Urgent environmental survey on groundwater pollution by sewer leakage after the 2016 Kumamoto Earthquake

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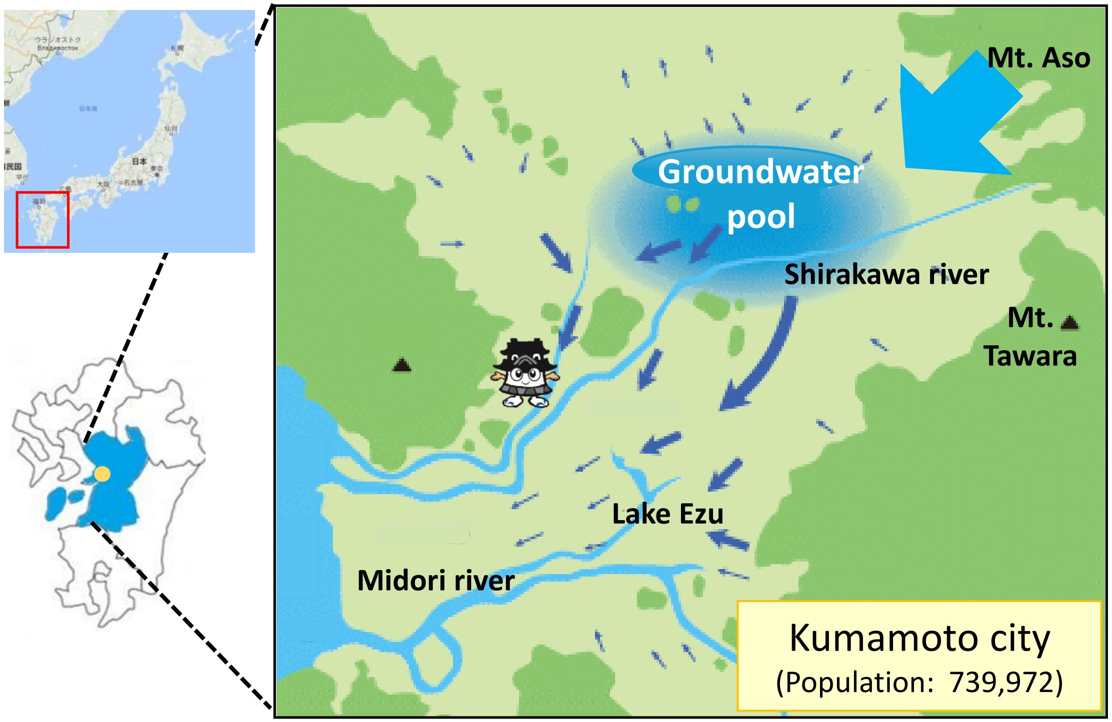
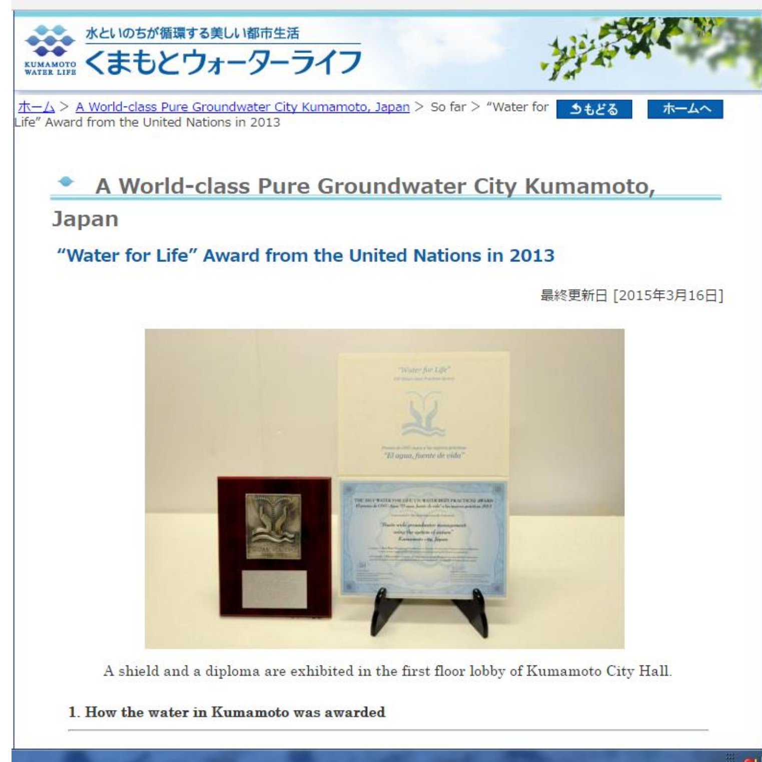
1) Kumamoto University, JAPAN
2) Kumamoto Prefectural University, JAPAN
3) Saga University, JAPAN
4) State University of New York, Albany, USA

Background



Groundwater flow in Kumamoto area[1)2)]


水といのちが循環する美しい都市生活
KUMAMOTO WATER LIFE
くまもとウォーターライフ

ホーム > A World-class Pure Groundwater City Kumamoto, Japan > So far > "Water for Life" Award from the United Nations in 2013
もどる ホームへ

A World-class Pure Groundwater City Kumamoto, Japan

"Water for Life" Award from the United Nations in 2013

最終更新日 [2015年3月16日]

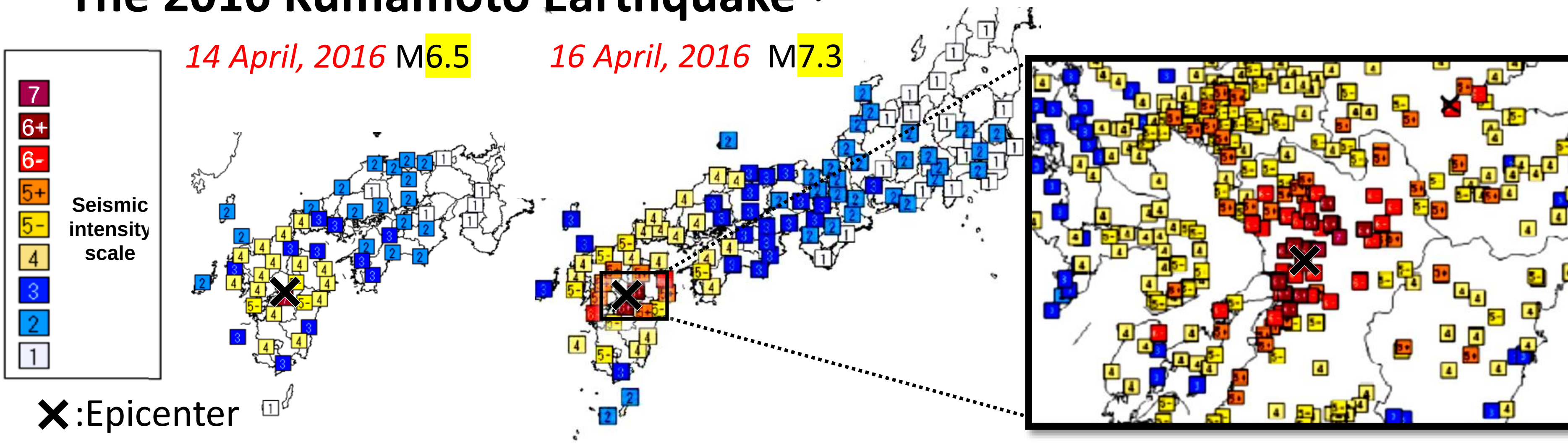
A shield and a diploma are exhibited in the first floor lobby of Kumamoto City Hall.

1. How the water in Kumamoto was awarded

1) http://www.kumamoto-waterlife.jp/ 2) Manabe (2010)

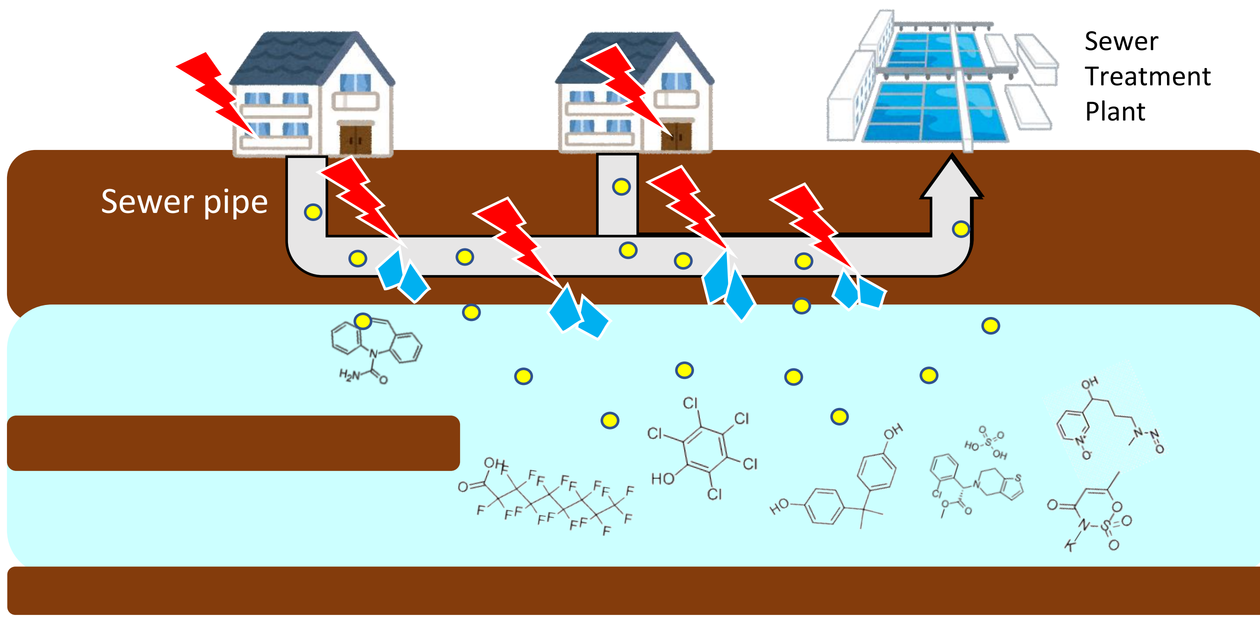
Background

The 2016 Kumamoto Earthquake[1)]



