Determination of radioiodine in environment and biological samples using Pyrolyser sytem for iodine separation

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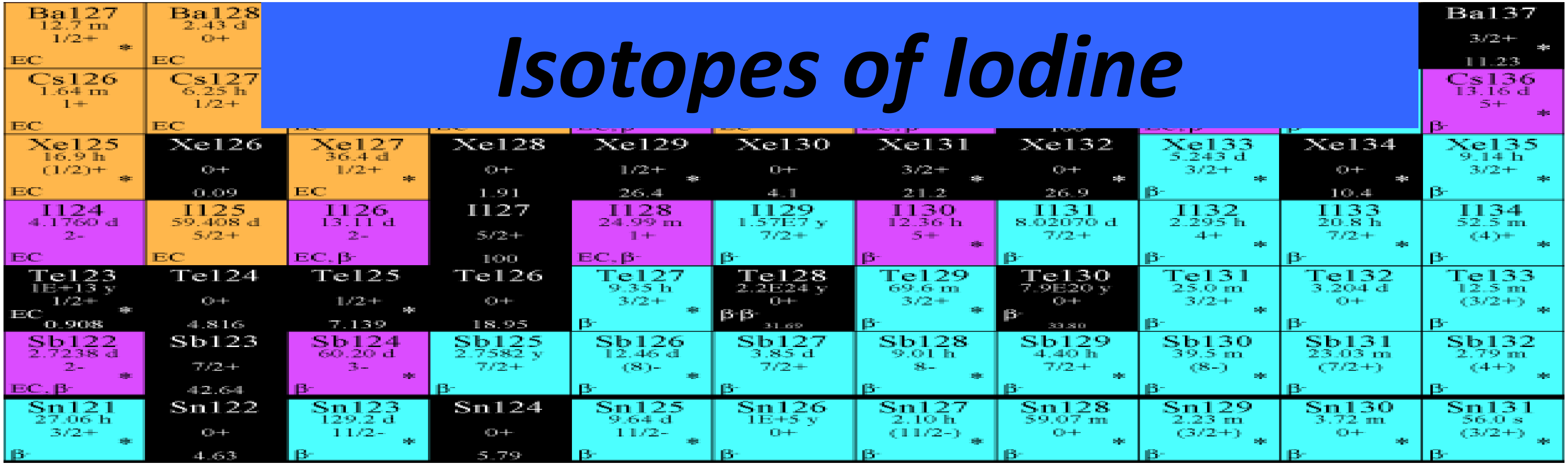
Isotopes of Iodine



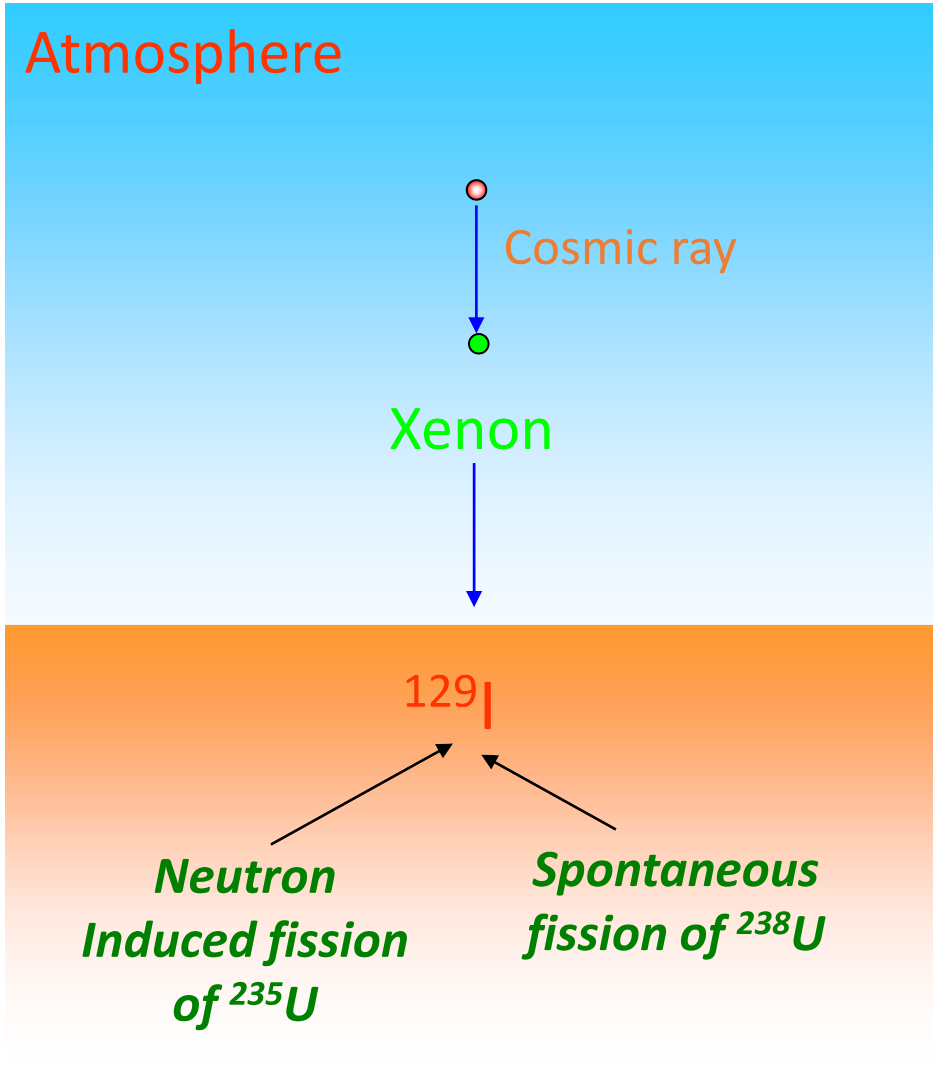
- ^{127}I (stable, 100% abundance)
- ^{131}I (8 d),
- ^{125}I (56 d),
- ^{126}I (13 d), ^{124}I (4.2 d)
- **^{129}I (15.7 ×10^{6} years)**
- others (< 1 day)

Properties of iodine

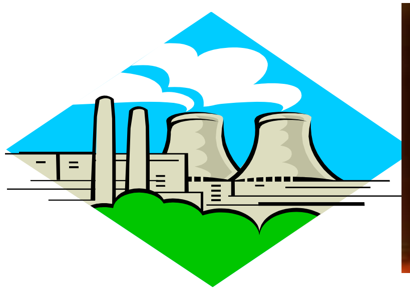
- Chemically active element with oxidation state of -1, 0, +1, +3, +4, +5, +7, e.g. I^-, I_2, IO^-, IO_3^-, IO_4^-
- The most popular species of iodine in the environment are: I^-, IO_3^- and organic iodine.
- Iodine is considered as a n volatile element, based on the volatile feature of the species of I_2, CH_3I, etc.
- Iodine is considered as a conservative element on the ocean, because it mainly presents as iodate, and iodide in some waters.

Source of ^{129}I

- **Natural process:**



- **Artificial process:**



$^{235}U(n, f)\ ^{129}I$

$^{239}Pu(n, f)^{129}I$

^{129}I level in environment

Source	Release/ inventory, kg	^{129}I/^{127}I ratio
Nature	250	10^{-12}
Nuclear weapons testing	63	10^{-10}~10^{-9}
Chernobyl accident	1.3~6	10^{-9}~10^{-6} (local)
Marine discharge of EU NRPs	5200	10^{-7}~10^{-6} (seawater)
Atmospheric emission from NRPs	800	10^{-6}~10^{-4} (local)

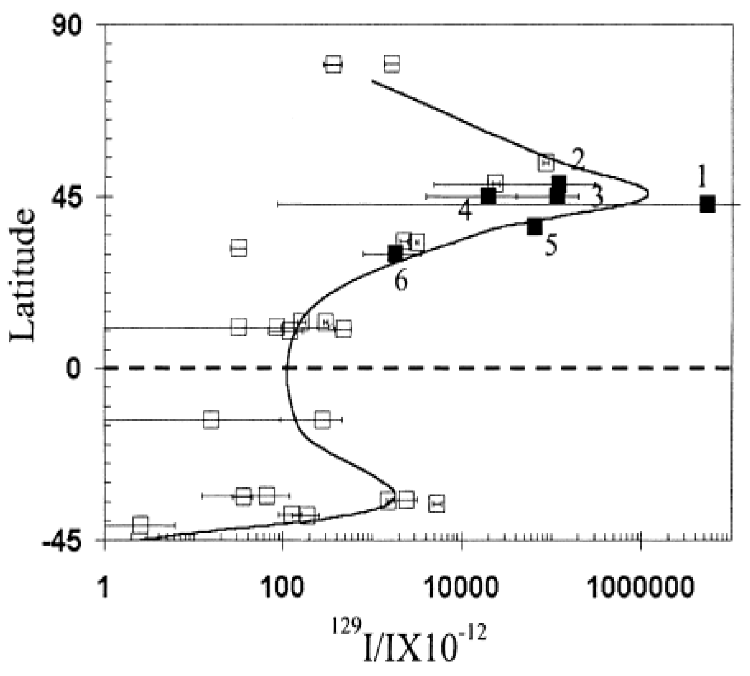
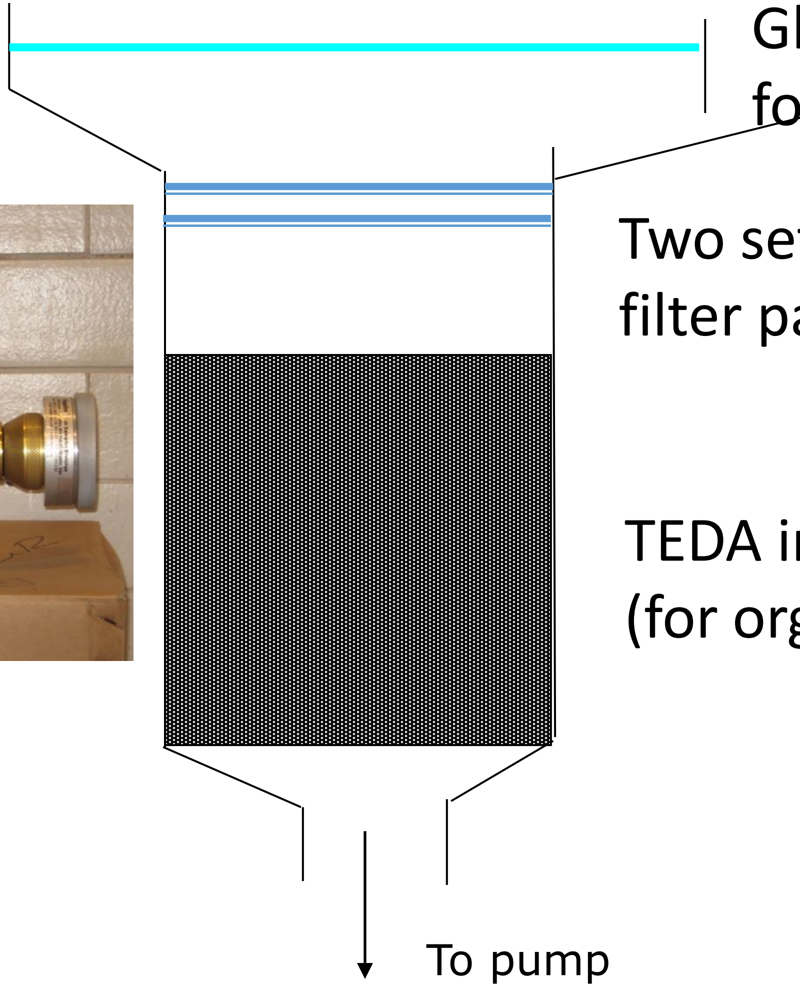


Fig. 1. ^{129}I/I ratios in surface waters as a function of latitude. Open squares indicate samples with associated errors (1 σ)

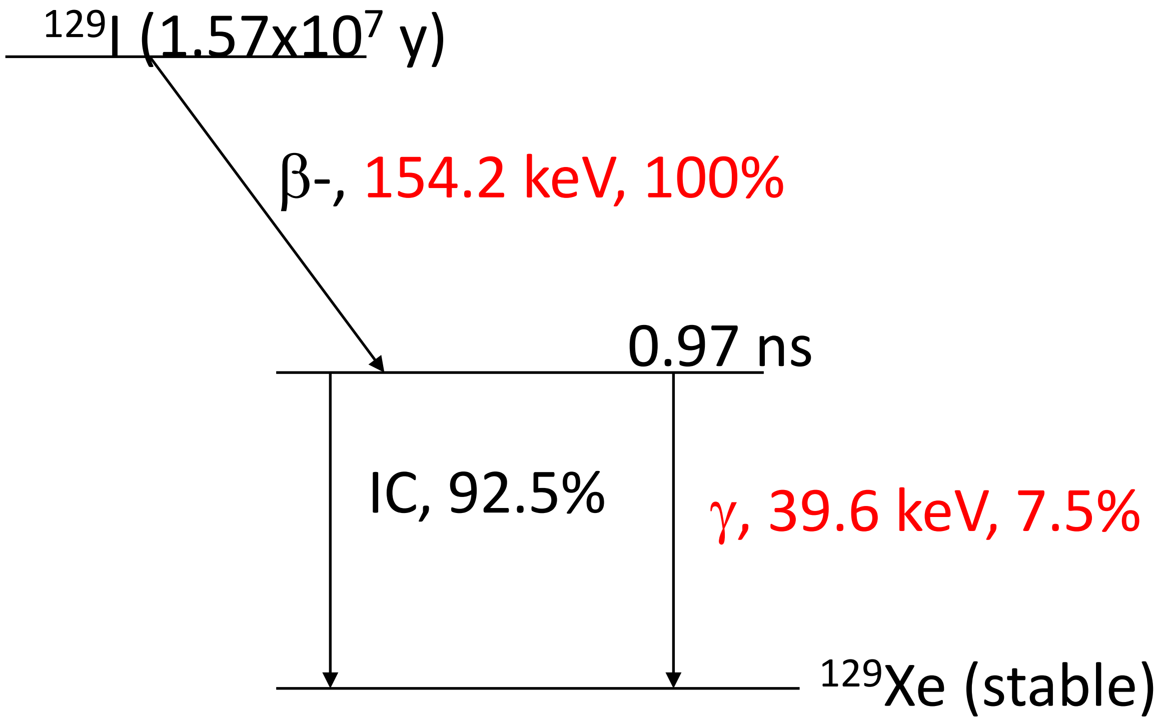
Speciation analysis of ^{129}I and ^{127}I in air

- Particle associated Iodine
- Inorganic gase iodine (I_2, HI, HIO)
- Organic gas iodine (CH_3I, C_2H_7I, CH_2BrI, etc.)

