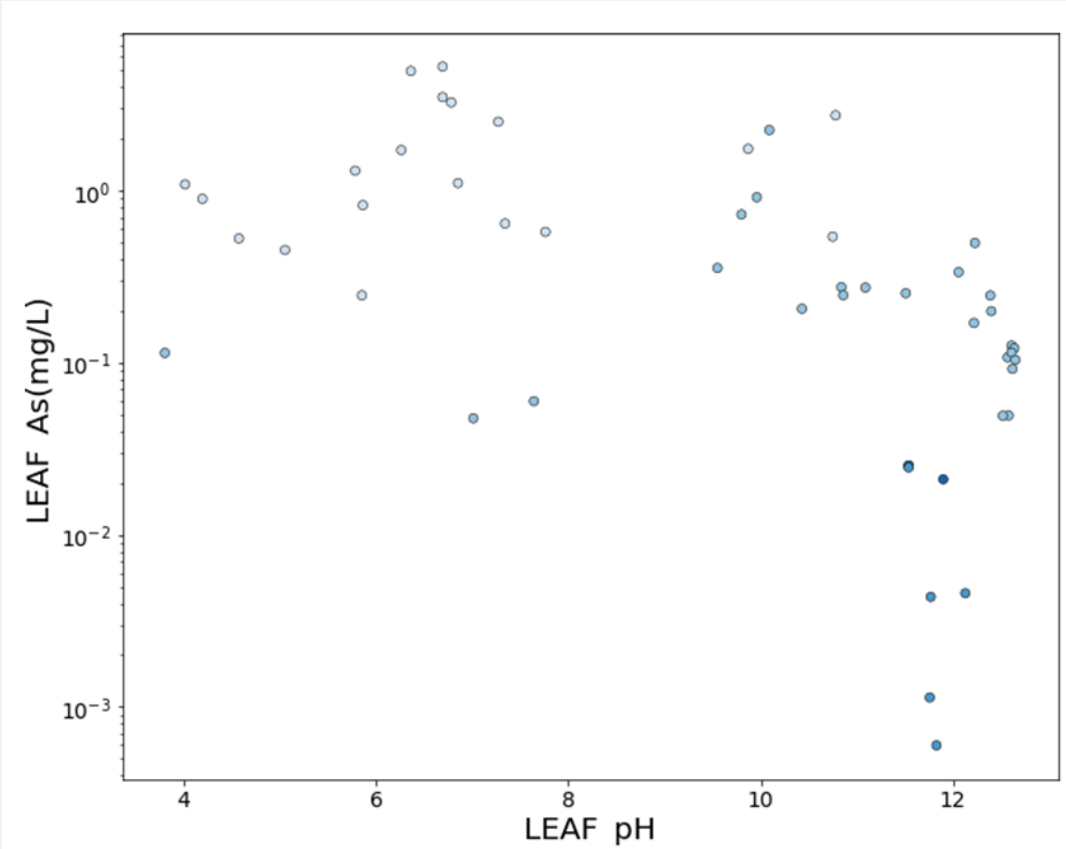

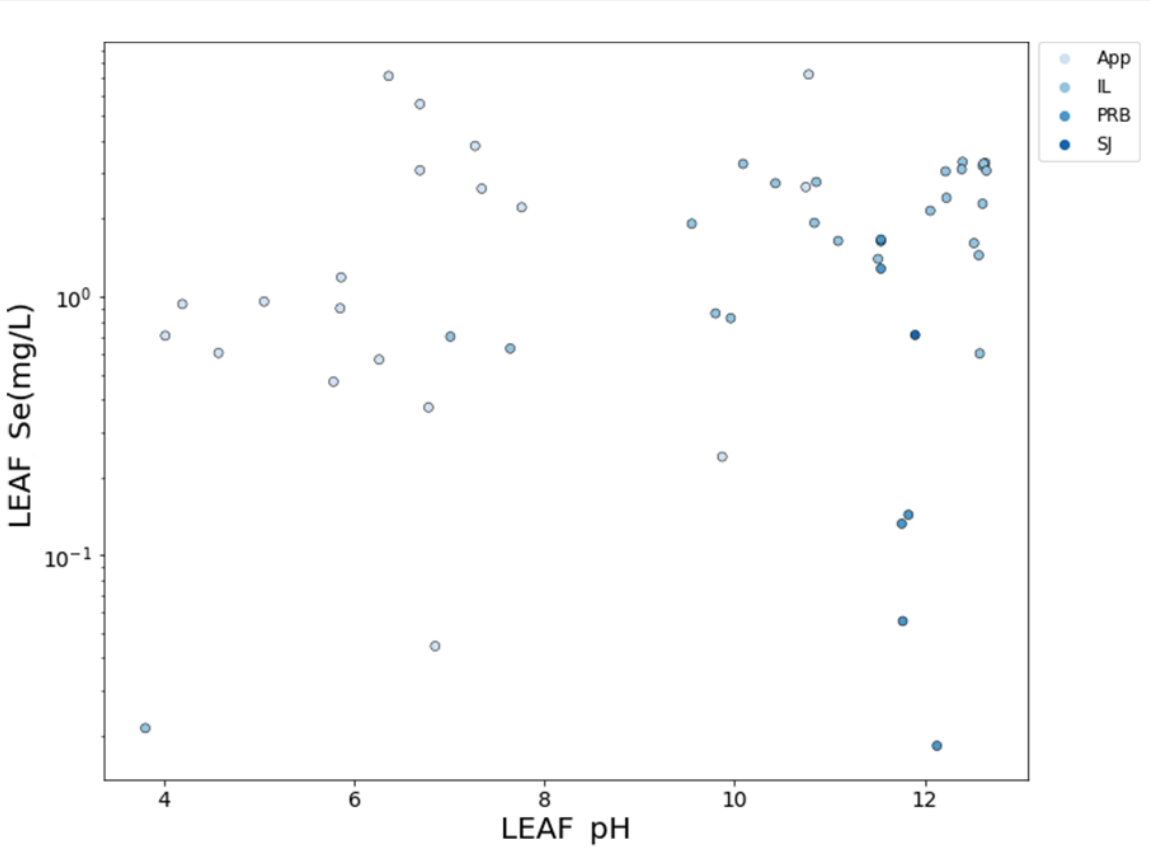

