Selective Gadolinium Filtration: History, Status, and Plans

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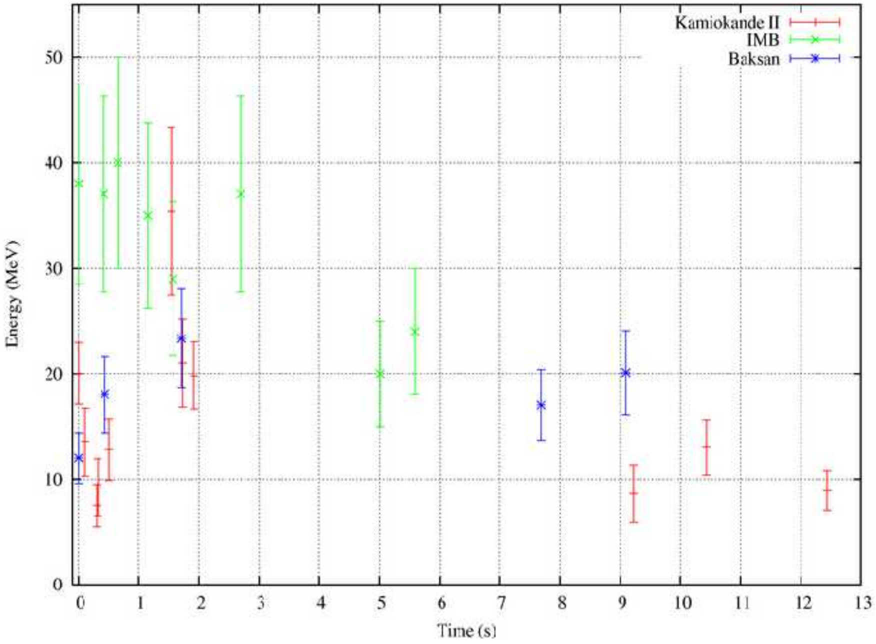
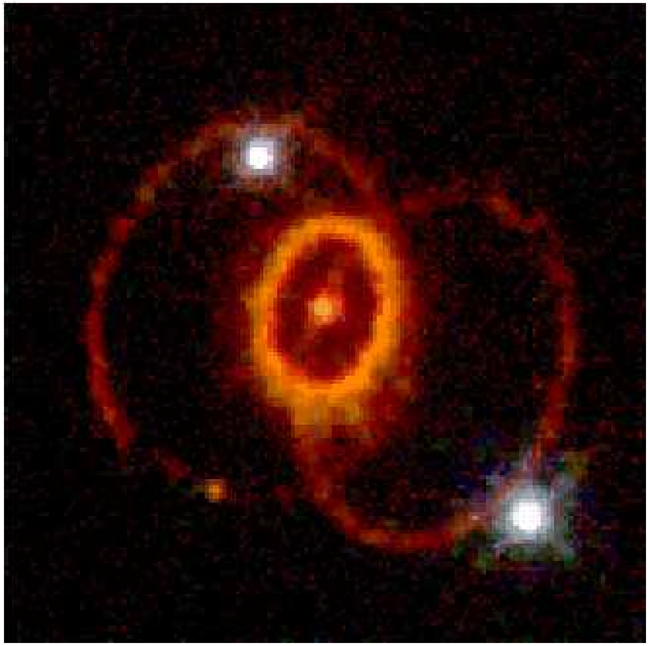
In order to understand the universe's evolution and our place in it, we need to understand as much as possible about SN explosions.

Neutrinos provide a window into core collapses' inner dynamics.

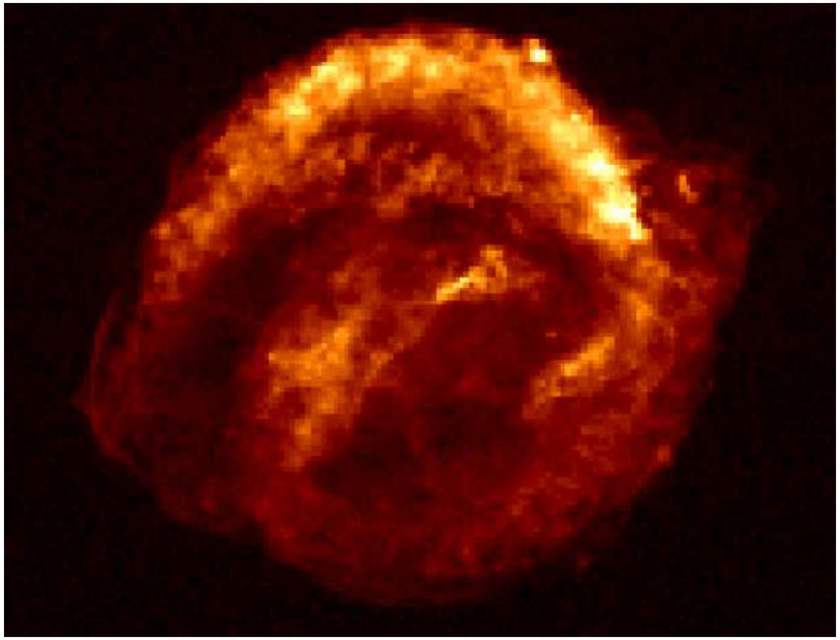
We had a dramatic demonstration of this in 1987:

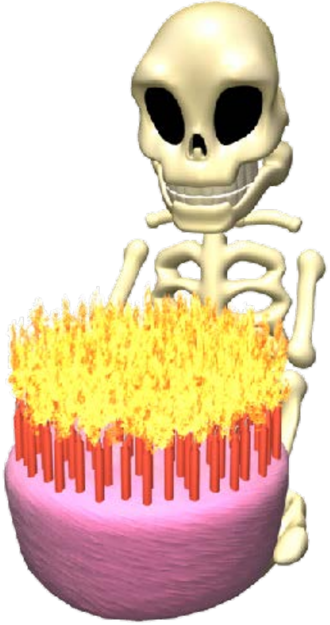




Based on this handful of neutrino events, on average
one paper has been published every ten days...
for the last 25 years!

We would very much like to collect some more supernova neutrinos!





But it has already been over a quarter century since SN1987A, and exactly 408 years and 56 days since a supernova was last definitely observed within our own galaxy.

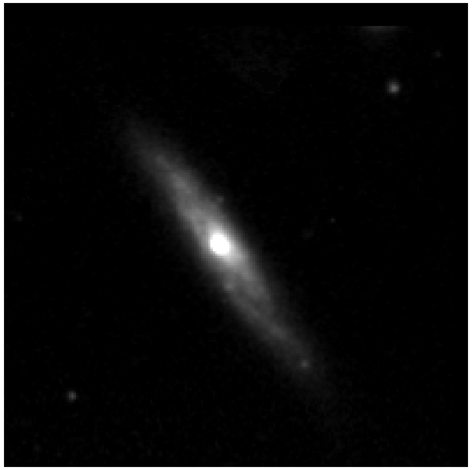
Yes, it's been a long, cold winter for SN neutrinos…

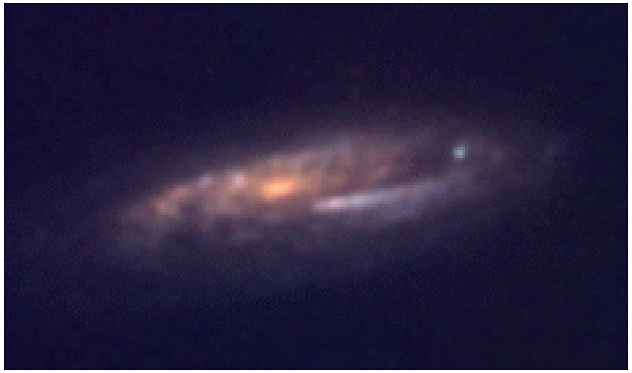
but there is hope!

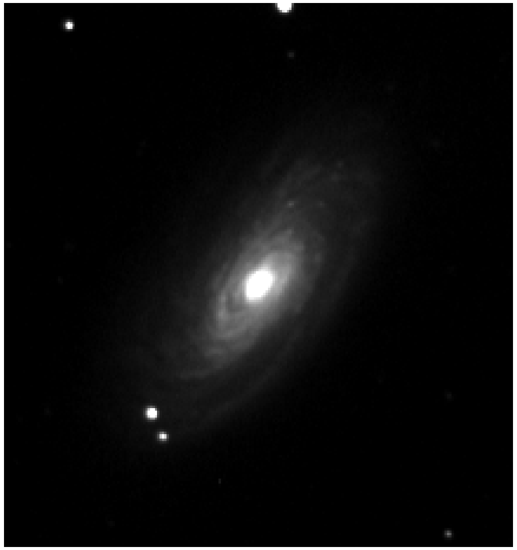
So, how can we be <u>certain</u> to see more supernova neutrinos without having to wait too long?

This is not the typical view of a supernova! Which, of course… is good.

Yes, <u>nearby</u> supernova explosions may be rare, but supernova explosions are extremely common.





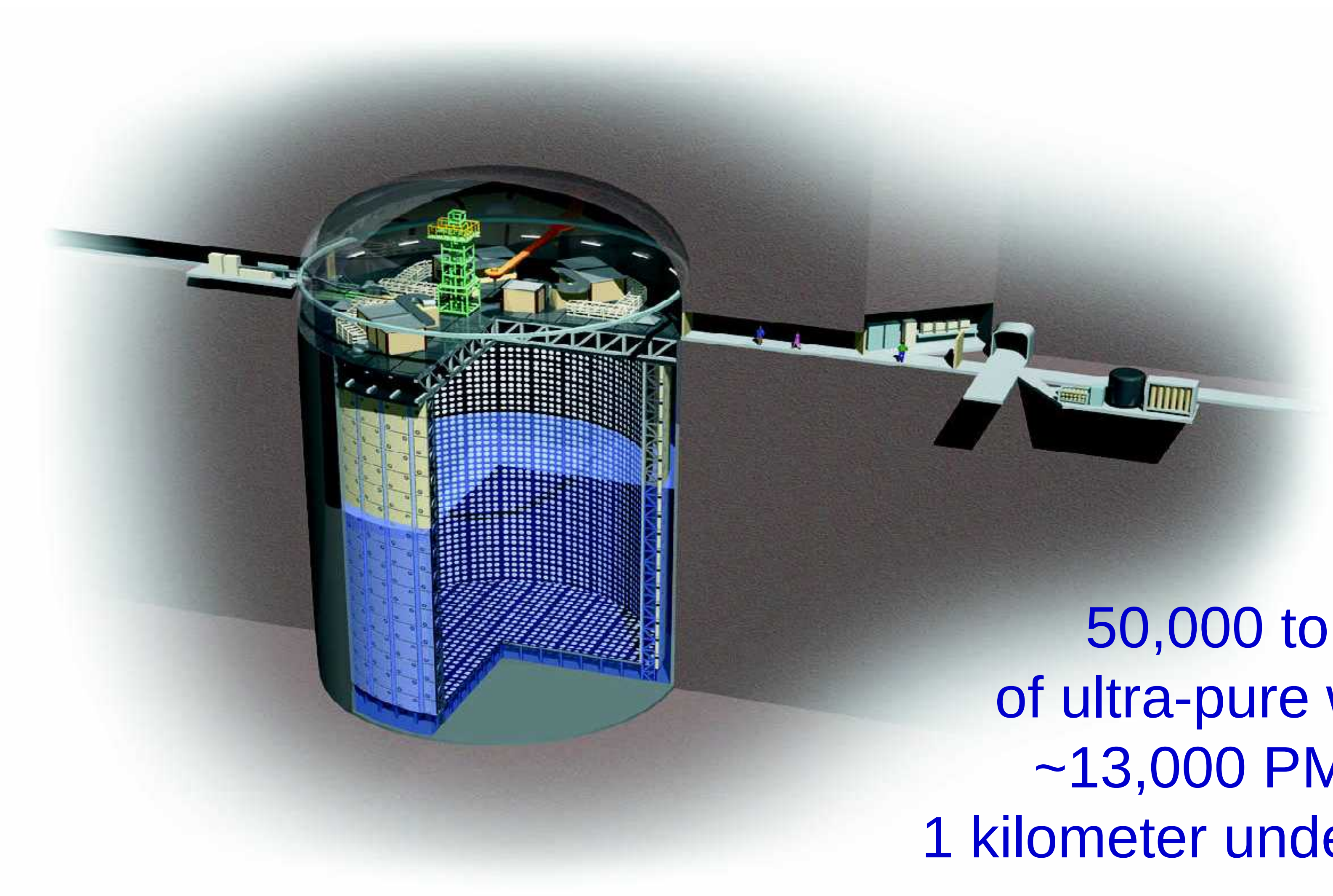


Here's how most of them look to us (video is looped).

There are ***thousands* of supernova explosions per hour** in the universe as a whole!

These produce a diffuse supernova neutrino background [DSNB], also known as the supernova relic neutrinos [SRN].

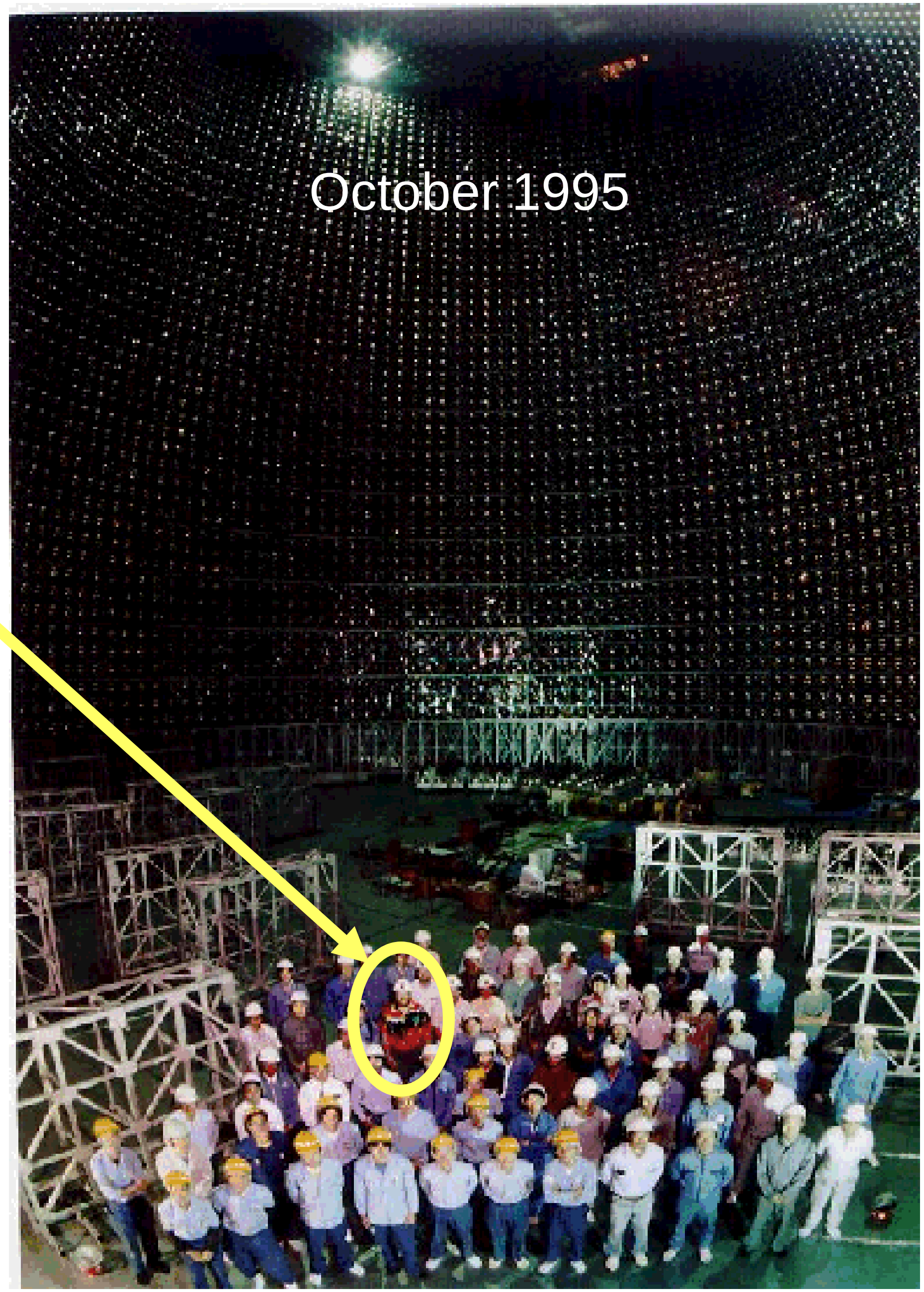
My beloved Super-Kamiokande – one of the best and most successful neutrino and proton decay detectors in the world – is nevertheless based on 30-year-old water Cherenkov technology.



50,000 tons
of ultra-pure water,
~13,000 PMT’s,
1 kilometer underground

I've been a part of Super-K (and wearing brightly-colored shirts) from its very early days…

January 1996



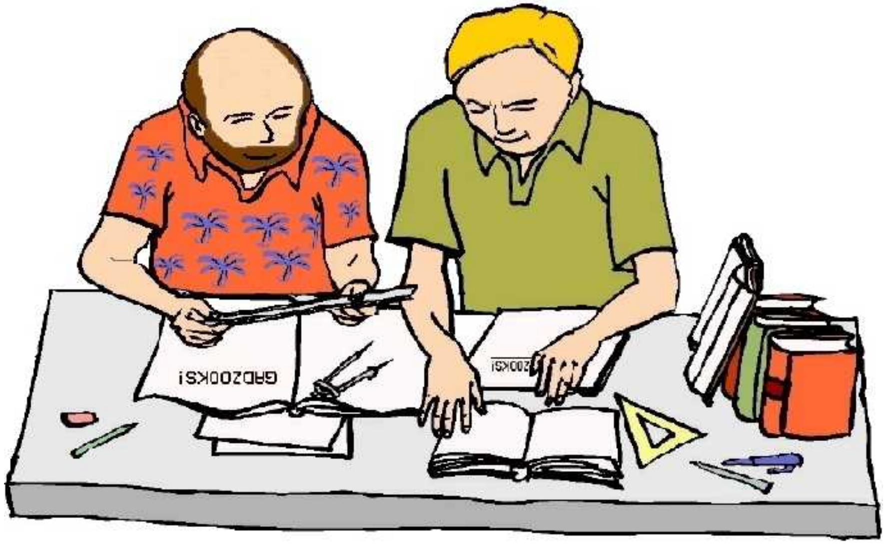
Super-K has now been taking data for over a decade. But what does the future hold?

On July 30th, 2002, at ICHEP2002 in Amsterdam, Yoichiro Suzuki, then the newly appointed head of SK, said to me,

"We must find a way to get the new physics."

גדוליניום

"Gadol" = Great!



Inspired by this call to action, theorist John Beacom and I wrote the original GADZOOKS!

(Gadolinium Antineutrino Detector Zealously Outperforming Old Kamiokande, Super!) paper.

It proposed loading big WC detectors, specifically Super-K, with water soluble gadolinium, and evaluated the physics potential and backgrounds of a giant antineutrino detector.

[Beacom and Vagins, *Phys. Rev. Lett.*, **93**:171101, 2004]

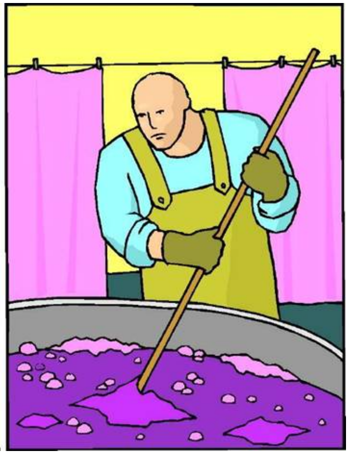
(162 citations → one every 18 days)

How can we identify neutrons produced by the inverse beta process (from supernovae, reactors, etc.) in really big water Cherenkov detectors?

$$\bar{\nu}_e + p \longrightarrow e^+ + n$$

Beyond the kiloton scale, you can forget about using liquid scintillator, ^{3}He counters, or heavy water!

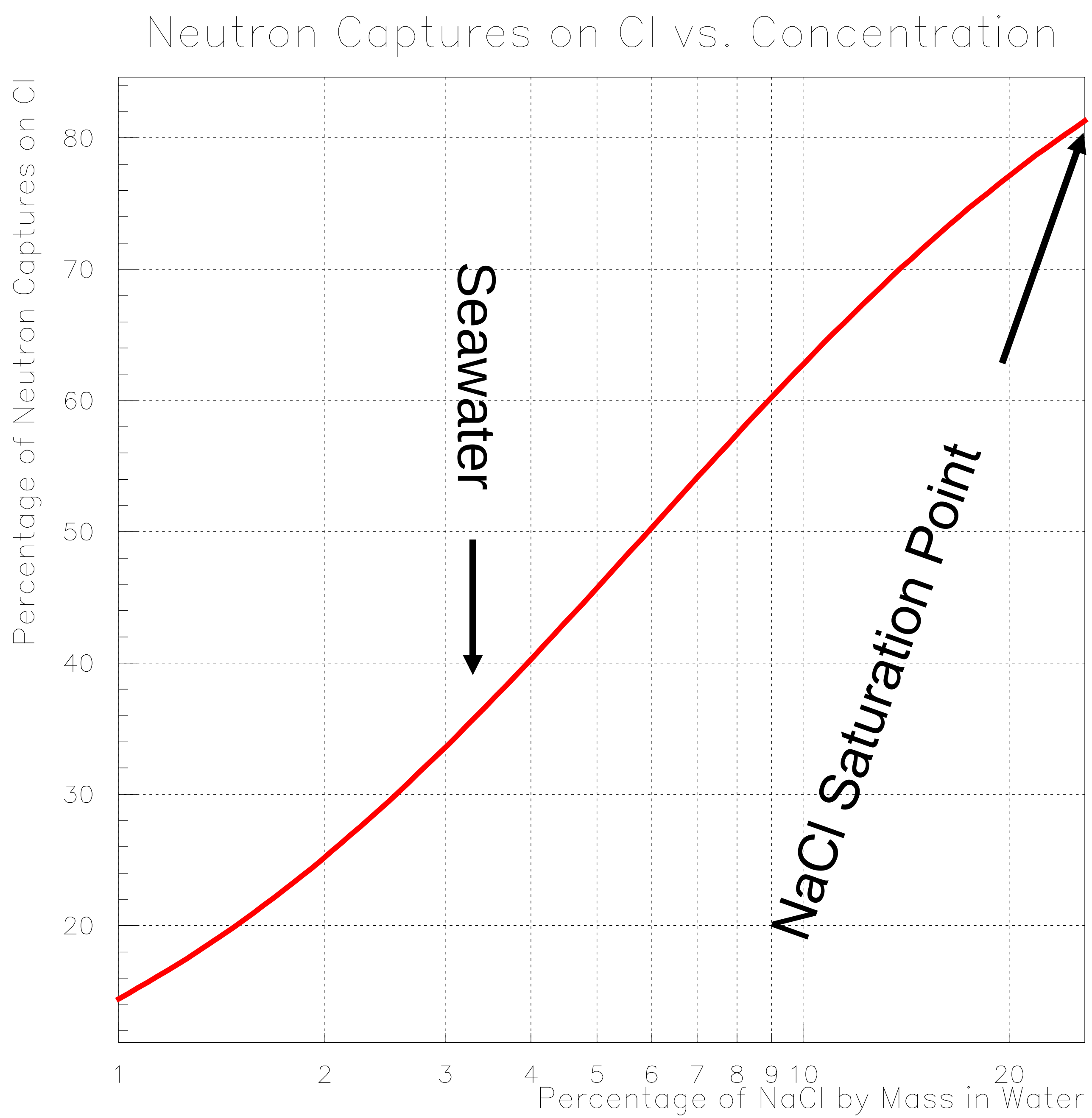
Without a doubt, at the 50 kton+ scale the only way to go is a solute mixed into the light water...



One thing's for sure: plain old NaCl isn't going to work!