Speciation method for ^{129}I in air



Decay of ^{129}I and its radiation



X-rays:
29.5 keV, 20.4%
29.8 keV, 37.7%
33.6 keV:10.1%

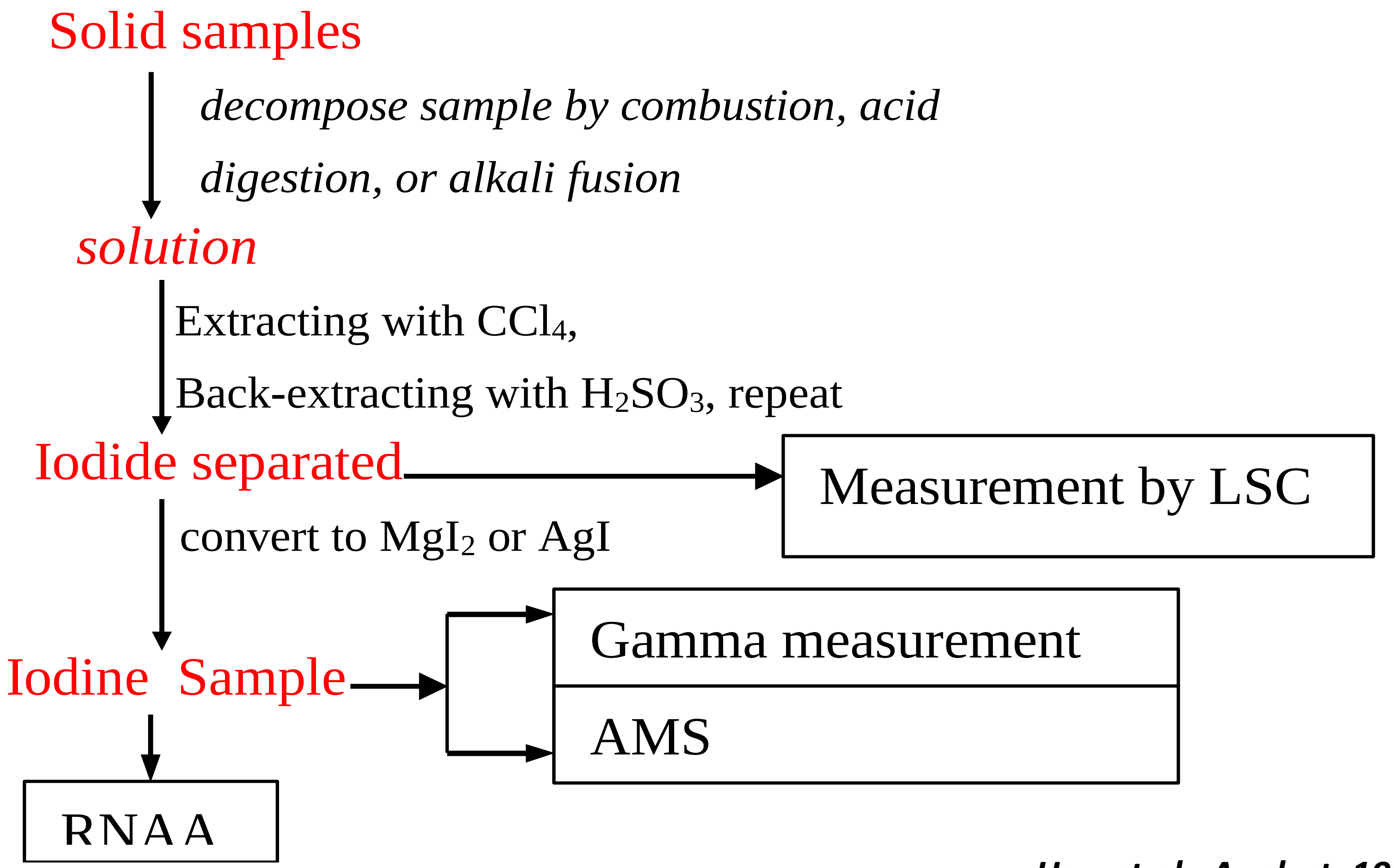
Measurement Method for ^{129}I and their detection limits

Method	Detection limit		
	^{129}I, atoms	^{129}I, mBq	$^{129}I/^{127}I$ Ratio
Liquid scintillation	10^{13}	10 mBq	
γ-spectrometry	10^{13}	10 mBq	
ICP-MS	2×10^{11}	0.4 mBq	10^{-6}
Radiochemical neutron activation analysis	10^{8}	0.2 mBq	10^{-10}
Accelerator mass spectrometry (AMS)	10^{5}	0.1 nBq	10^{-13}

Hou et al. ACA, 2007

Separation procedure of iodine from solid samples

(seaweed, soil, sediment, filter, vegetation, etc.)



Hou et al. Analyst, 1999

Separation of ^{129}I from concrete, soil and sediment by alkali fusion

Solid samples (seaweed, soil, sediment)

↓ add NaOH, iodine carrier, ash/fusion at 650°C for 2 h

ash or Fusee cake

↓ Leaching with H_2O, filter to remove residue (precipitation)

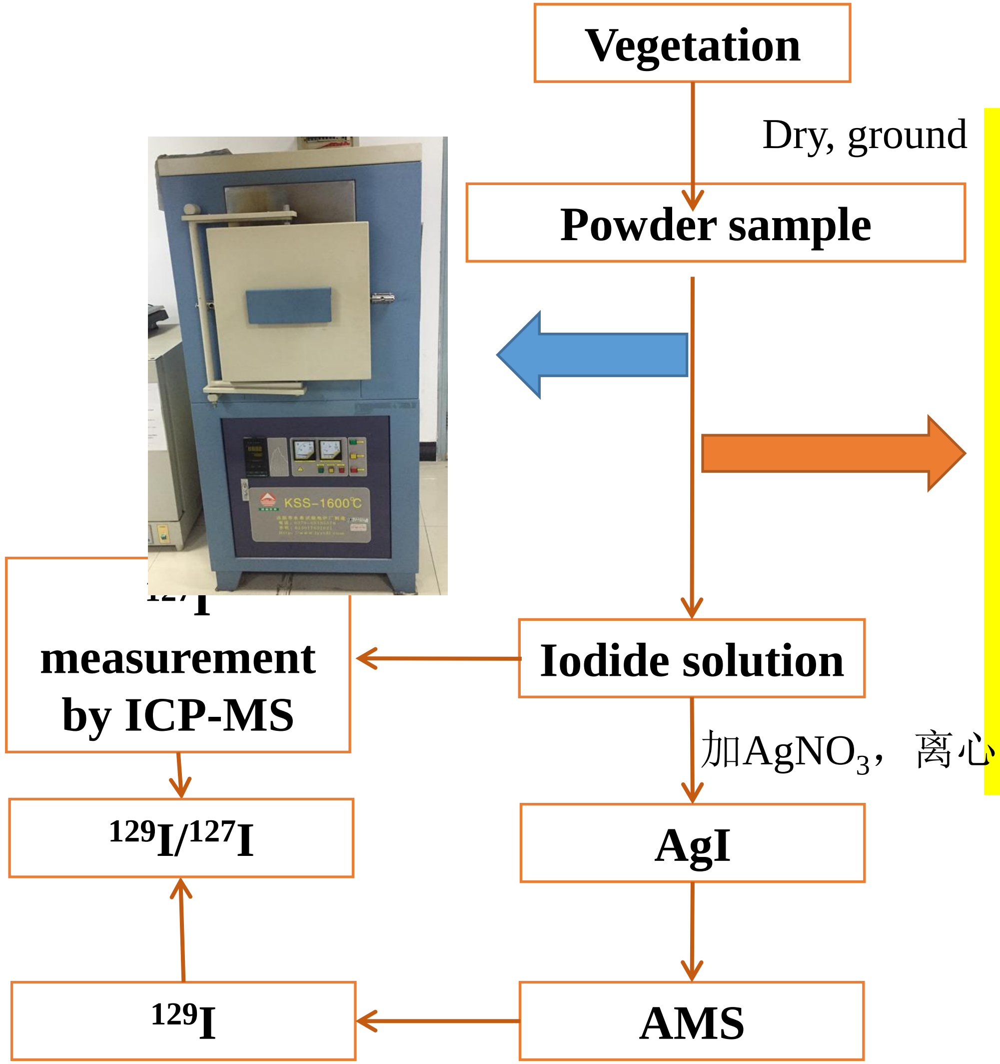
Solution with ^{129}I

↓ Convert iodine to iodide, extract with CCl_4, back extract with $NaSO_3$

Separated Iodine

Hou et al. Analyst, 1999

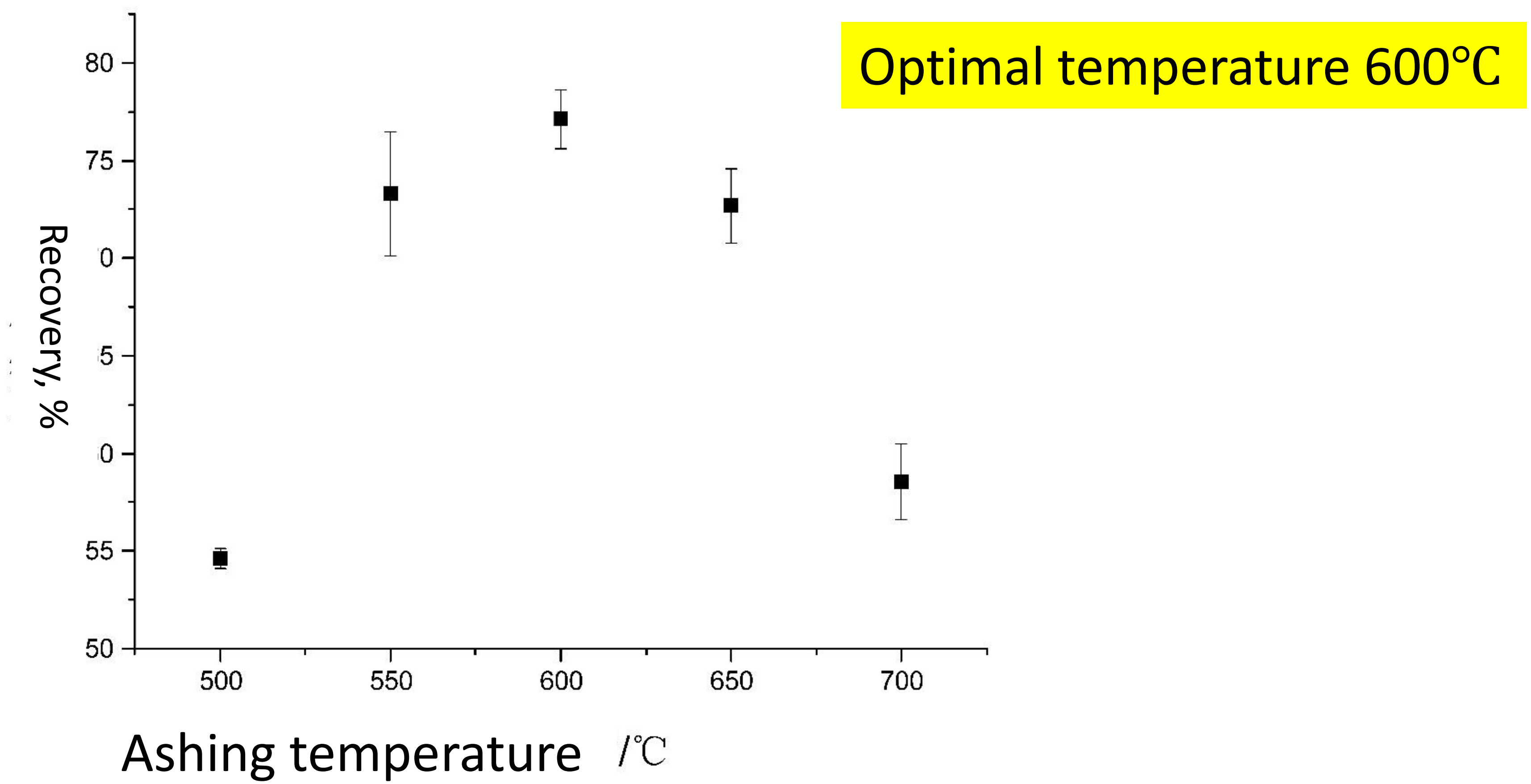
Separation of ^{129}I from vegetation by alkali fusion



Alkali fusion

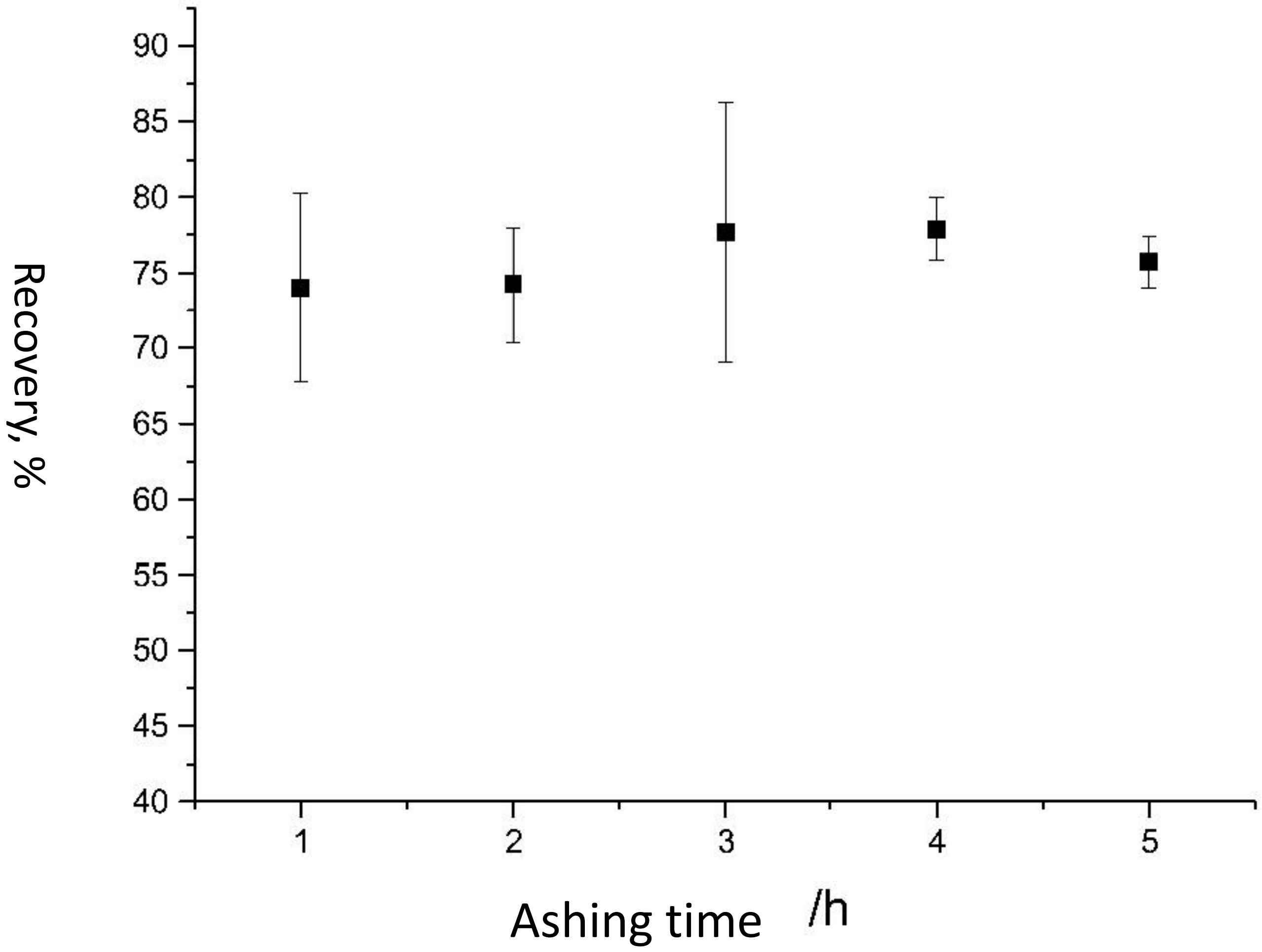
- Add NaOH, dry (ratio of sample to NaOH)
- Ashing (temperature)
- Leaching
- Iodine separation by solvent extraction