Broken manhole after the earthquake[2)]

(14,Nov. 2016 18:00)



1) http://www.jma.go.jp/jma/press/1604/16a/kaisetsu201604160330.pdf
2) http://www.mlit.go.jp/common/001136237.pdf

Objectives

1. Target analysis

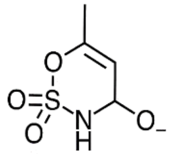
To evaluate the sewer pollution in groundwater by comparing persistent water-soluble compounds and nutritive salts concentrations before/after the 2016 Kumamoto Earthquake as chemical tracer.

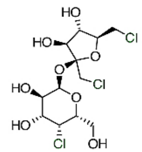
2. Non-target analysis

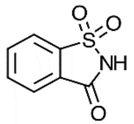
To investigate the occurrence sites of unknown chemicals in groundwater after the 2016 Kumamoto Earthquake by non-target analysis.

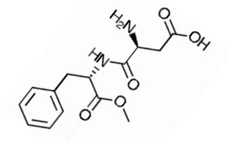
Target and Non-target analysis

Artificial Sweeteners (ASs)

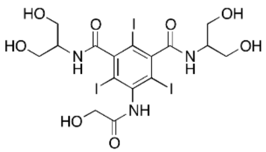

Acesulfame (ACE)

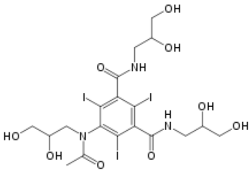

Sucralose (SUC)

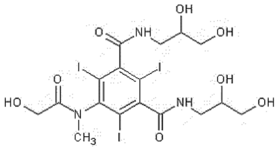

Saccharin (SAC)

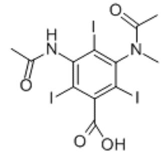

Aspartame (ASP)

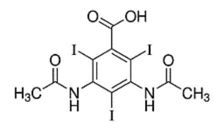
Iodinated X-ray contrast media (ICMs)*)

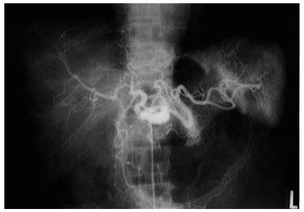

Iopamidol (IOP)


Iohexol (IHX)

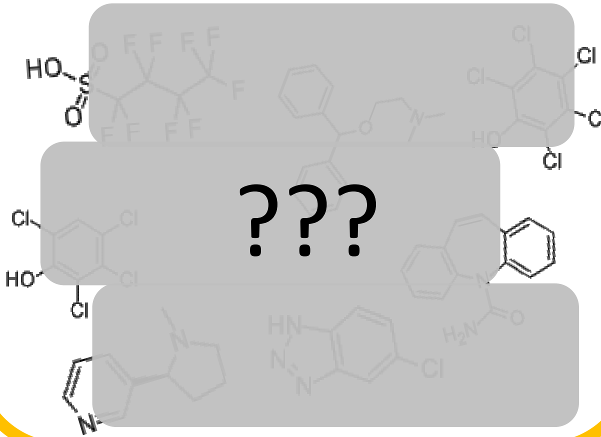

Iomeprol (IMP)

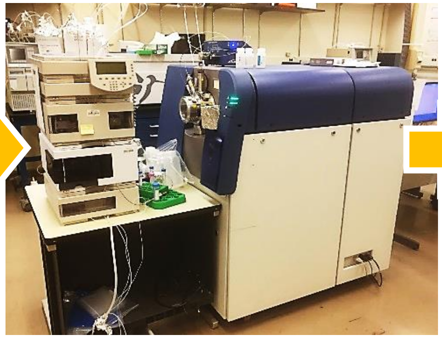

Metrizoic acid (MTA)


Diatrizoic acid (DIA)

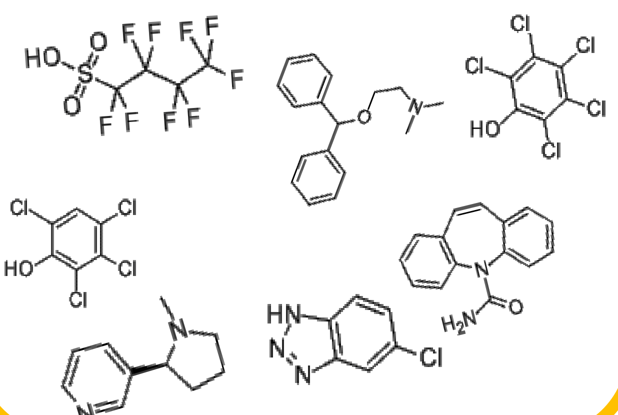


Non-target



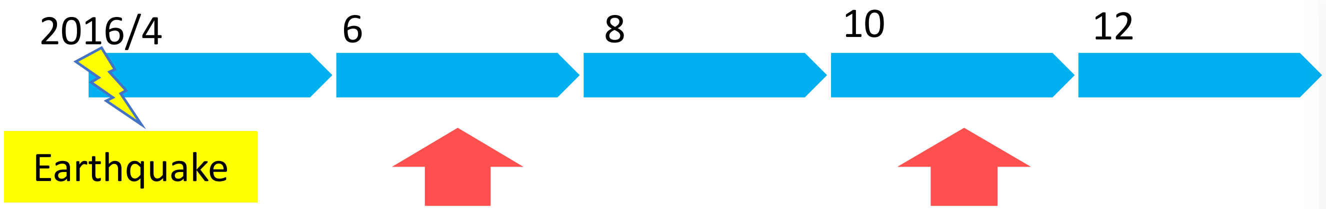

LC-TOFMS

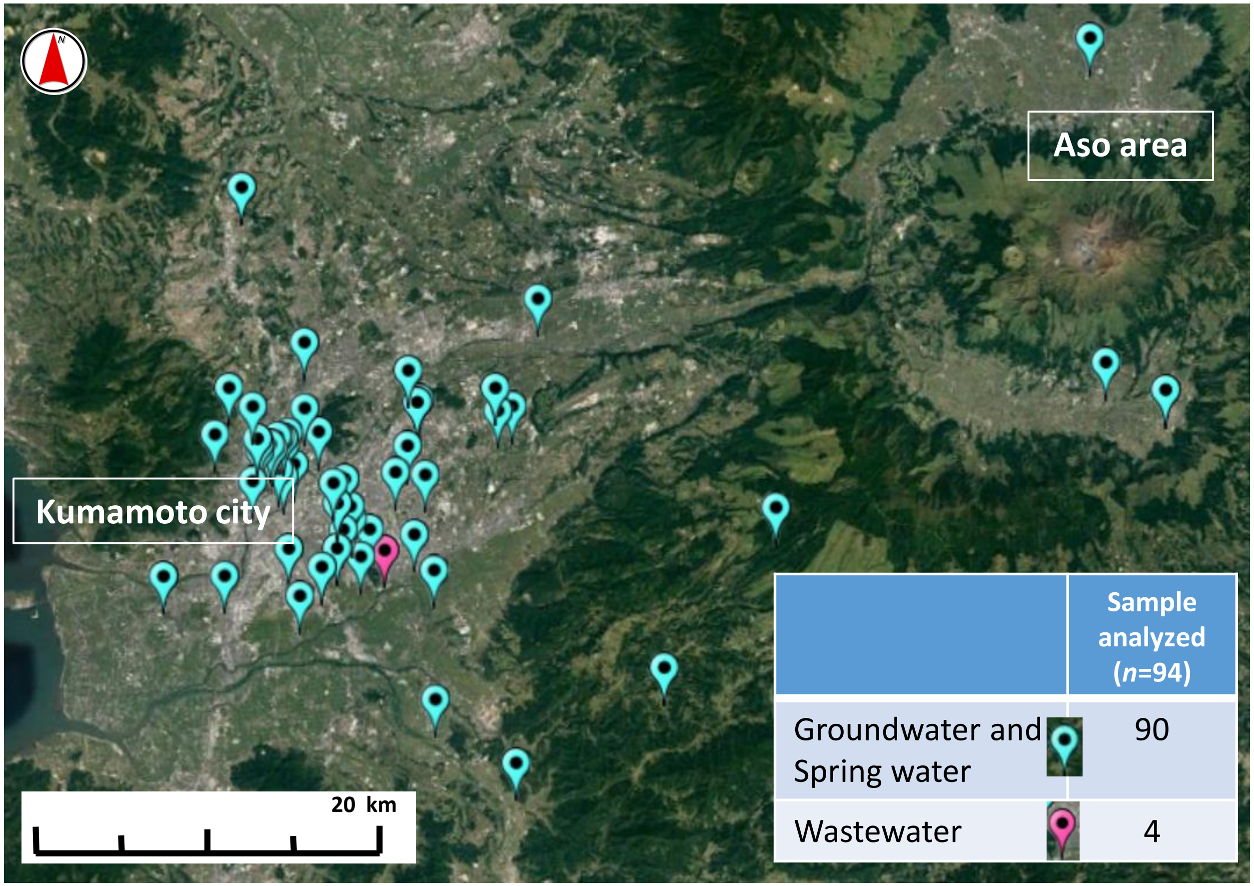
Identification



*) Yabuki *et al.* (2011)