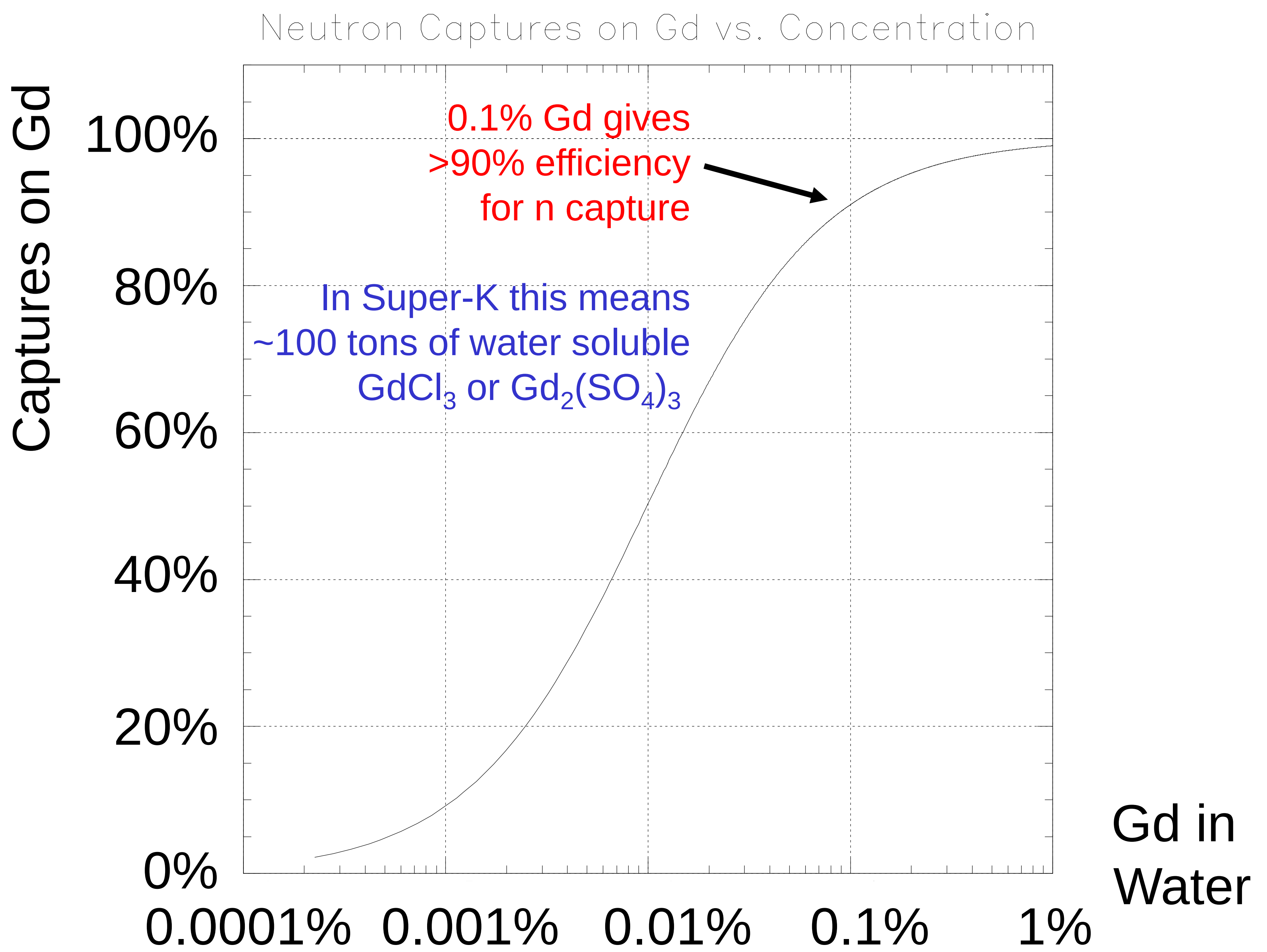
To get 50% neutron capture on Cl
(the other 50% will be on the hydrogen
in the water and essentially invisible)
you'll need to use 6% NaCl by mass:

→ *3 kilotons of salt for a 50 kton detector!* ←


Neutron Captures on Cl vs. Concentration
Percentage of Neutron Captures on Cl
80
70
60
50
40
30
20
1
2
3
4
5
6
7
8
9 10
20
Percentage of NaCl by Mass in Water
Seawater
NaCl Saturation Point

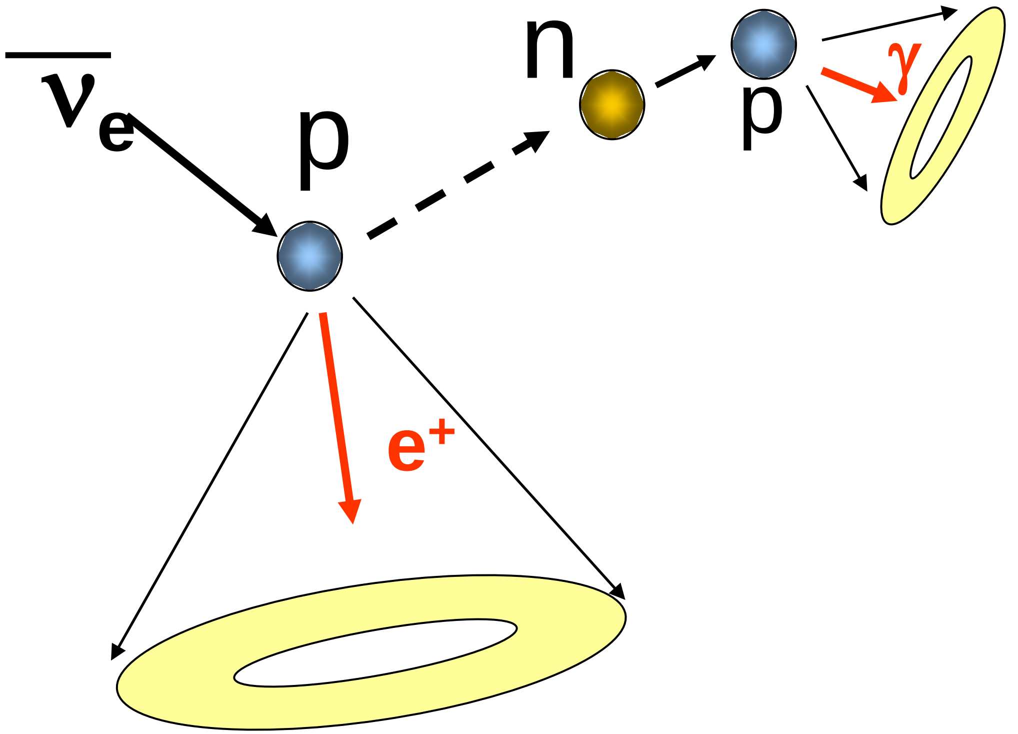
So, we eventually turned to the best neutron capture nucleus known – gadolinium.

- $GdCl_3$ and $Gd_2(SO_4)_3$, unlike metallic Gd, are highly water soluble
- Neutron capture on Gd emits a 8.0 MeV γ cascade
- 100 tons of $GdCl_3$ or $Gd_2(SO_4)_3$ in SK (0.2% by mass) would yield >90% neutron captures on Gd
- Plus, they are easy to handle and store.



Neutron Captures on Gd vs. Concentration
Captures on Gd
100%
80%
60%
40%
20%
0%
0.0001%
0.001%
0.01%
0.1%
1%
Gd in Water
0.1% Gd gives >90% efficiency for n capture
In Super-K this means ~100 tons of water soluble $GdCl_3$ or $Gd_2(SO_4)_3$

Basically, we said, "Let's add 0.2% of a water soluble gadolinium compound to Super-K!"



Positron and gamma ray vertices are within ~50cm.

But, um, didn't you just say 100 *tons?*
What's <u>that</u> going to cost?

In 1984: $4000/kg –> $400,000,000
In 1993: $485/kg –> $48,500,000
In 1999: $115/kg –> $11,500,000
In 2006: $5/kg –> $500,000

These low, low prices are for real.

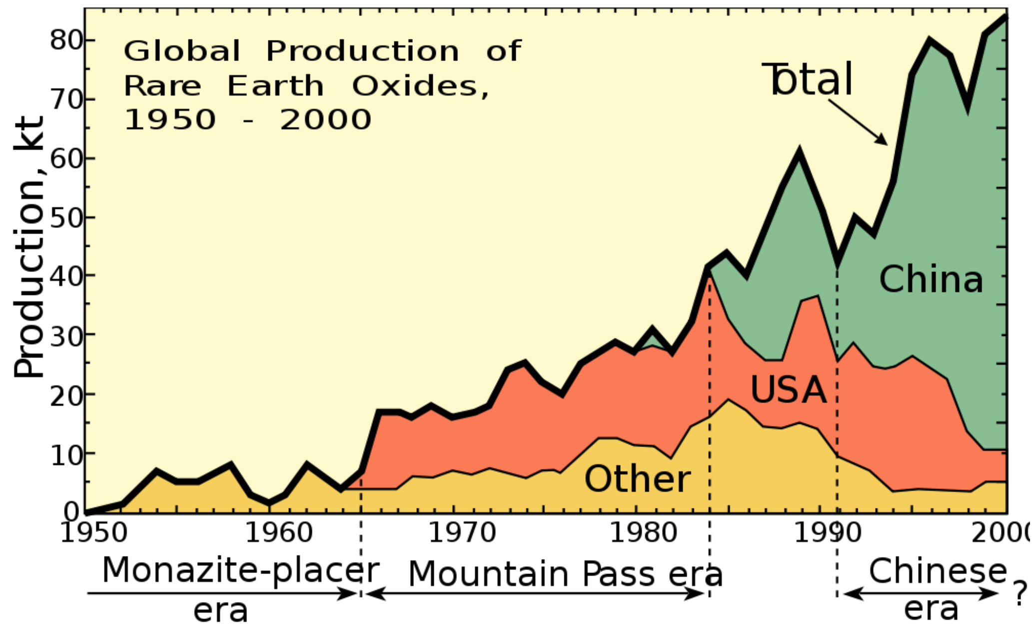
Back in 2005, $24,000 bought me 4,000 kg of $GdCl_3$.

Shipping from Inner Mongolia to Japan was included!

But since China dominates the world's rare earth production, what if they cut off the supply of gadolinium or force up its price?

Although China currently produces >90% of the world's rare earths, they control only 37% of the proven reserves. In fact, the Mountain Pass mine in California was the world's main source of rare earths for decades:

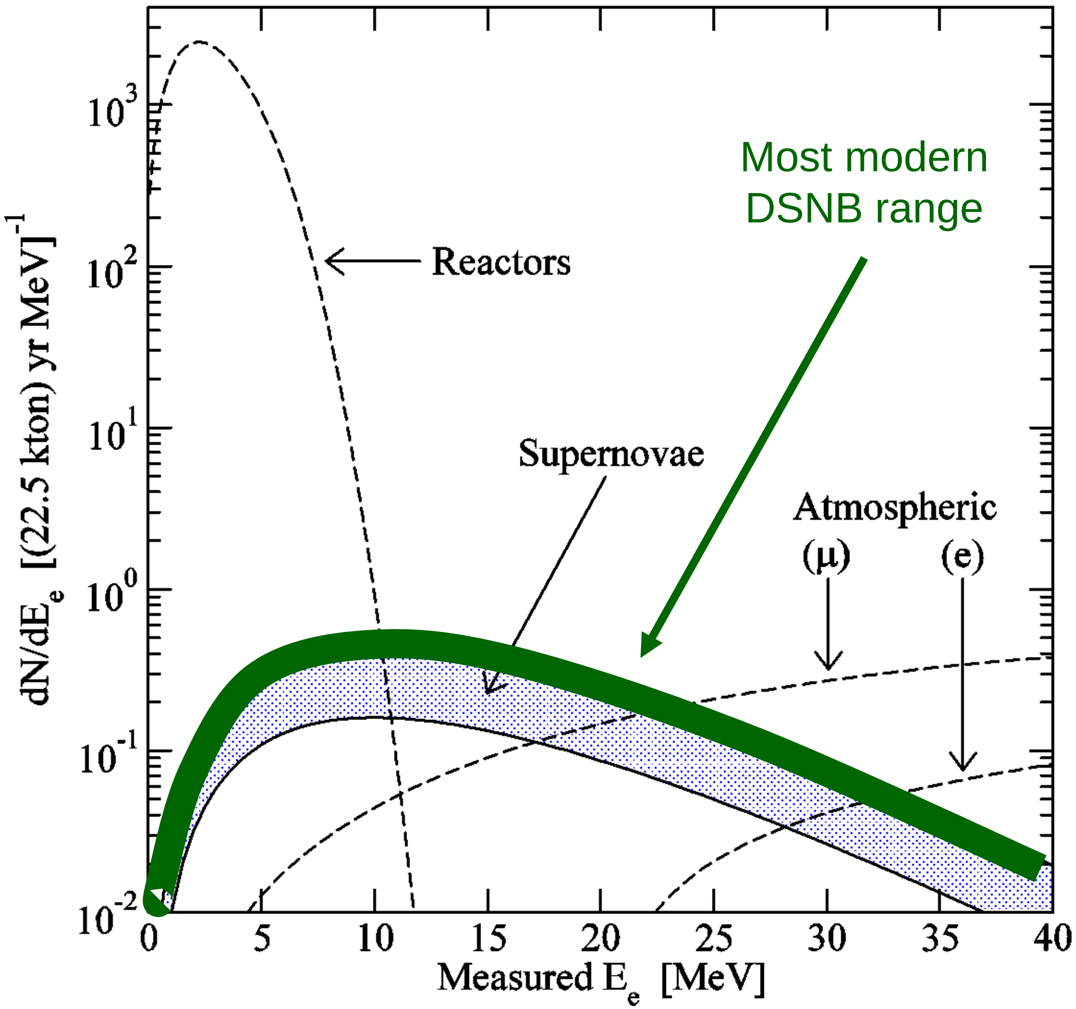
After China undercut prices in the 1990's, the California plant was shuttered. However, given the strategic importance of various rare earth elements, it is now being reopened. As of next year California's production will once again exceed that of China.



The fact is that the so-called "rare" earths are not rare at all.

They are about as abundant on Earth as are "common" elements such as zinc, copper, nickel, and tin. With healthy international competition, there is no need to be concerned about their long-term supply or cost.

Here's what the coincident signals in Super-K with $GdCl_3$ or $Gd_2(SO_4)_3$ will look like (energy resolution is applied):



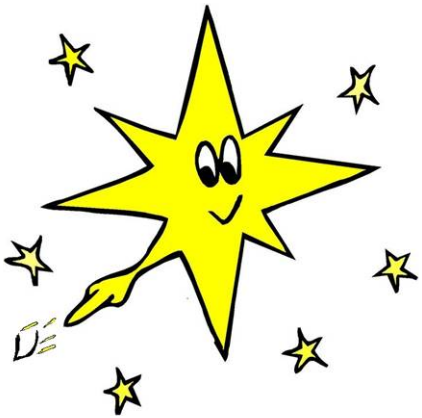
$\bar{\nu}_e + p \rightarrow e^+ + n$

spatial and temporal separation between prompt e^+ Cherenkov light and delayed Gd neutron capture gamma cascade:

λ=~4cm, τ=~30μs

→ A few clean events/yr in Super-K with Gd

In a nutshell: adding 100 tons of soluble Gd to Super-K would provide at least two brand-new signals:



1) Discovery of the diffuse supernova neutrino background [DSNB], also known as the "relic" supernova neutrinos (up to 5 events per year)

2) Precision measurements of the neutrinos from all of Japan's power reactors (thousand[s of] events per year)

Will improve world average precision of Δm^2_{12}

In addition to two guaranteed new ν signals - SN and reactor - adding gadolinium to a big WC would provide a variety of other interesting possibilities:

- Sensitivity to very late-time black hole formation
- Full de-convolution of a galactic supernova's ν signals
- Early warning of an approaching SN ν burst
- Proton decay background reduction (5X)
- New long-baseline flux normalization (T2K)
- Matter- vs. antimatter-enhanced atmospheric ν samples

All of this would work even better in a much larger detector.

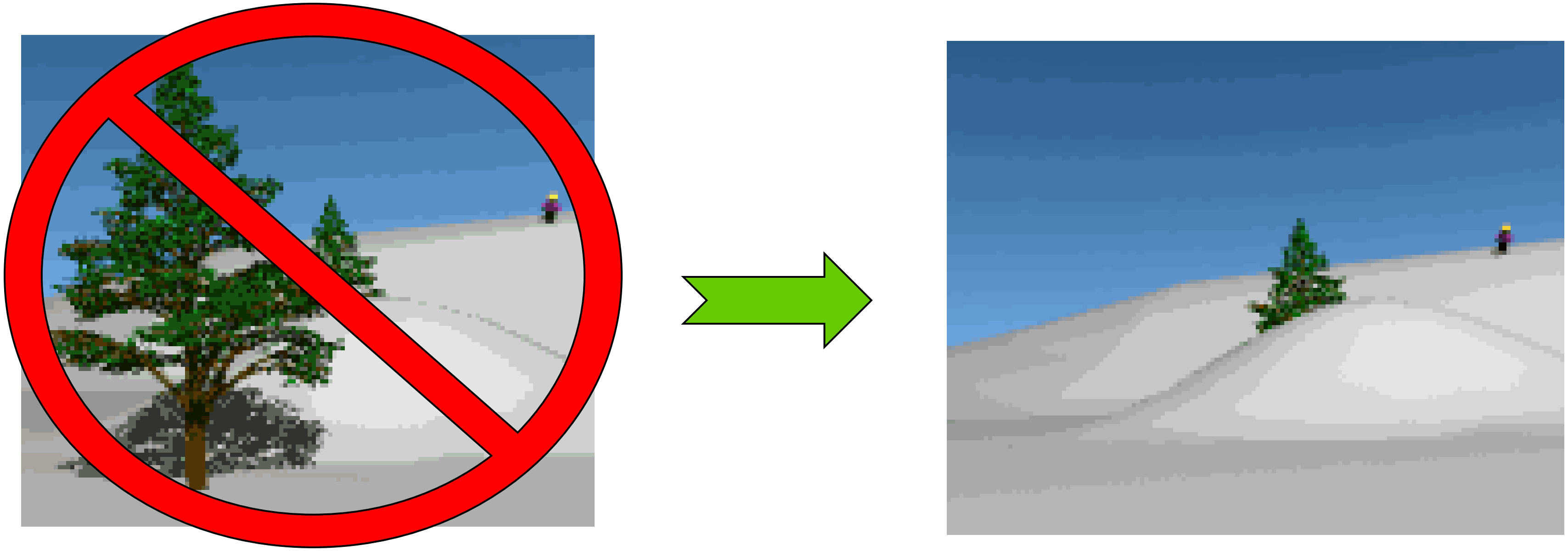
Indeed, any such massive (and massively expensive) new project will *need* to have many new physics topics to explore!

Now, Beacom and I never wanted to merely propose a new technique – we wanted to make it work!

[Snowbird photo by A. Kusenko]

Suggesting a major modification of one of the world's leading neutrino detectors may not be the easiest route…

…and so to avoid wiping out, some careful hardware studies are needed.

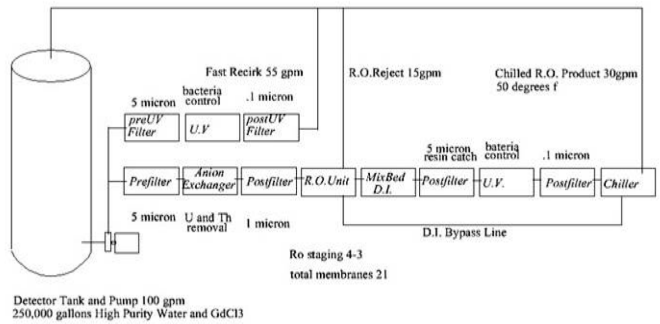


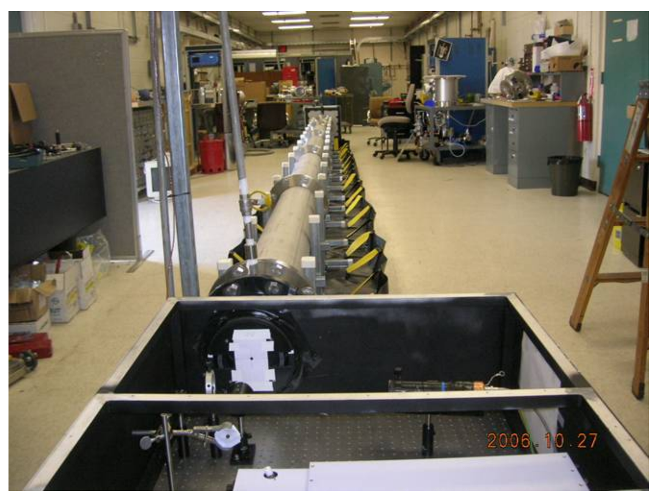
- What does gadolinium do the Super-K tank materials?
- Will the resulting water transparency be acceptable?
- Any strange Gd chemistry we need to know about?
- *How will we filter the SK water but retain dissolved Gd?*

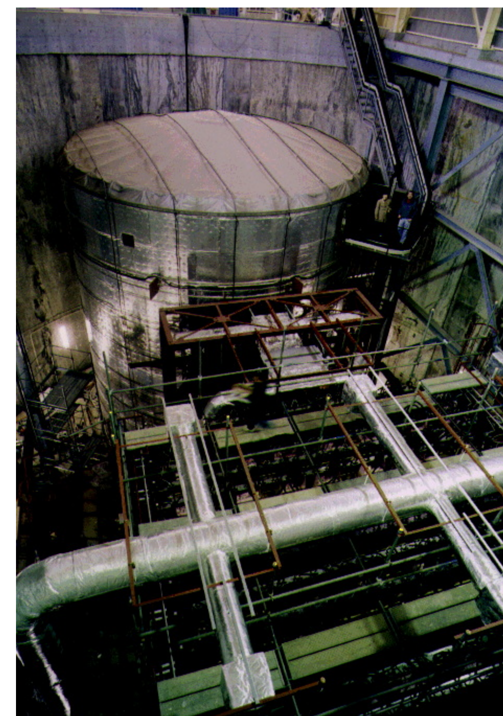
As a matter of fact, I very rapidly made two discoveries regarding $GdCl_3$ while carrying a sample from Los Angeles to Tokyo:

1) $GdCl_3$ is quite opaque to X-rays
2) Airport personnel get very upset when they find a kilogram of white powder in your luggage

Over the last eight years there have been a large number of Gd-related R&D studies carried out in the US and Japan:







Now, to make GADZOOKS! work, we will have to:

Dissolve the gadolinium sulfate in the water

→ Easy and fast (pH control)

Remove the gadolinium efficiently and completely when desired

→ Also easy and fast (pH control)

Keep pure water pure yet retain gadolinium in solution

→ The tricky part; need a selective Gd filtration system

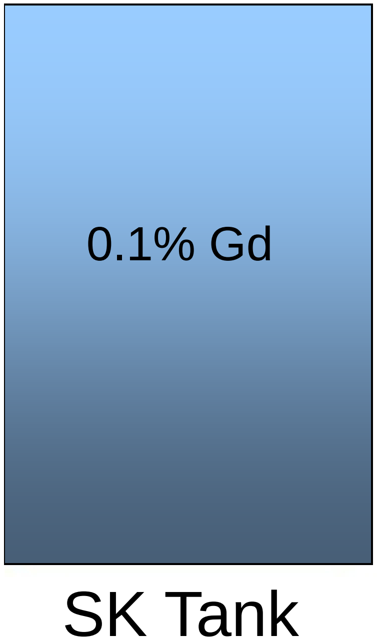
Super-K's water is incredibly clean. Almost all of the particulate matter, as well as dissolved gasses, biological agents, and dissolved ions, has been removed by continuous recirculation through the SK water system.

But our goal is to add 0.2% of water soluble gadolinium, about 100 tons, to the clean SK water. Currently gadolinium sulfate, $Gd_2(SO_4)_3$, is our leading candidate. In the past, $GdCl_3$ was also studied in considerable detail, but it is now considered too corrosive for direct contact with the SK tank material and welds.

So, our task is to determine how we can continue to keep the SK water perfectly clean, yet *not* remove the gadolinium.

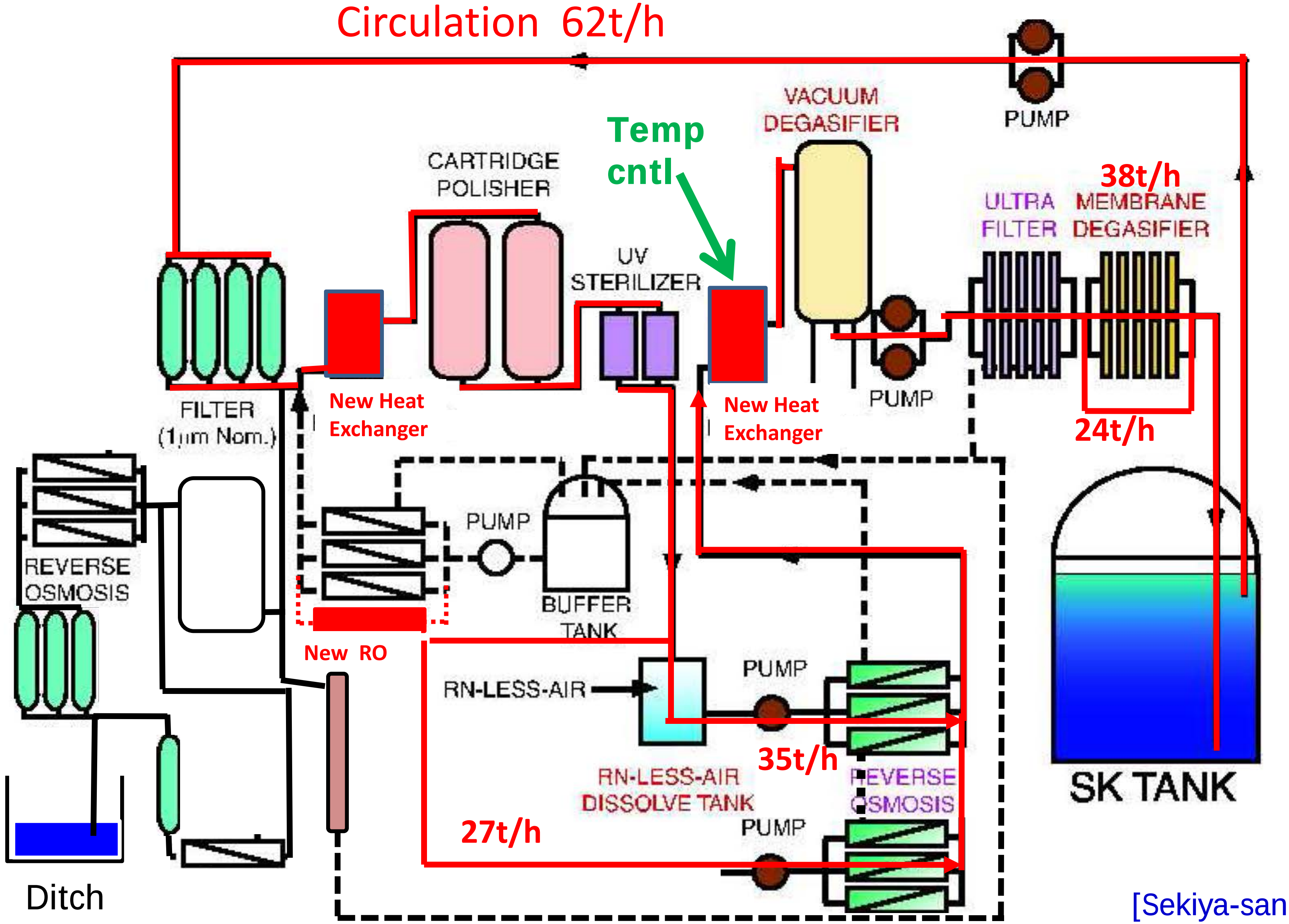
This is what we call "selective filtration."