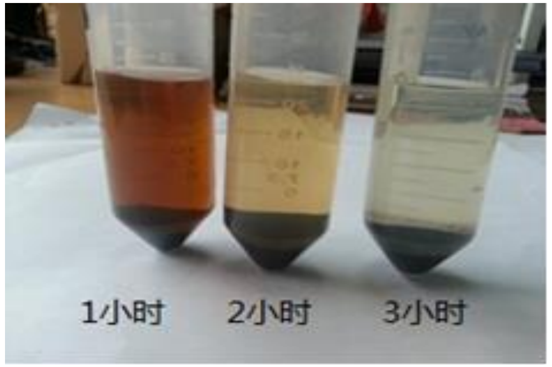
Ashing temperature



Effect of ashing temperature on the recovery of iodine in teh ashing

Ashing time

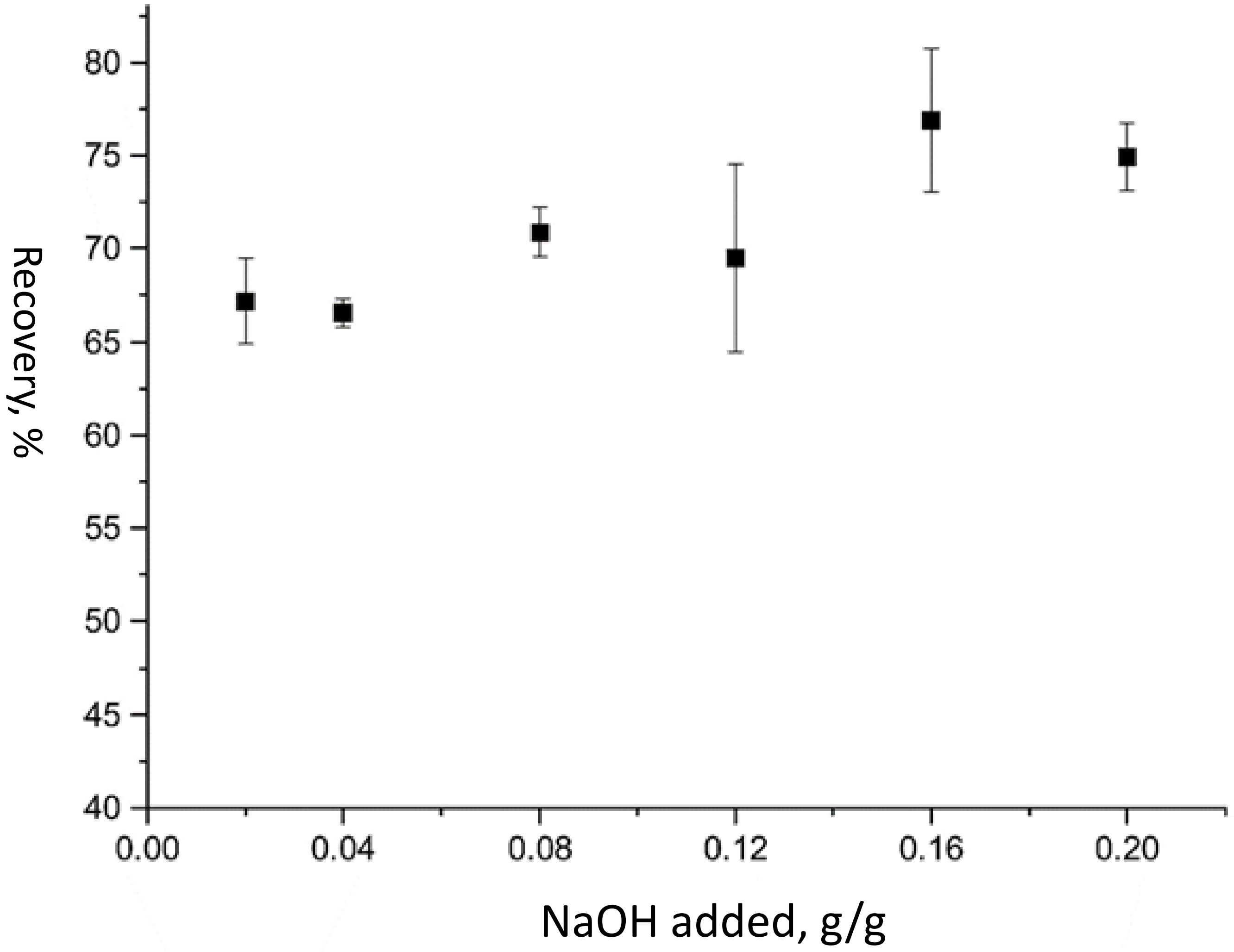




Effect of ashing time on the recovery of iodine

Akalin anount added

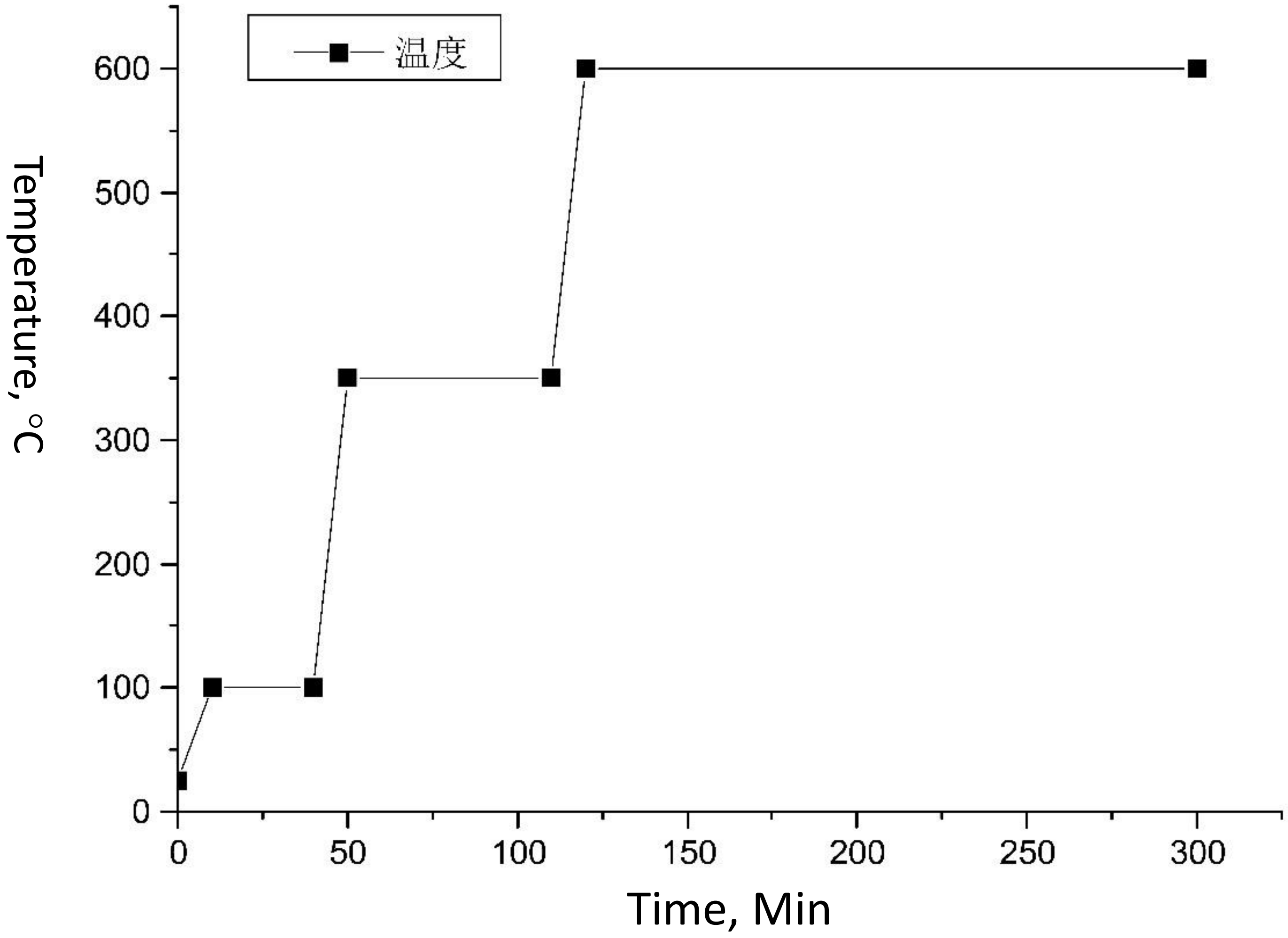
NaOH=1g:0.16g



Effect of NaOH added on the recovery of iodine

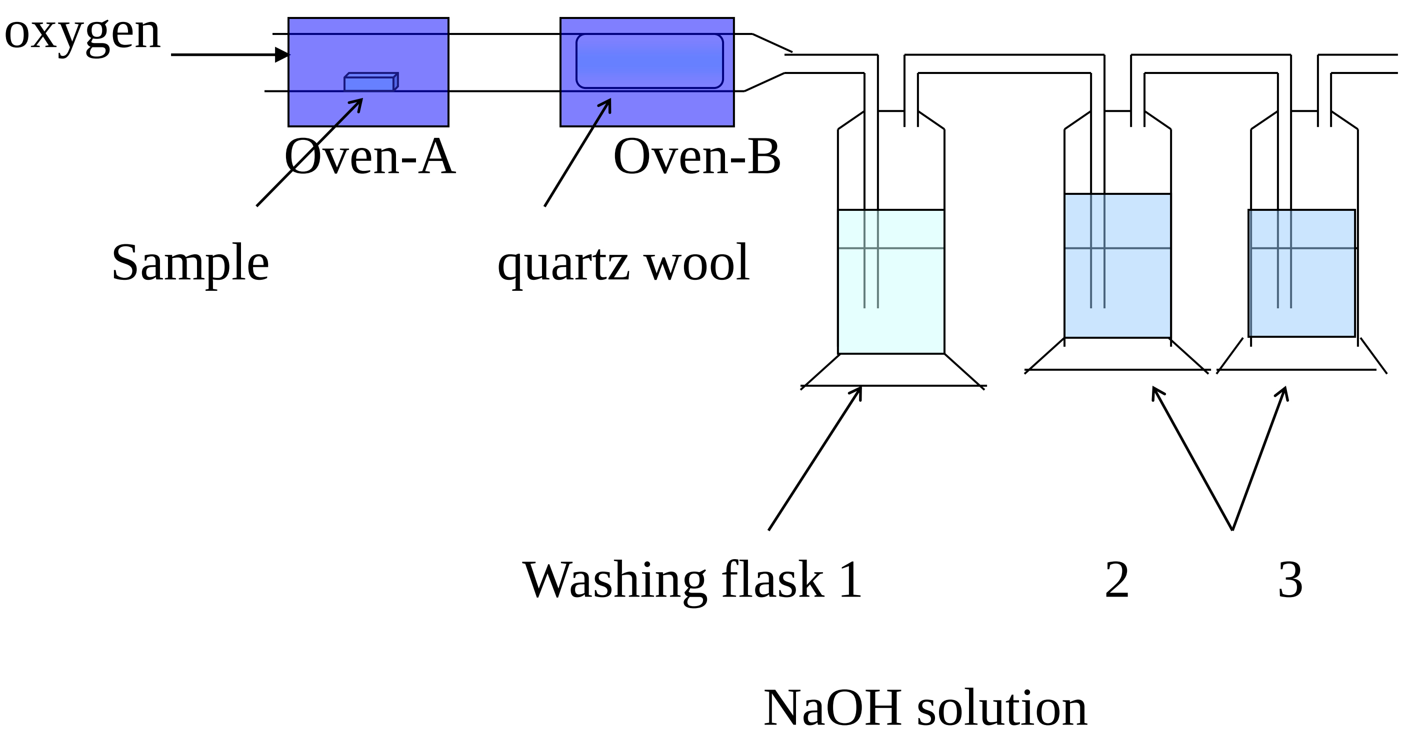
Wang & Hou JRNC 2020

Temperature arising protocol

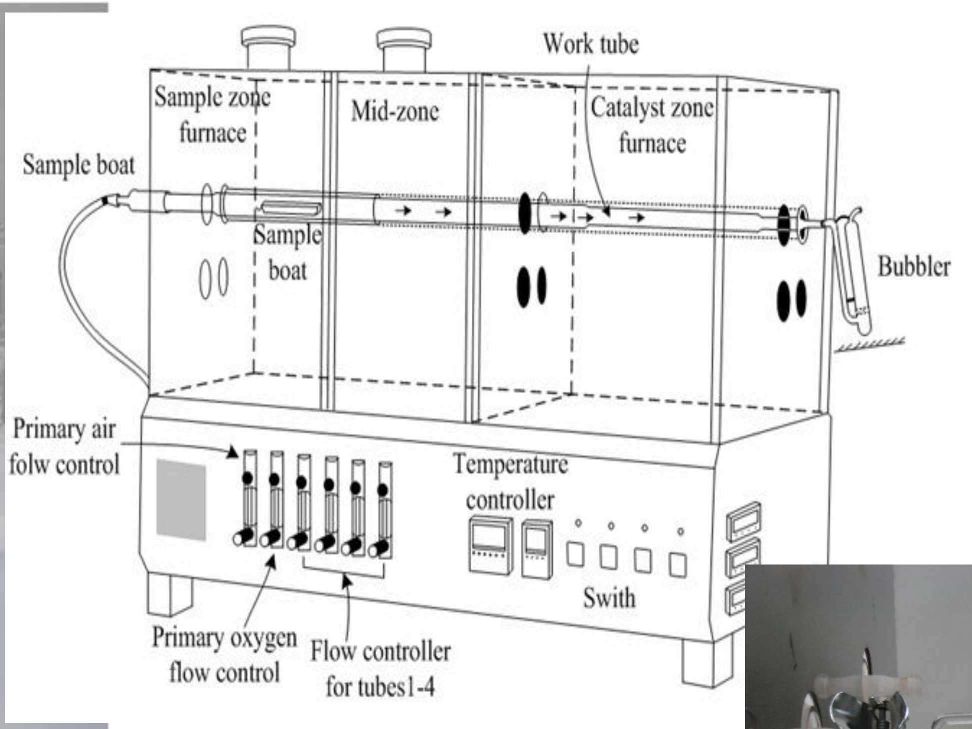


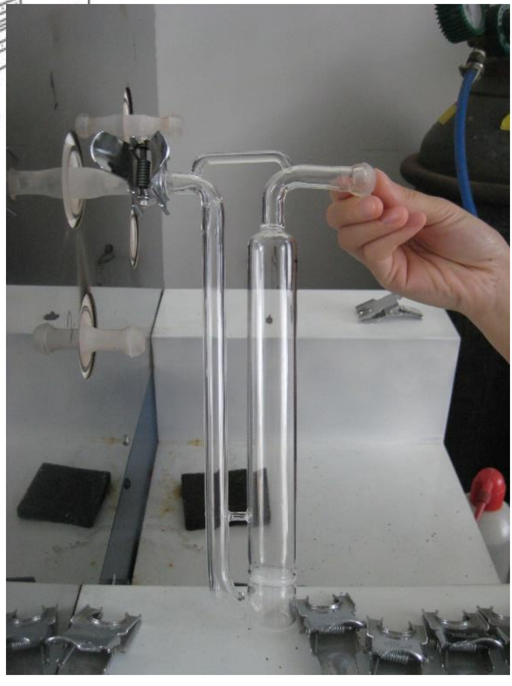
Separation of iodine from solid sample by combustion

Separation of ^{129}I from soild samples by combustion method