Samples





	Sample analyzed (n=94)
Groundwater and Spring water	90
Wastewater	4

Method

Target analysis *)

Aqueous sample (2 L)

Filtration

Adjustment of pH (<2.8)

← Add acesulfame-*d4*

Solid Phase Extraction

Centrifugation

Elution

Dryness and reconstitution

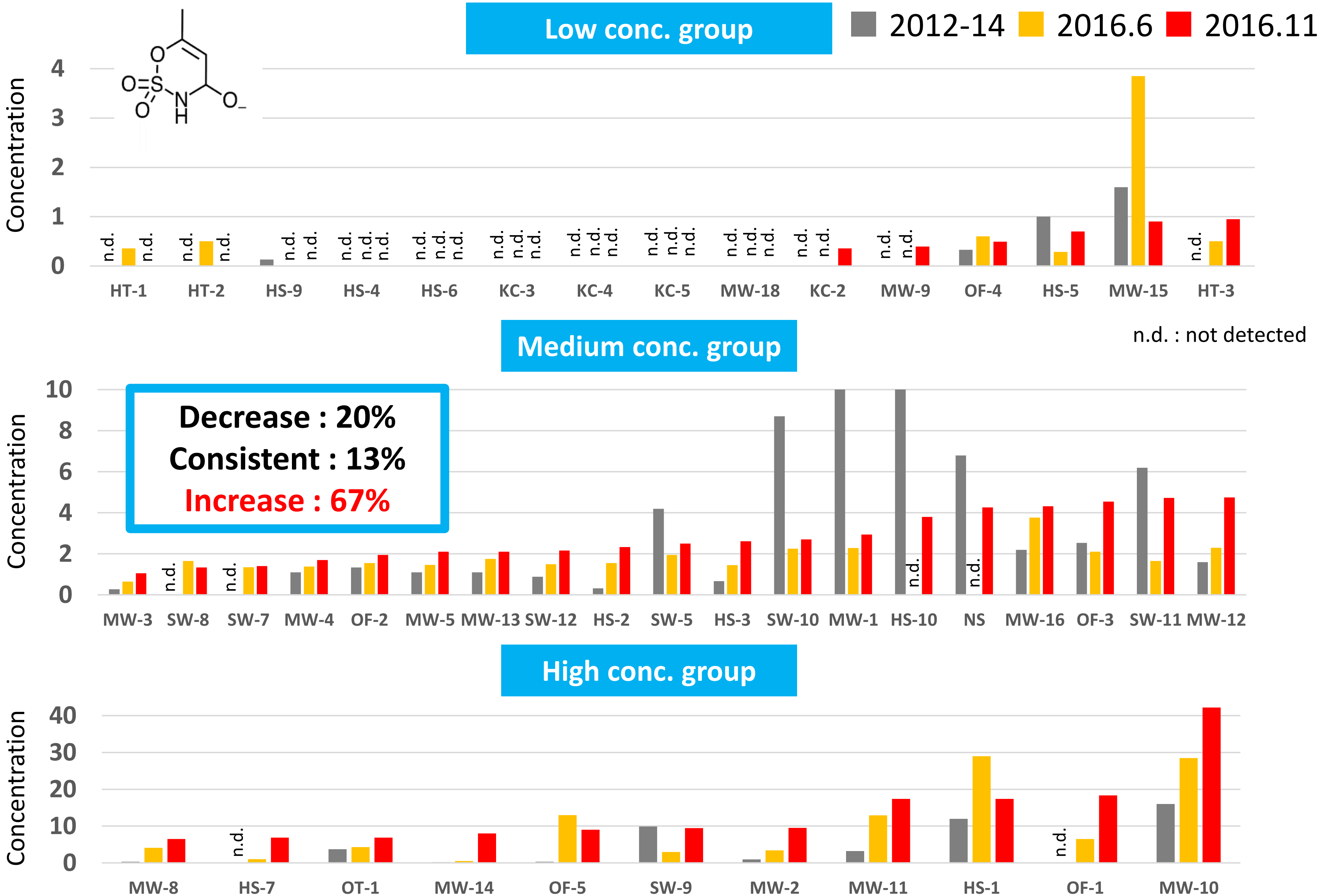
Non-target analysis

HPLC-MS or HPLC-MS/MS

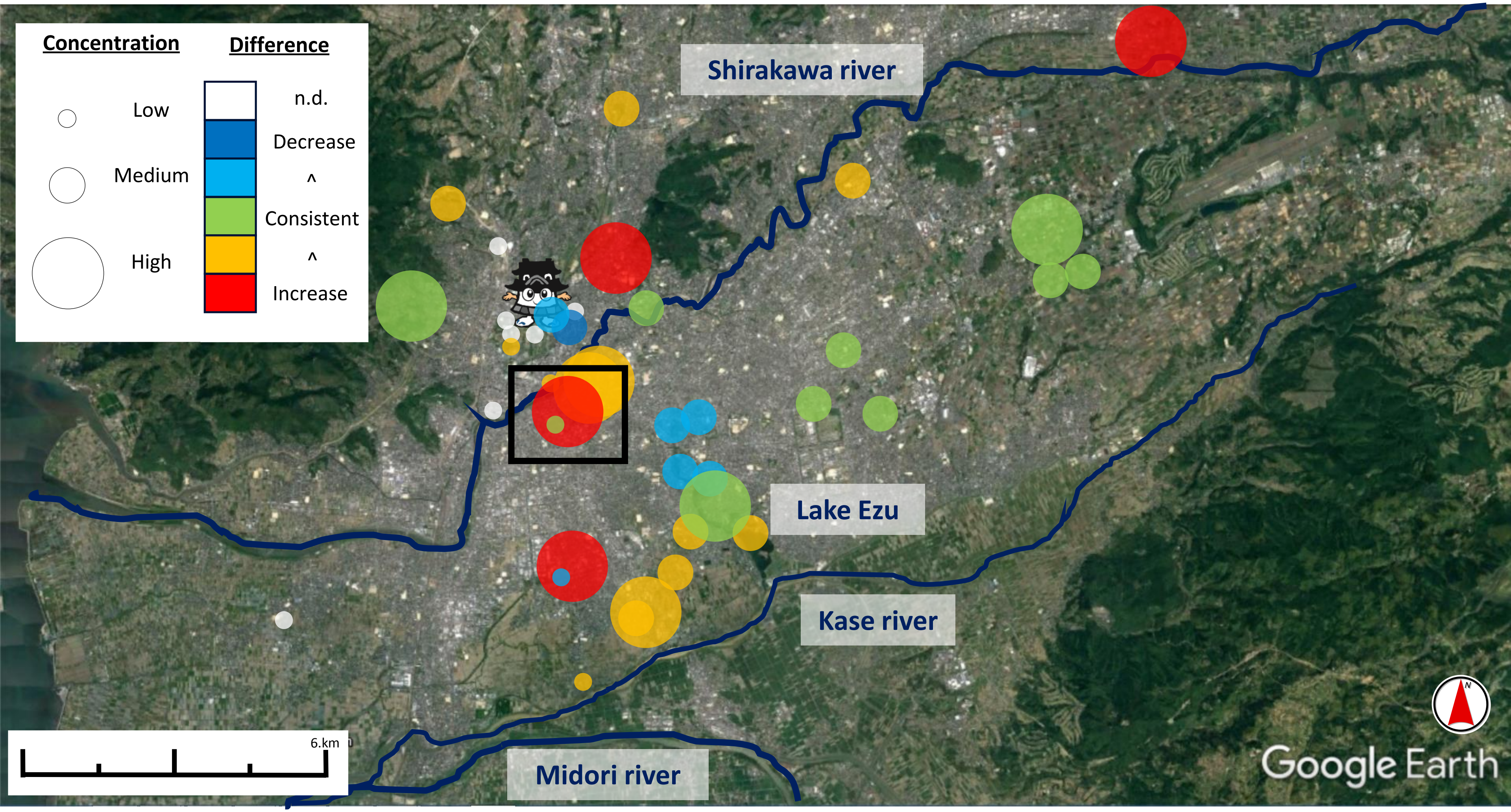
LC-TOFMS and HPLC-MS/MS

*) Watanabe *et al.* (2016)