In highly schematic form, we would like the SK water system with selective Gd filtering to work something like this:



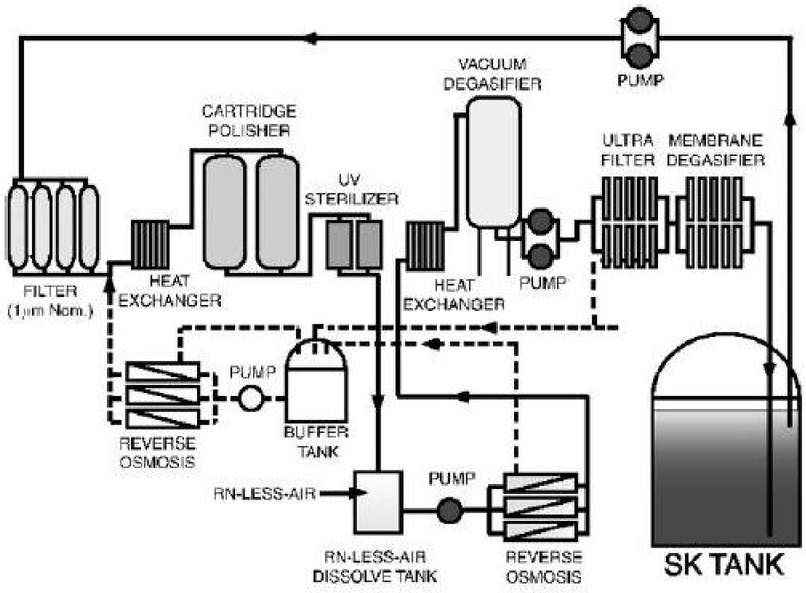
SK Water System

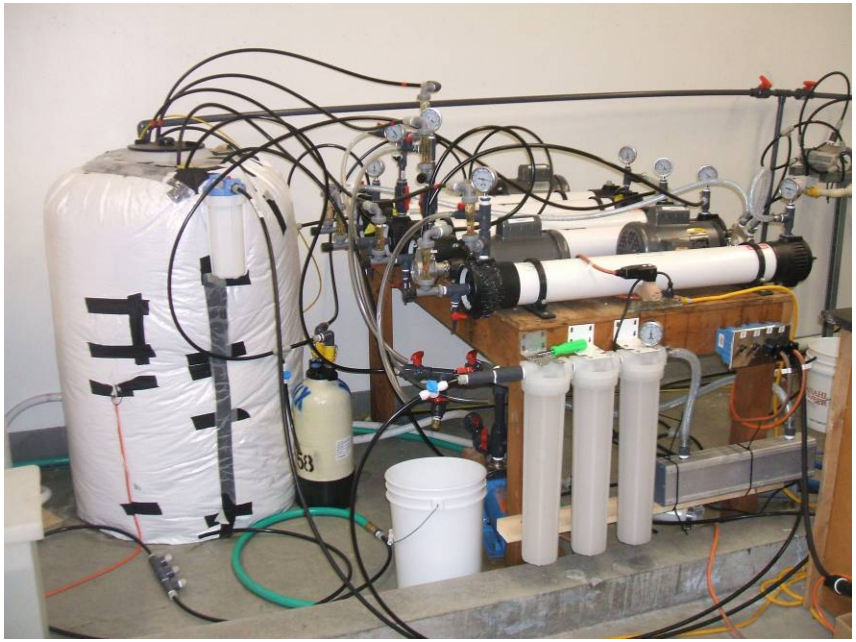
Since Jul. 2 2008



[Sekiya-san]

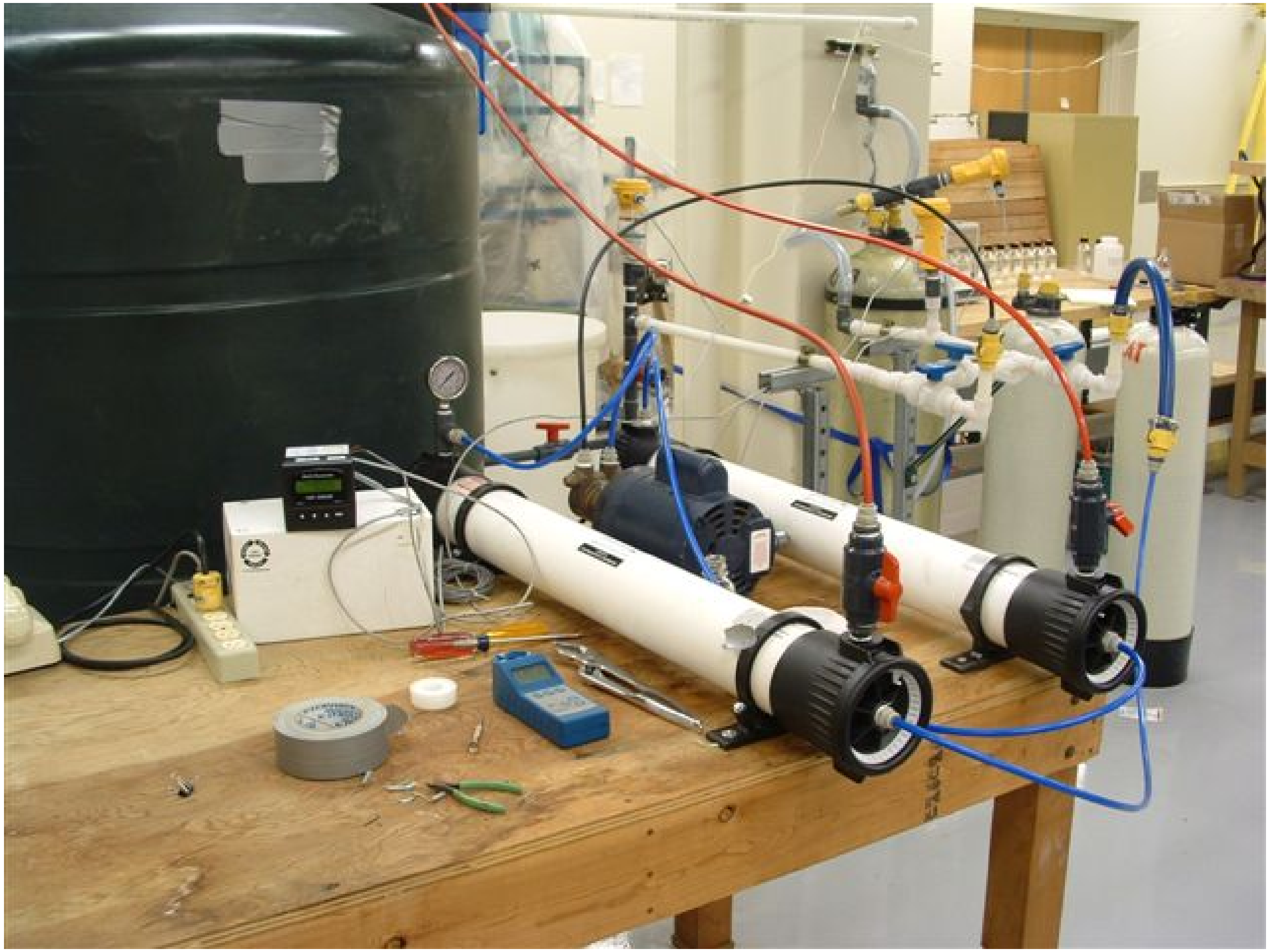
Water system studies have been under way at UCI for some time (since late 2003):



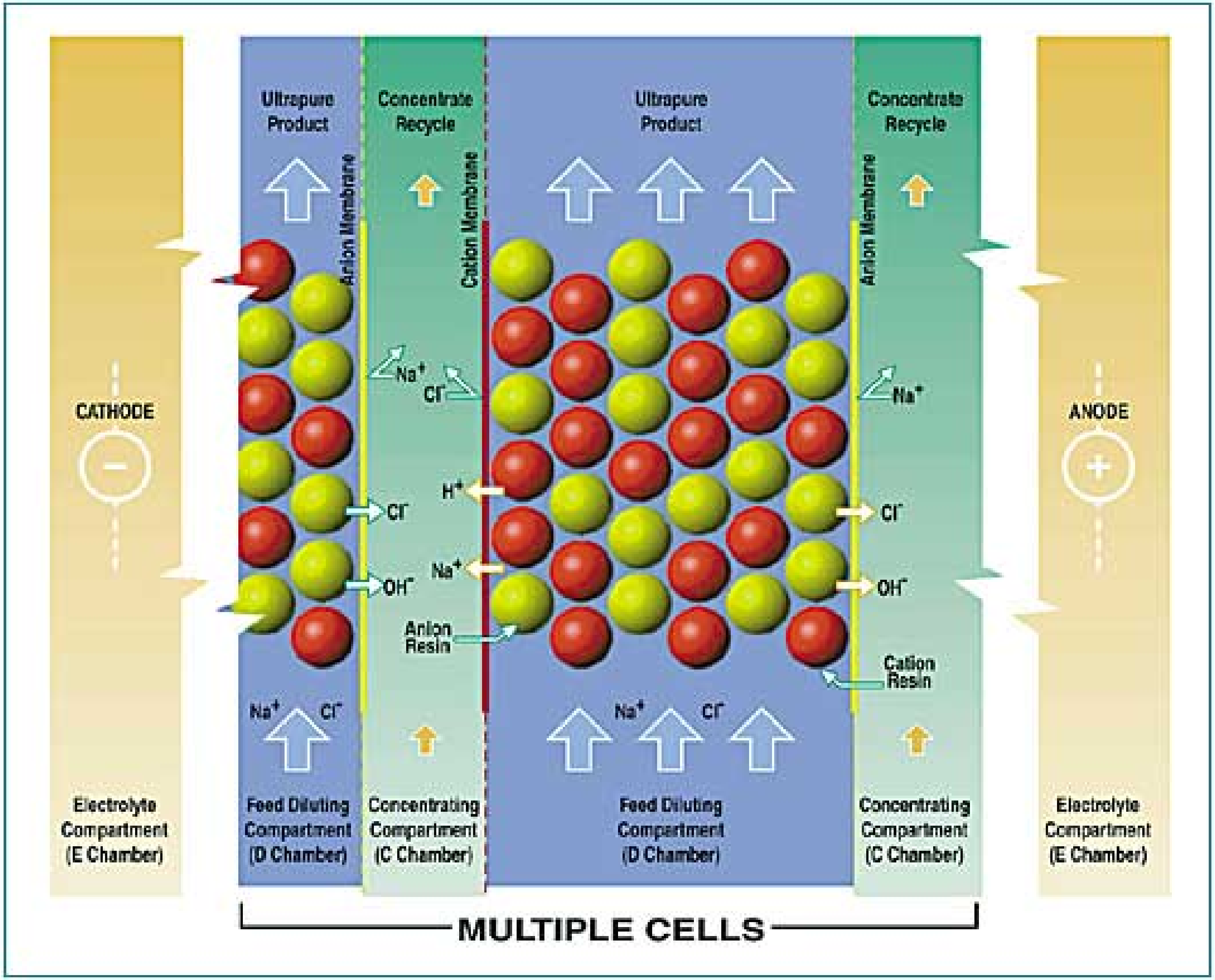


- We are replicating the conditions in SK as closely as possible (chiller, degasifier, UV, etc.)
- Components of the SK system are being checked for Gd retention and/or fouling
- Gd removal technologies are being investigated
- Long-term filtering stability will be verified in Gd test tank

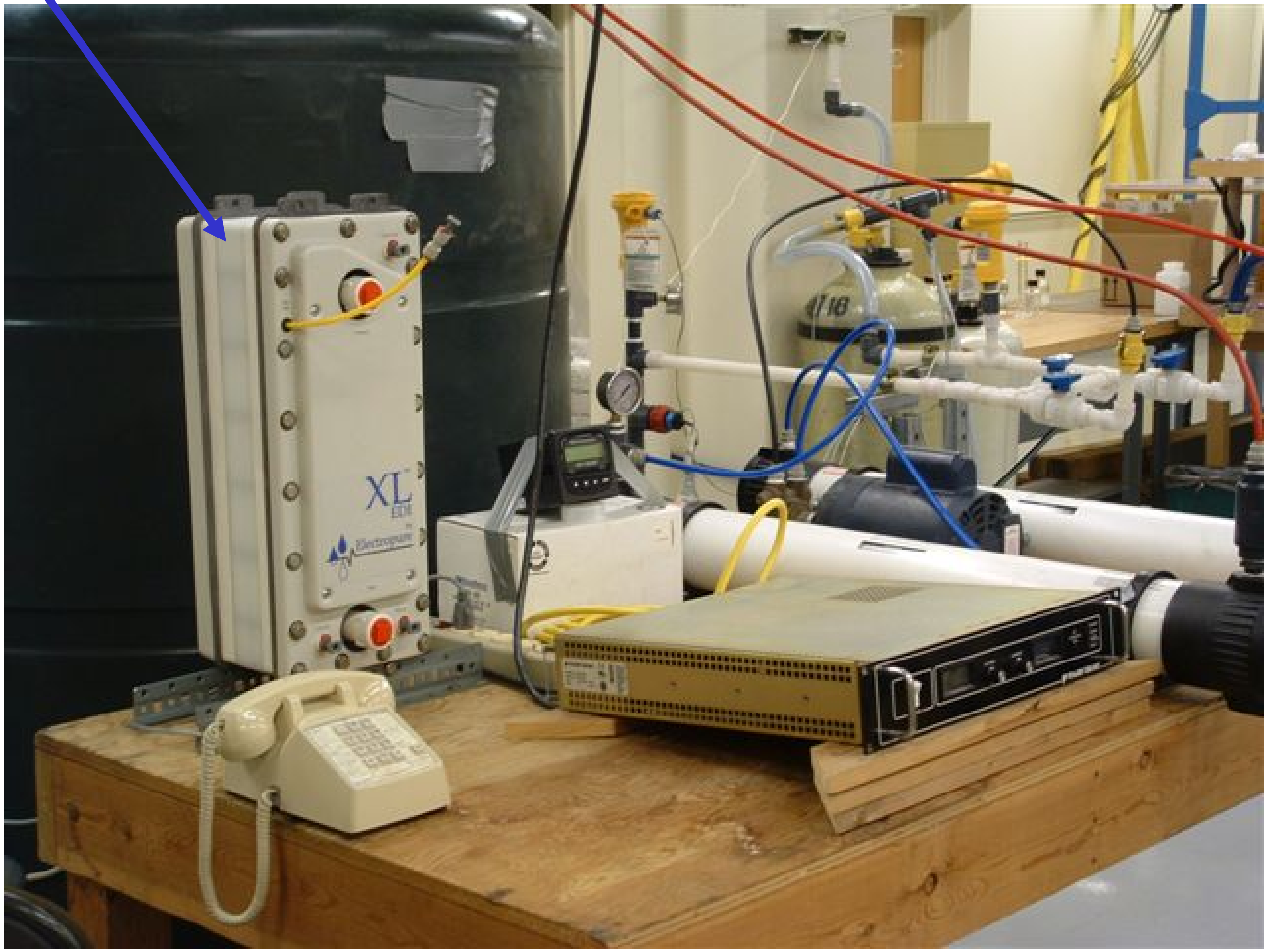
At first we tried using just reverse osmosis to remove the Gd, but it was initially only ~95% efficient (single pass)



Then we learned of a new technique called electrodeionization (EDI):



In combination with a single RO stage, EDI removed ~99.95% (per pass) of the Gd and returned it to the holding tank.



But EDI unfortunately had two really big problems:

1) It split $GdCl_3$ into gaseous chlorine…

Highly toxic!

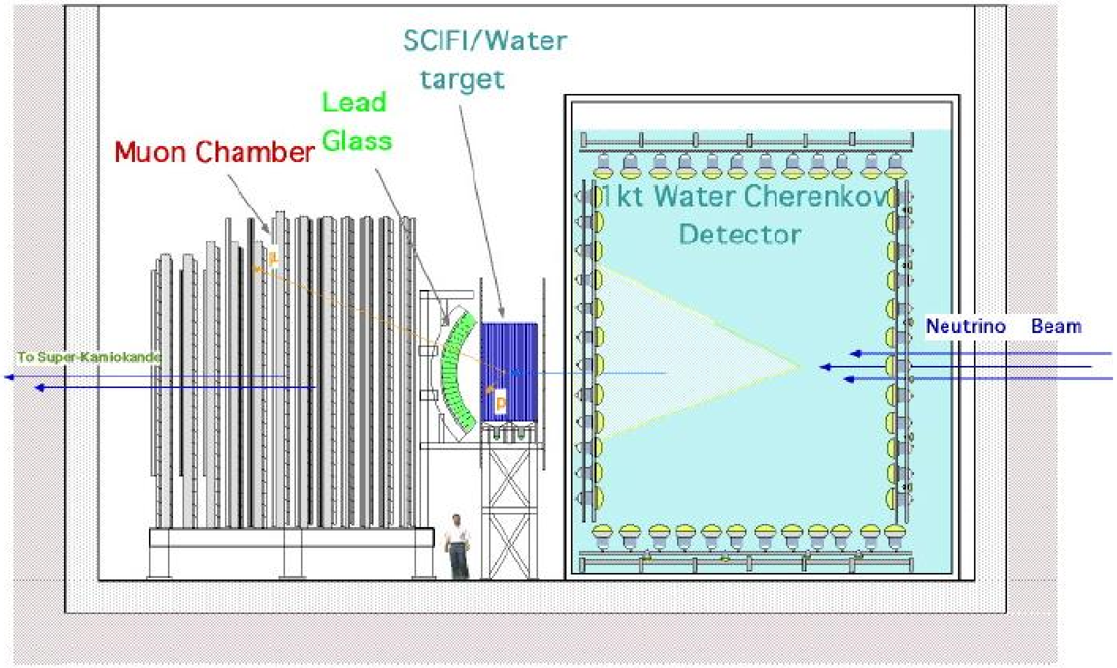
1) It split H_20 into gaseous hydrogen…

Highly explosive!

So we were forced to abandon our EDI studies.

Instead, we focused on careful tuning of the RO flows and pressures for maximum efficiency.

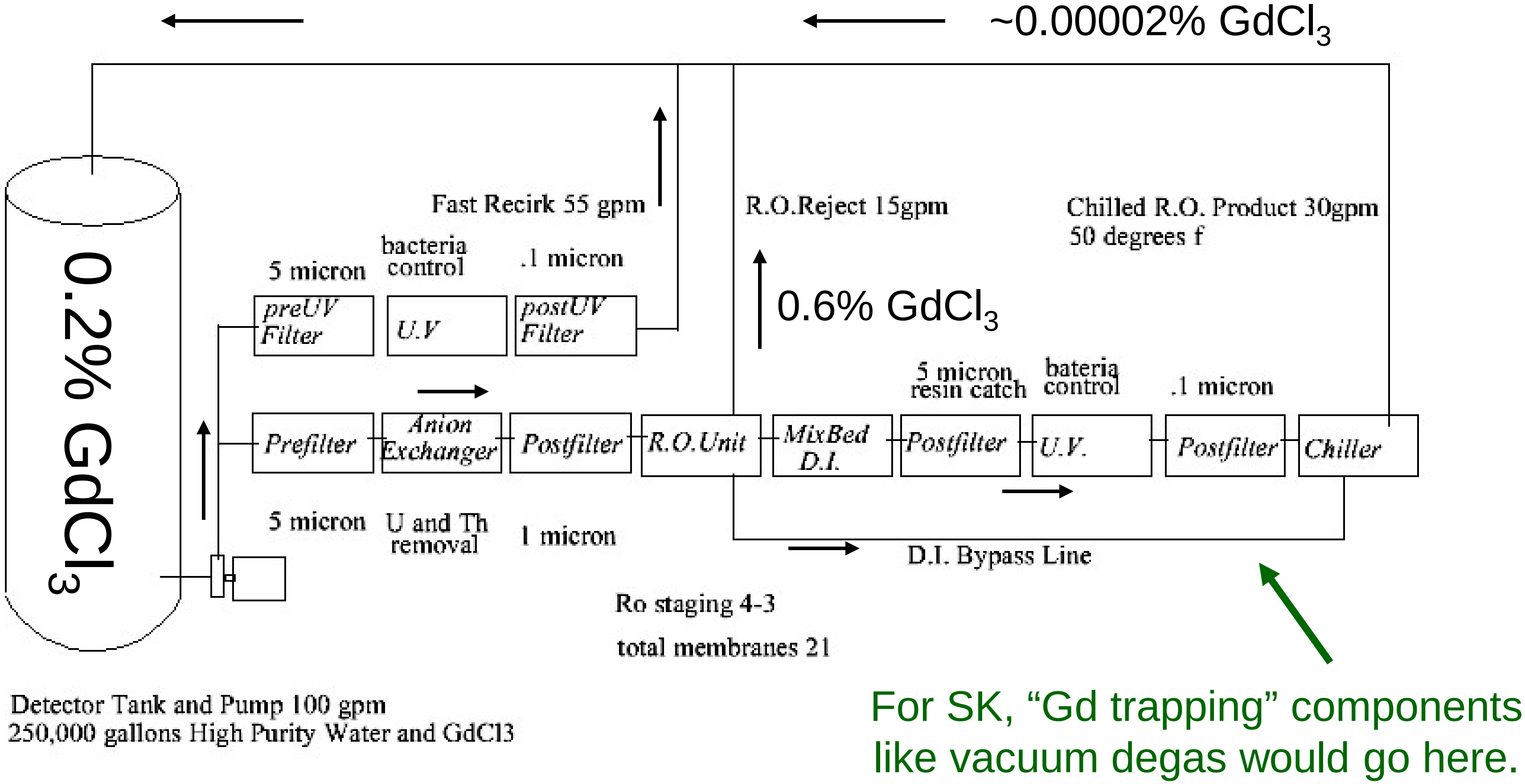
We demonstrated (and confirmed at K2K's kiloton detector) that a well-tuned reverse osmosis (RO) system removes ~99.9% of the $GdCl_3$ in a single pass and returns it to the detector.



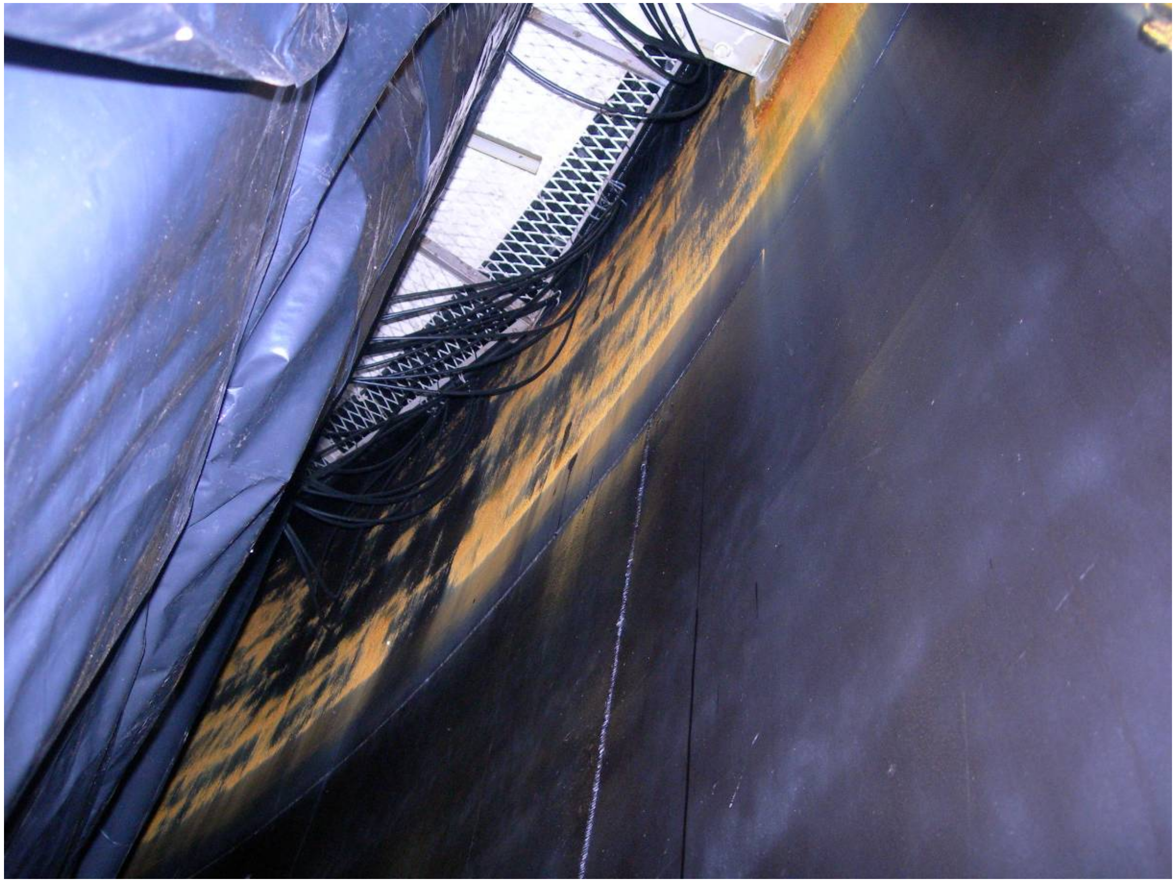
But RO removes just about everything else, too…

How can we avoid recirculating unwanted water contaminants back into SK along with the $GdCl_3$?

This was our schematic for the rebuilt K2K 1 kton water system (2005-2006):



The entire one kiloton volume was recirculated every two days.



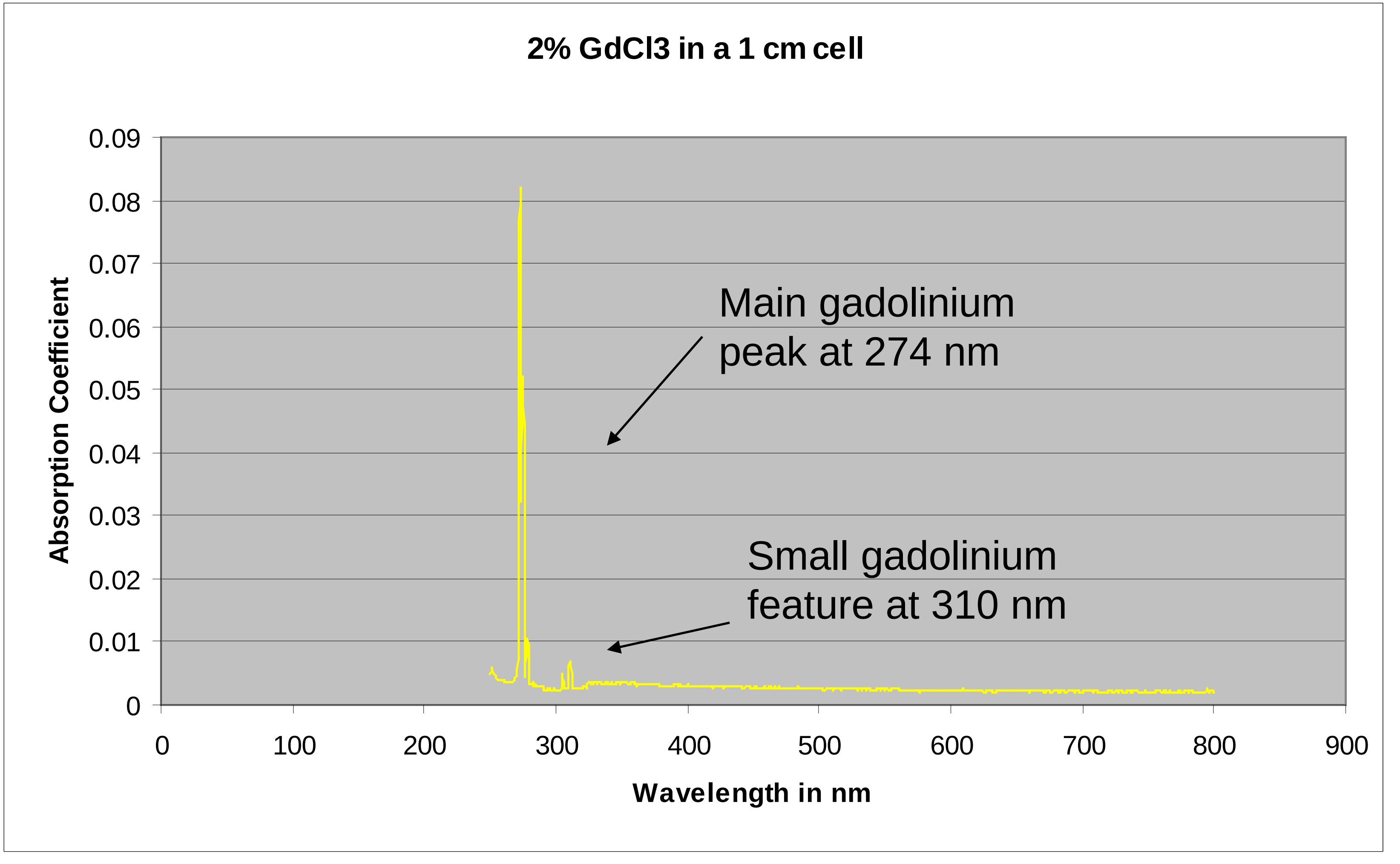
Unfortunately, eight years of exposure to ultra-pure water had led to large areas (~20% of the total surface) of corrosion. The $GdCl_3$ rapidly began lifting this pre-existing rust into solution.