Separation of iodine from solid samples using Pyrolyser





- The working tube bubbler and joint are quartz materials to avoid the reaction of I_2 with plastic material
- Combustion is controled by the gas composition and flow rate
- No catalyst materials

Separation of iodine from soil/sediemnt using combustion

Parameters influencing the separation of iodine by combustion:

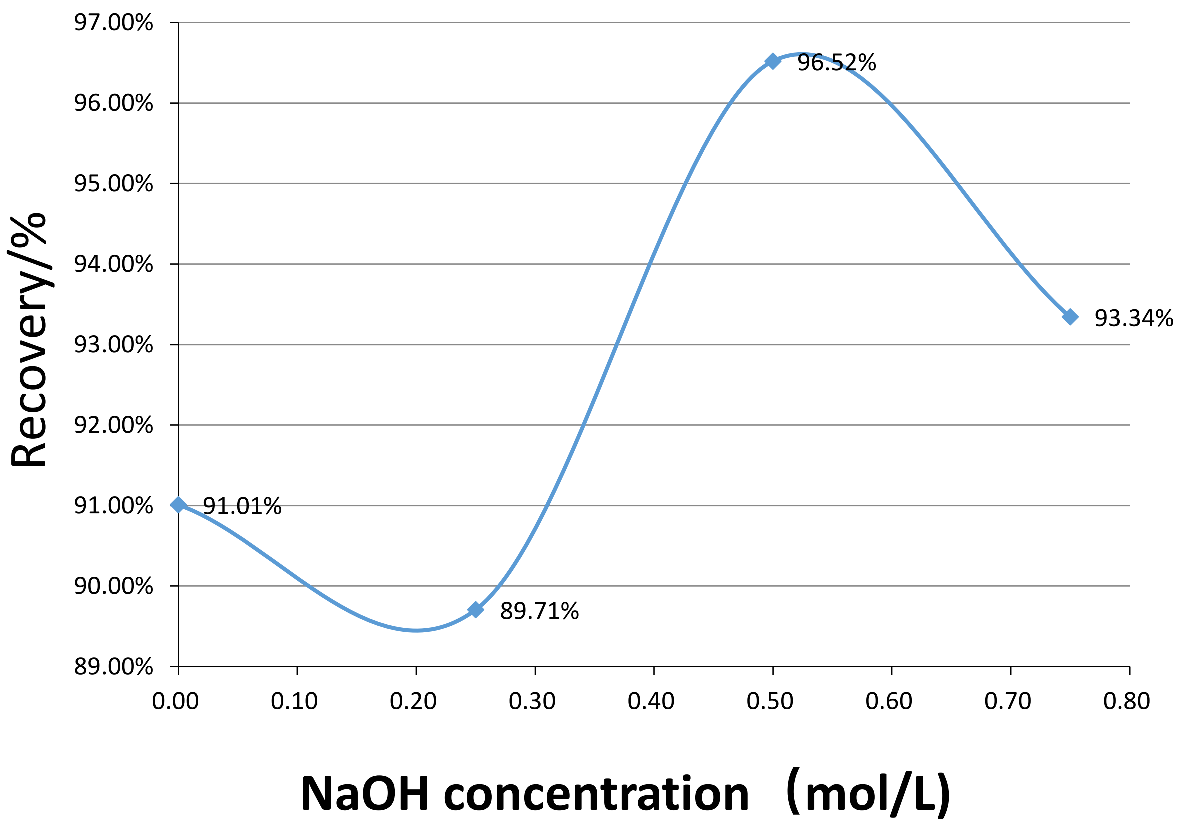
- Trap solution and the concentrations
- combustion protocol and temperature
- Combustion time time
- Carrier gases and flow rate

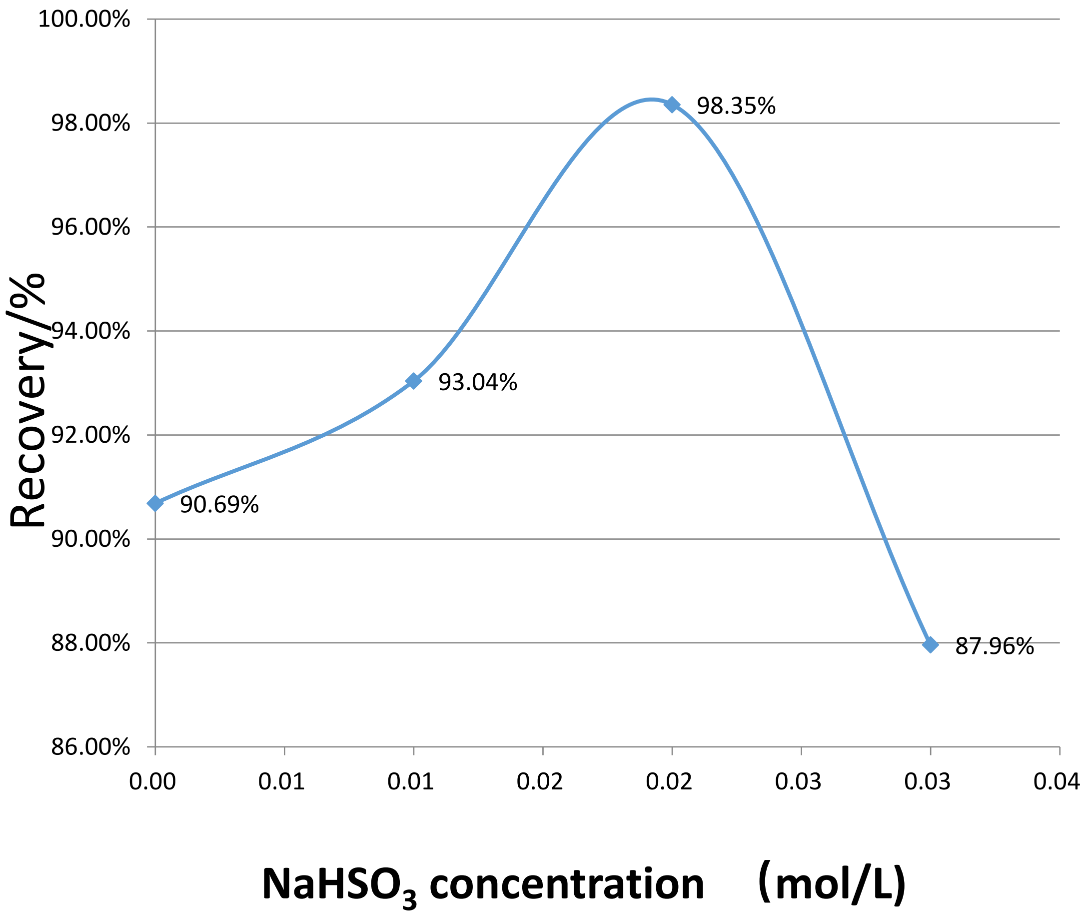
$$3I_2 + 6NaOH = 5NaI + NaIO_3 + 3H_2O$$