Acesulfame concentrations in groundwater

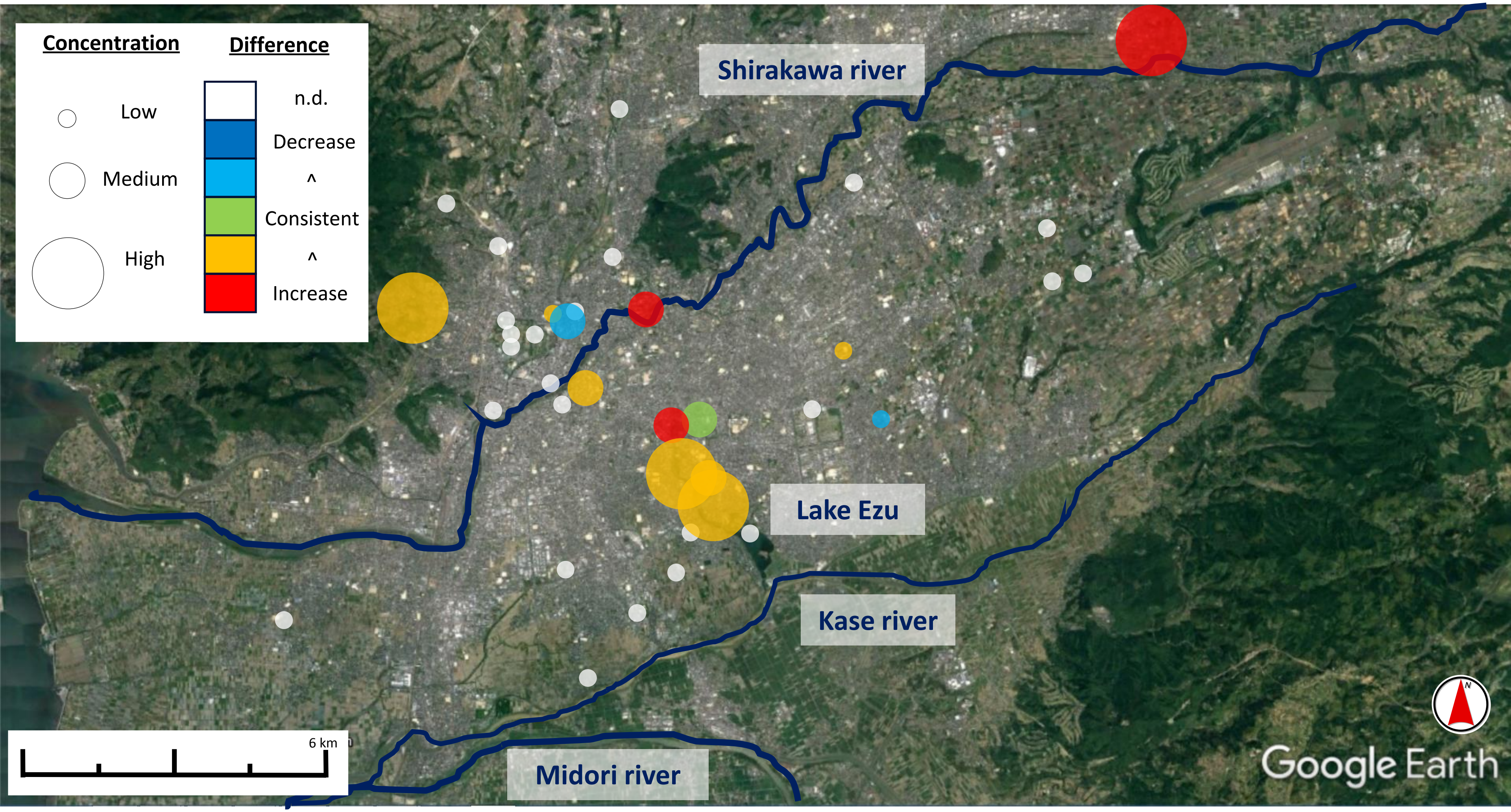


Comparison of acesulfame concentrations before/after the earthquake



Distribution of ACE concentrations varies, depending on sampling sites. → **Possibility of sewer pipes breaking**

Comparison of sucralose concentrations before/after the earthquake



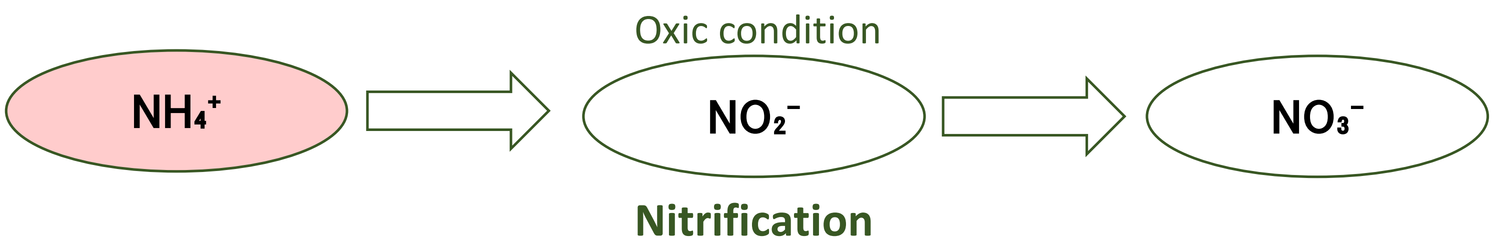
Sweeteners contained in wastewater may be heterogeneous, depending on sampling sites.

Background

- About one million peoples in and around the city entirely depend on groundwater for drinking purpose ⇒ nitrogen: a major environmental problems

＜Major source of nitrogen in GW＞

- **Leakage from sewage pipes**
- Application of chemical fertilizers
- Application and loading of manure



- **Chemical fertilizers and manure**

  Active nitrification in surface soil

  in aquifer ⇒ Nitrate (NO_3^-)

- **Sewage**

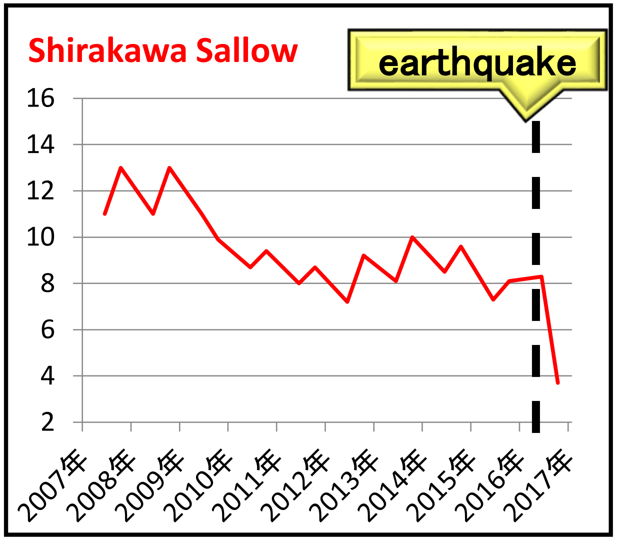
  Settled up under the surface

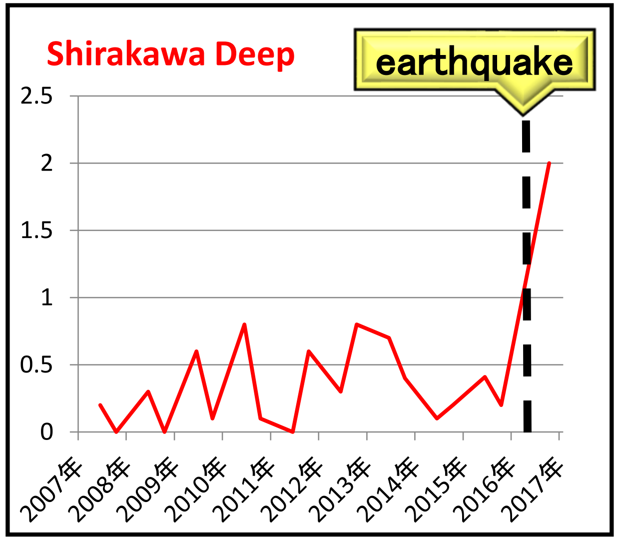
  in aquifer ⇒ Ammonium (NH_4^+)

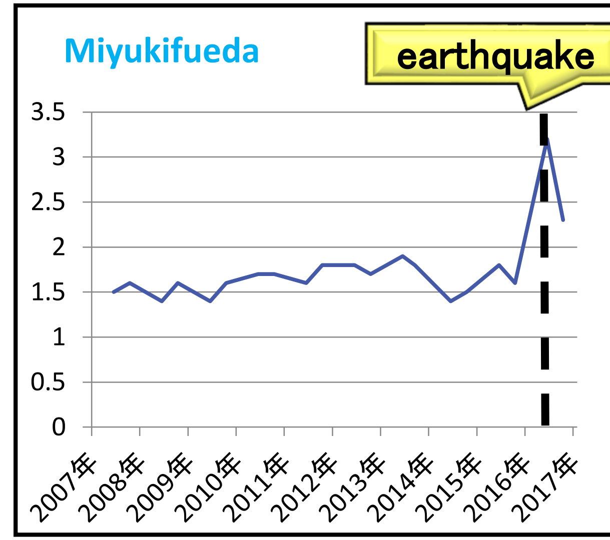
To check environmental status (NH_4^+・NO_2^-・NO_3^-)