We needed a new Gd compound!

To select the best gadolinium compound we have to balance optical and mechanical effects:

Name	Formula	Pros	Cons
Gadolinium Chloride	$GdCl_3$	Low Cost High Solubility Safety Transparency	Corrosion
Gadolinium Nitrate	$Gd(NO_3)_3$	Low Cost High Solubility Low Corrosion	Absorbs UV
Gadolinium Sulfate	$Gd_2(SO_4)_3$	Transparency Low Corrosion	Low pH Lower Solubility

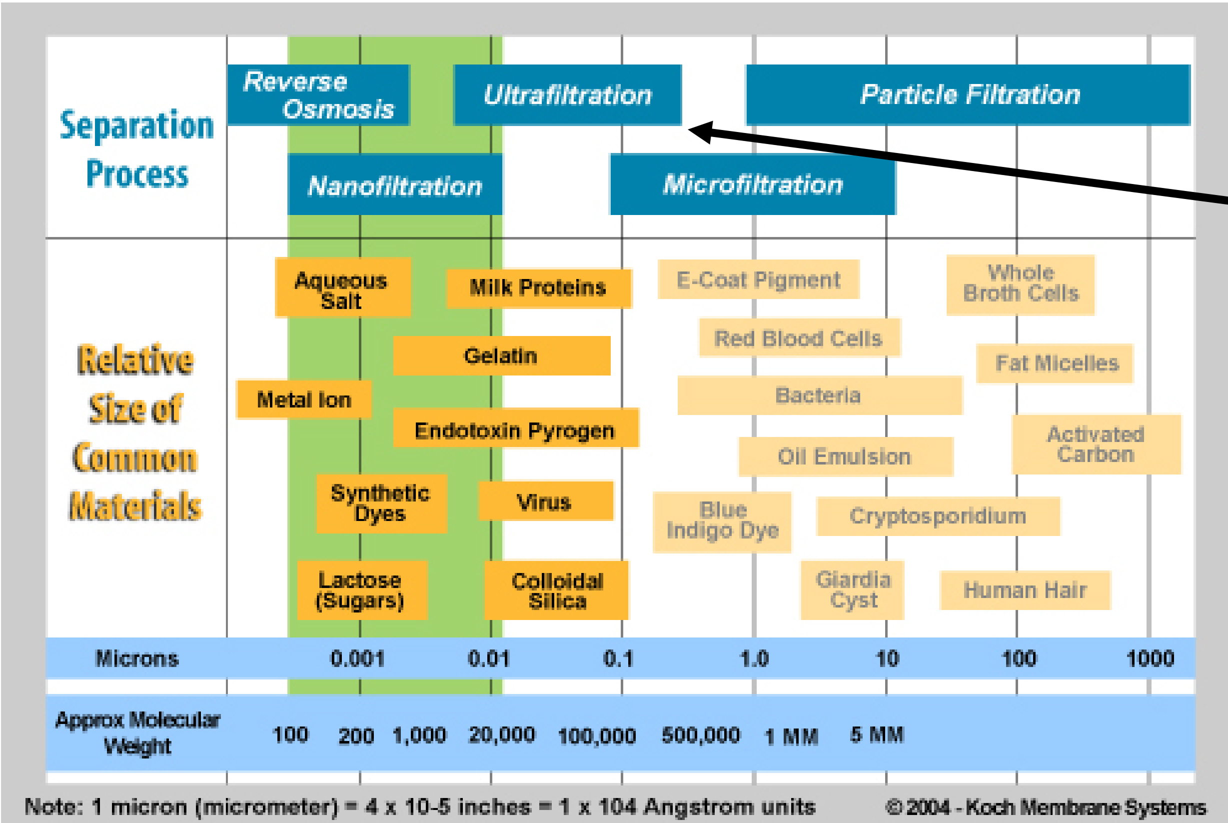
Measured using a Lambda 900 UV/VIS spectrophotometer



An absorption coefficient of 0.01 means 98% of the light survives

This plot corresponds to an attenuation length of ~70 meters @ 0.2%

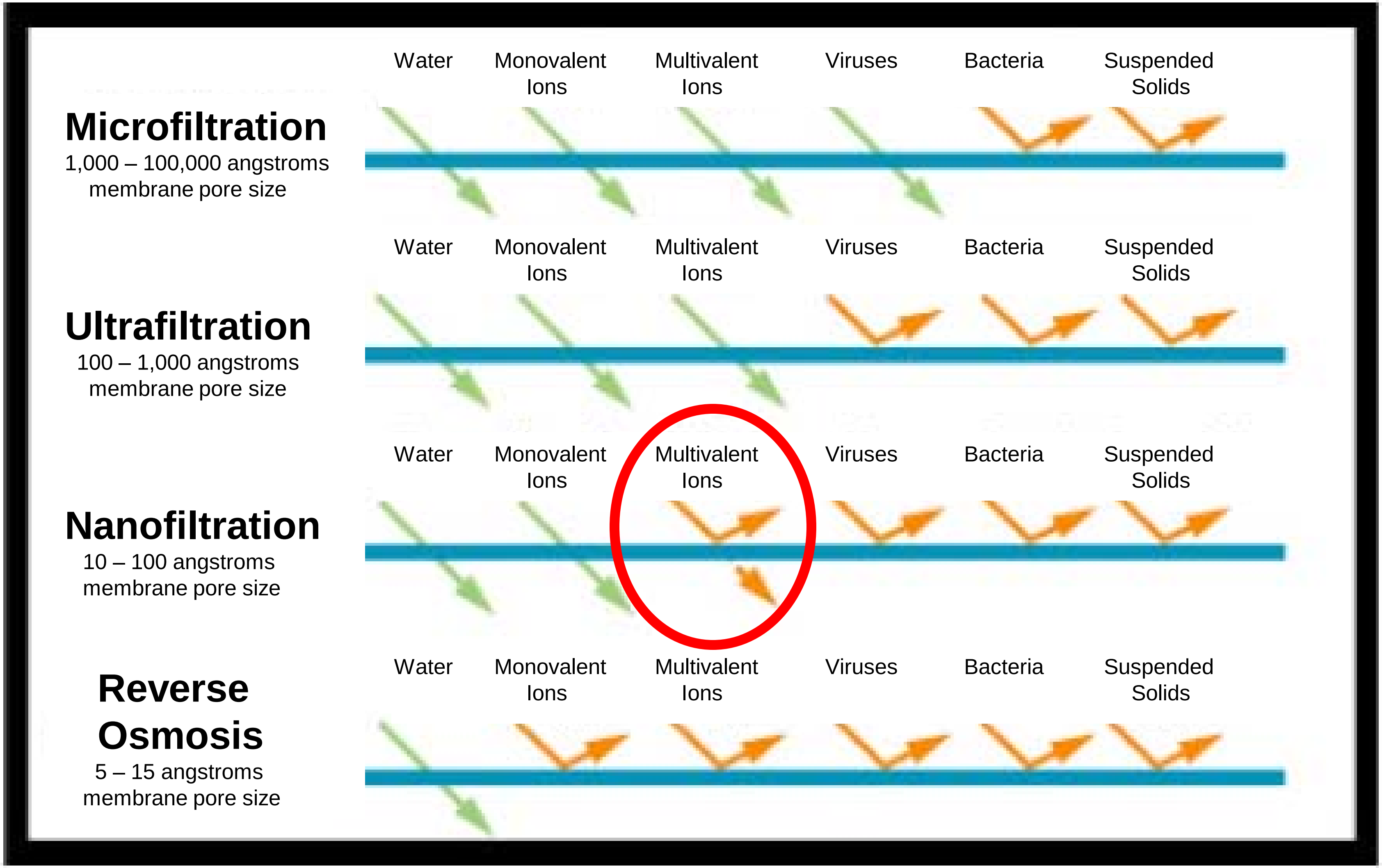
But what we *really* want is true selective filtration.



Adding nanofiltration (NF) to the SK water system should make this possible.

Membrane-based Filtering Technologies

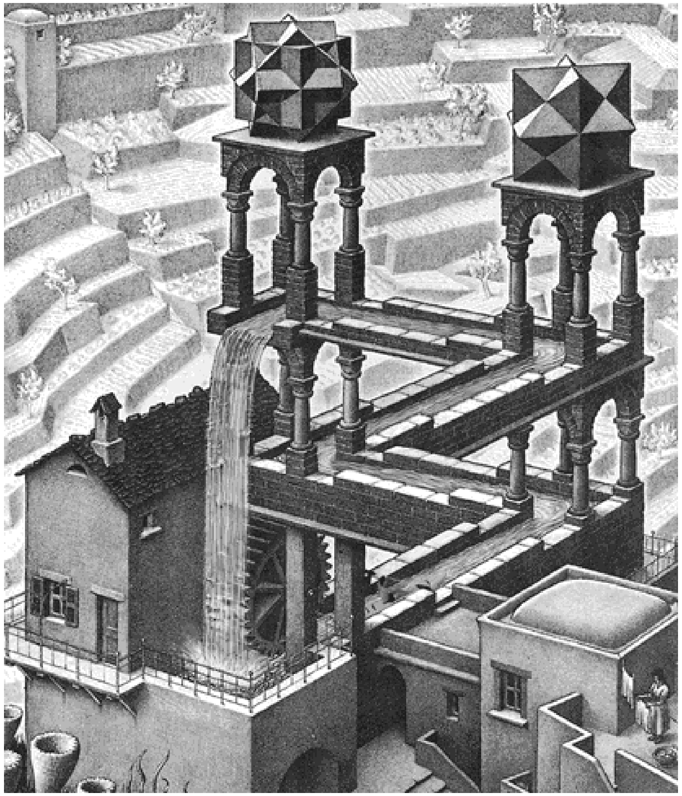
$$Gd_2(SO_4)_3 \rightarrow 2\ Gd^{3+} + 3\ (SO_4)^{2-}$$



The Essential Magic Trick

→ We must keep the water in any Gd-loaded detector perfectly clean… *without removing the dissolved Gd.*

→ I've developed a new technology:
"Molecular Band-Pass Filtration"
Staged nanofiltration selectively retains Gd while removing impurities.



Amazingly, the darn thing works!

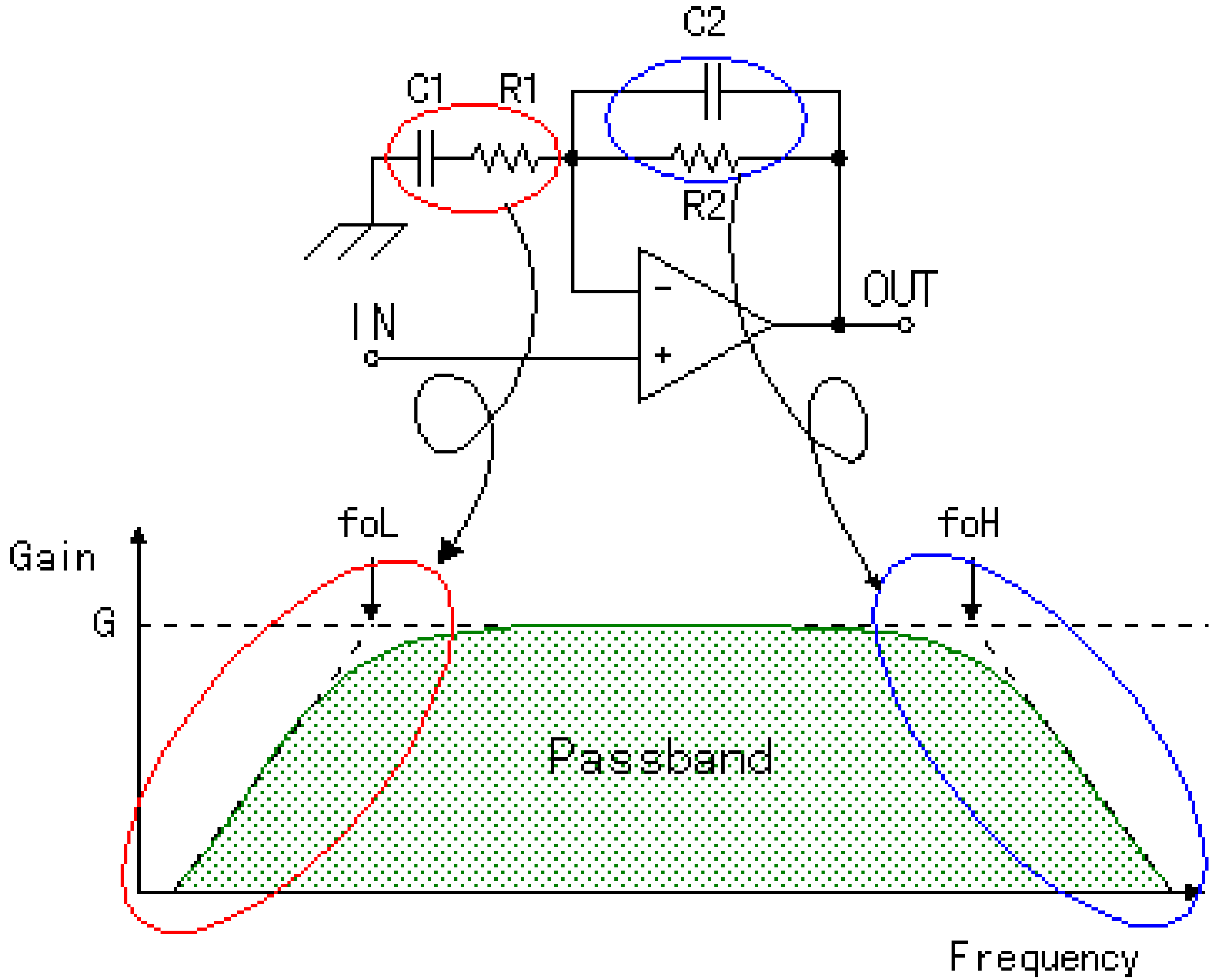
This technology will support a variety of applications, such as:

→ Supernova neutrino and proton decay searches

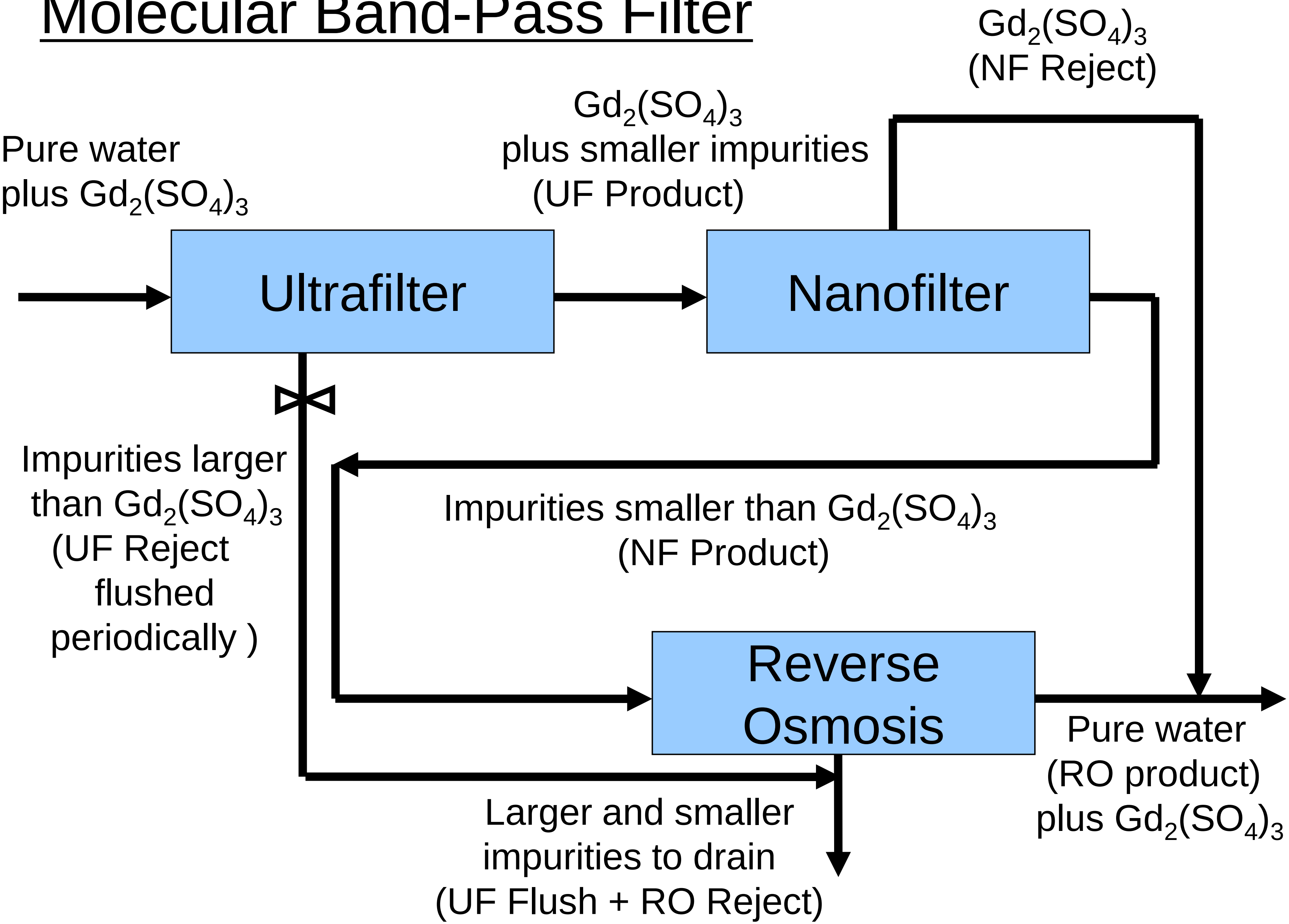
→ Remote detection of clandestine fissile material production

→ Efficient generation of clean drinking water without electricity

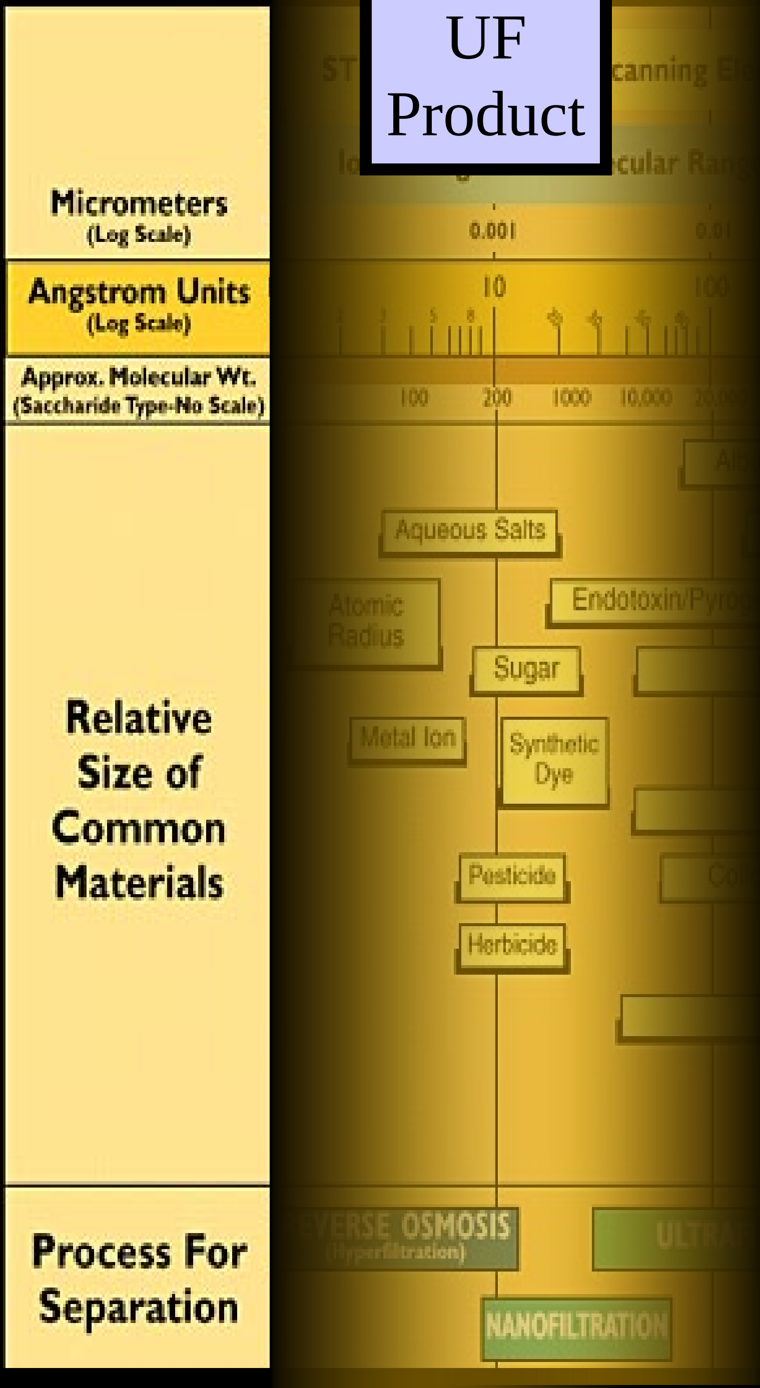
Electrical Band-Pass Filter



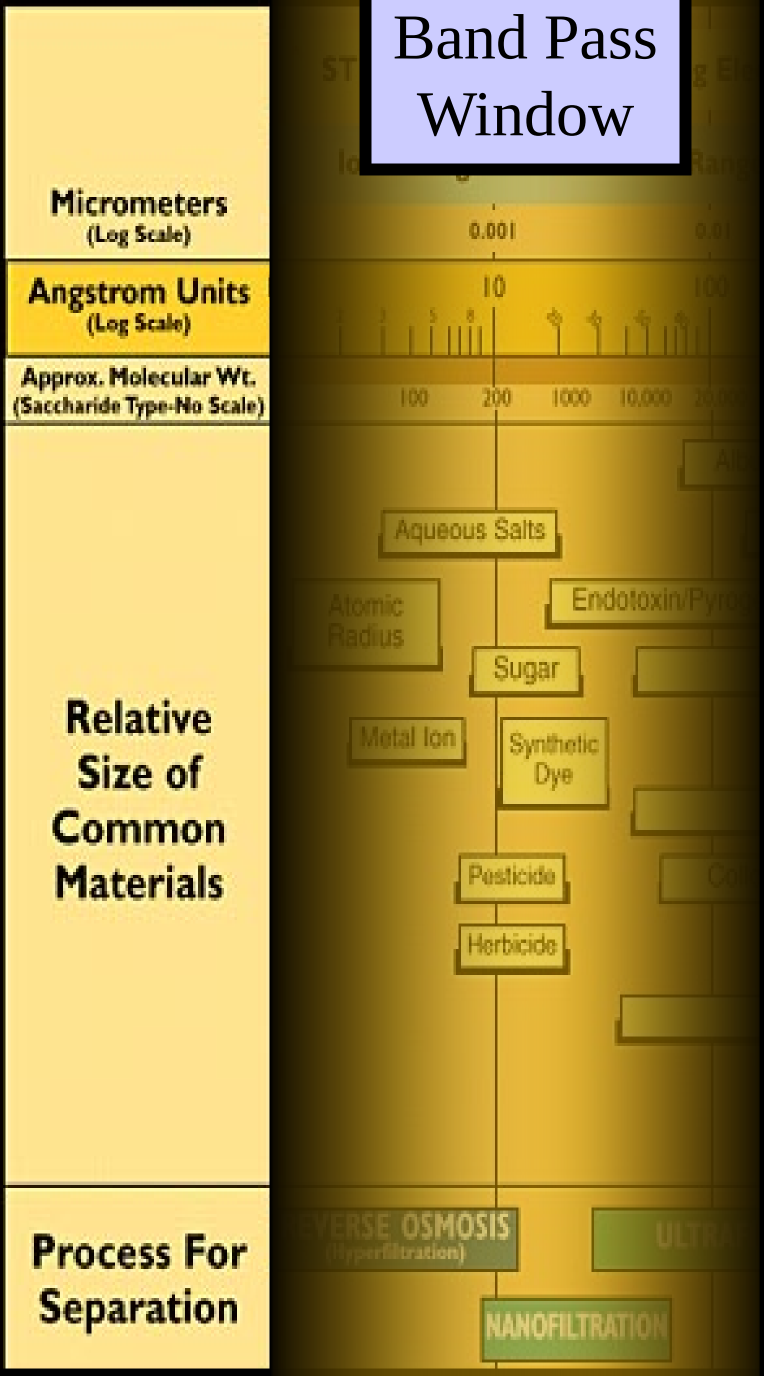
Molecular Band-Pass Filter



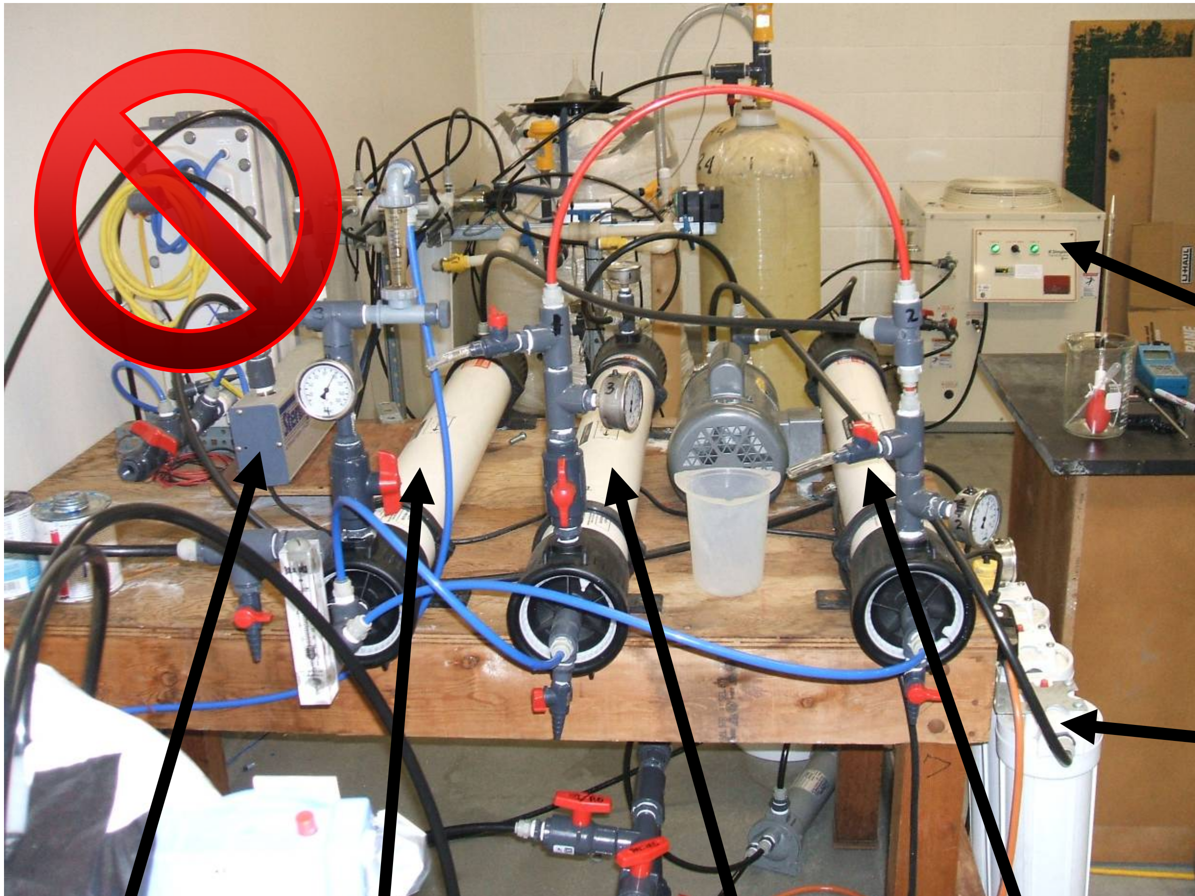
UF Product


Micrometers
(Log Scale)
0.001
Angstrom Units
(Log Scale)
10
100
Approx. Molecular Wt.
(Saccharide Type-No Scale)
100
200
1000
10,000
Relative Size of Common Materials
Aqueous Salts
Atomic Radius
Sugar
Metal Ion
Synthetic Dye
Pesticide
Herbicide
Process For Separation
NANOFILTRATION