$$2I_2 + 2SO_3^{2-} + O_2 \rightarrow 4I^- + 2SO_4^{2-}$$

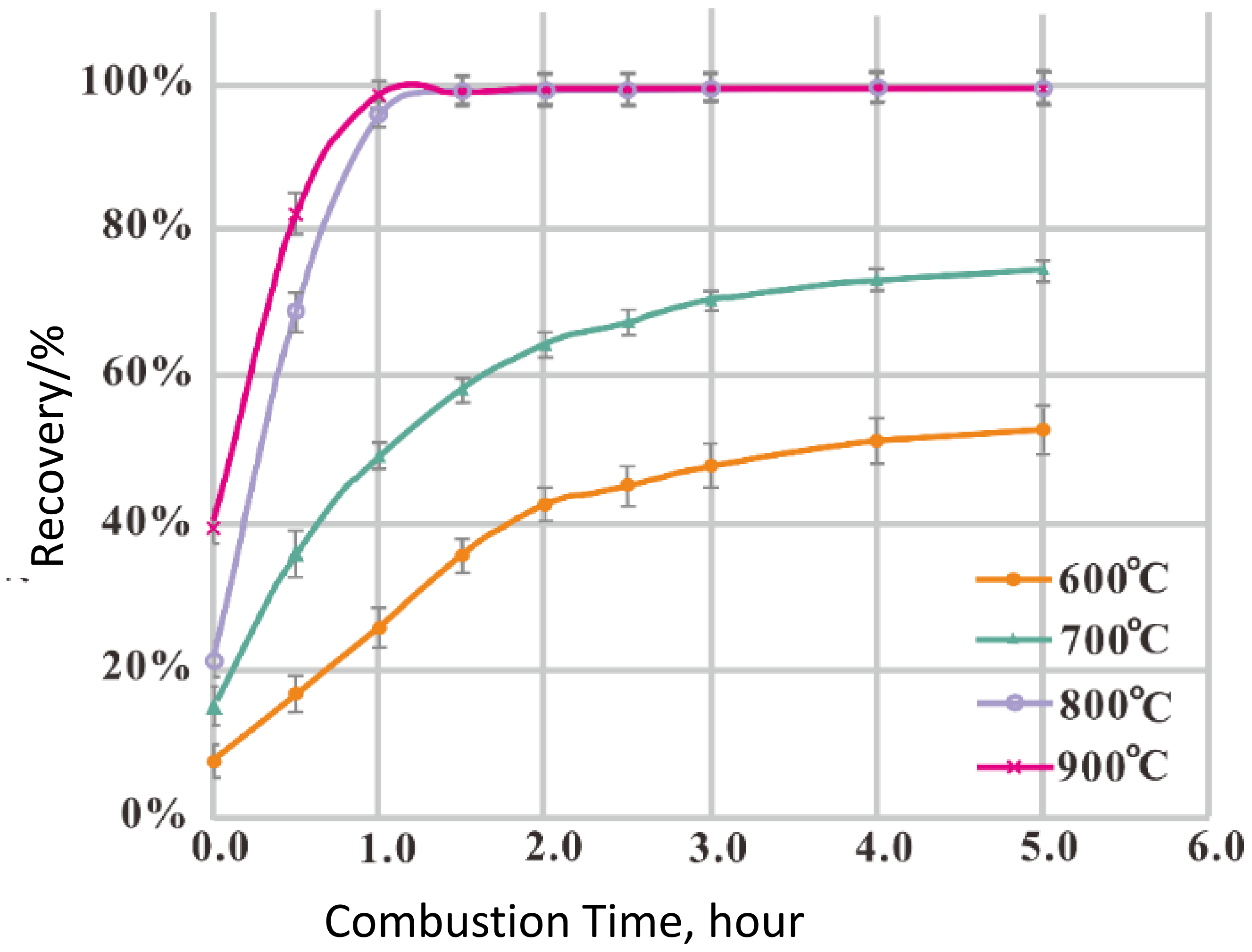
$$I^-(IO_3^-) + O_2 \rightarrow I_2$$

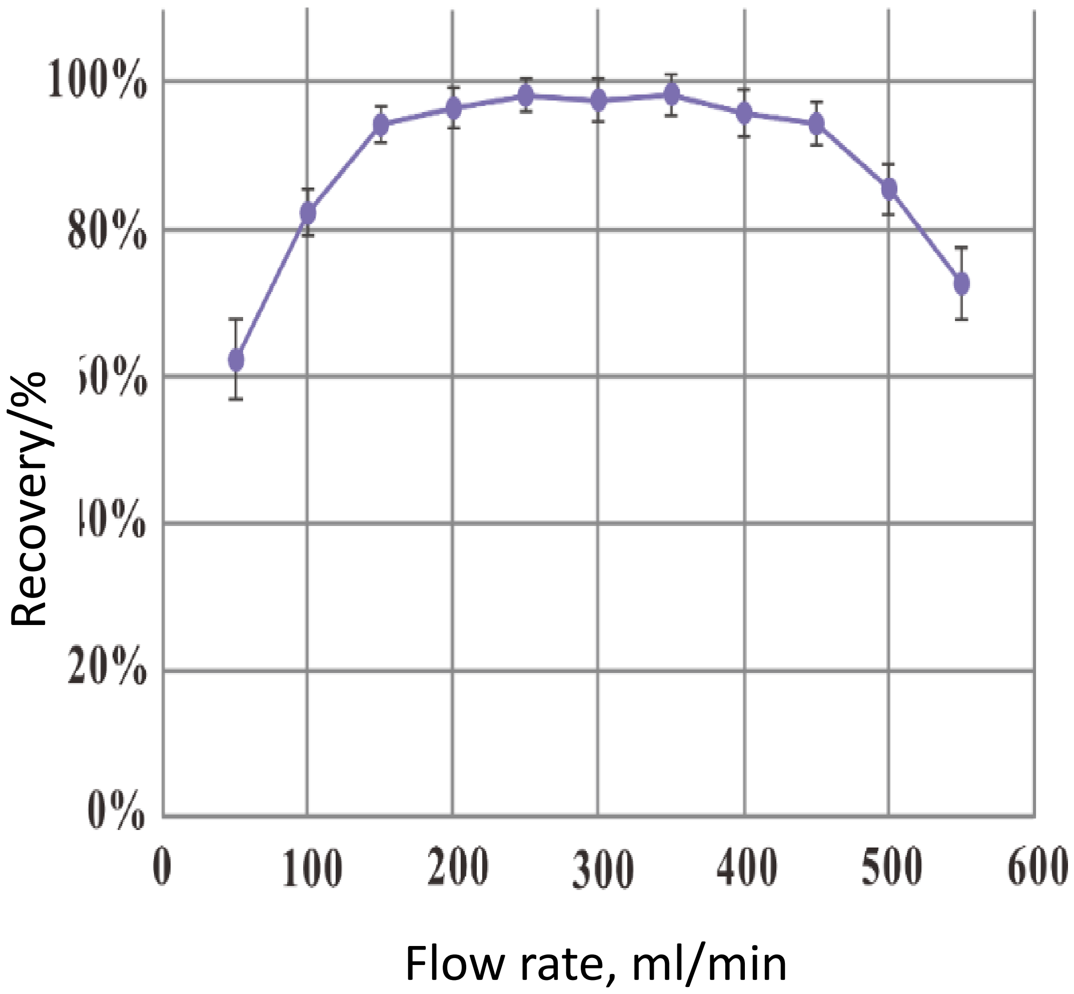
Separation of iodine from soil/sediemnt using combustion





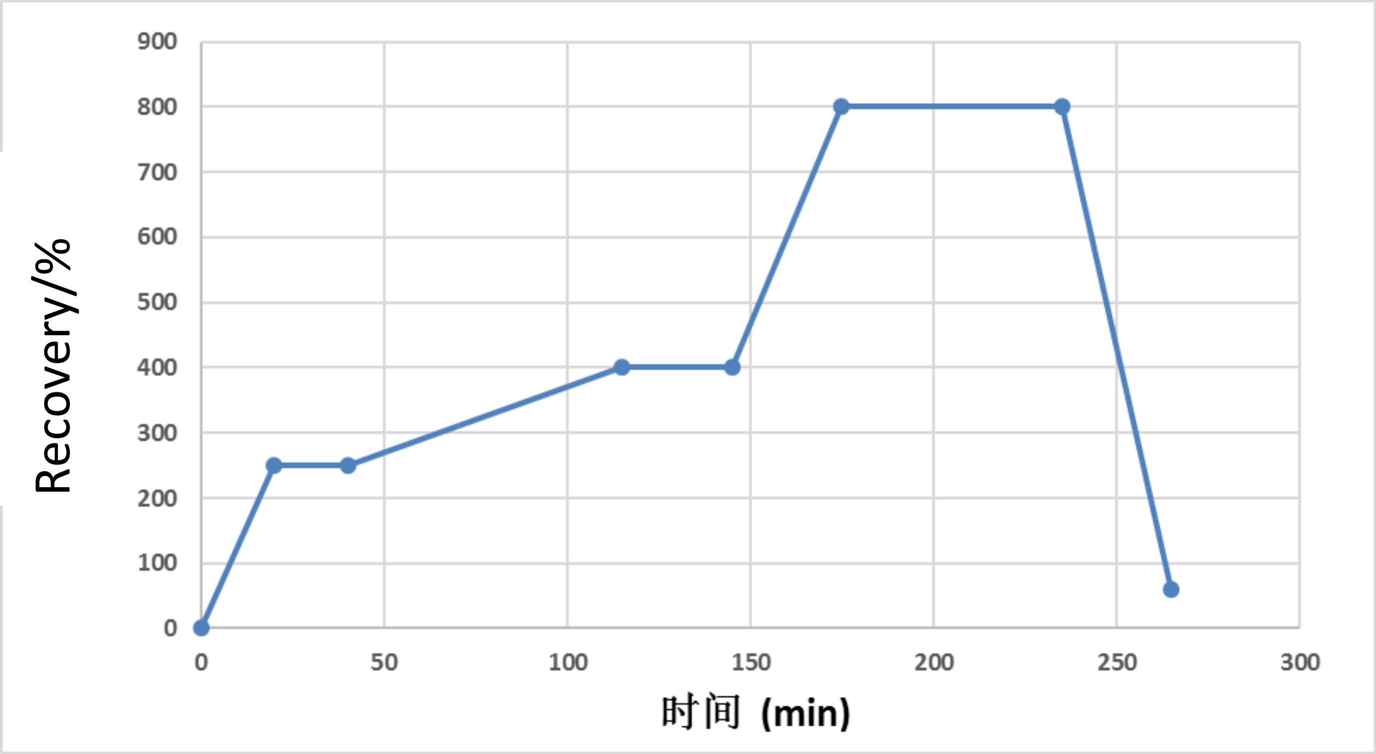
Separation of iodine from soil/sedimnt using combustion