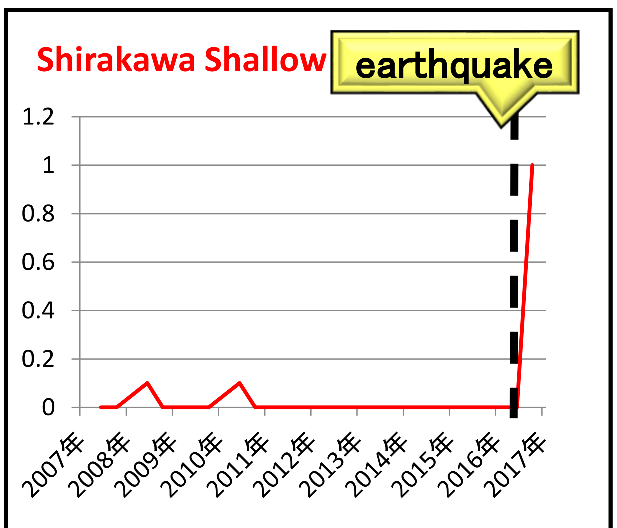
To begin, we checked long term monitoring data

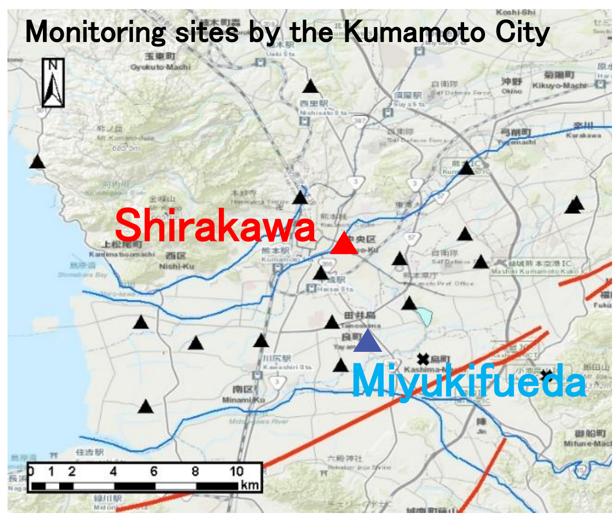
Significant change in NO_3^- and NO_2^- ⇒ 2 sites











Increasing in NH_4^+

⇓

Leaking of sewage

Effect of sewage leaking in nitrogen pollution

<sewage water before traetment>

NO_2^-	0.00 mg/L
NH_4^+	13.13 mg/L
NO_3^-	0.00 mg/L

⇒ nitrogen in sewage water exists as NH_4^+ from

＜Nitrogen compounds in groundwater＞

	Max
NH_4^+	0.74（mg/L）
NO_3-N＋NO_2-N	6.16（mg/L ）

（June）

	Max
NH_4^+	0.89（mg/L）
NO_3-N＋NO_2-N	6.89 （mg/L）

（November）

NH_4^+・・・ low level below than 1 mg/L.
NO_3−N＋NO_2−N ＜ 10 mg/L ⇒ Less than environmental standard values.
Systematic increase in nitrogen after the earthquake was not found.

Effect of sewage leaking in nitrogen pollution

<sewage water before traetment>

NO_2^-	0.00 mg/L
NH_4^+	[illegible] mg/L
NO_3^-	0.00 mg/L

⇒ nitrogen in sewage water exists as NH_4^+ form

<Nitrogen compounds in groundwater>

	Max
NH_4^+	0.74 (mg/L)
NO_3-N＋NO_2-N	6.16 (mg/L)

(June)

	Max
NH_4^+	[illegible] (mg/L)
NO_3-N＋NO_2-N	6.89 (mg/L)

(November)

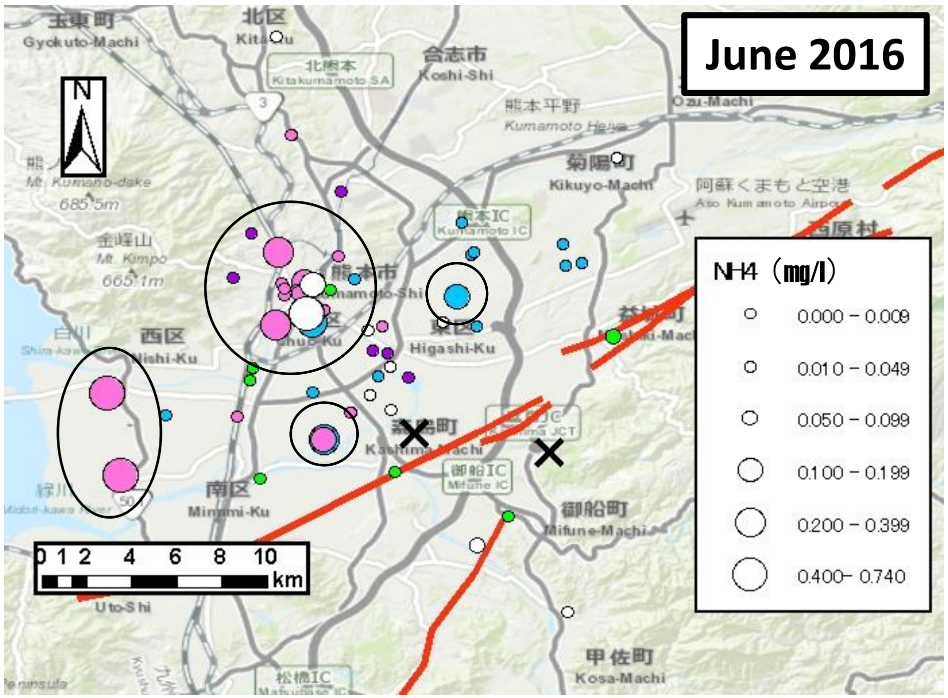
…was not significant at this moment as changing the groundwater nitrogen pollution status; some signals of sewage leaking was detected

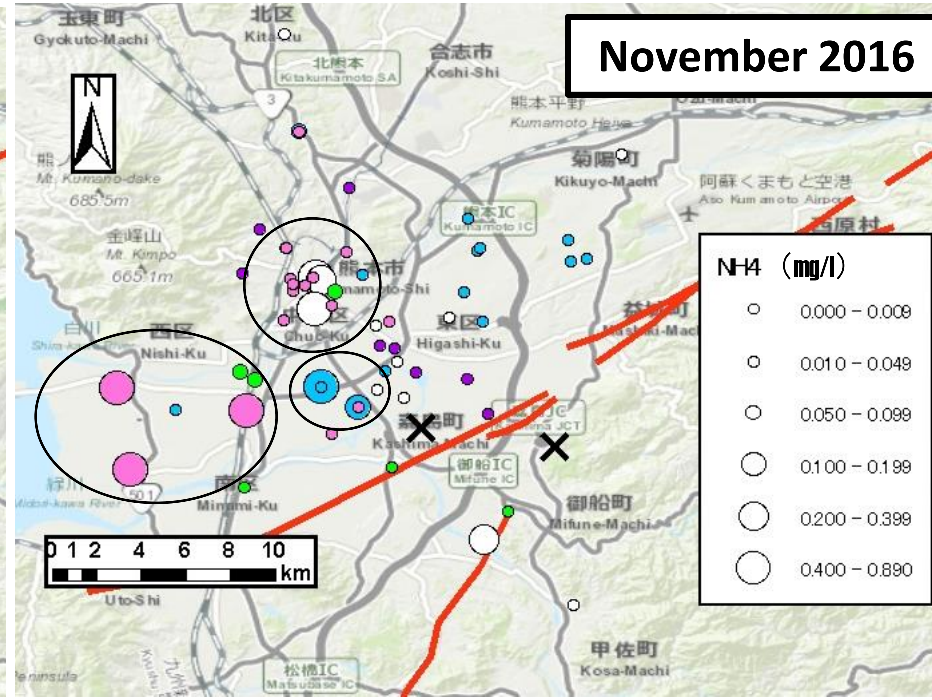
NH_4^+・・・ low level below than 1 mg/L.

NO_3−N＋NO_2−N < 10 mg/L ⇒ Less than environmental standard values.

Systematic increase in nitrogen after the earthquake was not found.

Possible sewage leaking area (NH_4^+)





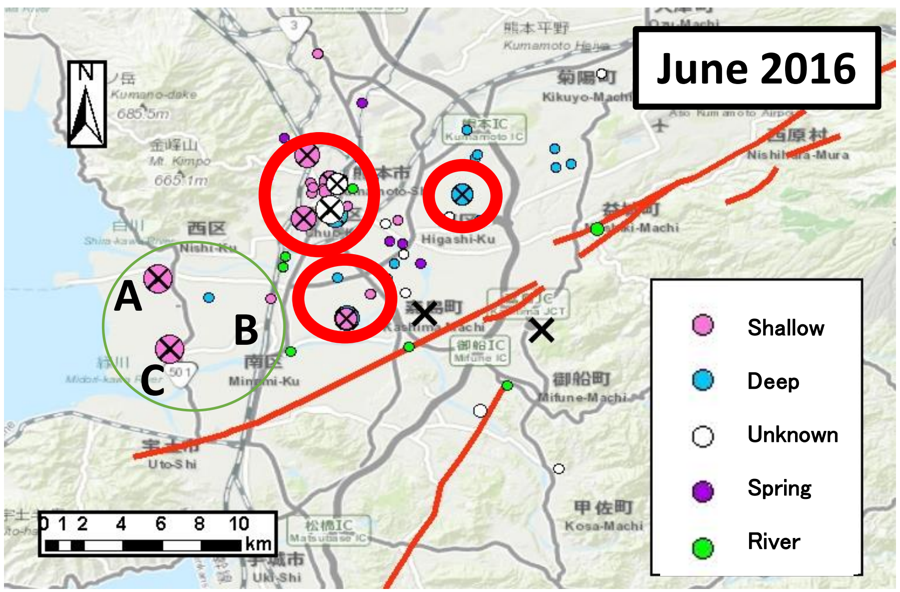
- Low (< 1 mg/L) concentration; there are some NH_4^+ in groundwater
  ⇒ Sewage leaking or natural source (Ariake marine clay)