Band Pass Window



Selective Filtration Prototype Setup @ UCI



Chiller

0.2 → 5 Micron Filters

UV Sterilizer

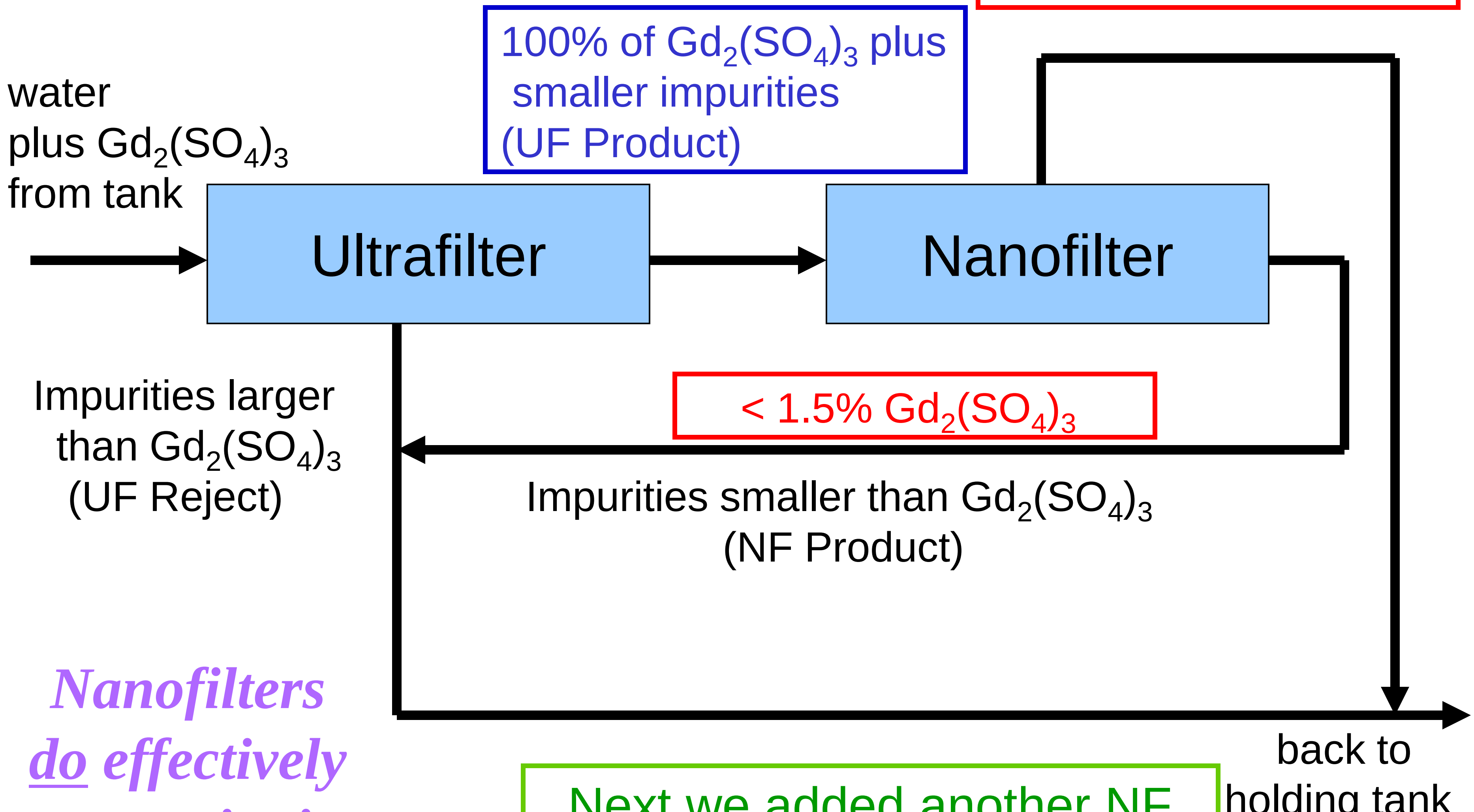
Ultrafilter

Nanofilter

Reverse Osmosis

Initial Test of Nanofilter

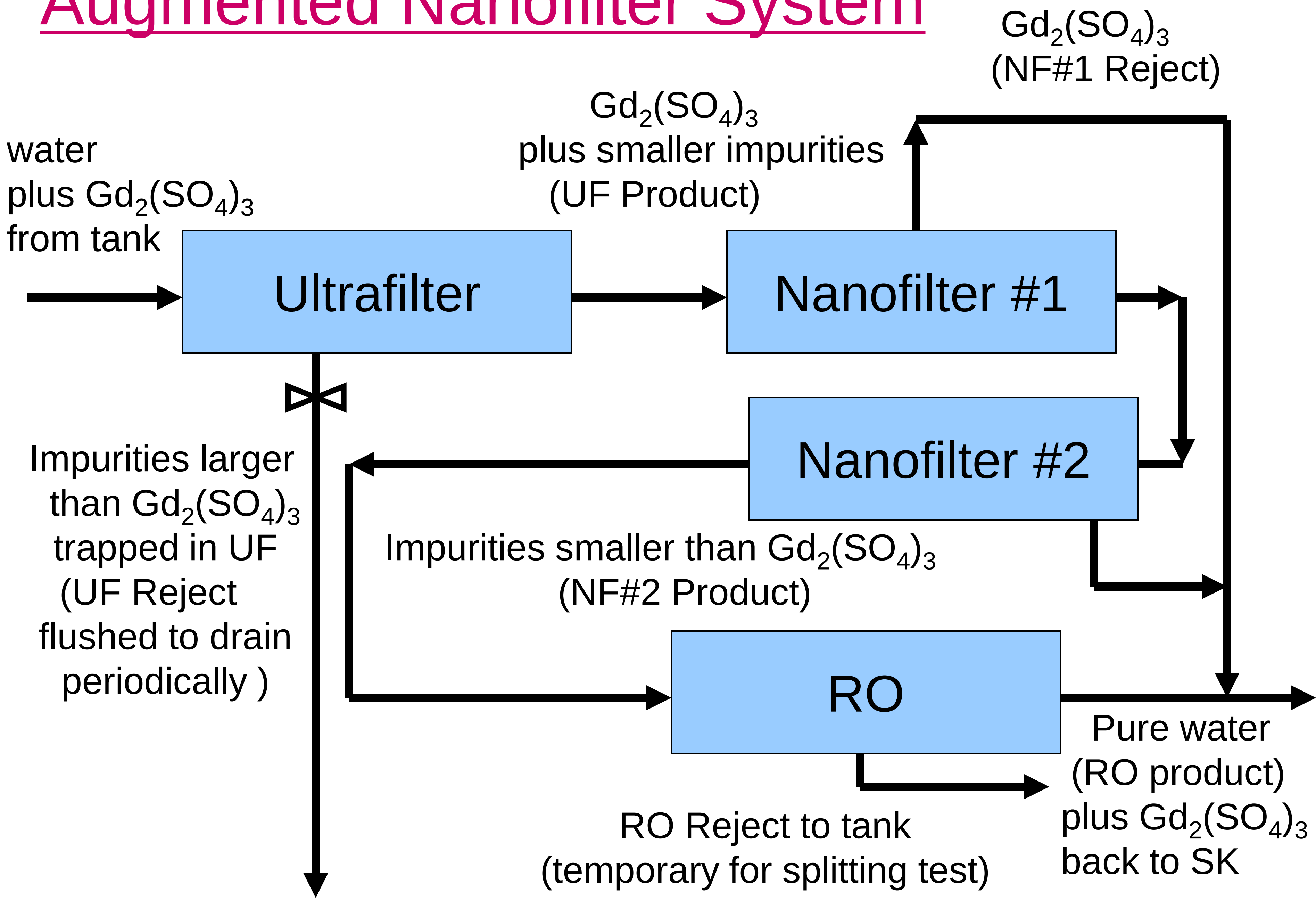
> 98.5% $Gd_2(SO_4)_3$
(single stage NF Reject)



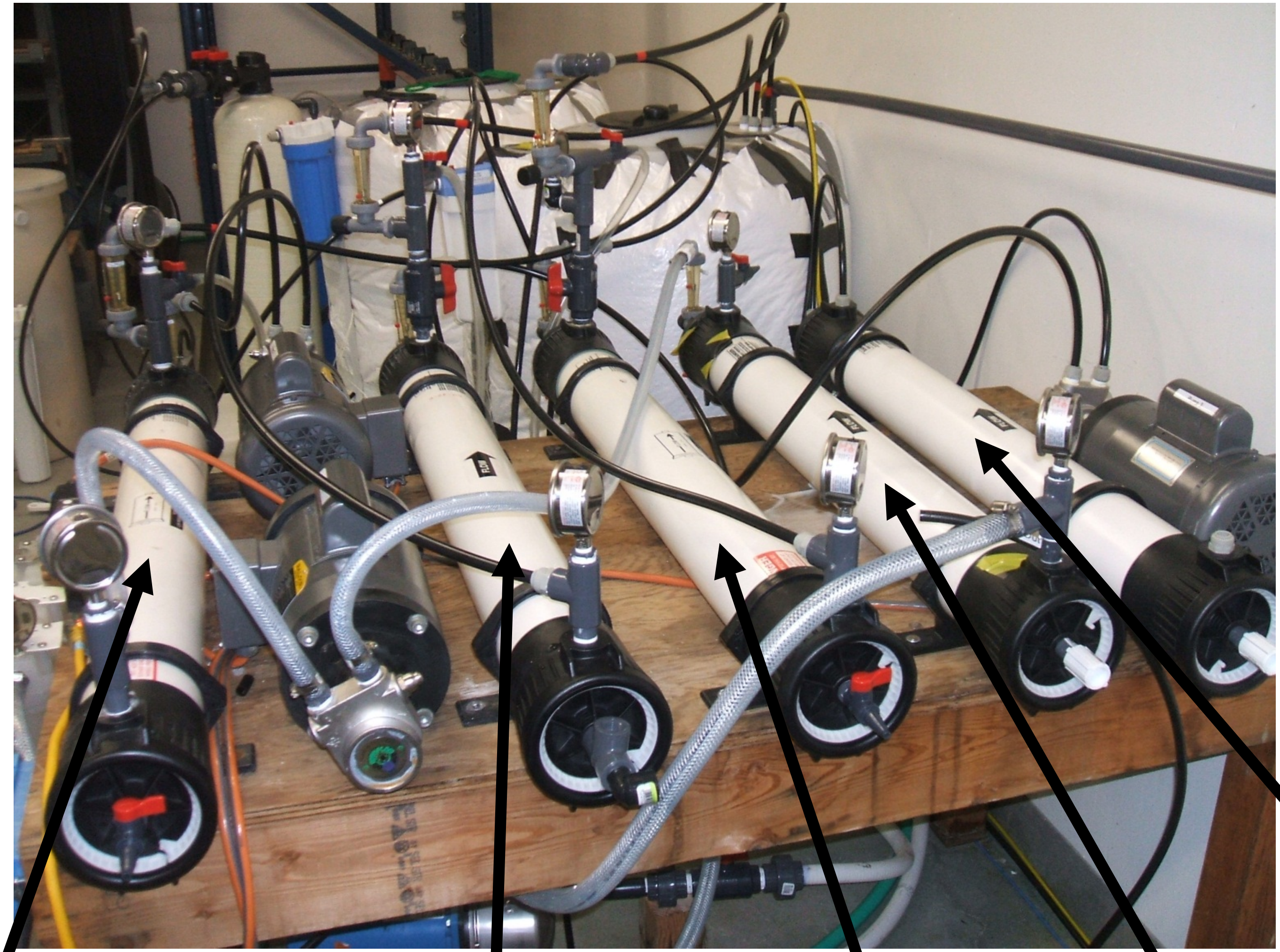
Nanofilters <u>do</u> effectively remove ionic Gd and SO_4

Next we added another NF stage to see if we could get even better separation

Augmented Nanofilter System



Current Selective Filtration Setup @ UCI



Membrane Pre-Flush

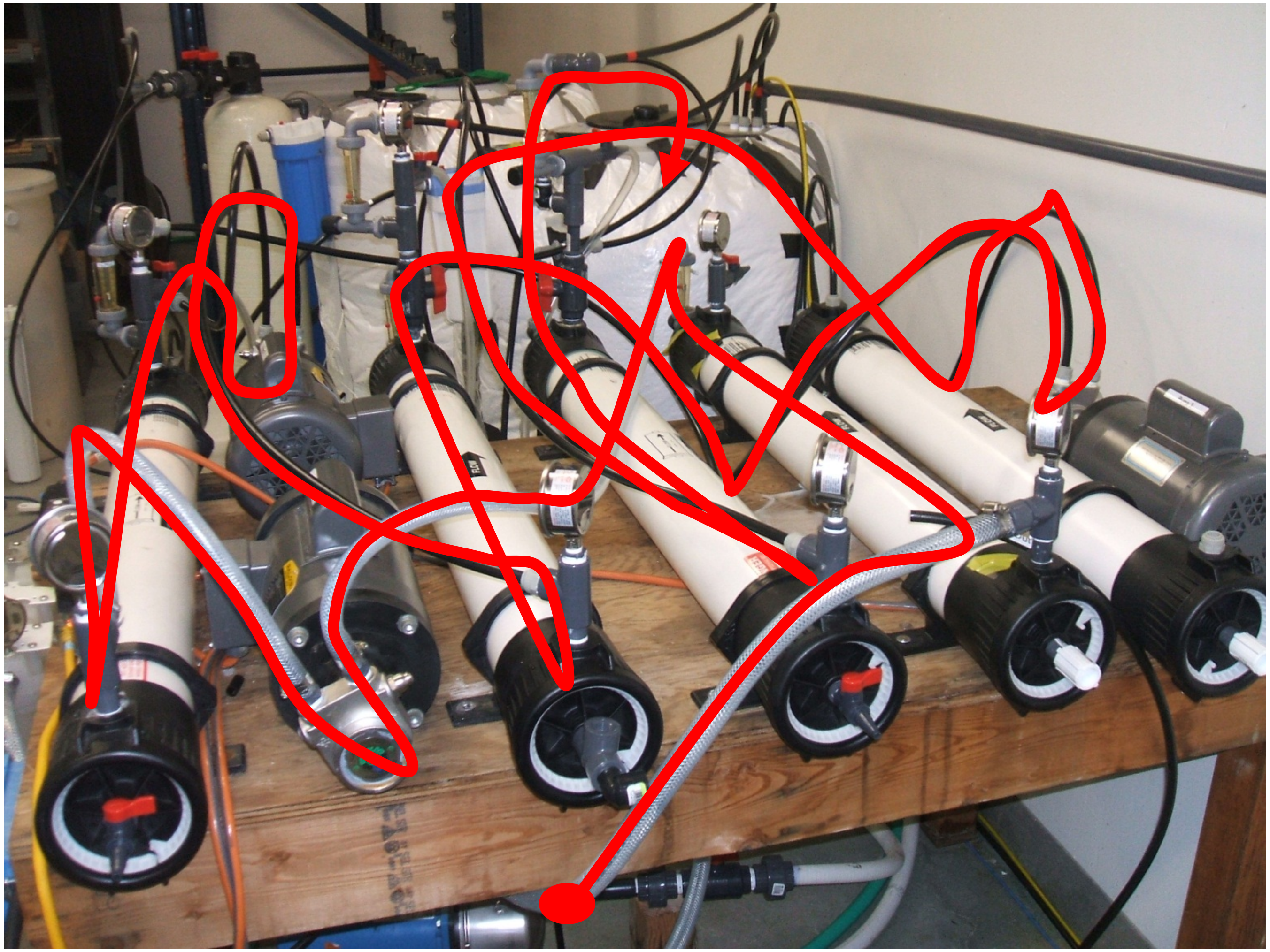
Nanofilter #1

Nanofilter #2

Reverse Osmosis

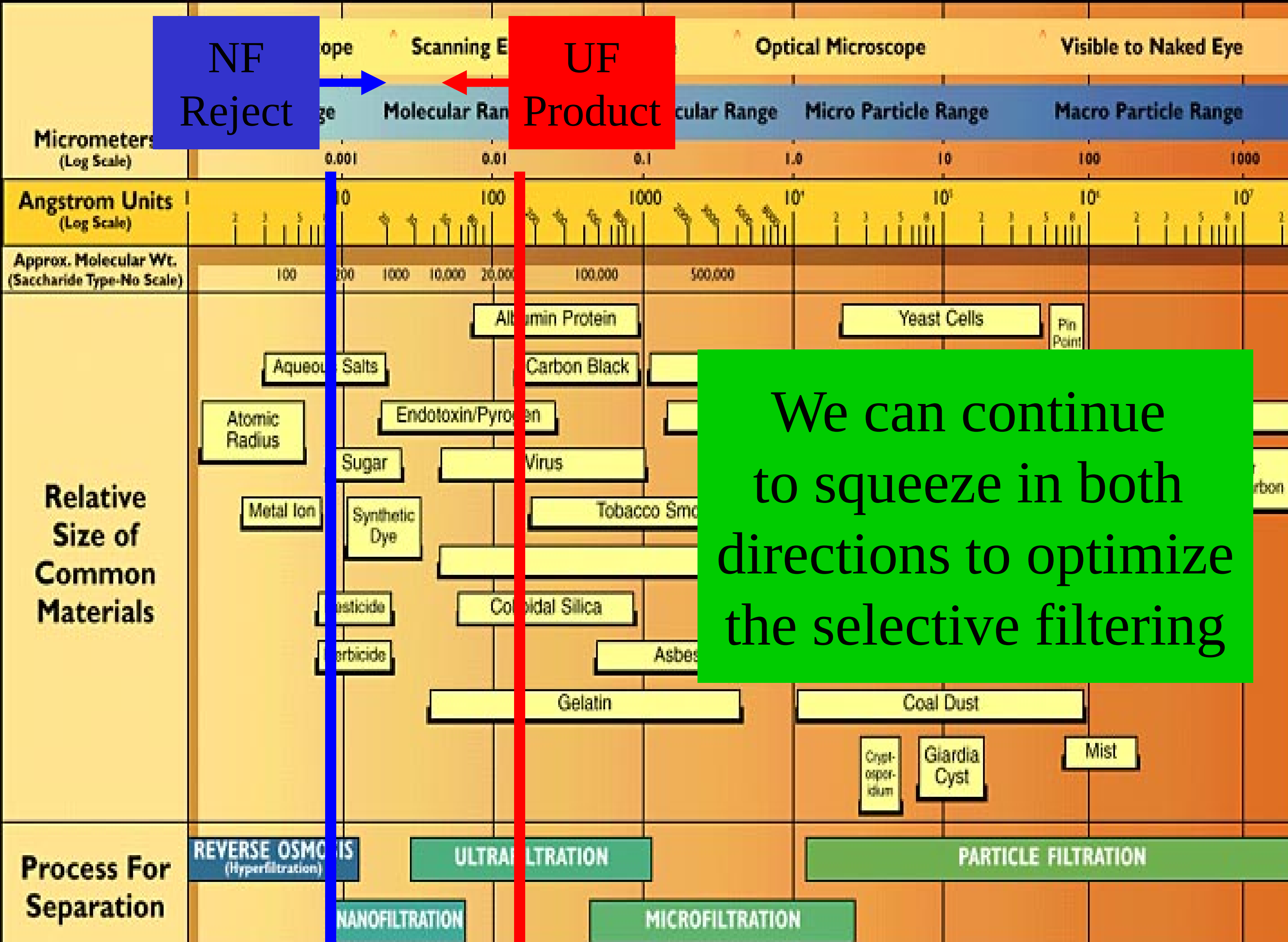
Ultrafilter

Current Selective Filtration Setup @ UCI

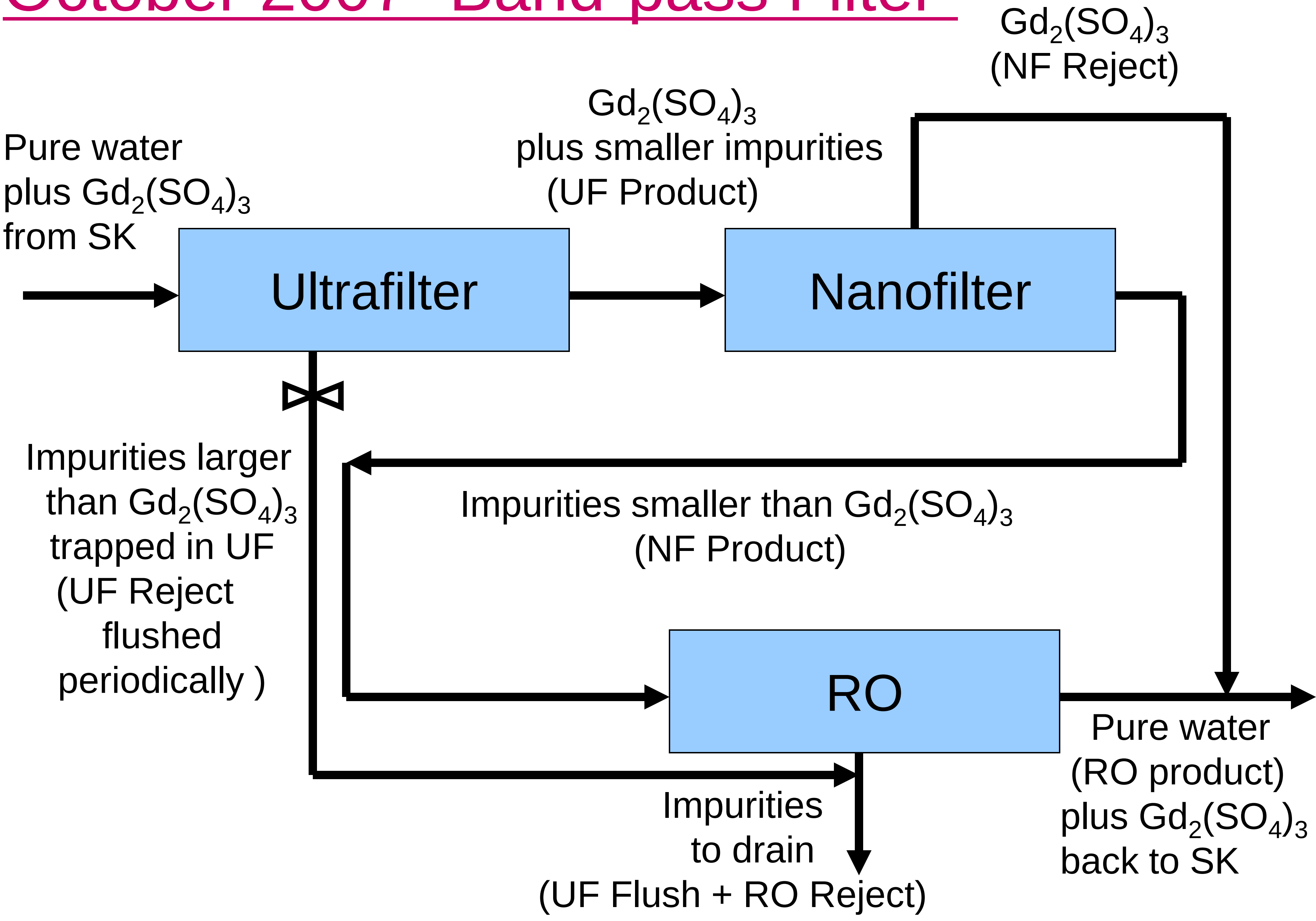


Membrane
Pre-Flush

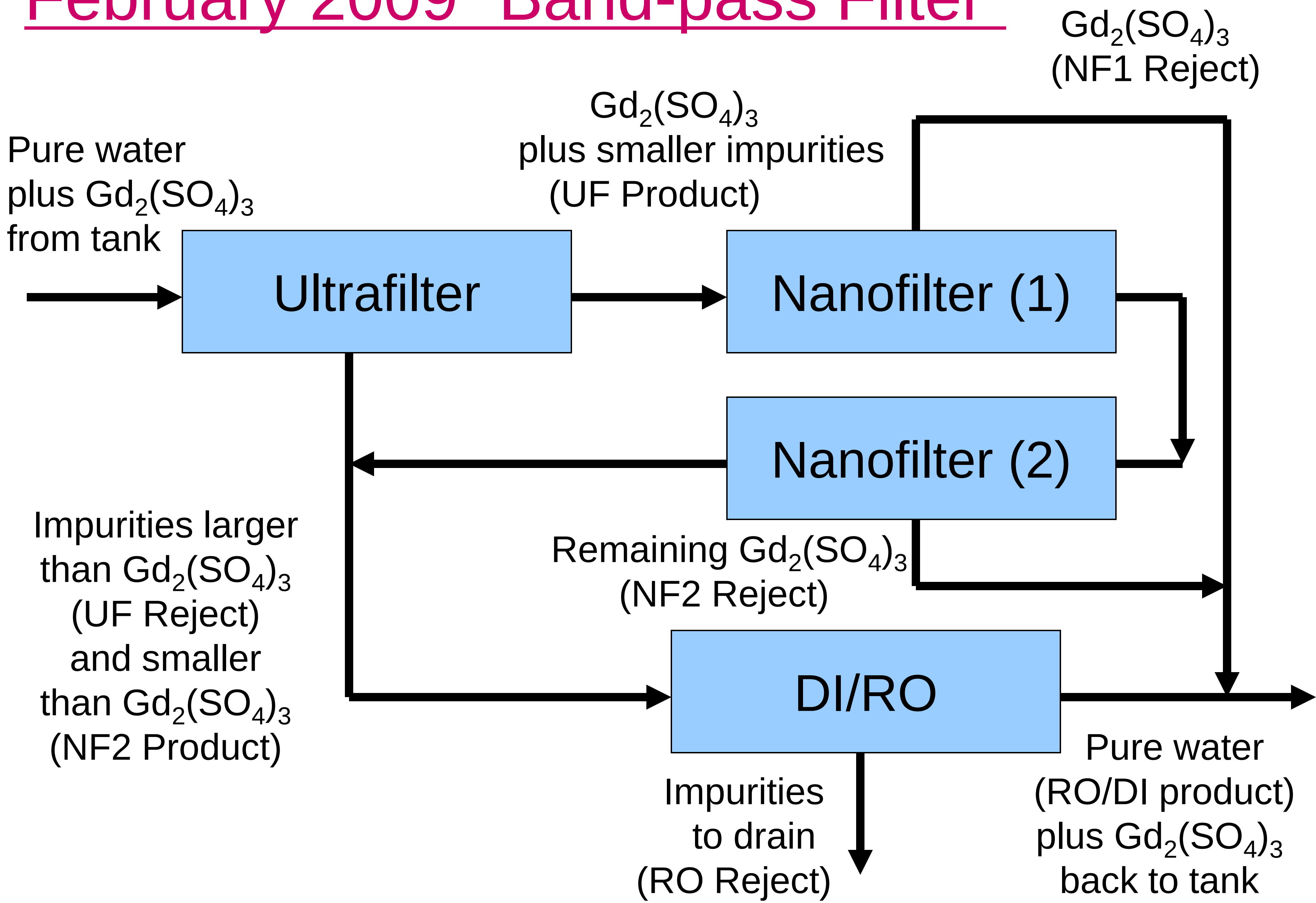
Nanofilter #1 Nanofilter #2 Reverse Osmosis Ultrafilter