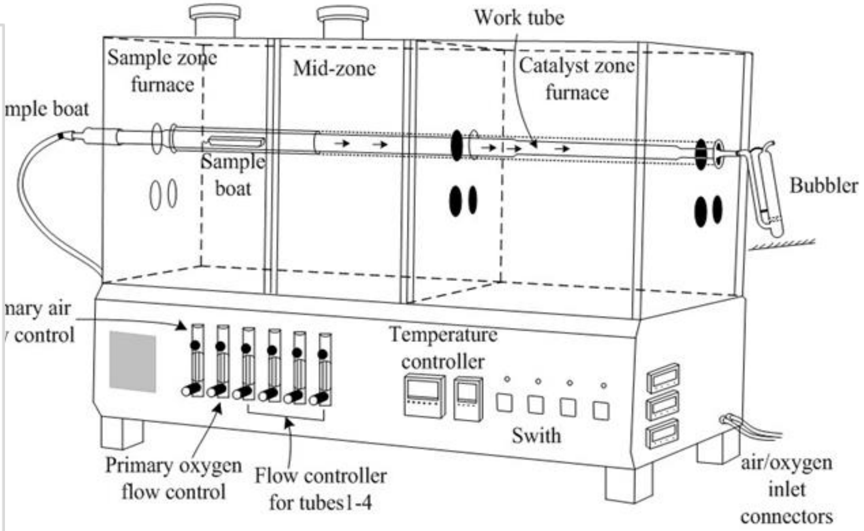




Zhang & Hou. JRNC, 2018

Separation of iodine from soil/sedimnt using combustion

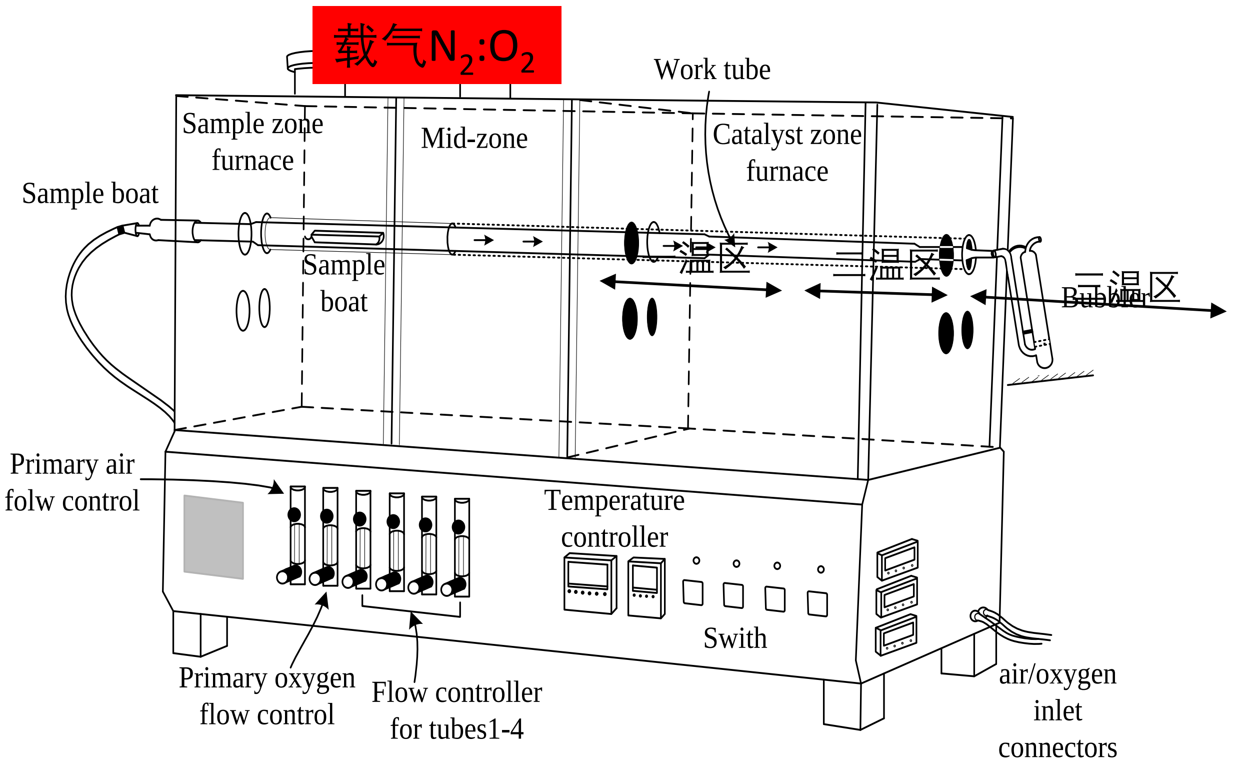




Temperarture rising, min

Zhang & Hou. JRNC, 2018

Combiustion of vegetation sample for iodine separation



Experimental condition

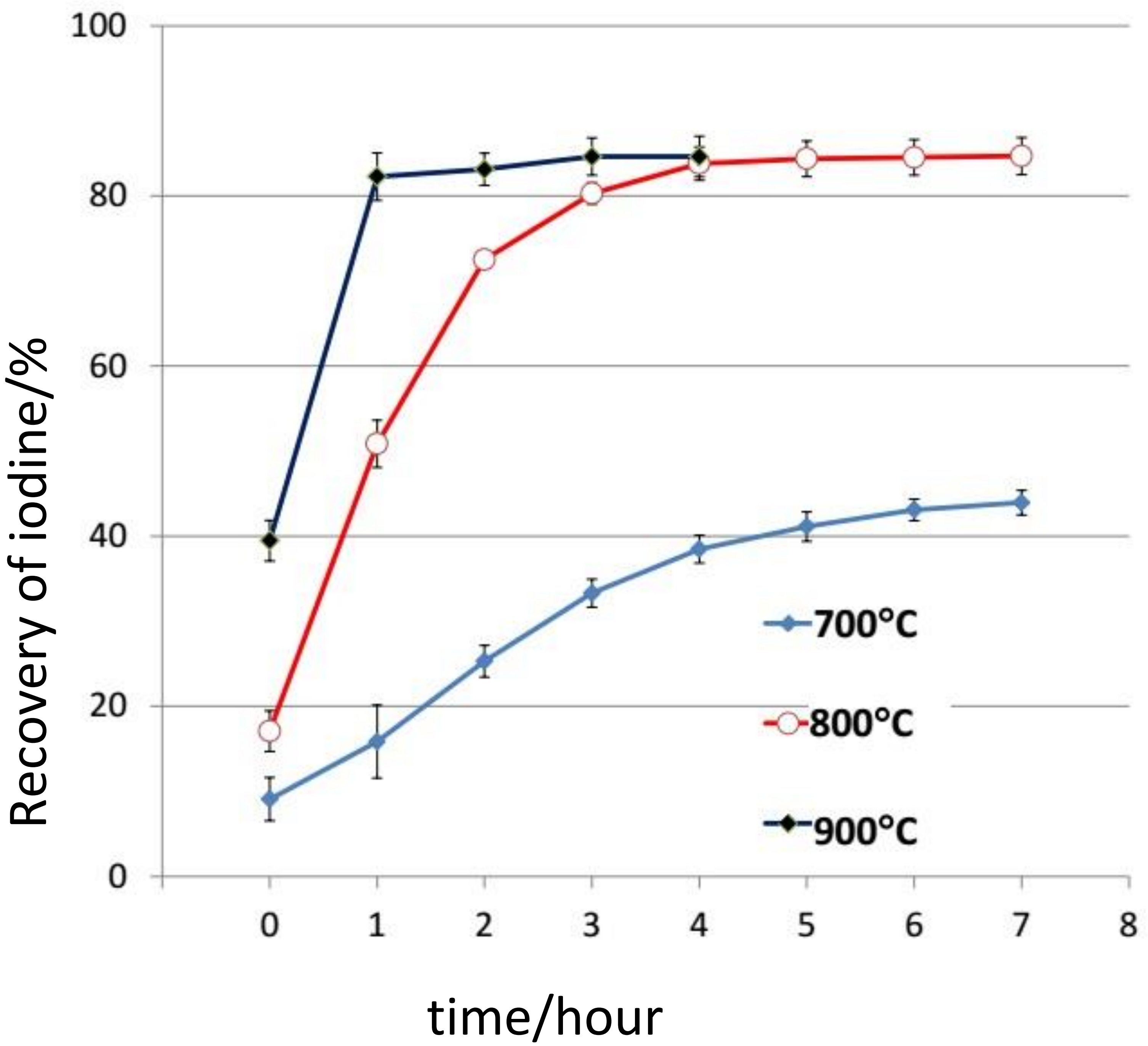
Igniting point： 220-300℃

- Temperature ramp： 1-1.5℃/min

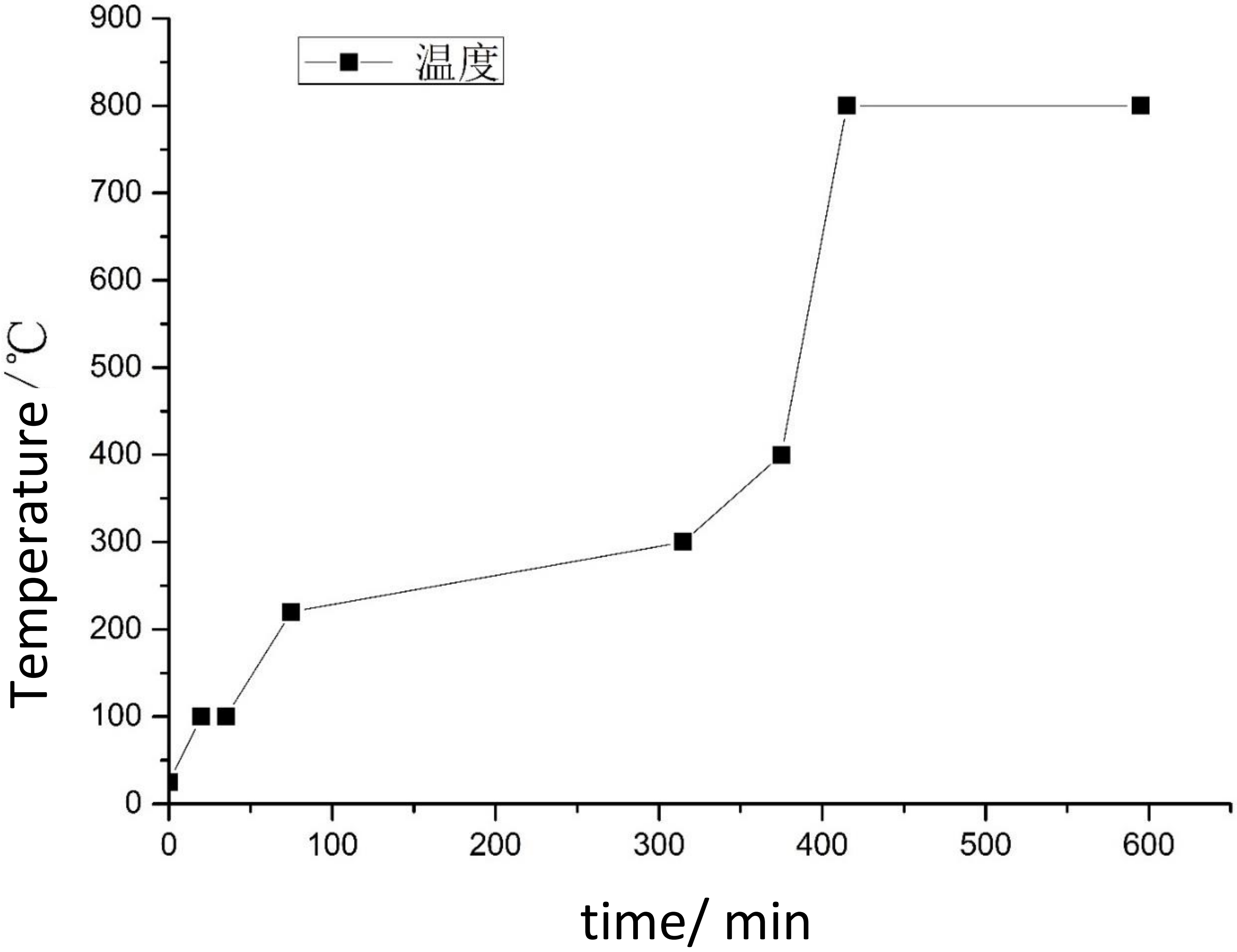
Assistant gas O_2： N_2=1:2-1:3，

- Gas flow rate： 100-200ml/min
- Temperature in Zone 3 : :900℃
- Mass of sample: 5-10 g

Influence of temperature and duriation

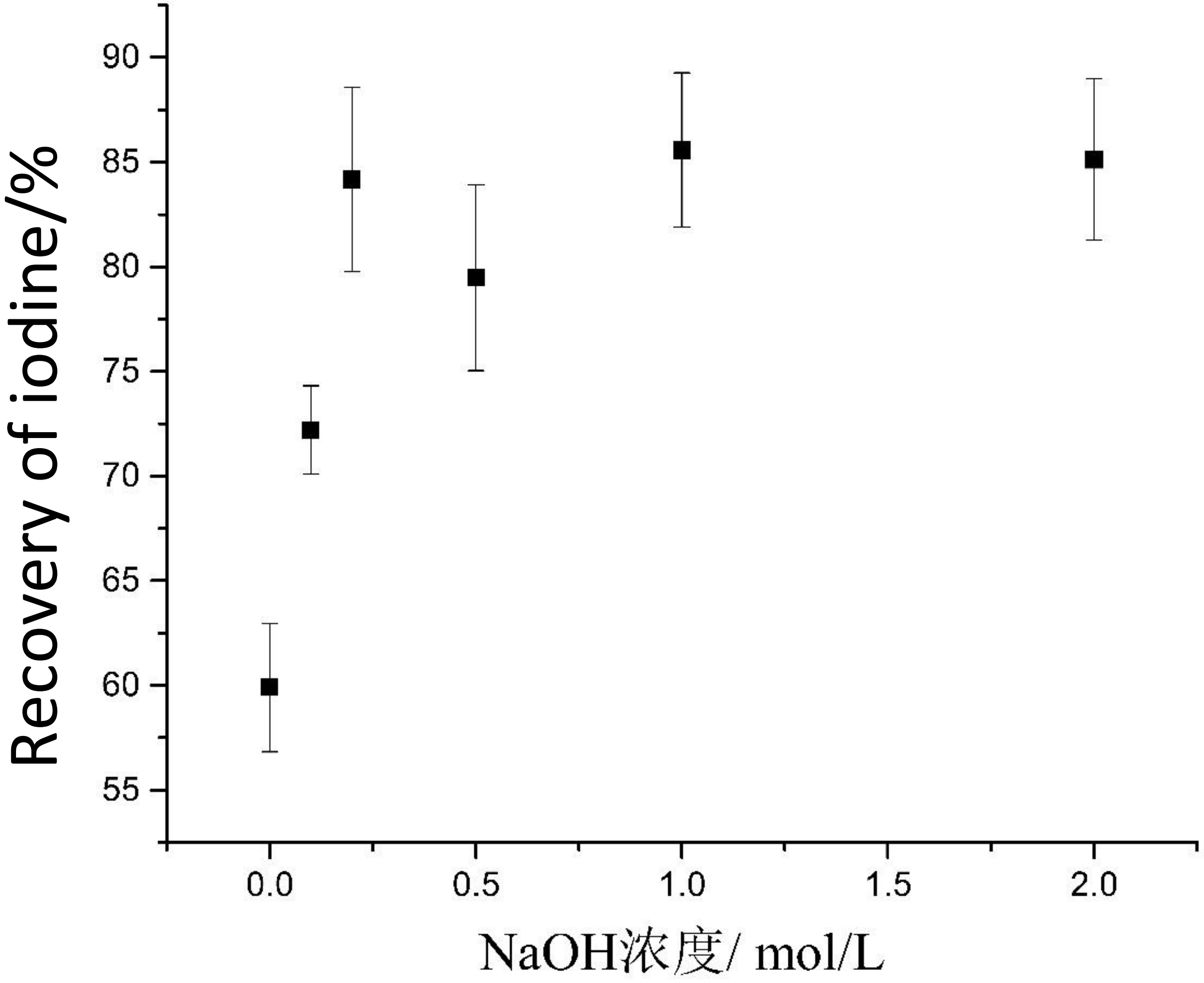


Temperature ramp protocol



Trapping solution

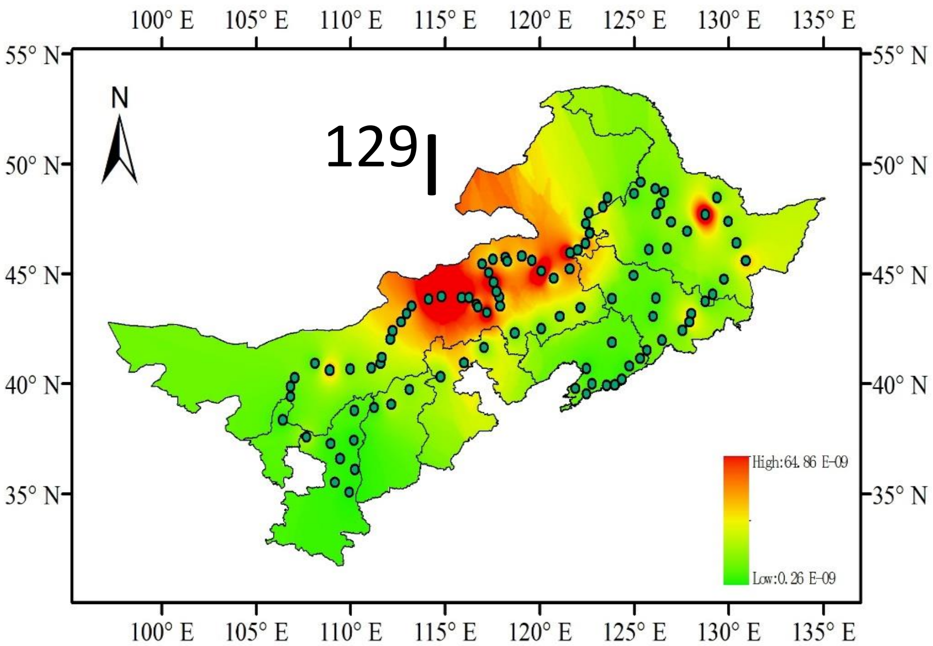
NaOH
– $NaHSO_3$ (0.05M)

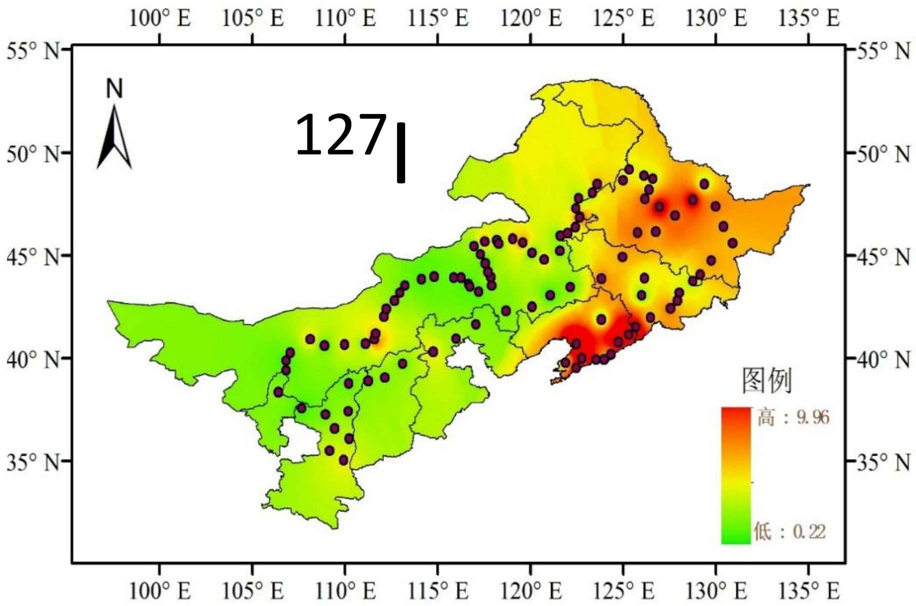


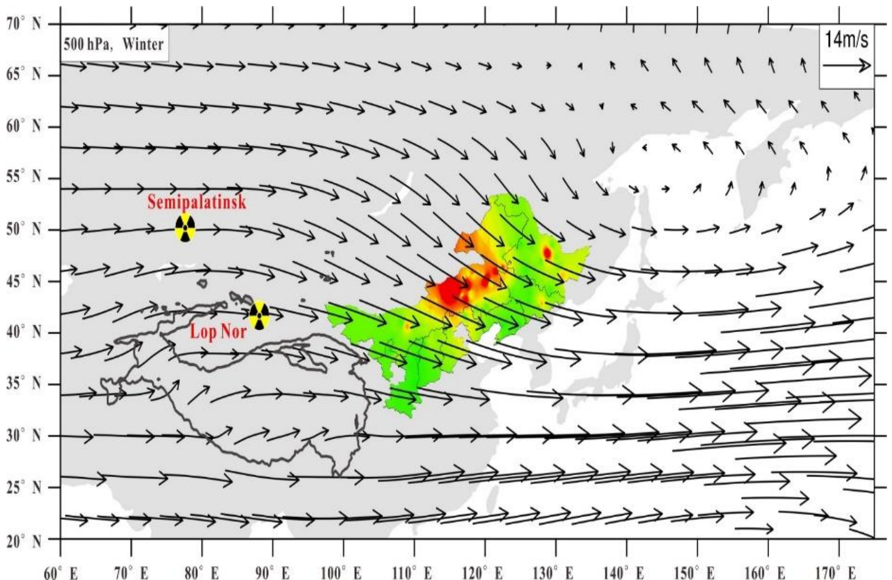
Comparison of combustion and alkalin fusion for extraction of iodine from vegetation samples