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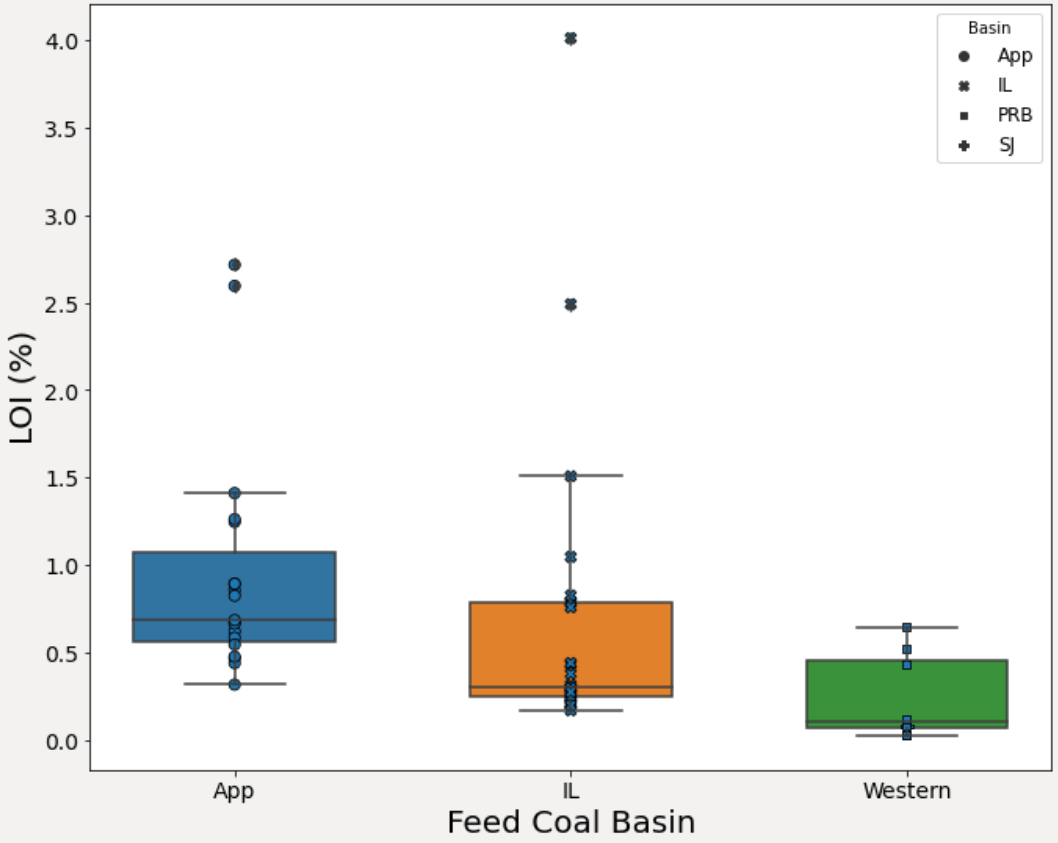