NF Reject
UF Product
We can continue to squeeze in both directions to optimize the selective filtering
Scanning E
Optical Microscope
Visible to Naked Eye
Molecular Ran
cular Range
Micro Particle Range
Macro Particle Range
Micrometers
(Log Scale)
0.001
0.01
0.1
1.0
10
100
1000
Angstrom Units
(Log Scale)
Approx. Molecular Wt.
(Saccharide Type-No Scale)
100
200
1000
10,000
20,000
100,000
500,000
Relative Size of Common Materials
Albumin Protein
Yeast Cells
Pin Point
Aqueous Salts
Carbon Black
Atomic Radius
Endotoxin/Pyrogen
Sugar
Virus
Metal Ion
Synthetic Dye
Tobacco Smo
Colloidal Silica
Gelatin
Coal Dust
Crypt-osporidium
Giardia Cyst
Mist
Process For Separation
REVERSE OSMOSIS
(Hyperfiltration)
ULTRAFILTRATION
PARTICLE FILTRATION
NANOFILTRATION
MICROFILTRATION

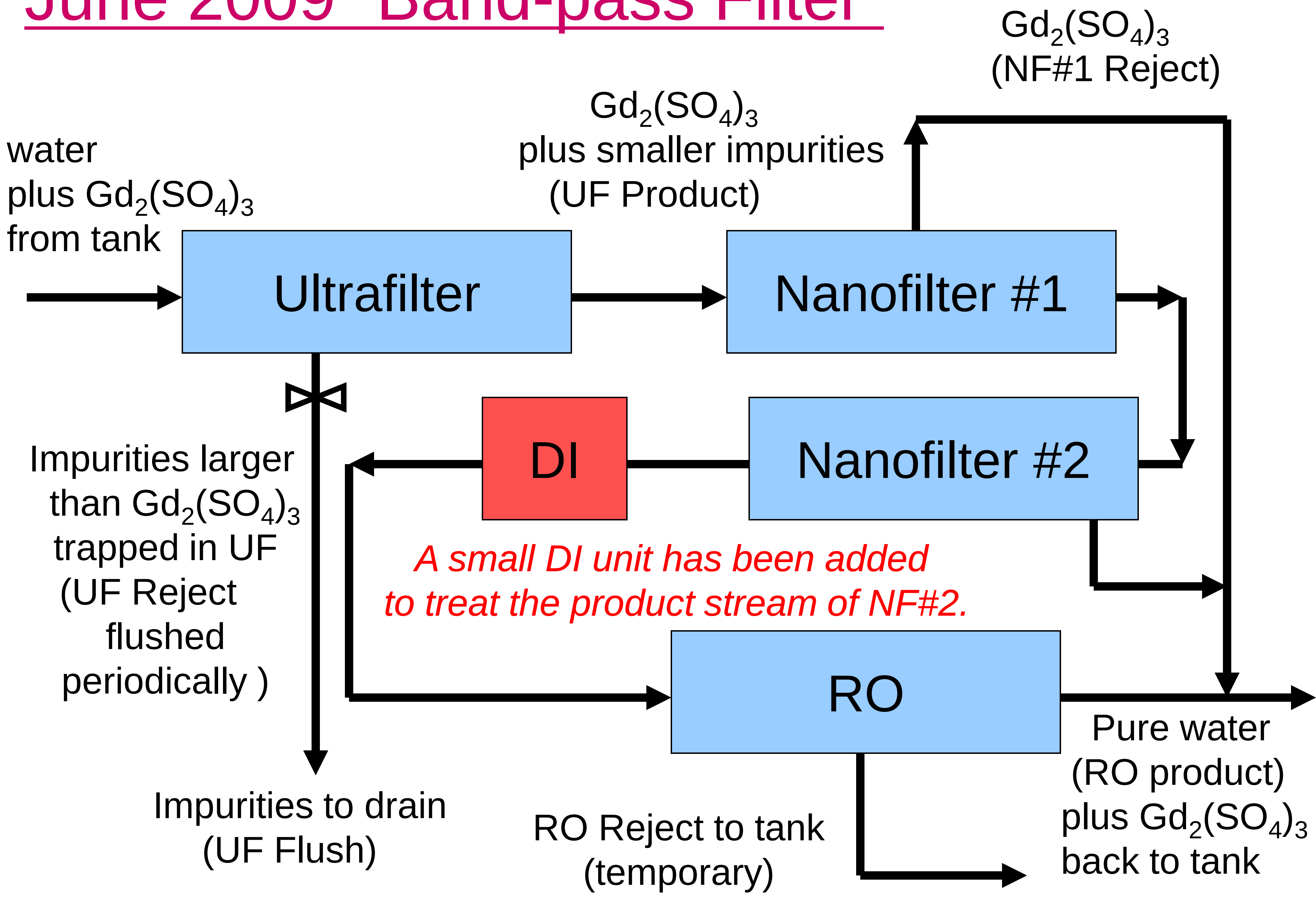
October 2007 “Band-pass Filter”



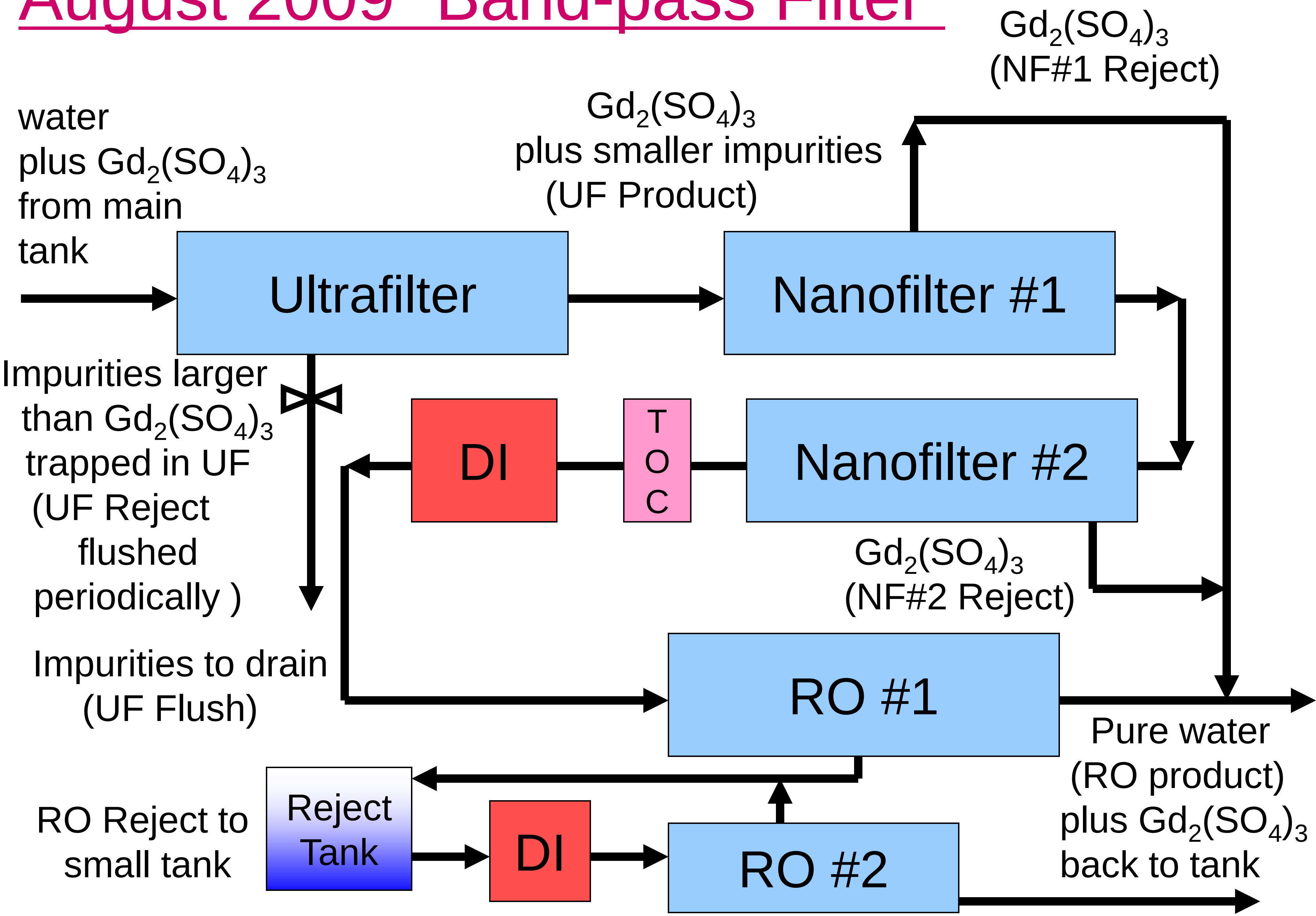
February 2009 “Band-pass Filter”



June 2009 "Band-pass Filter"



August 2009 “Band-pass Filter”



Augus…

This design works well. It is the world's first operational selective filtration system.

→ Water quality is indefinitely maintained/improved, with or without gadolinium.

→ There is <60 ppb loss of Gd per cycle.

However, the prototype system at UCI processes just 0.2 tons of water per hour.

It must be industrialized to be of use in SK…

water plus Gd_2(S… from main tank

Impurities la… than Gd_2(S… trapped in… (UF Rejec… flushed… periodical…

Impurities… (UF Flu…

RO Rejec… small tank

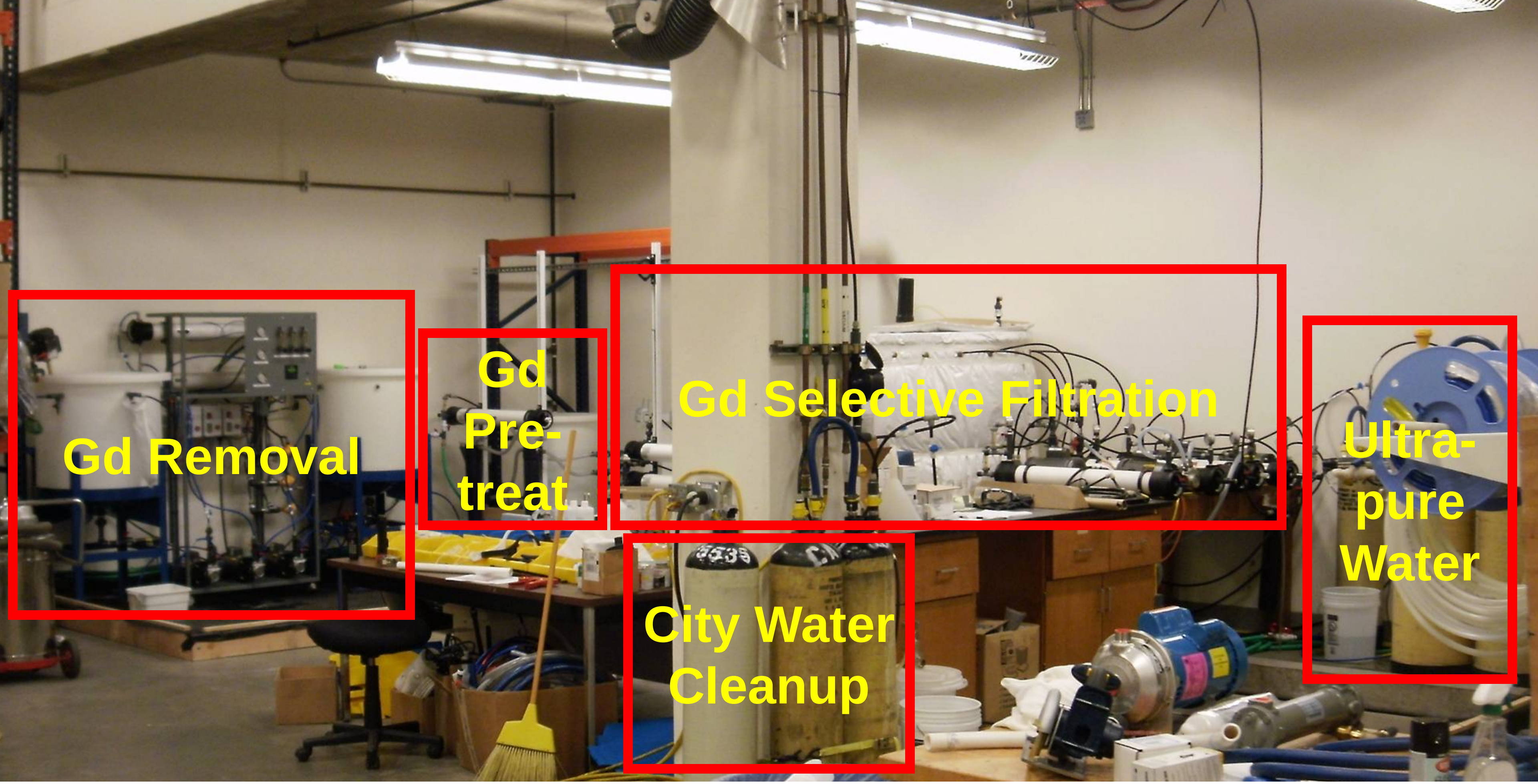
…$SO_4)_3$ … Reject)

…Pure water …RO product) …us $Gd_2(SO_4)_3$ back to tank

…Tank

DI

RO #2



Water Systems at UCI

(not shown - a material emanation soak system and an improved ultrapure water system)

In 2008 I underwent a significant transformation…

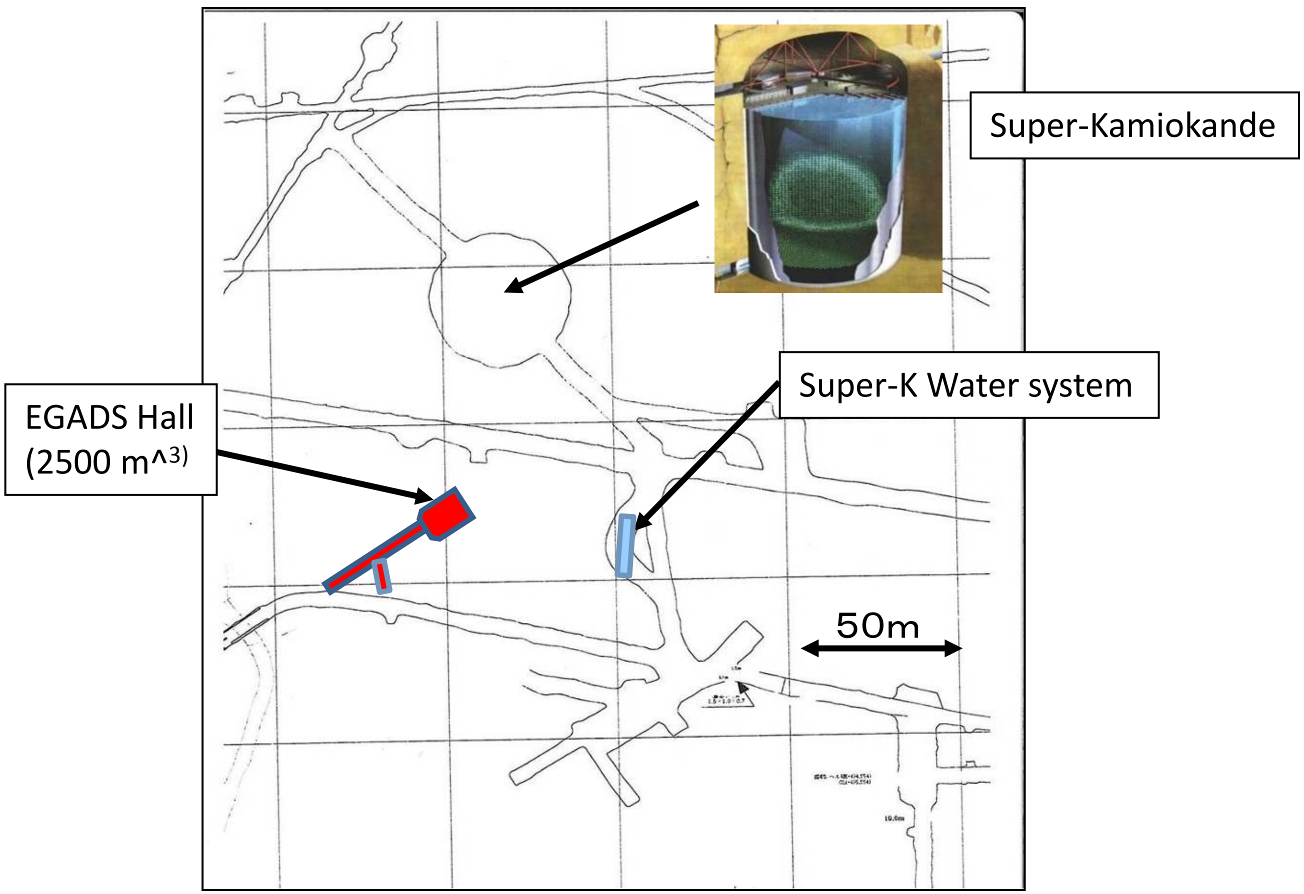
I joined UTokyo's newly-formed IPMU as their first full-time *gaijin* professor, though I still retain a "without salary" position at UCI and continue Gd studies there.

I was explicitly hired to make gadolinium work in water!

INSTITUTE FOR THE PHYSICS AND MATHEMATICS OF THE UNIVERSE

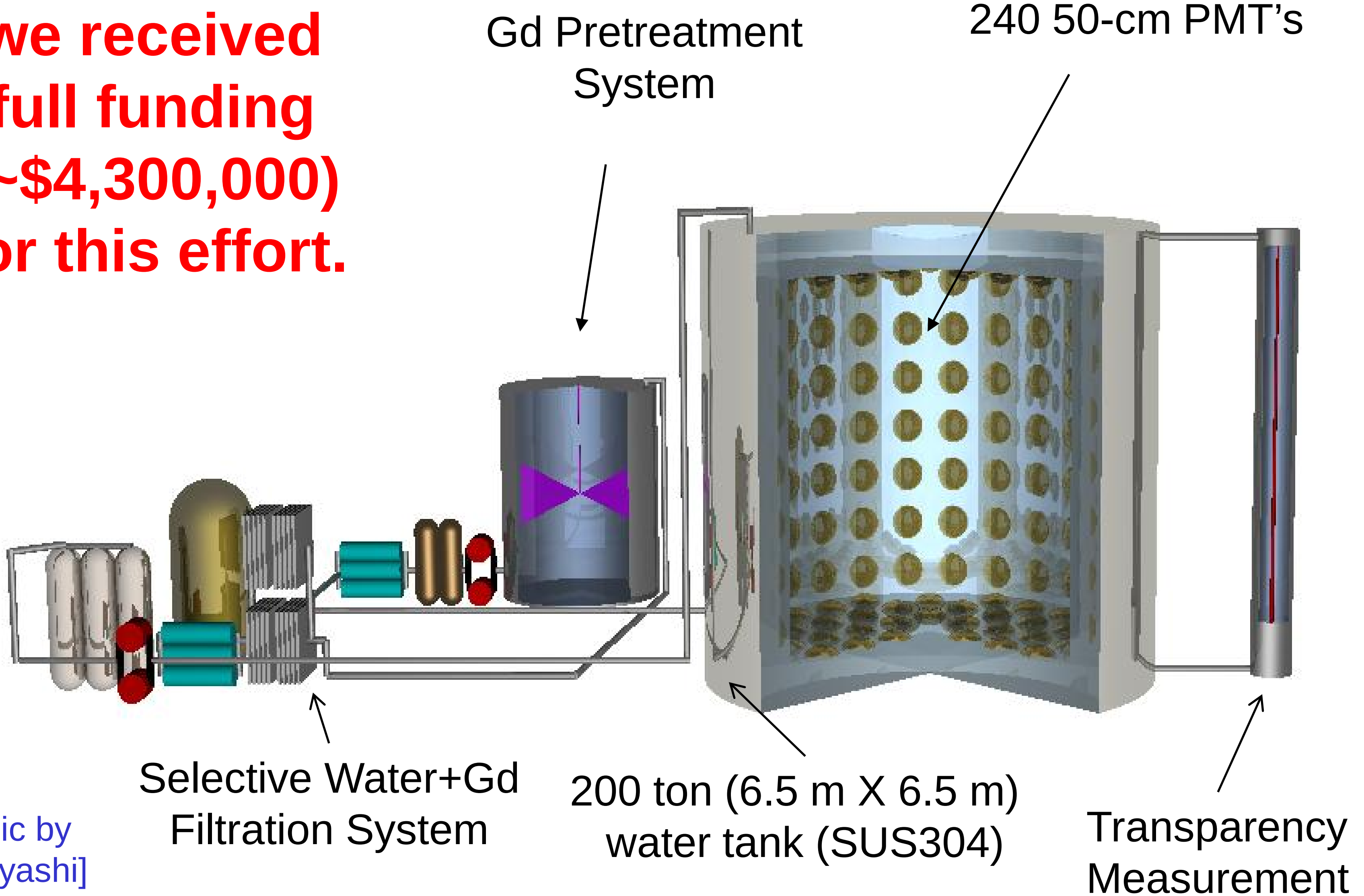
A dedicated Gd test facility has been built in the Kamioka mine, complete with its own water filtration system, 50-cm PMT's, and DAQ electronics.

This 200 ton-scale R&D project is called EGADS – Evaluating Gadolinium's Action on Detector Systems.