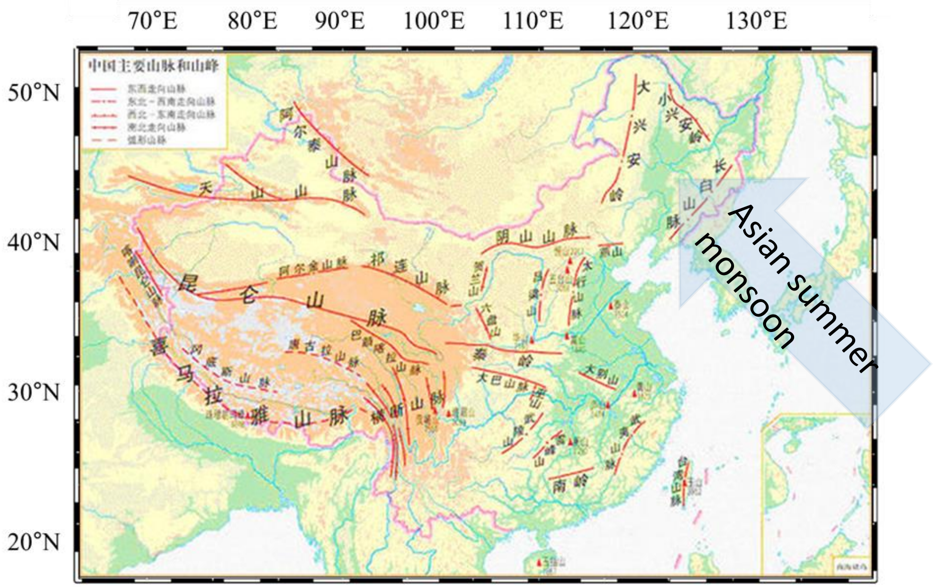
Sample	Sampling site	^{127}I conc, ppm	-^{129}I conc, 10^6 atoms/g		-^{129}I/^{127}I ratio, x10^{-9}		Recovery of iodine, %	
			Combustion	Fusion	Combustion	Fusion	Combustion	Fusion
Pine needle	Xi'an	0.49±0.02	11.9±2.42	9.30±0.94	4.77±0.71	4.01±0.40	78.5±3.1	45.8±2.9
Lichens	宝鸡	6.06±0.28	262±27.1	258±23.3	9.14±0.94	8.98±0.81	83.7±0.5	71.5±9.8
Spinach	宝鸡	1.73±0.08	61.3±6.30	56.5±5.31	7.44±0.76	6.86±0.64	86.7±4.1	77.0±2.2
Grass	西安	0.76±0.04	8.23±1.12	8.64±0.63	2.29±0.31	2.41±0.22	70.7±4.0	81.8±7.4
Seaweed	福建	3978±196	1840±130	1740±113	0.098±0.008	0.087±0.007	73.5±4.0	91.4±4.0

^{129}I in surface soil in Northeast China





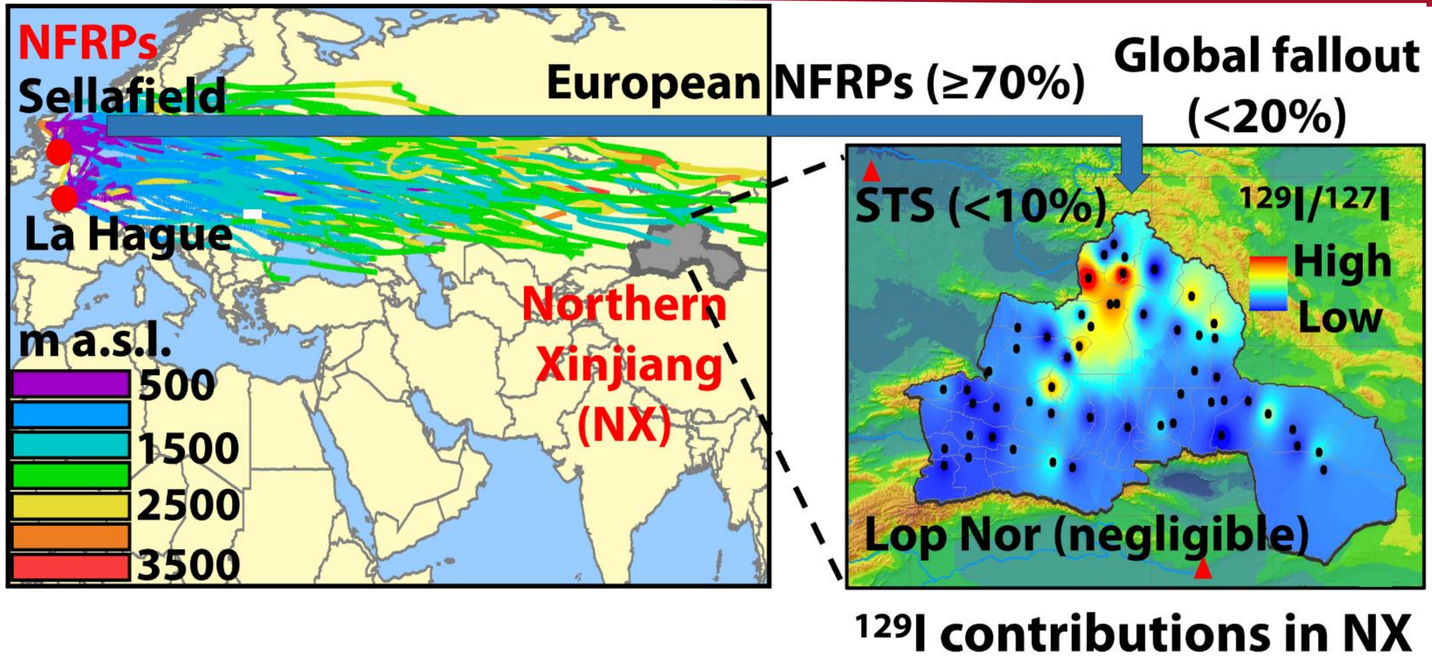


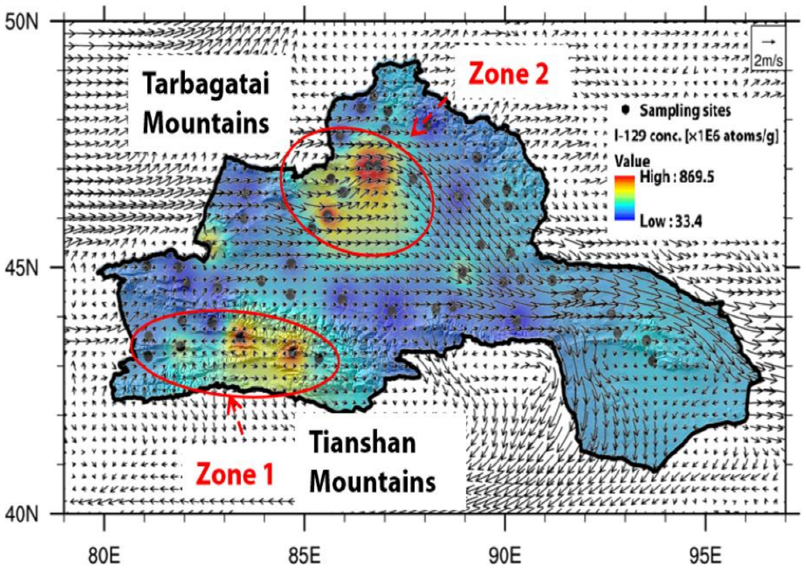


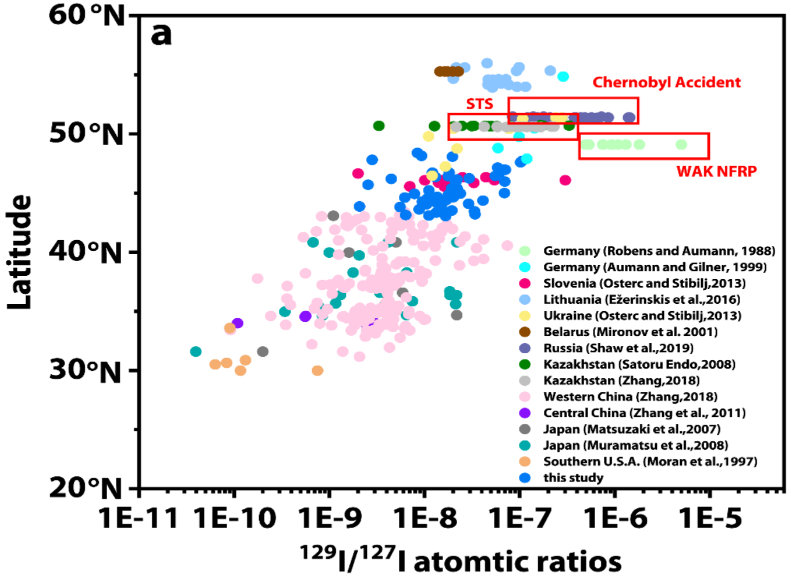
- Different distribution of ^{129}I ad ^{127}I
- The dominant westward wind pass through the Semipalatinsk test site and arid area, transport ^{129}I to east
- The topography of mountains and vegetation coverage make ^{129}I deposited and reserved in this region

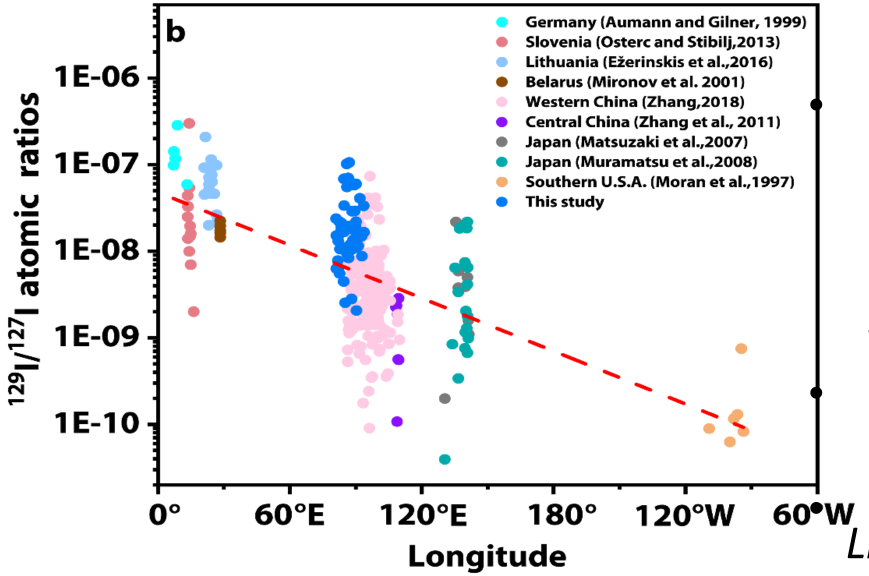
Wang, Hou, et al. STOTEN, 2024

^{129}I in surface soil in the Northwest China





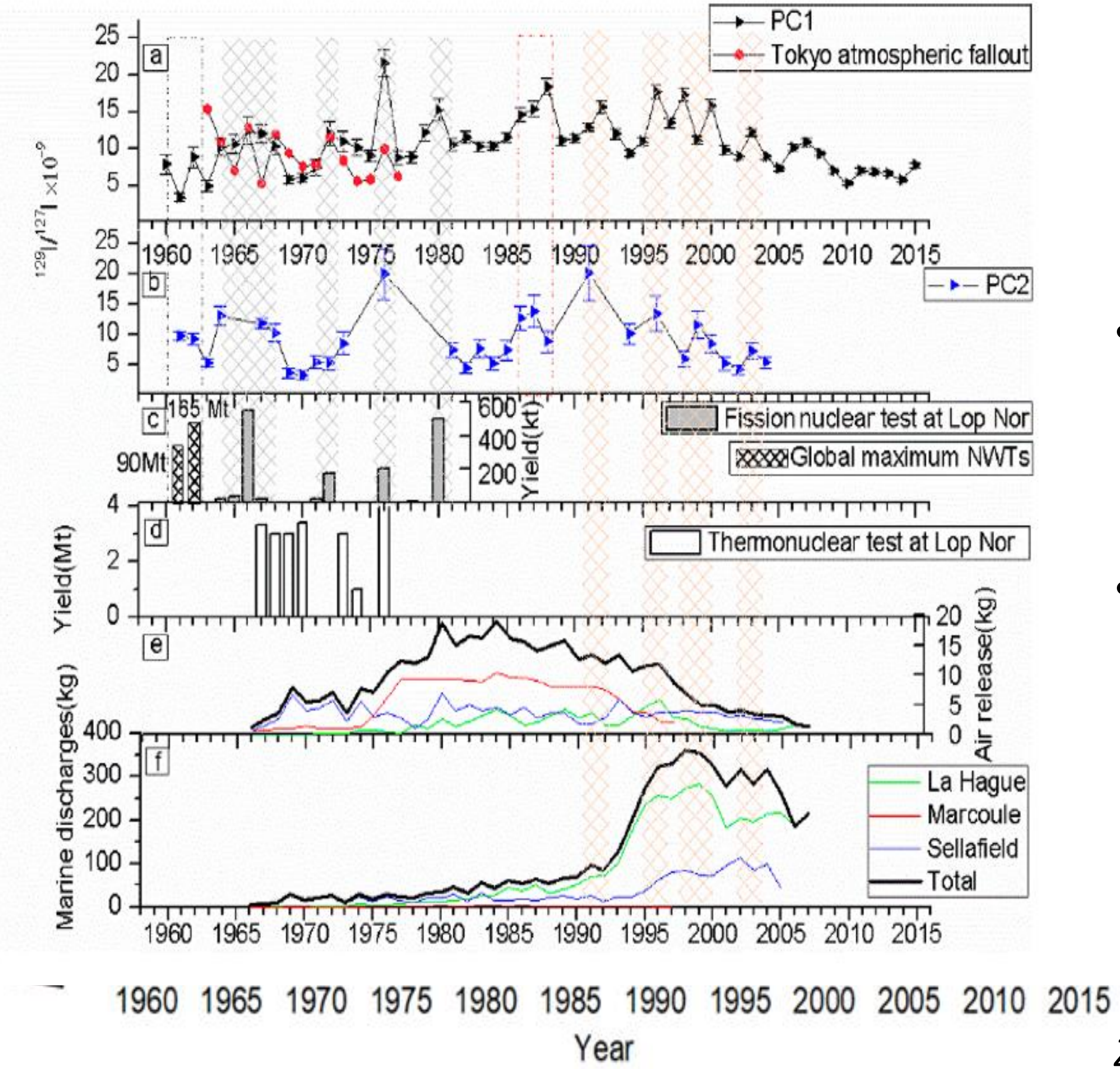




- ^{129}I in North China mainly originated from the European reprocessing plant through re-emission of marine discharged ^{129}I (>70%) driven by westlines.
- Global fallout (<20%) and regional deposition of wespons tests in Semiplatinsk (<10%) also contributed. No significant contribution from nuclear test in Lop Nor site.
- Topography dominate the transport pathway.