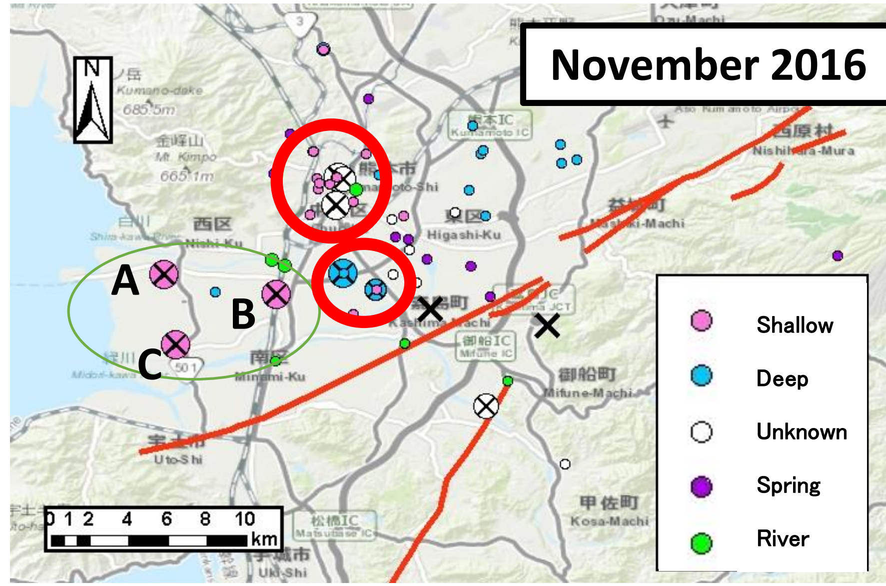
Evaluating factors

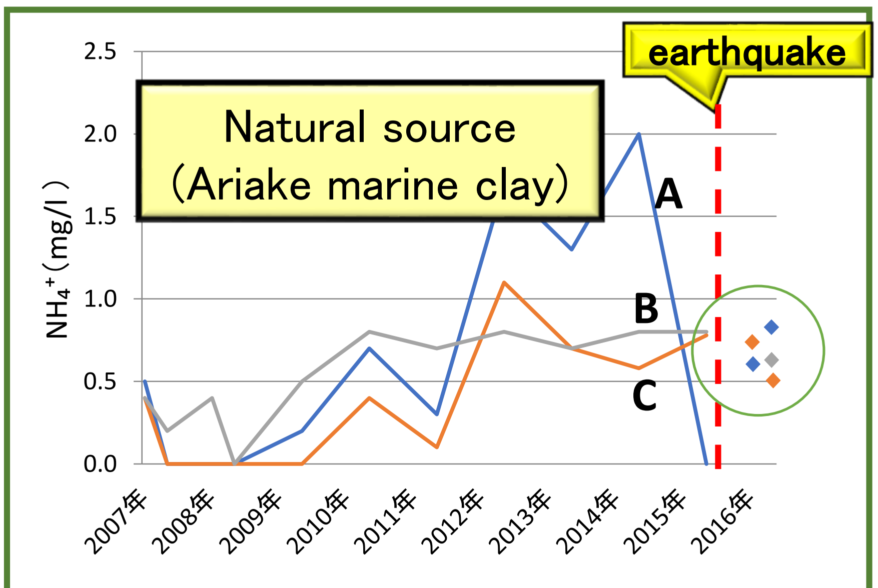
- ✓ Long term monitoring data
- ✓ Land-use
- ✓ Geology

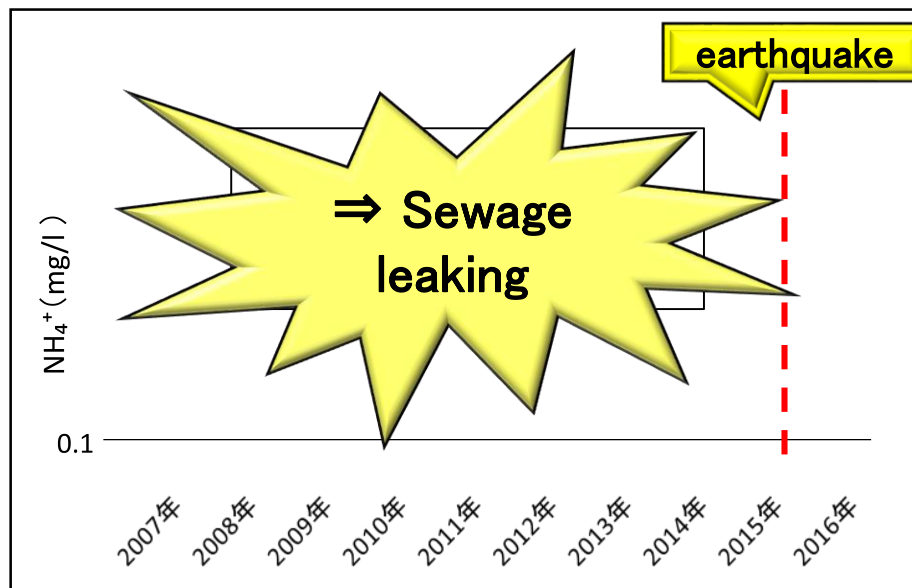
Possible sewage leaking area (NH_4^+)

＜Whole Data＞



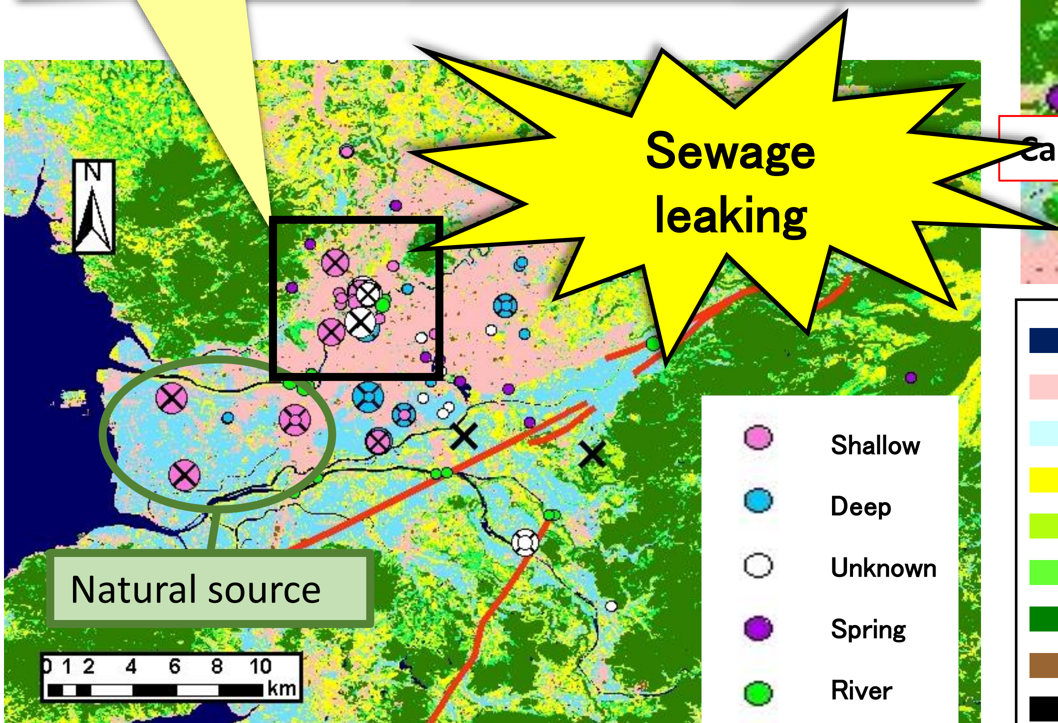


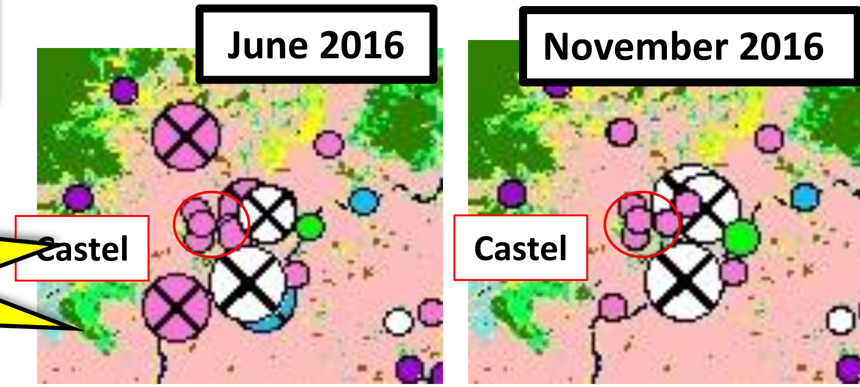




Possible sewage leaking area (NH_4^+)

Corresponding to the results of acesulfame concentrations.