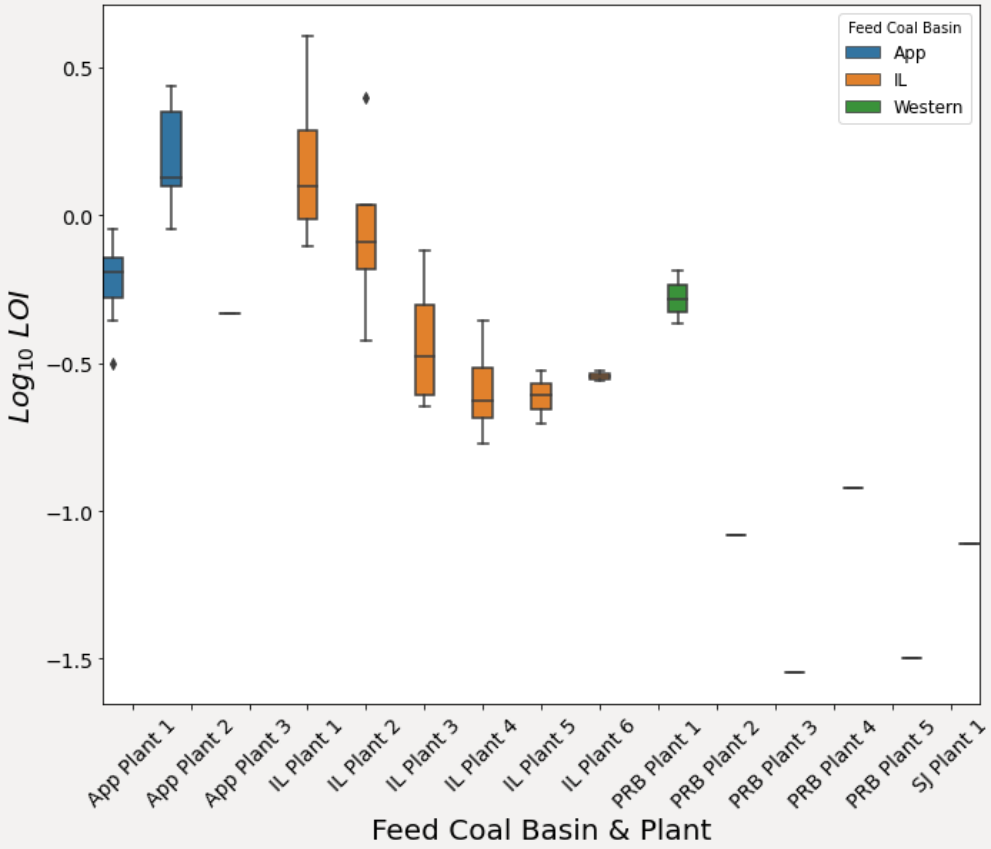
3. Functional Predictor Variables