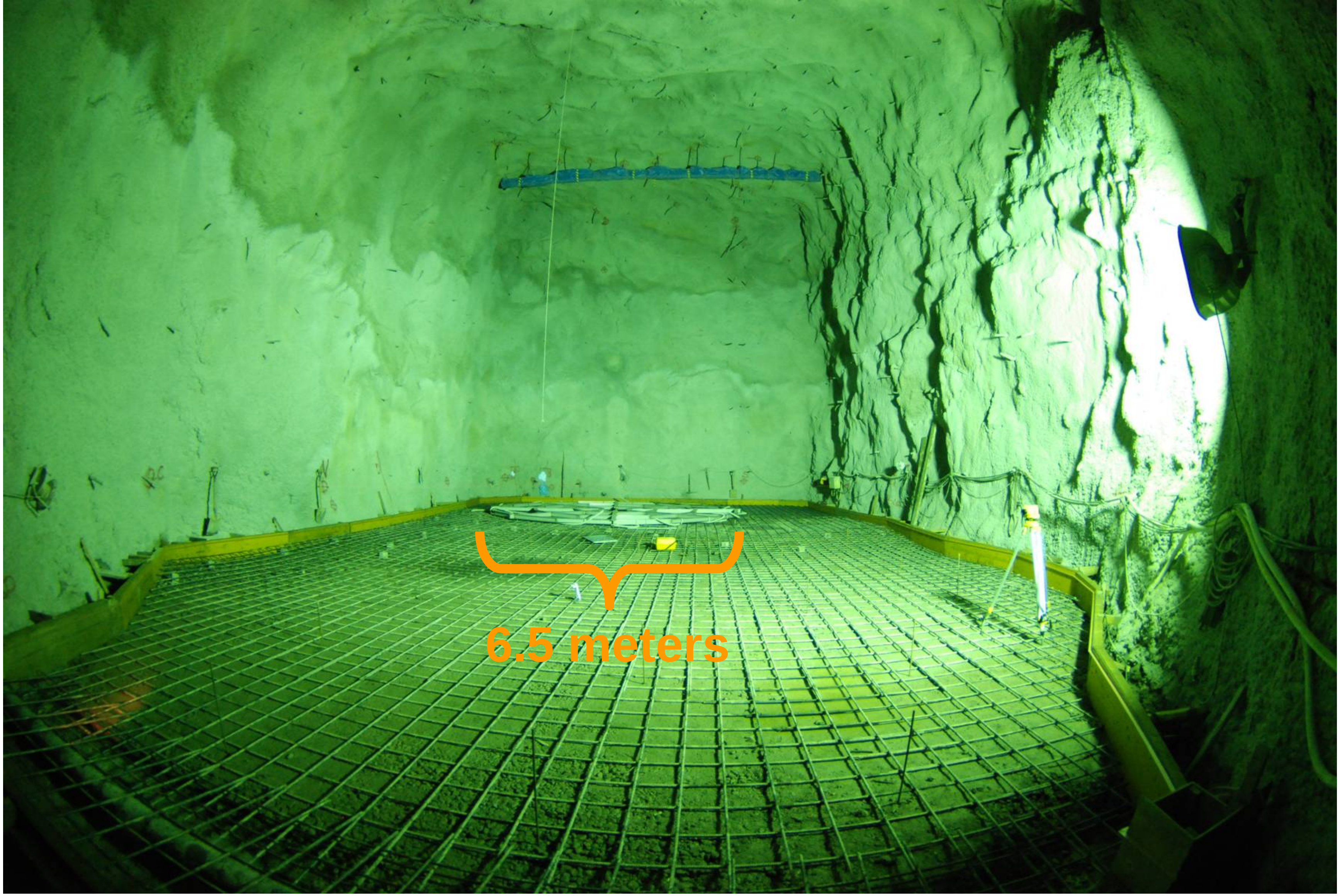


EGADS Facility

In June of 2009 we received full funding (~$4,300,000) for this effort.

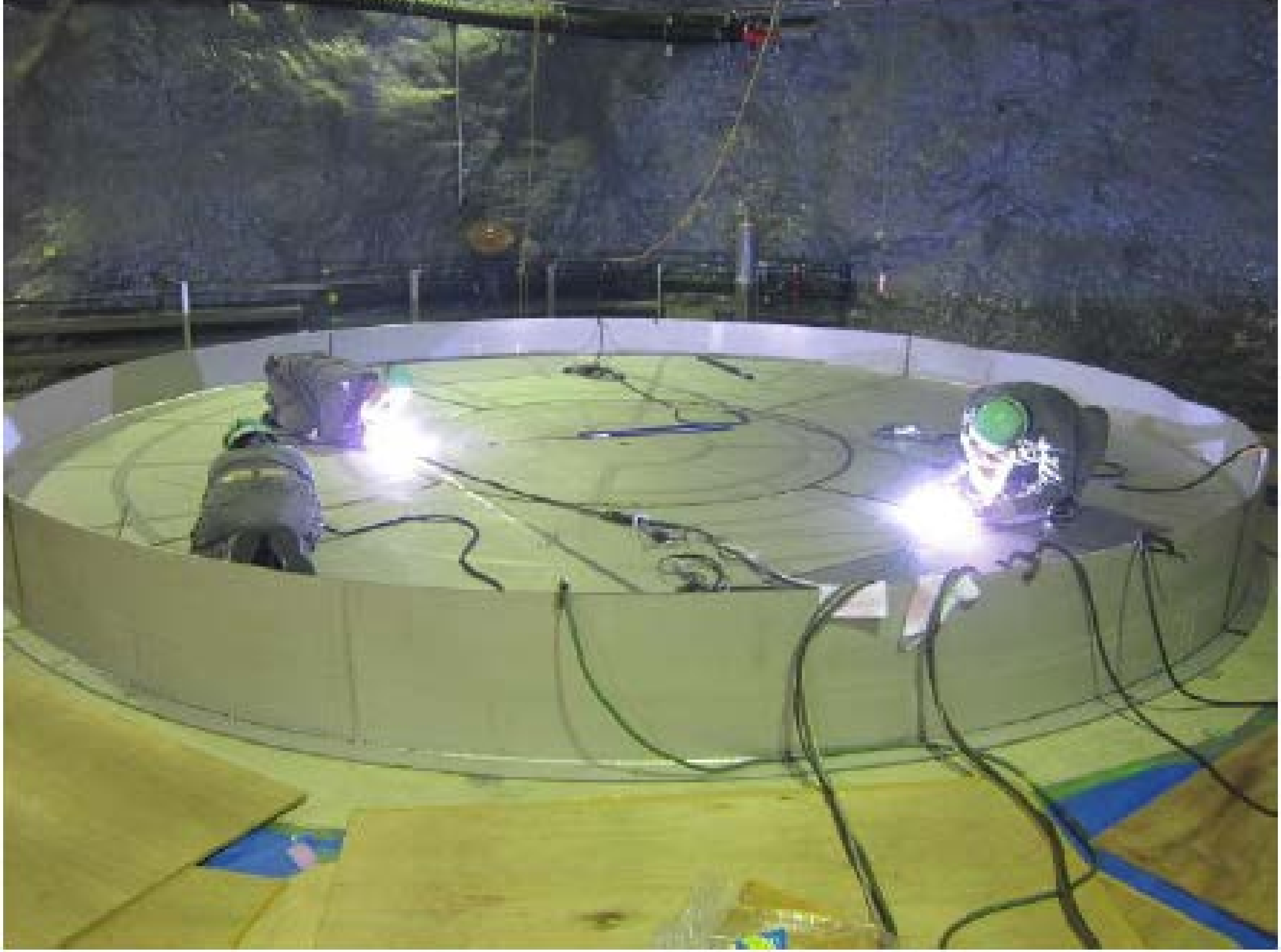


[graphic by A. Kibayashi]



EGADS Cavern as of December 14, 2009

EGADS Cavern as of February 27, 2010



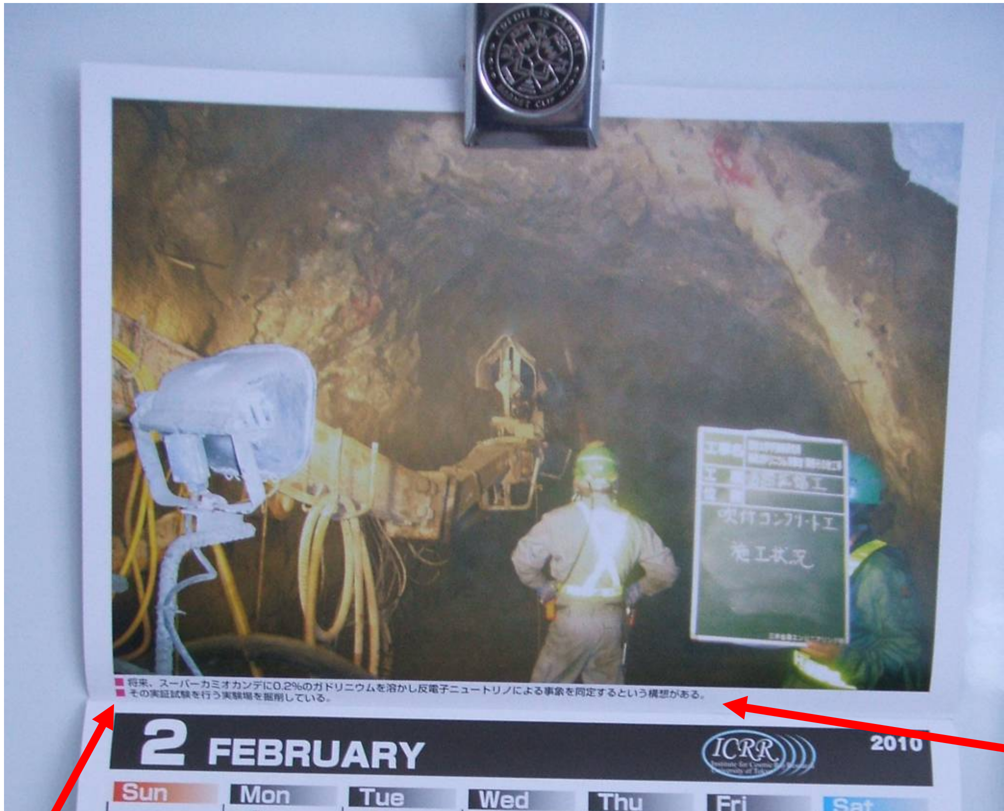
EGADS Cavern as of April 16, 2010

EGADS Cavern as of April 28, 2010

EGADS Cavern as of June 8, 2010

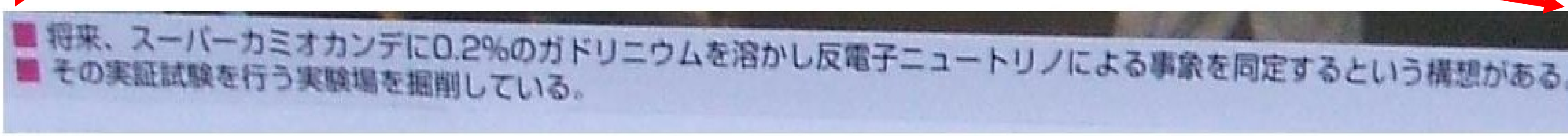
EGADS Cavern as of December 10, 2010

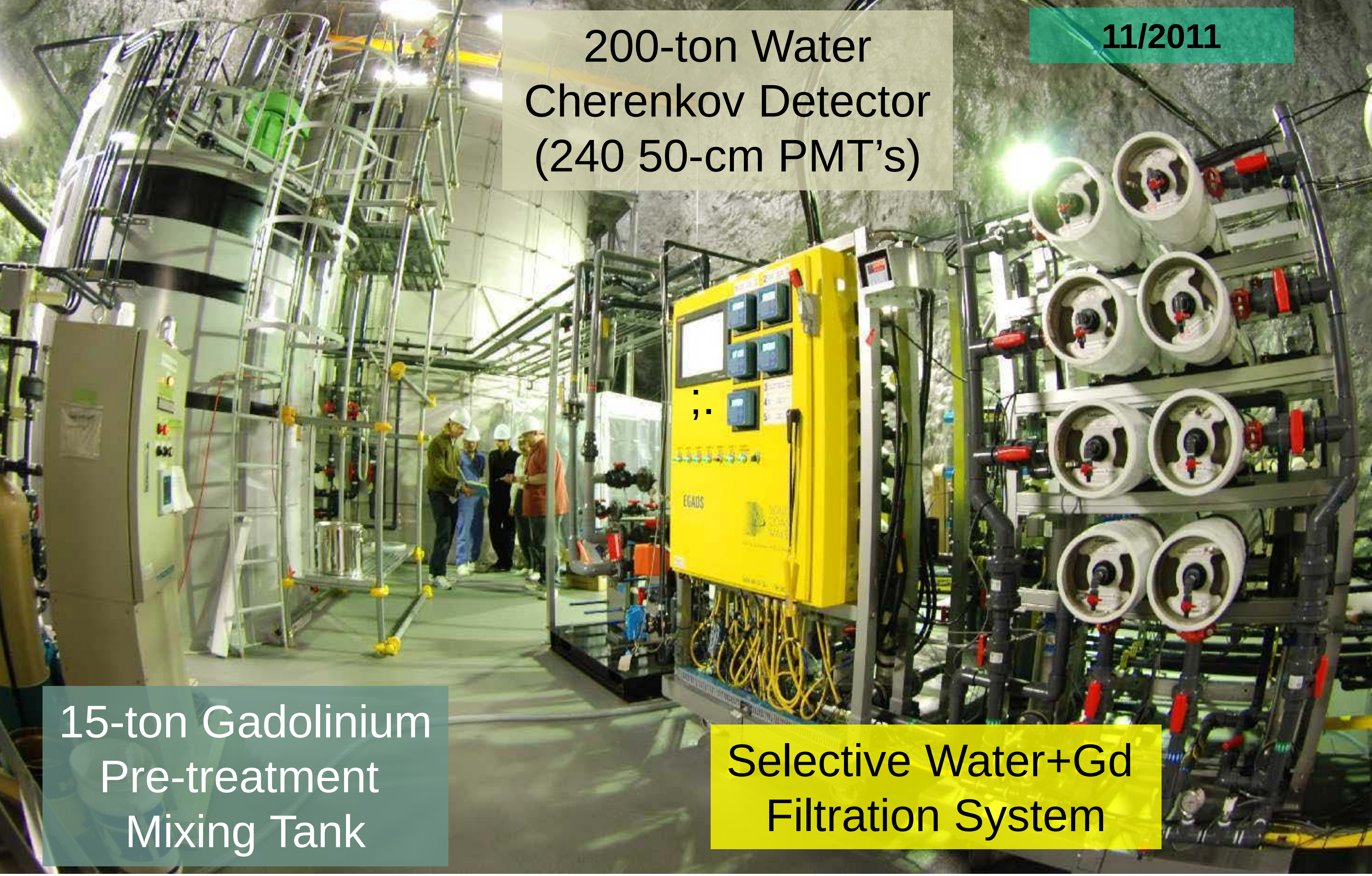
Just another Thanksgiving weekend; Nov. 25th, 2011



Here's the official Institute for Cosmic Ray Research [ICRR] calendar:

EGADS was Miss February in 2010, and Miss March in 2012!

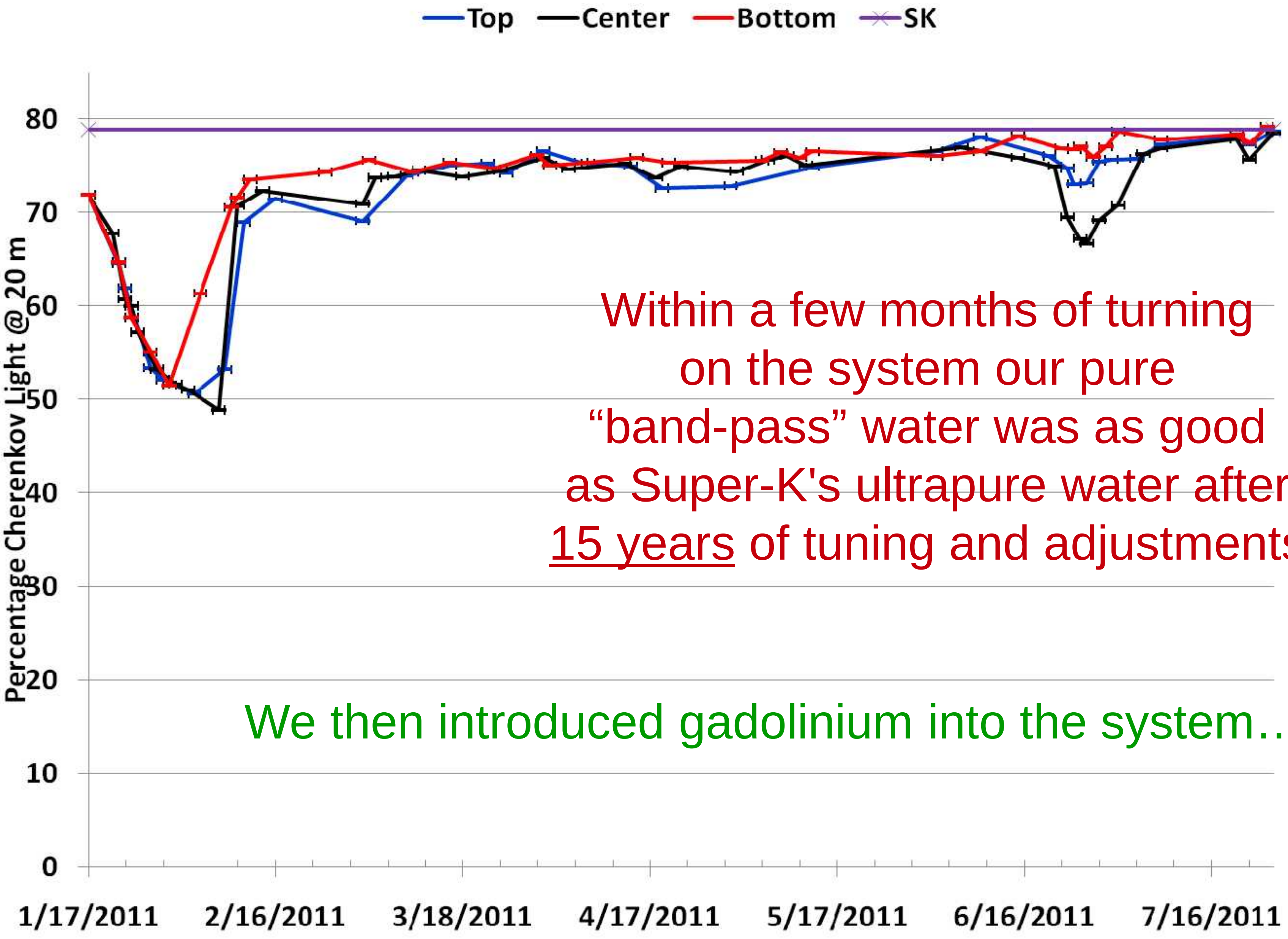




By next year, EGADS will have shown conclusively whether or not gadolinium loading of Super-Kamiokande will be safe and effective.

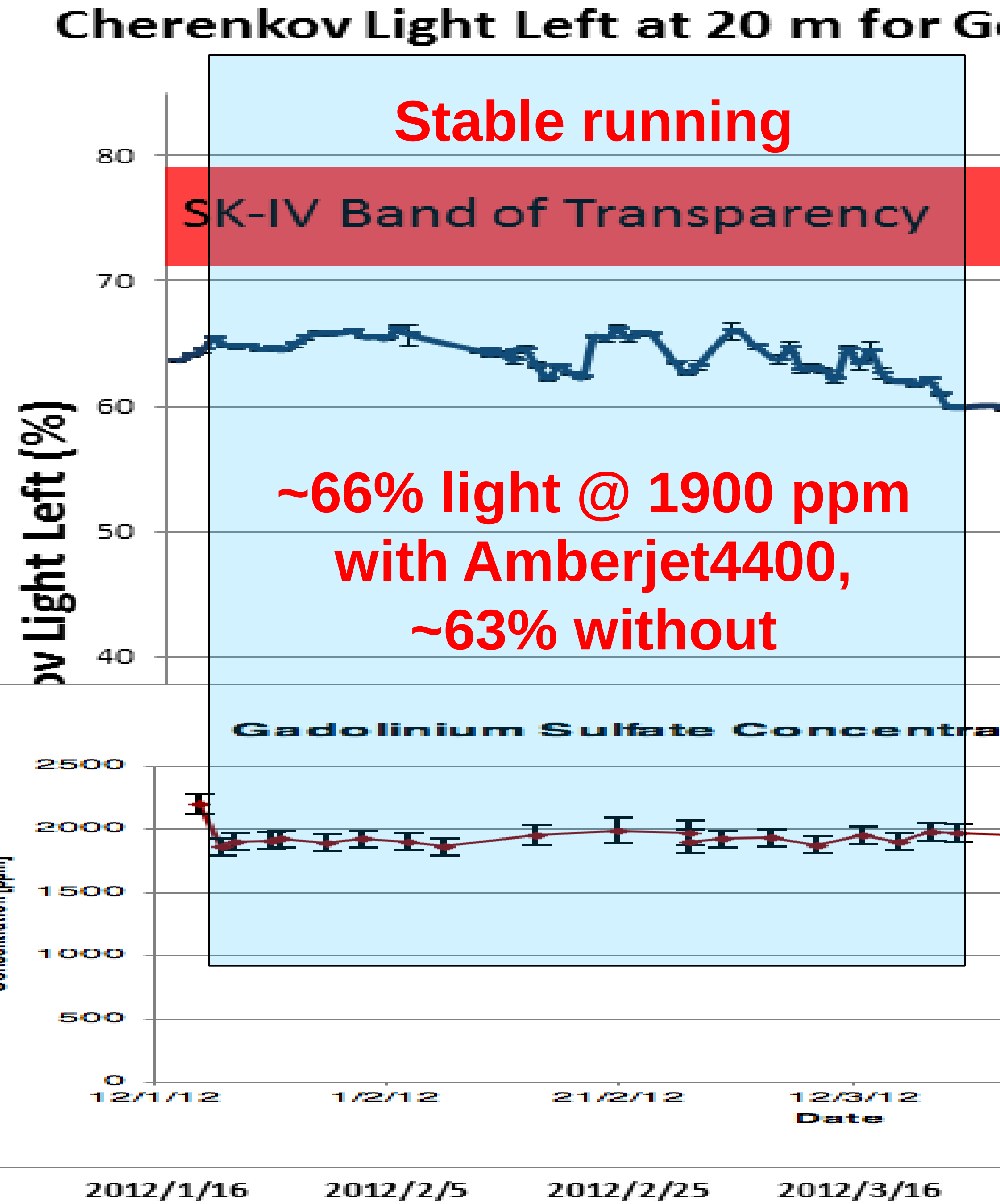
<u>**If so, this is the likely future of *all* water Cherenkov detectors.**</u>

Cherenkov Light Remaining at 20 m (200-ton tank)



Within a few months of turning on the system our pure "band-pass" water was as good as Super-K's ultrapure water after 15 years of tuning and adjustments!

We then introduced gadolinium into the system…



Studies continue, but we have already achieved stable light levels of 66% at 20 meters with fully Gd-loaded water.

This should be compared to a range of 71%→79% for "perfect" pure water in SK-IV.

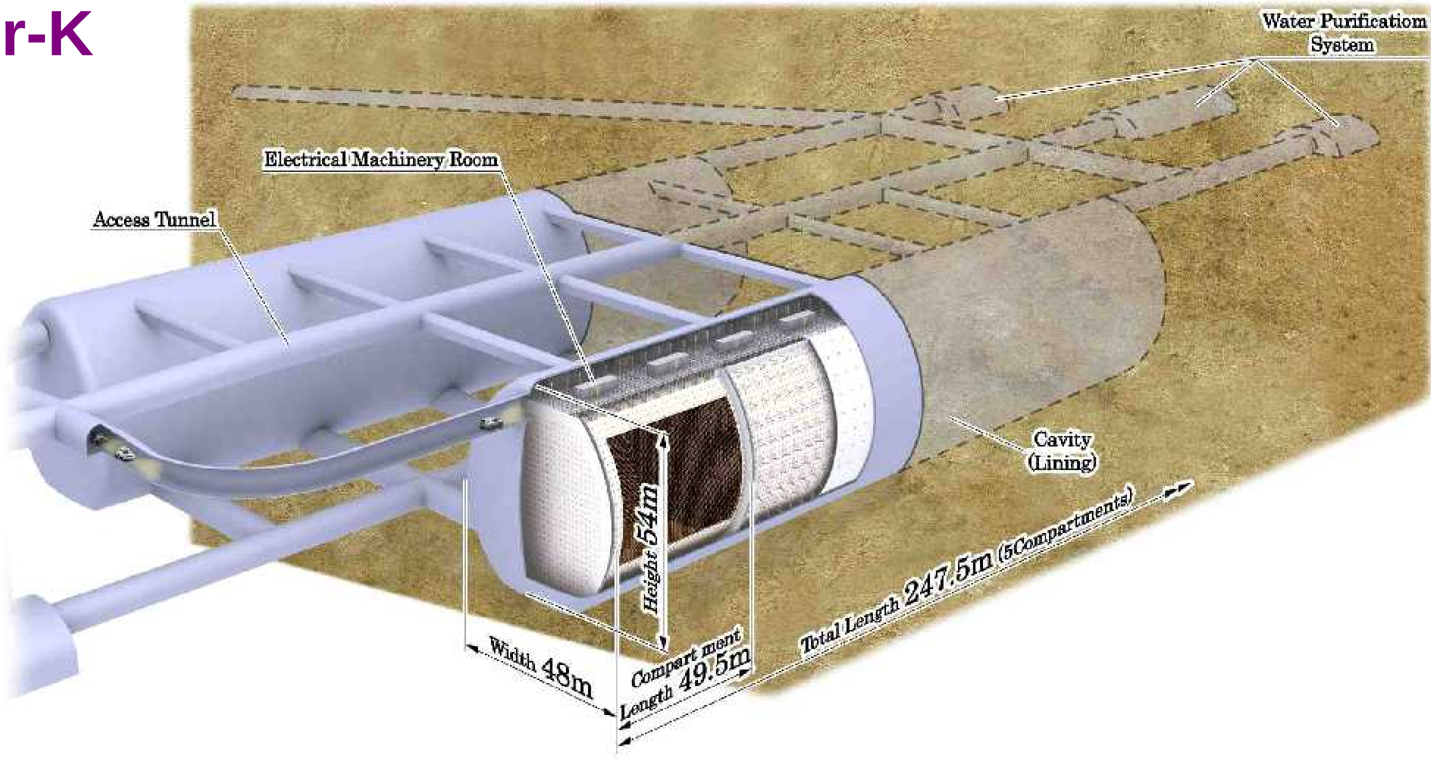
→ No detected Gd loss after >100 complete turnovers.←

Gadolinium loading is part of the executive summary!