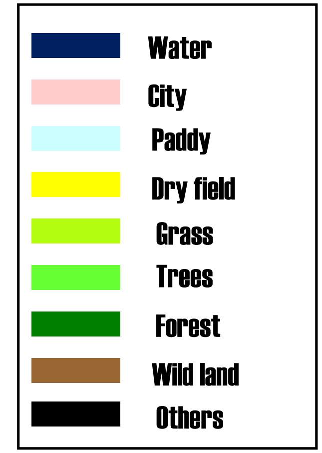


Kumamoto Castel
: No sewage pipes
No sewage leakings

Possible sewage leaking area (NH_4^+)

❖ Less contributions from natural (Ariake marine clay) sources

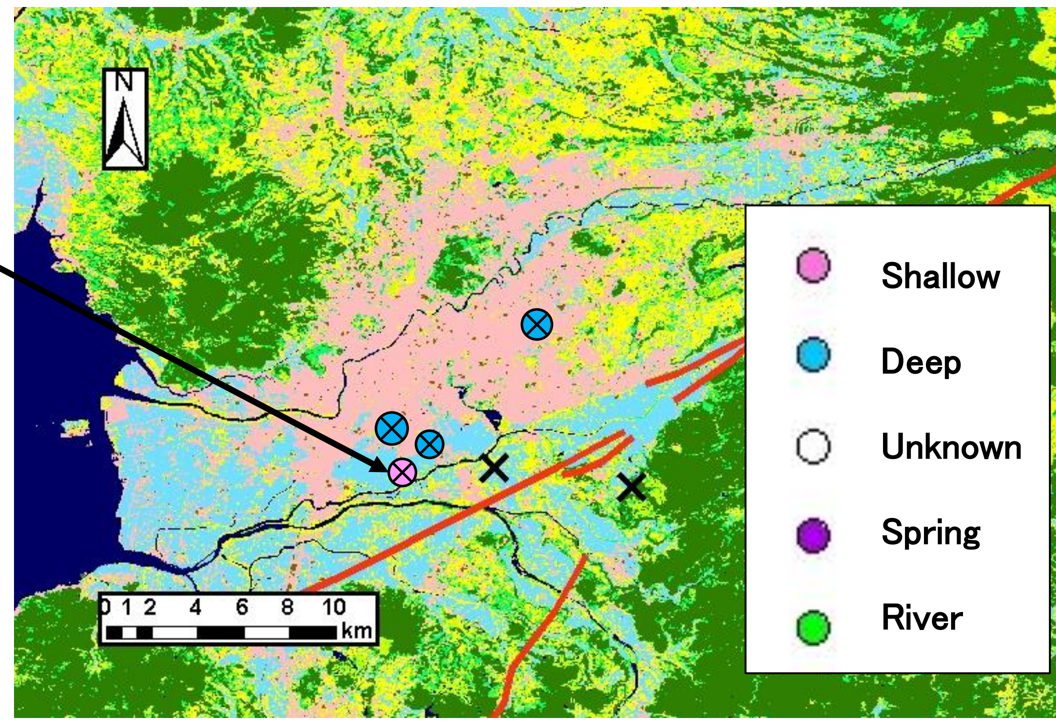
❖ Land-use = residensial area



❖ Increasing in acesulfame concentration

High possibility

Sewage leaking



Sumarry

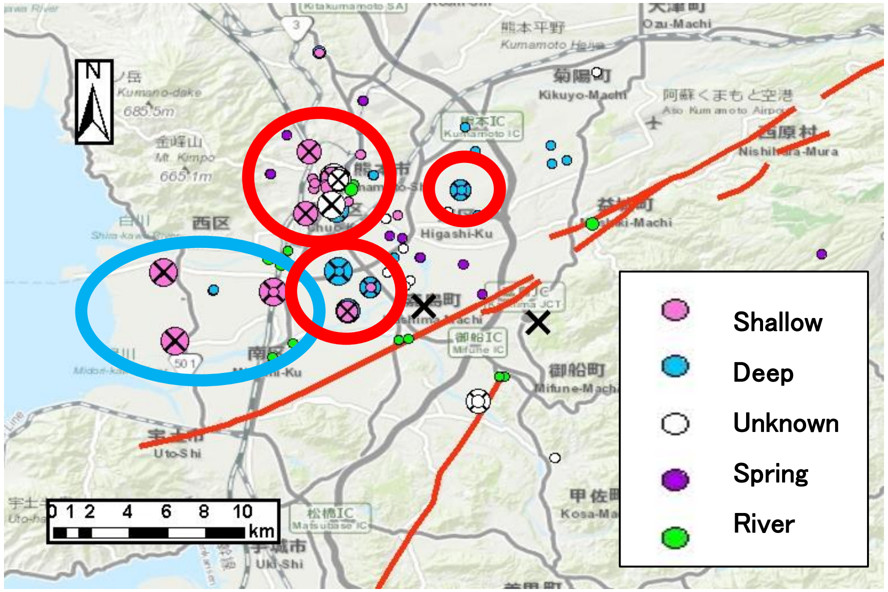
Major ions

✓ There are no significant change except in the Shirakawa site.

Nitrogen compounds

✓ There are no significant change except in the Shirakawa and Miyukifueda site.
✓ There are no sites exceeding nitrogen environmental standard values within our sampling campaign.
✓ NH_4^+ was existed only in low level < 1 mg/L.

No significant nitrogen pollutions by sewage pipe destruction



Natural sources

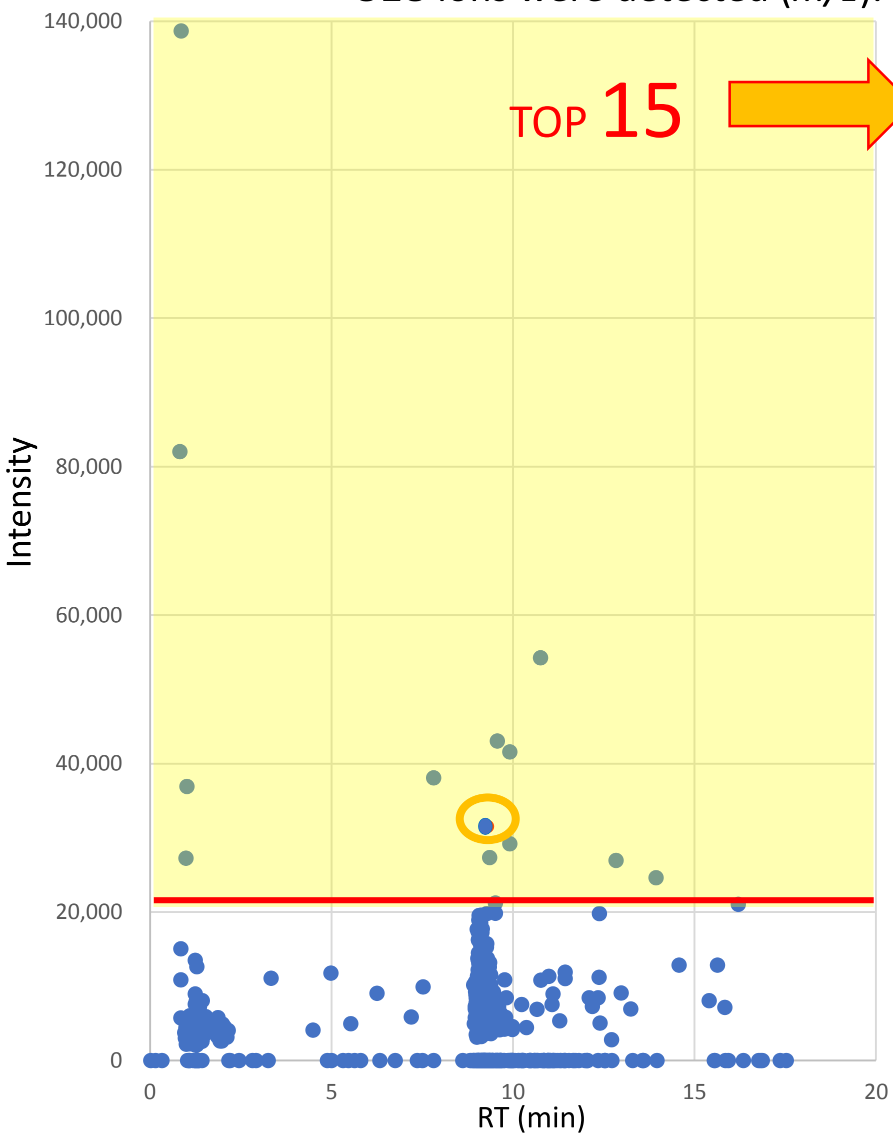
Possibility of sewage water leaking

Next scope: need to consider…
○ Groundwater flow change
○ Sewage pipe broken point

Non-target analysis -Selection of candidate chemicals

Groundwater sample (MW-10)

818 ions were detected (*m/z*).