Loss on ignition (LOI) selected to present unburned carbon content, also contains geophysical information





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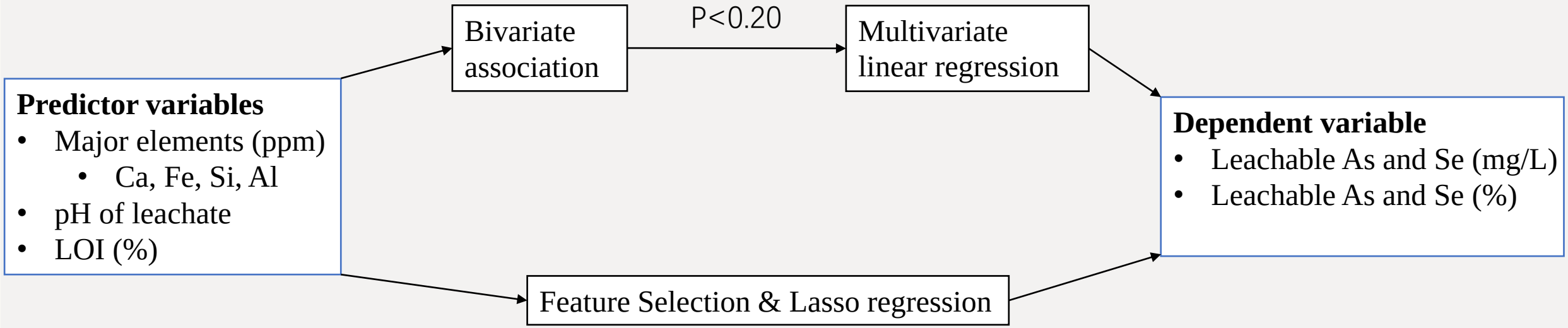
4. Regression models

Multivariate linear regression



Lasso linear regression

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4. Regression models

	Leachable As (mg/L) bivariate analysis				Leachable As (mg/L) multivariate analysis			
	β	p value	95% CI		β	p value	95% CI	
Intercept	\	\	\	\	10.731	0.608	-31.123	52.585
Log10 Fe	-0.404	0.702	-2.508	1.701	\	\	\	\
Log10 Ca	-4.222	0.000	-4.996	-3.449	-5.070	0.000	-6.742	-3.398
Log10 Al	6.251	0.004	2.150	10.351	4.280	0.068	-0.322	8.883
Log10 Si	20.375	0.000	14.000	26.749	-2.366	0.583	-10.973	6.241
LOI	0.585	0.115	-0.148	1.318	0.004	0.991	-0.758	0.766
pH	-0.369	0.000	-0.538	-0.199	0.168	0.114	-0.042	0.377
R^2	\				0.779			

	Leachable As (%) bivariate analysis				Leachable As (%) multivariate analysis			
	β	p value	95% CI		β	p value	95% CI	
Intercept	\	\	\	\	-35.300	0.212	-91.438	20.839
Log10 Fe	-1.227	0.161	-2.962	0.507	-0.658	0.484	-2.536	1.220
Log10 Ca	-2.730	0.000	-3.643	-1.816	-1.759	0.040	-3.433	-0.086
Log10 Al	2.843	0.125	-0.820	6.506	-1.537	0.622	-7.771	4.698
Log10 Si	16.023	0.000	10.408	21.638	9.940	0.018	1.773	18.107
LOI	0.640	0.038	0.036	1.244	-0.117	0.830	-1.209	0.974
pH	-0.102	0.220	-0.267	0.063	\	\	\	\
R^2	\				0.496			