Liu, Hou, et al. ES&T, 2023, 57:10070-10078

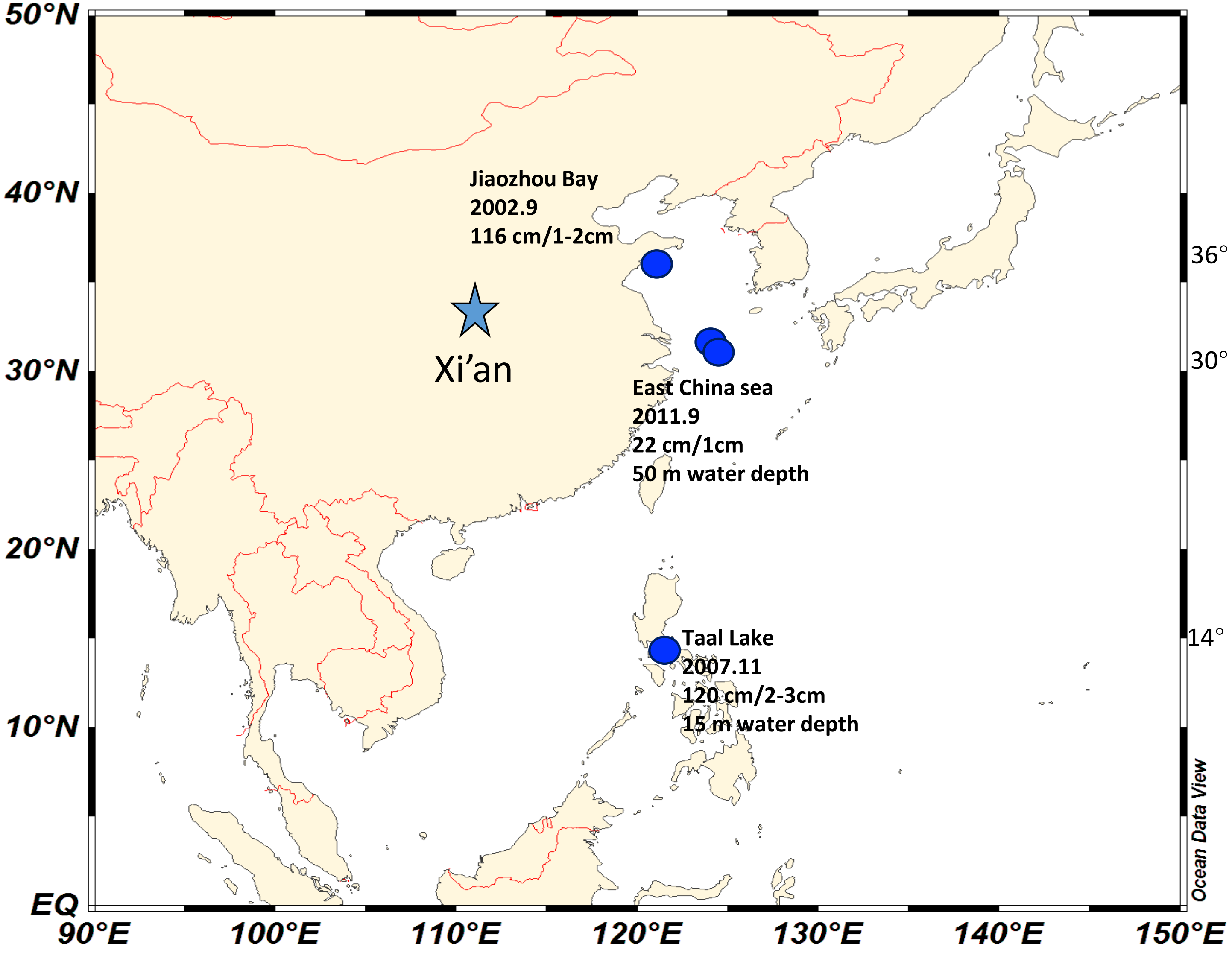
Variation of $^{129}I/^{127}I$ in tree rings from Northwest China (Qinghai)



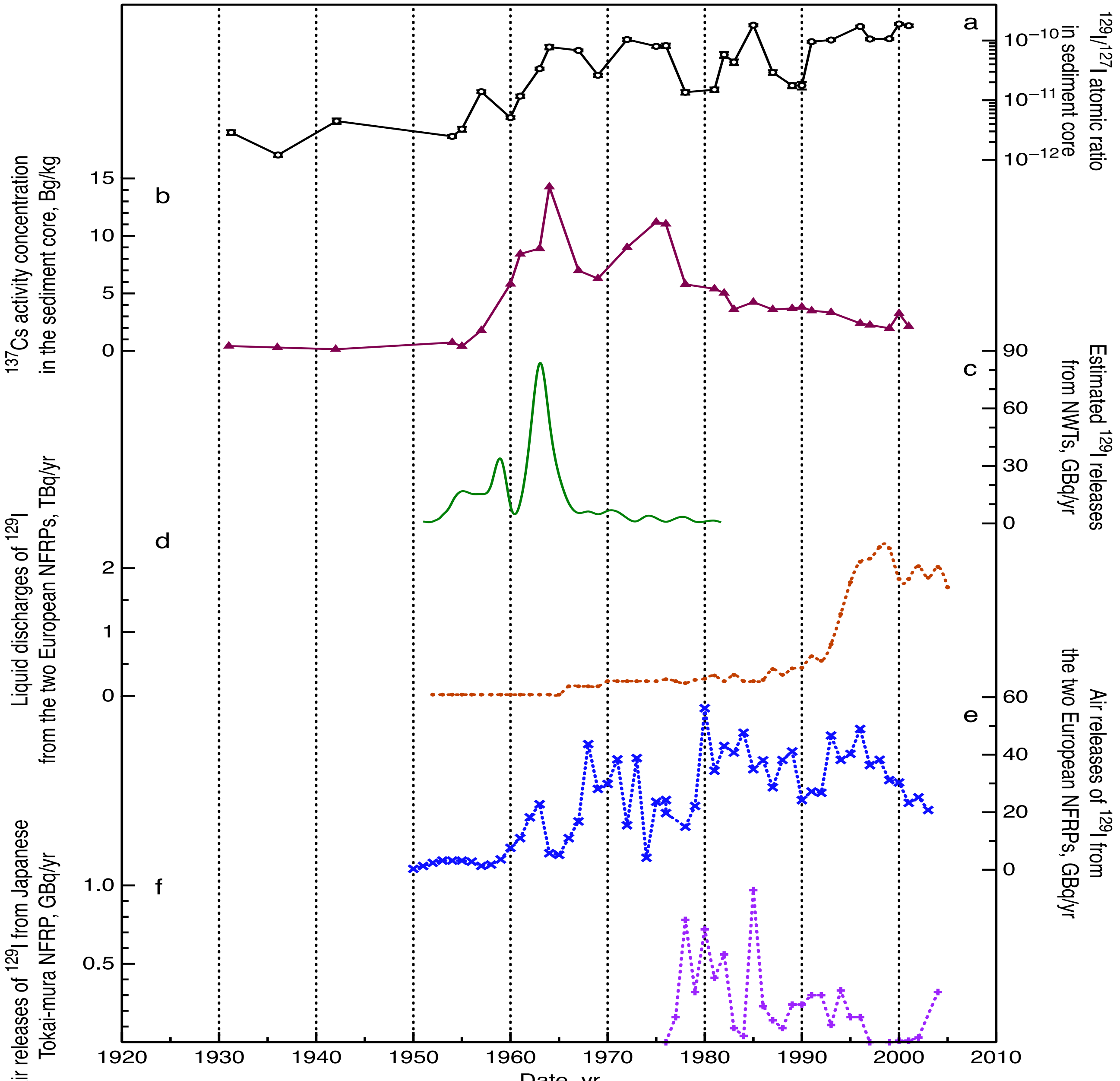
- Nuclear weapons tests including Chinese tests were recorded in the tree ring.
- Constant high ^{129}I level was observed after 1980, matched well with the increased discharges of ^{129}I from European reprocessing plants.

Zhao, Hou, Zhou, EST, 2019

Level of variation of ^{129}I in sediment cores at different latitudes in Asia



^{129}I profile in sediment core from Jiaozhou Bay, Yellow Sea

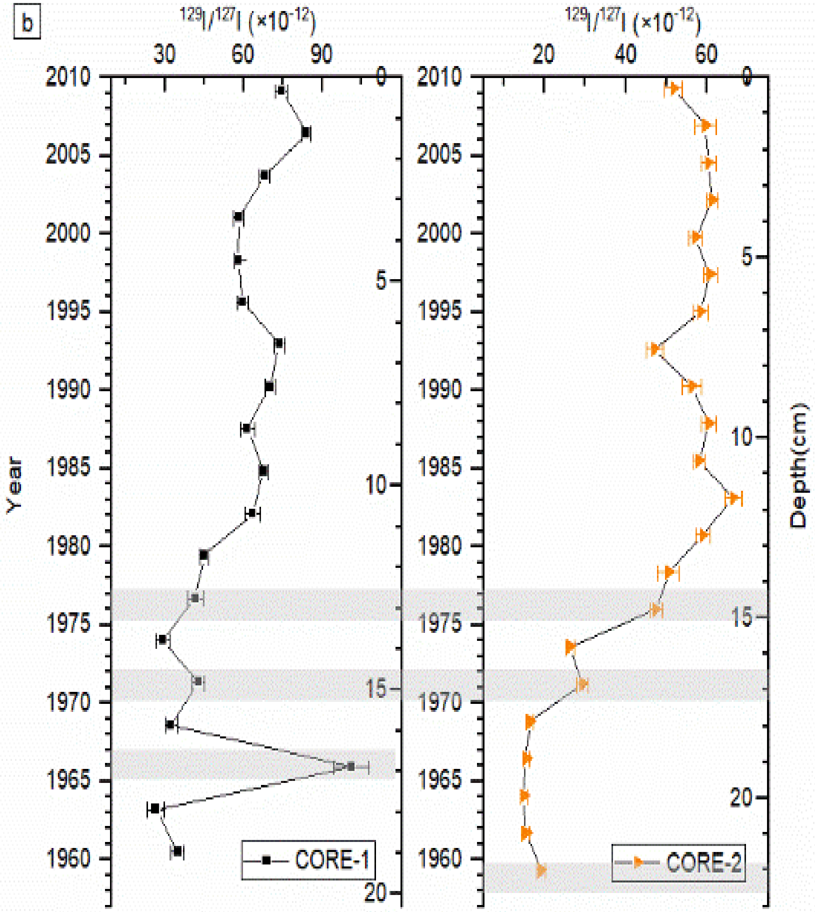


- **European reprocessing releases dominates the present ^{129}I in North China.**
- **Re-emission from European seawater is an important source of ^{129}I in North China.**
- **Signals of weapons testing at PPG and Chernobyl accident signal was clearly recorded in the sediment core.**

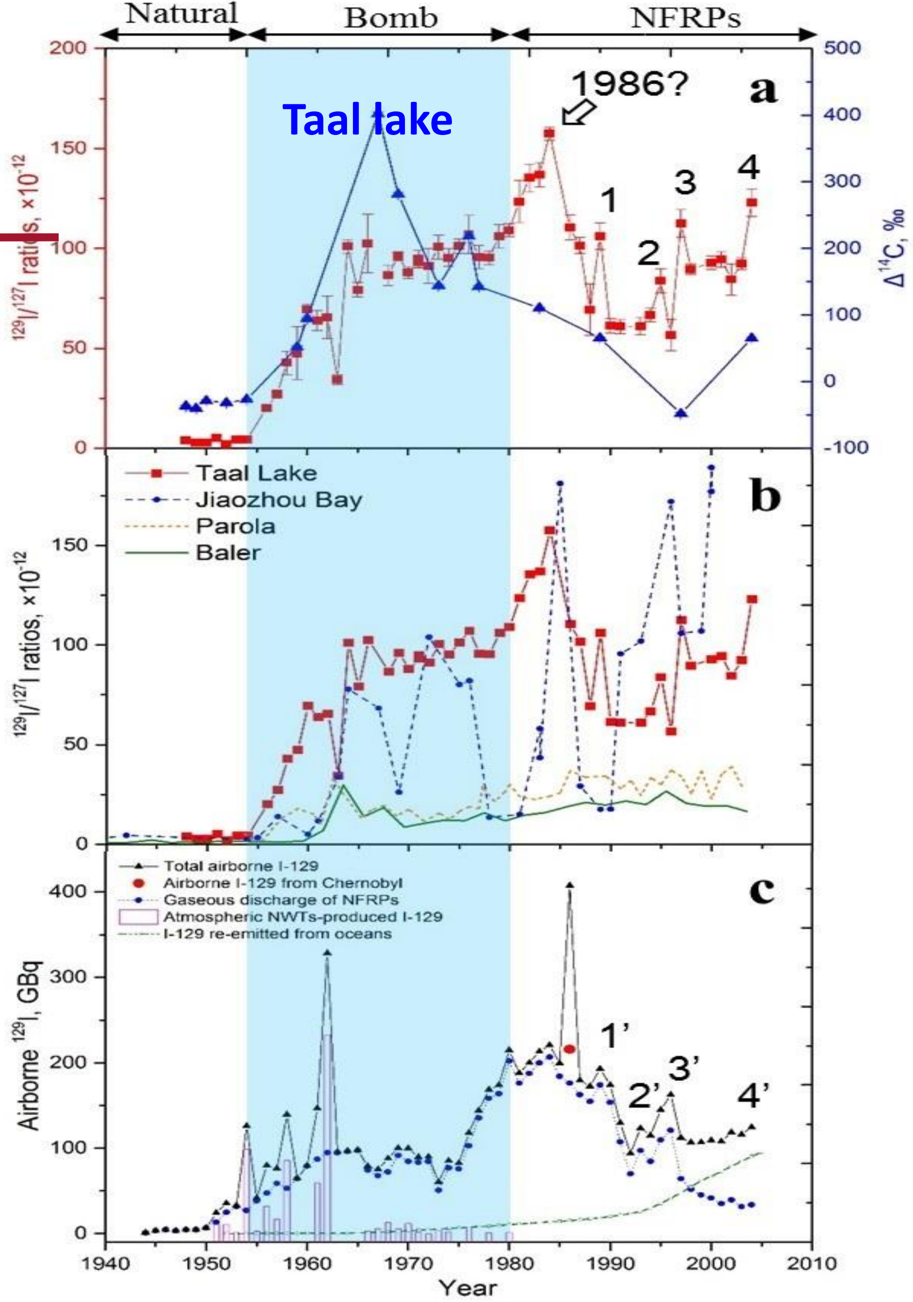
Fan, Hou, et al. JER, 2016

$^{129}I/^{127}I$ profile in sediment cores from the East China Sea and Taal lake, Philipines

East China Sea



Zhao, Hou, et al. Envir. Poll. 2019



Zhang, Hou, et al. Chemosphere, 2018

Thanks you for your attention