#	Mass (Da)	RT (min)	Intensity	#	Mass (Da)	RT (min)	Intensity
1	194.9276	0.86	138716	9	209.1549	9.92	29184
2	96.9608	0.83	82035	10	498.9293	9.35	27345
3	251.2012	10.8	54274	11	209.9492	0.99	27243
4	297.9586	9.57	43031	12	61.9897	12.8	26974
5	267.1963	9.91	41579	13	291.1957	13.9	24608
6	290.0206	7.81	38087	14	261.0069	9.51	21202
7	61.9900	1.01	36918	15	61.9900	16.2	21065
8	412.9652	9.29	31522				

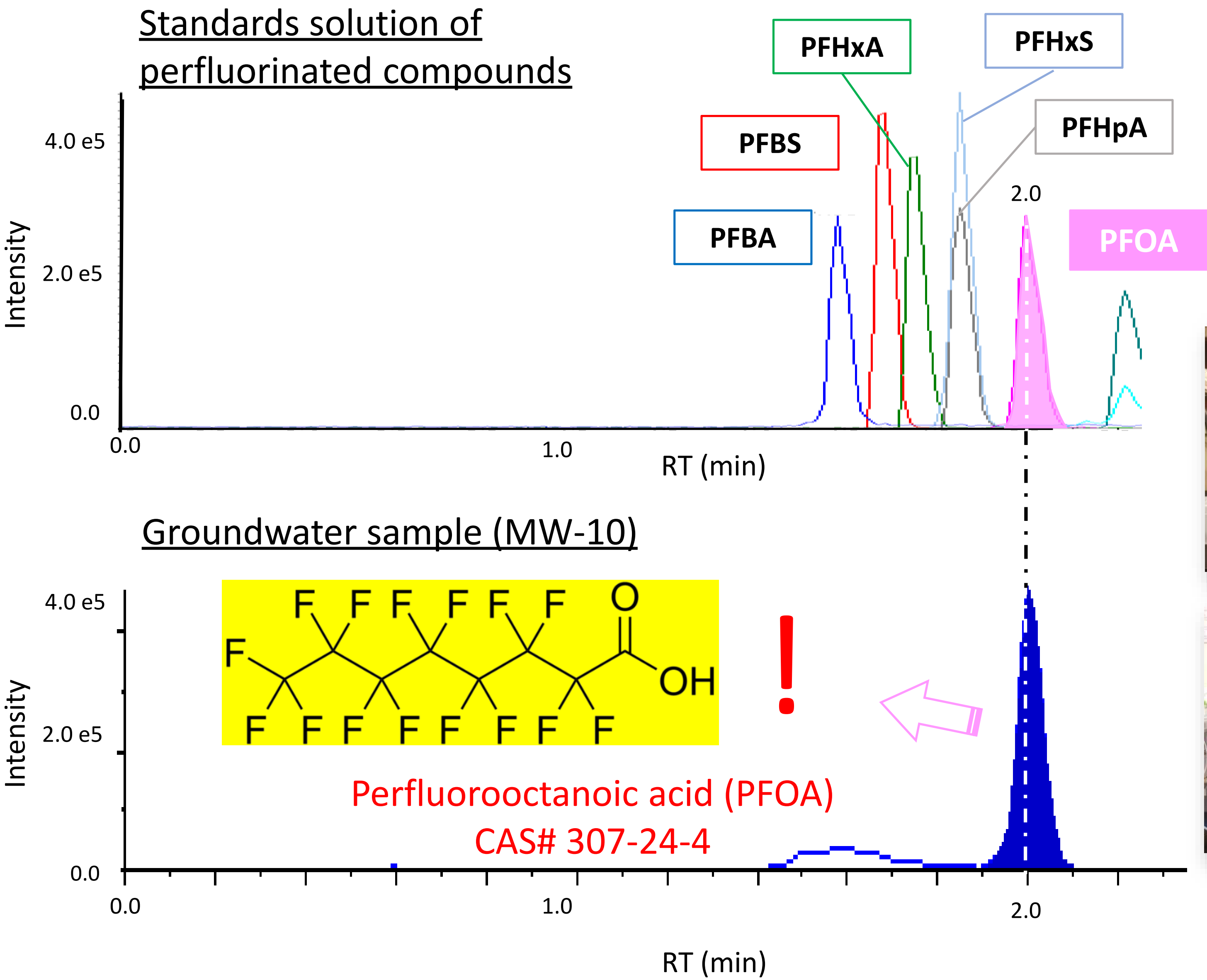
Found elemental compositions   Find Any   Find

Hit	Formula	m/z	RDB	ppm	MS Rank	MSMS ppm	MSMS Rank	Found
1	*C8ClF4N9O5*	*412.9653*	*10.5*	*-1.1*	*1*	*1.7...*	*1*	*NA/NA*
2	C8H2ClF7N6O4	412.9642	7.0	1.6	3	1.8...	2	NA/NA
3	C8H2N10O9S	412.9654	13.0	-1.5	2	2.8...	3	NA/NA
4	C8HF11N2O5	412.9637	4.0	2.7	5	2.9...	4	NA/NA
5	*C8HCl2N13O4*	*412.9657*	*13.5*	*-2.2*	*4*	*9.9...*	*6*	*NA/NA*
6	C8HF15O2	412.9664	1.0	-3.9	6	4.7...	5	NA/NA

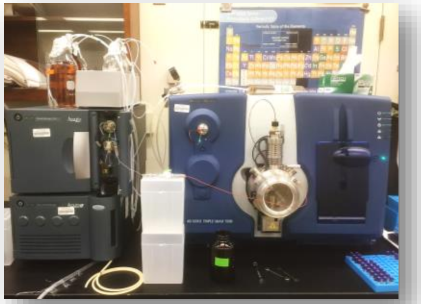
?

Perfluorooctanoic acid (PFOA)
CAS# 307-24-4

PFOA target analysis

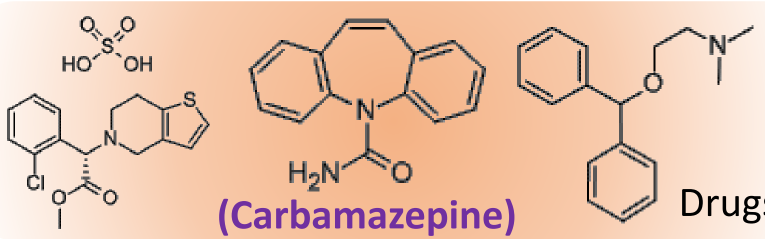


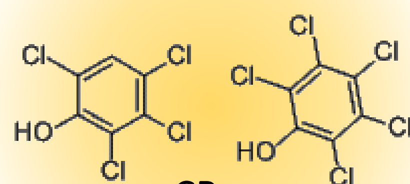
HPLC-MS/MS

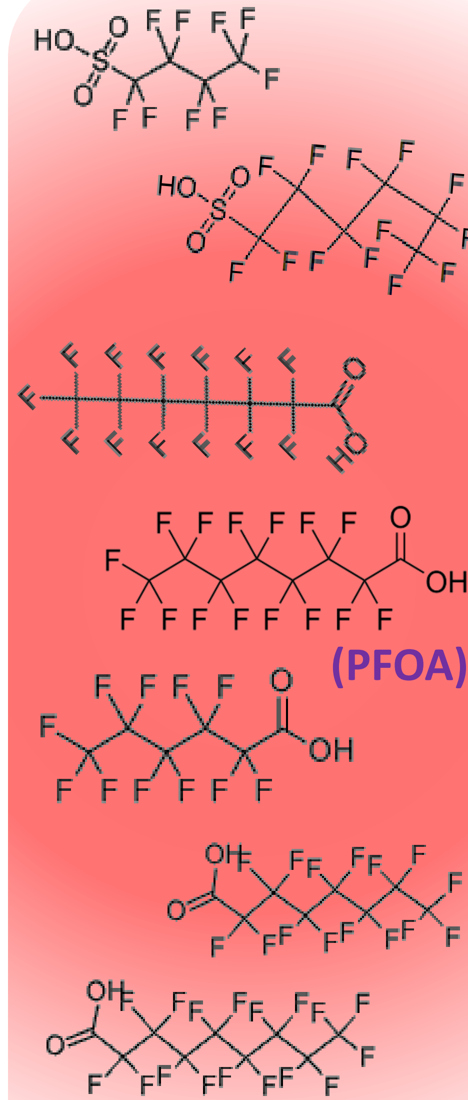


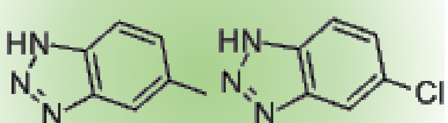
Identification of chemicals by target analysis

	# of analyzed	# of detection	DF (%)
Perfluoro related compounds (PFCs)	13	7	54
Drugs	98	3	3
Chlorophenols (CPs)	8	2	25
Benzotriazoles	5	2	40
Bisphenols	10	1	10
Tobacco smoke compounds (NCs)	4	0	0
Parabens	11	0	0
Organophosphates (OPs)	6	0	0
Caffeine (CFs)	3	0	0
Benzothiazoles	4	0	0
Benzophenones	6	0	0
	168	15	

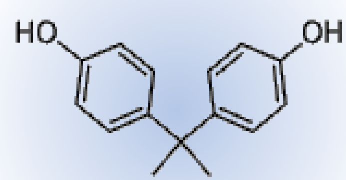








Other chemicals
(n=8)



Conclusion

Persistent water-soluble chemicals, such as artificial sweeteners and X-ray contrast media and some nutritive salts significantly increased in some locations after the Kumamoto earthquake.

Occurrence of sewer pollution in groundwater after the earthquake in Kumamoto downtown area.

<u>Future studies</u>

Spatial and temporal trends of groundwater pollution.

Investigation on the occurrence of unknown and hazardous chemicals.

Record of water quality under normal condition for comparison before/after disaster.