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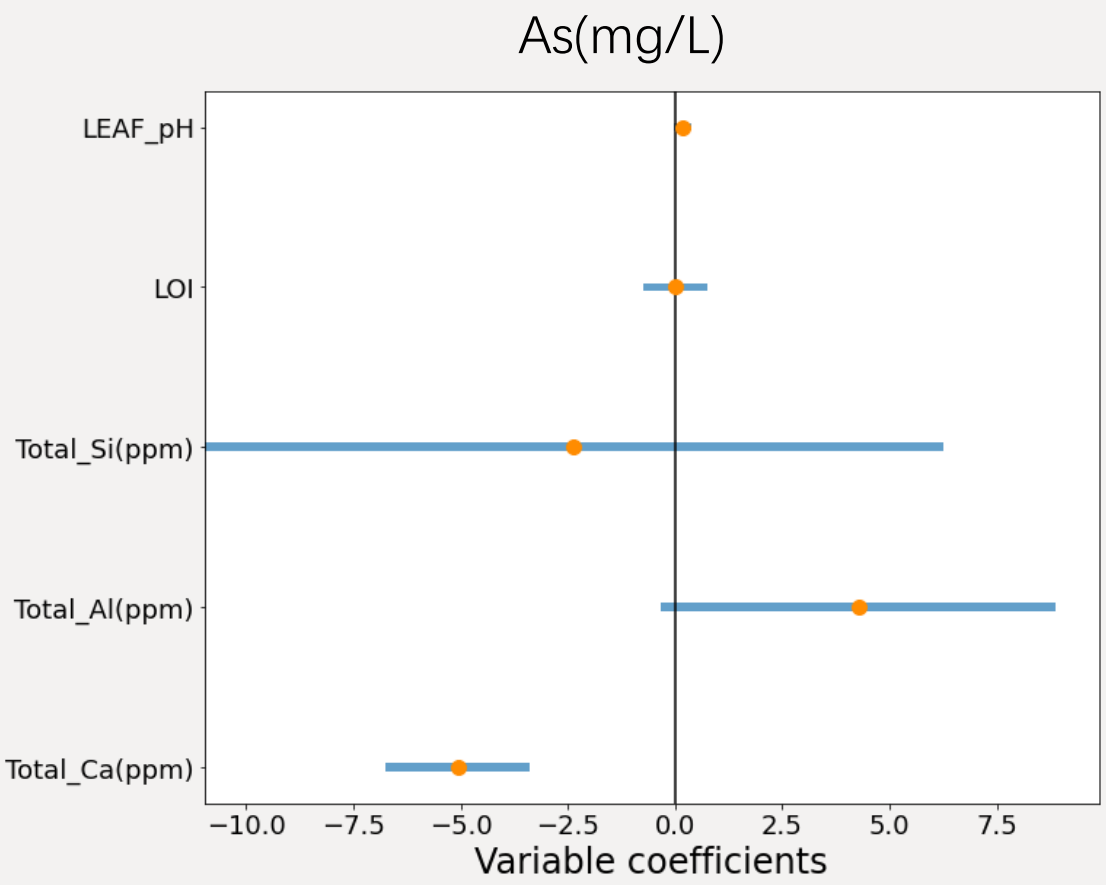
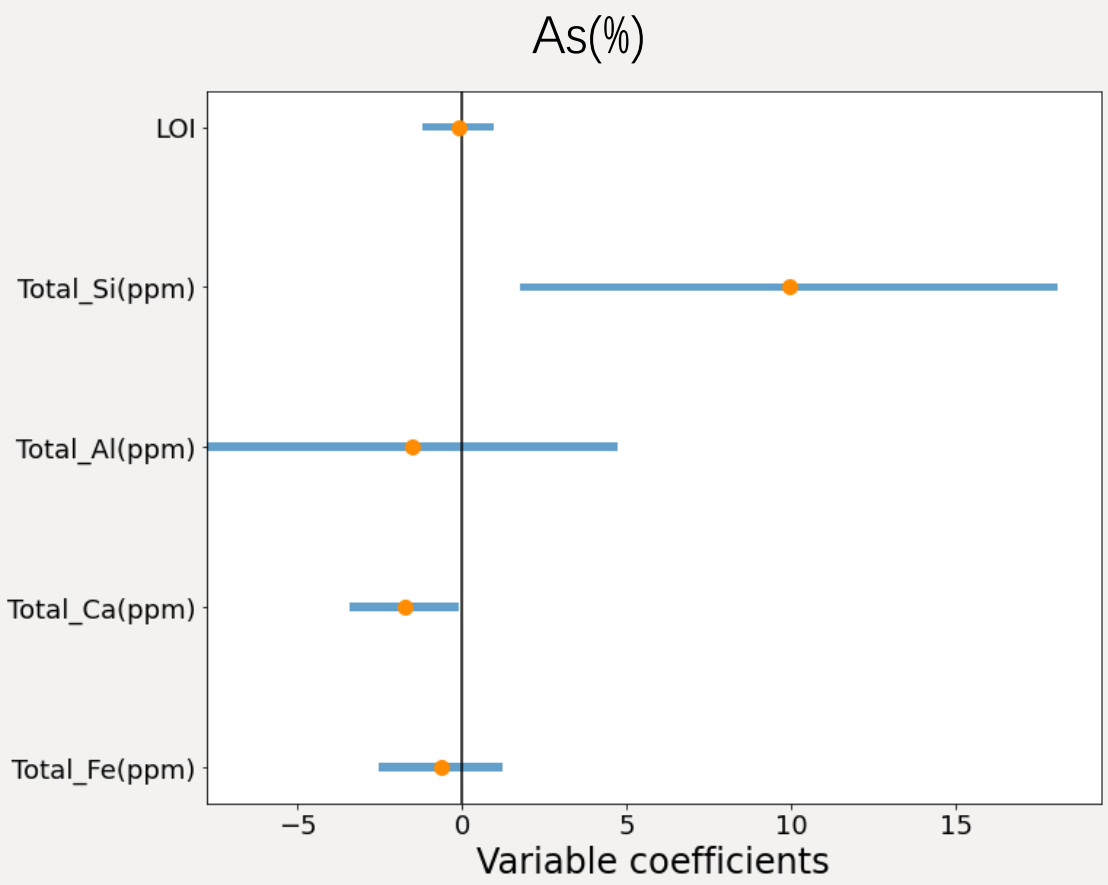
4. Regression models





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4. Regression models

	Leachable As (mg/L) lasso regression	Leachable As (%) lasso regression
	β	β
Intercept	0.449	4.153
Log10 Fe	7.992	-5.575
Log10 Ca	-60.768	-57.952
Log10 Al	40.592	37.169
Log10 Si	0.000	0.000
LOI	0.000	2.206
pH	0.000	6.831
R^2	0.785	0.699

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5. Conclusion

Correlations between the Leachability of arsenic and selenium and coal combustion characteristics

- Chemical variables
  - Major elements concentration in coal fly ash
  - pH of leachate
- Geographic information
  - Coal feedstock
  - LOI

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5. Conclusion

Predictive regression models to foresee leachable arsenic and selenium

Pros:

- all the predictor variables involved in the model are the physical and chemical properties of the major elements
- can be efficiently measured by a portable X-ray fluorescence spectrometer or optical emission spectrometer in-site

Cons:

- Lack of generality
  - Focus on 3 feed coal basin
  - Arsenic concentration under EPA's limit (10ppb)

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Thank you

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