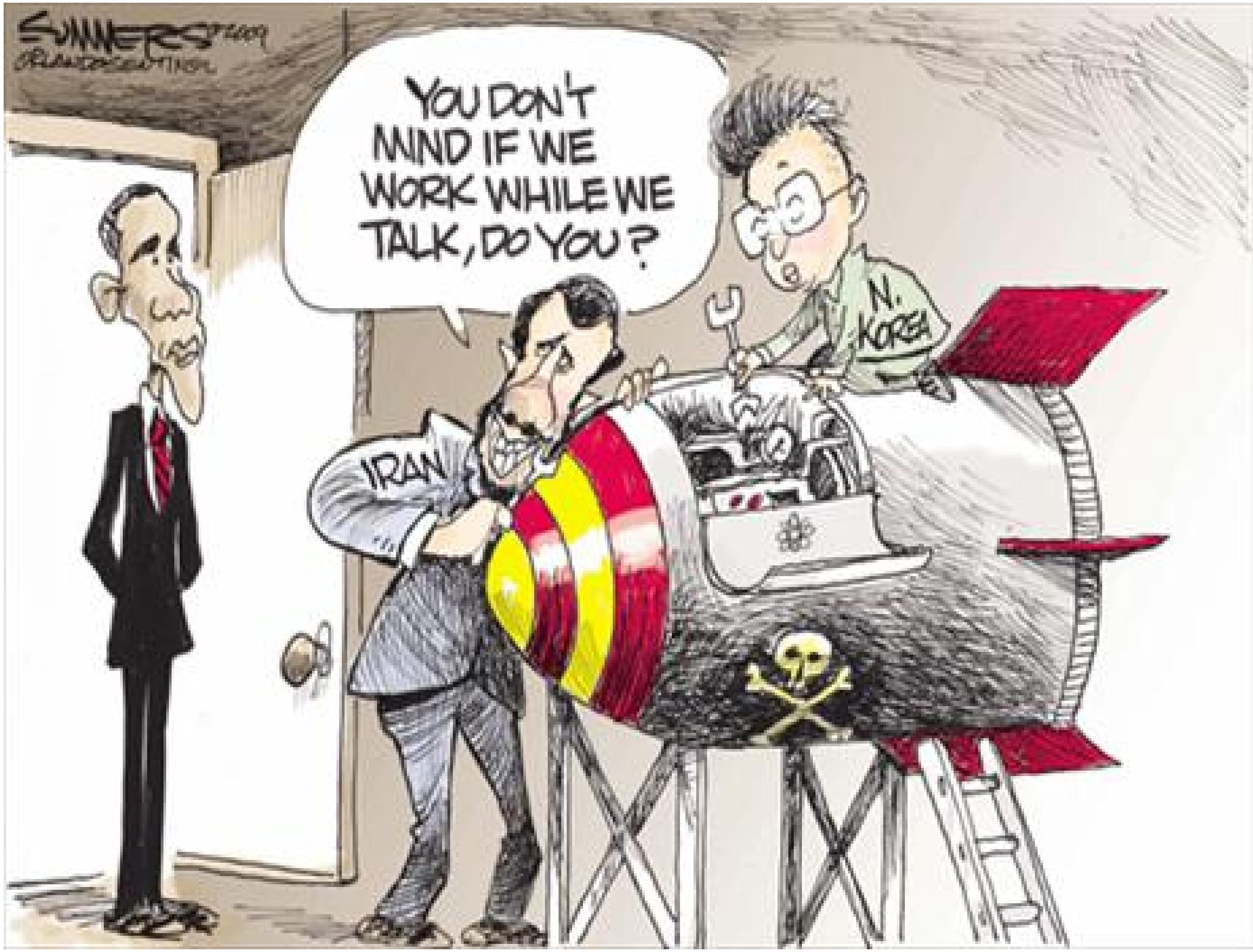
Last year, the official [Hyper-Kamiokande] Letter of Intent appeared on the arXiv:1109.3262

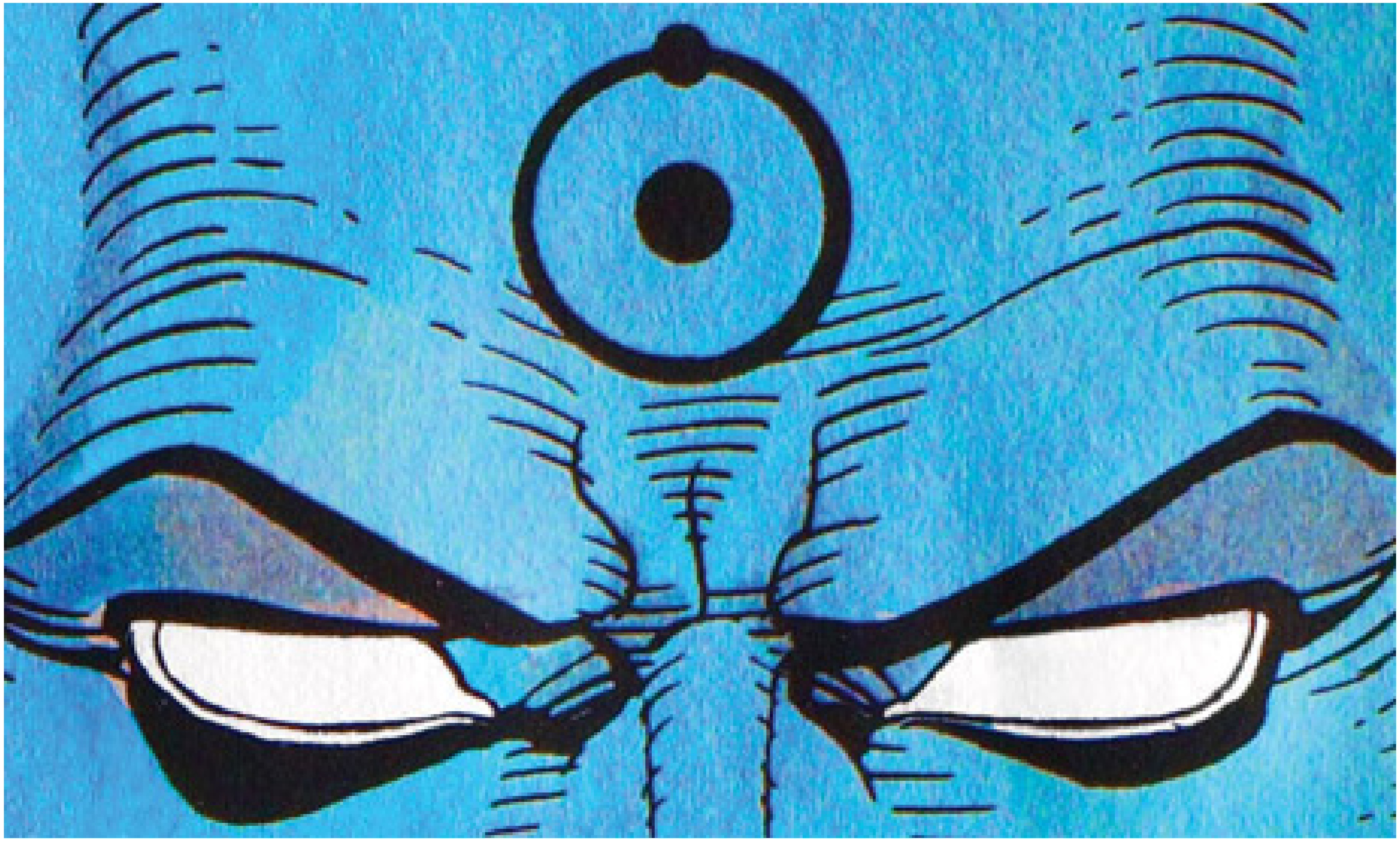
1.0 Mton total water volume
0.56 Mton fiducial volume
(25 X Super-K)

With Gd, Hyper-K should collect SN1987A-like numbers of supernova neutrinos...
every month!



Of course, very large scale anti-neutrino detection just might have another application or two…

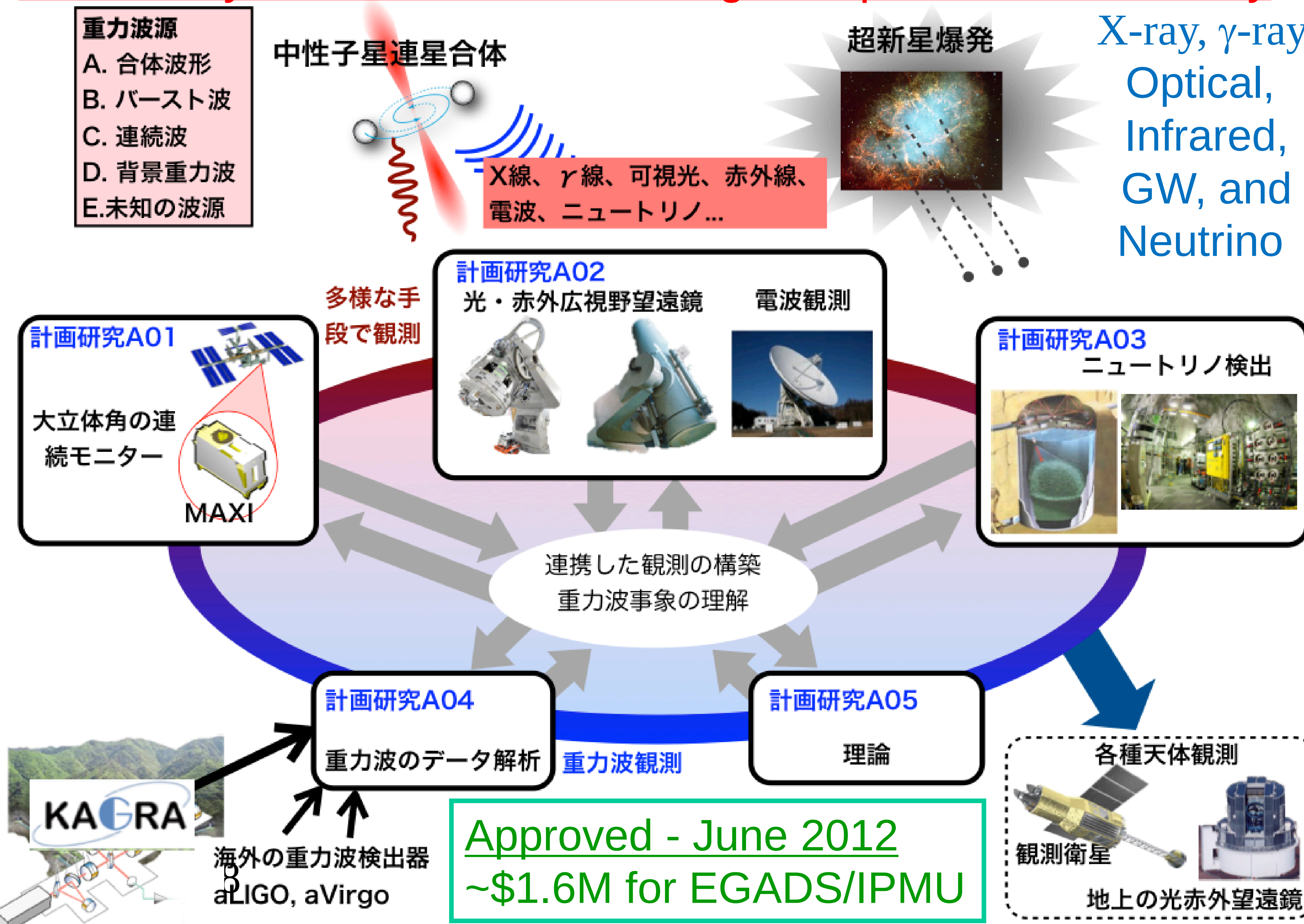




WATCHMAN: WATer CHerenkov Monitor of Anti-Neutrinos

A newly-funded US National Security initiative

Also newly funded: Multi-messenger Supernova Astronomy



X-ray, γ-ray, Optical, Infrared, GW, and Neutrino

Approved - June 2012
~$1.6M for EGADS/IPMU

計画研究 A03：なんとかかんとかの研究

Special features of SN neutrinos and GW's

- Provide image of core collapse itself (identical t=0)
- Only supernova messengers which travel without attenuation to Earth (dust does not affect signal)
- Guaranteed full-galaxy coverage

What is required for maximum SN ν information?

- Sensitivity to nearby explosions (closes gap in Super-Kamiokande's galactic SN ν coverage)
- Deconvolution of neutrino flavors via efficient neutron tagging

By converting an existing R&D facility (EGADS) into the world's most advanced SN ν detector, we could collect

3,690 ν events @ 3,000 light-years

369,000 ν events @ 300 light-years

By 2015 we expect to be ready to detect supernova neutrinos with EGADS from anywhere in our galaxy, and produce immediate alerts to the world.

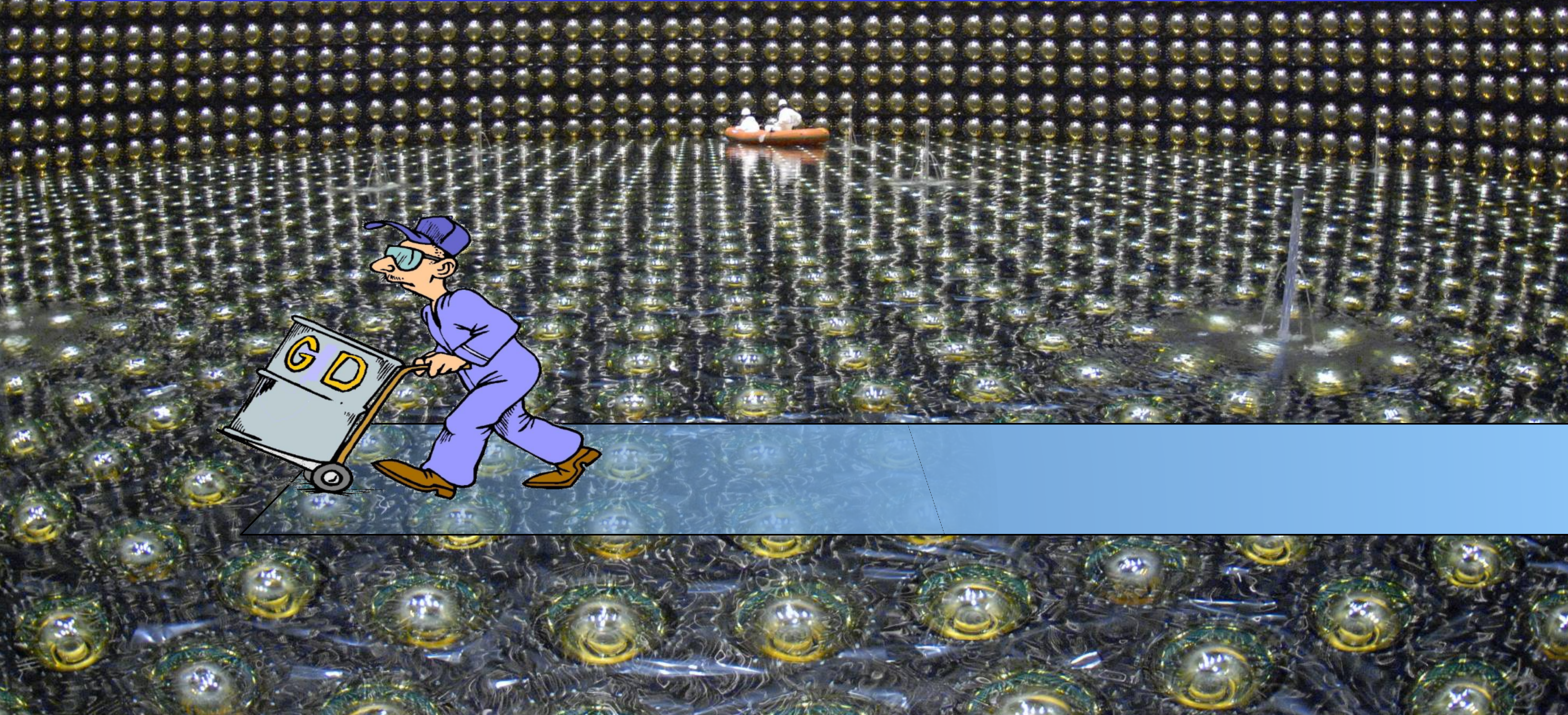
→ **No politics!** ←

By 2016 it is likely we will be adding Gd in Super-K.

In conclusion:

Water Cherenkov detectors have a long, proud history in neutrino physics and proton decay searches.

Now – with EGADS and gadolinium – the <u>next</u> thirty years can be as productive and exciting as the 1st thirty.



Supplementary Slides

At Super-K, a calibration source using $GdCl_3$ has been developed and deployed inside the detector:

Am/Be source

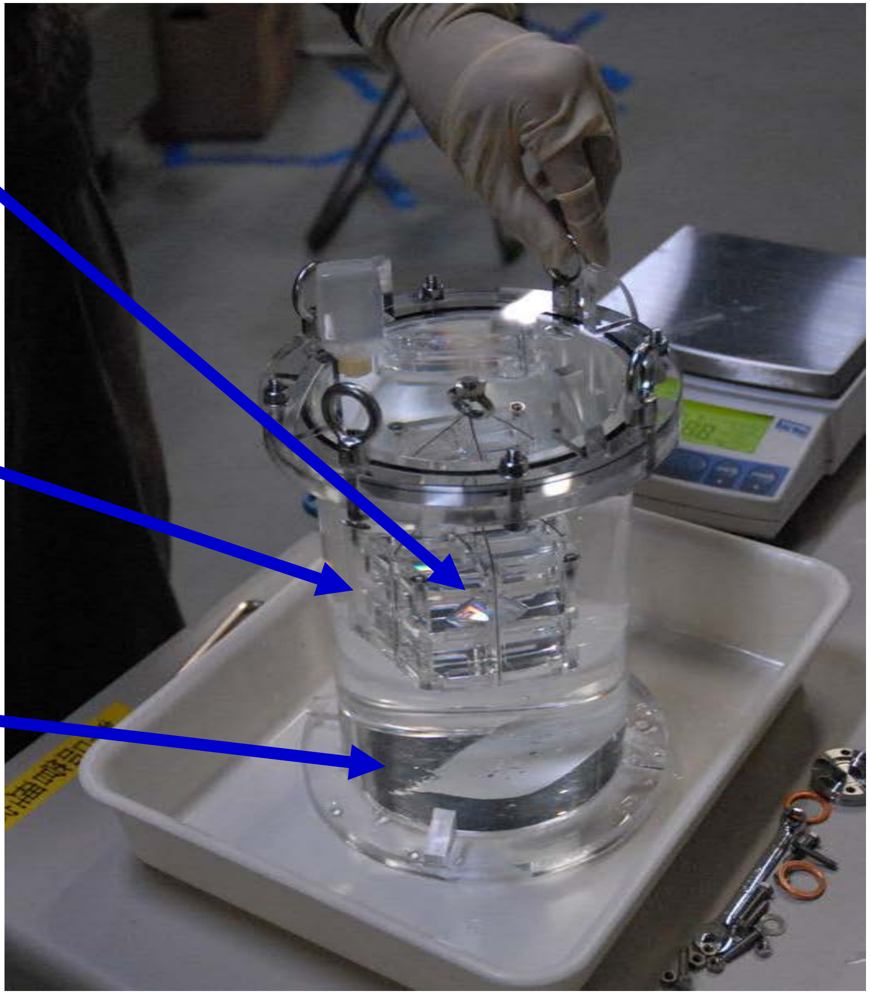
$$\alpha + {}^{9}\mathrm{Be} \longrightarrow {}^{12}\mathrm{C}^{*} + n$$

$$ {}^{12}\mathrm{C}^{*} \longrightarrow {}^{12}\mathrm{C} + \gamma(4.4\ \mathrm{MeV})$$

Inside a BGO crystal array

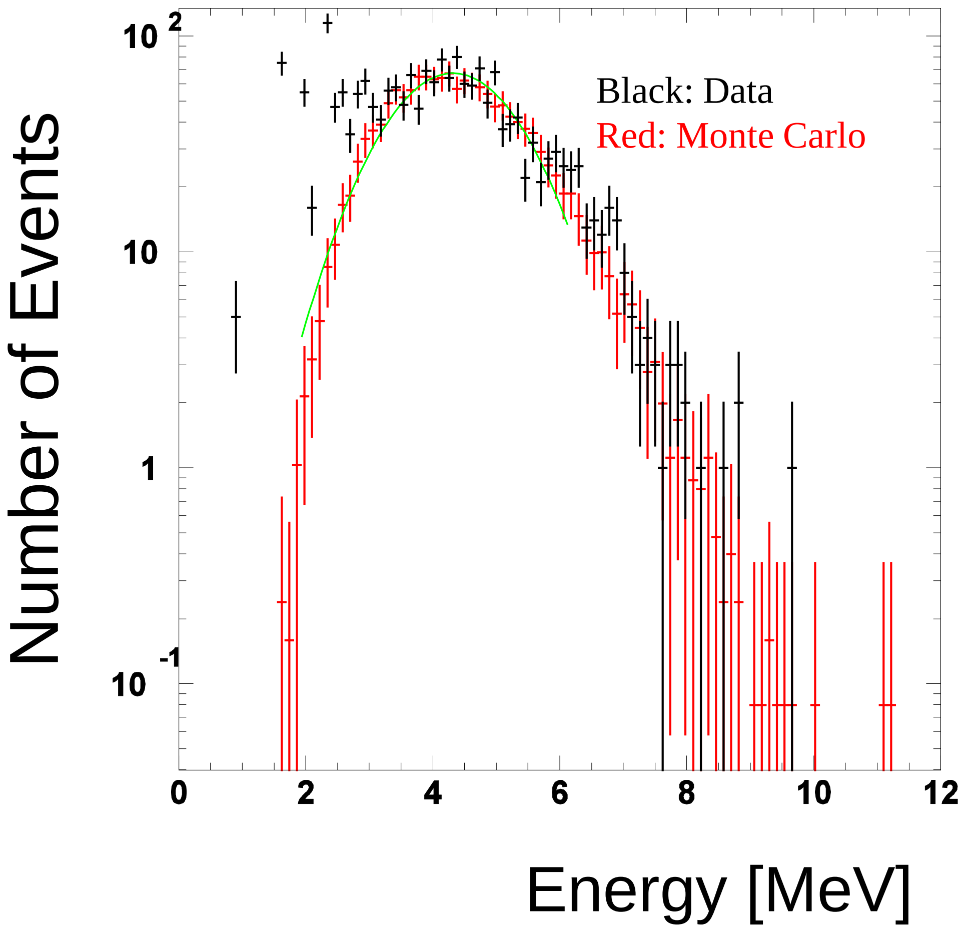
(BGO = $Bi_4Ge_3O_{12}$)

Suspended in 2 liters of 0.2% $GdCl_3$ solution



Data was taken starting in early 2007.

We made the world's first spectrum of $GdCl_3$'s neutron capture gammas producing Cherenkov light:

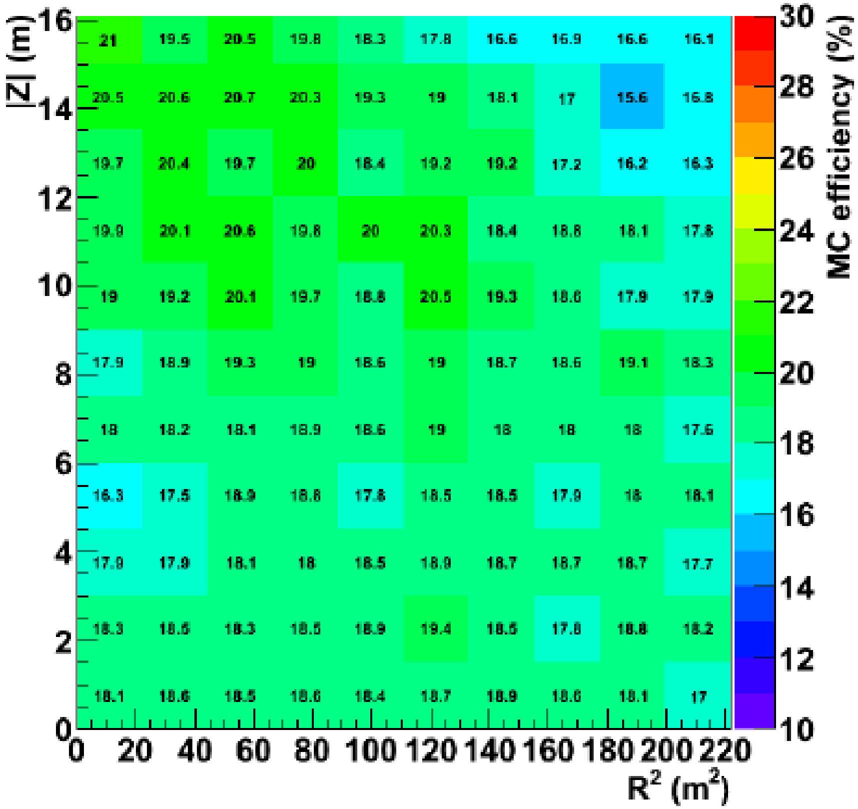


First $GdCl_3$ "in" SK!

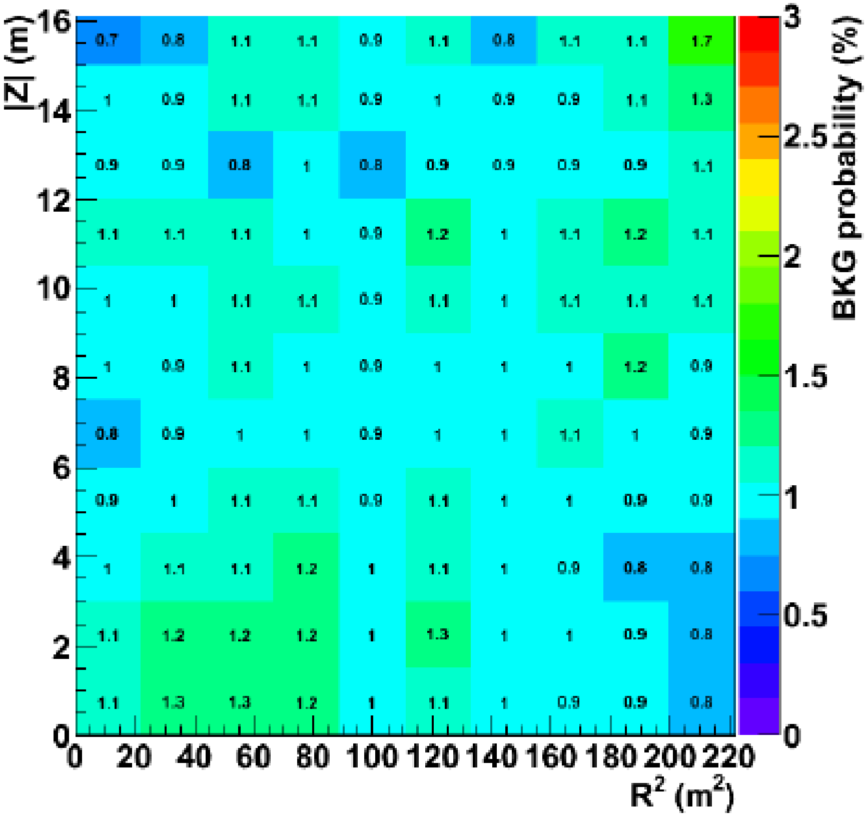
A paper on neutron tagging in Super-K, signed by the entire Collaboration was published:
*Astropart.Phys **31**:320 (2009)*

A study of 2.2 MeV gamma tagging efficiency vs. position in SK

σ= 0.4% in each cell



σ= 0.1% in each cell



- MC efficiency is 18.6%, bkg. probability is 1.0% / 500 us.

For comparable case of Gd in a 20% coverage HK

There is much less background which lives around 4.5 MeV, and the n-capture time window is reduced by a factor of five.

Therefore, cuts can be relaxed → signal efficiency >50%.