Australian Government

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Megavolt Accelerator Systems for Environmental Monitoring.

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http://www.ansto.gov.au/iba

Science. Ingenuity. Sustainability.

Urbanisation of the Globe

- As of 2008, more than 50% of the world's population lives in cities.
- Between 1950 and 2005 the world's population increased from 2.5 billion to 6.6 billion.
- It turned 7 billion in 2012 and will be 9.1 billion in 2050.
- Urbanisation is increasing at a higher rate in developing countries.
- Over 85% of the growth in world population between 2000 and 2024 is expected in urban areas of developing countries.

This urbanisation will have dramatic impacts on anthropogenic and biogenic emissions, alter atmospheric composition and chemistry.

Source: Zhu et al WMO/ IGAC Impacts of Megacities on Air Pollution and Climate, GAW Report No. 205, September 2012.

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Fine Particle PM2.5 Air Pollution Studies:

- Air pollution is driven by urbanisation.
- Everyday ~100,000 people globally move into cities!
- Since 2008, more than 50% of the world's population lived in cities.
- Cities become megacities with populations > 10 million. Currently more than 37 megacities globally. 8 of top 10 in Asia.
- Generating more fine particle pollution, shortens life expectancy. The average life expectancy in an Asian megacity is reduced by 5-6 years by pollution.
- Even annual average PM2.5 levels of 10µg/m^3 have health impacts. Some megacities PM2.5 >50µg/m^3
- Accelerator –based Ion Beam Analysis (IBA) PIXE, PIGE, RBS can characterize and identify sources of fine particle PM2.5 pollution.
- Couple IBA data with recent powerful statistical source fingerprinting and apportionment techniques.
- Combine source fingerprinting with meteorological data to trace long range transport (LRT) of PM2.5.

Manila on a 'clear day'

Manila on a smoggy day

Source: Zhu et al WMO/ IGAC Impacts of Megacities on Air Pollution and Climate, GAW Report No. 205, September 2012.

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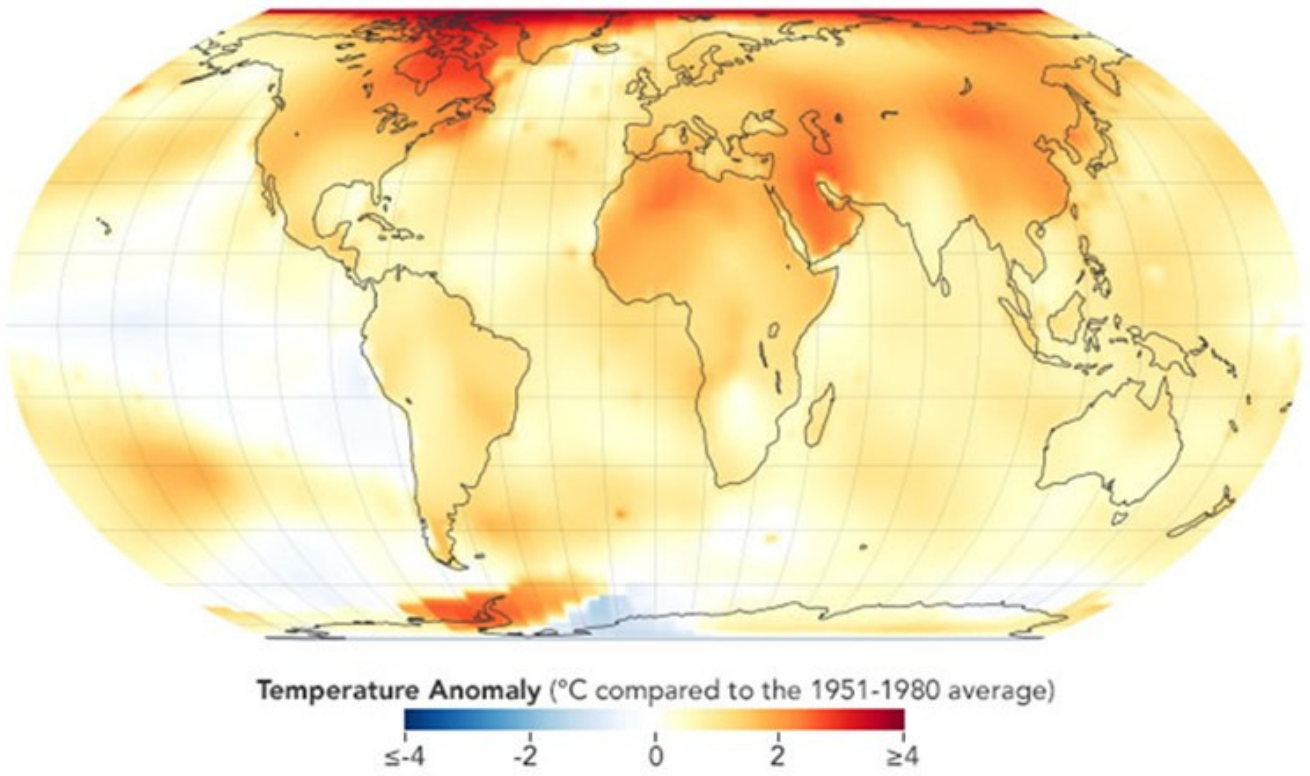
Global Climate Change is Here!



- 2021 was the 6th warmest year since 1880.
- Eight of the top 10 warmest years on our planet occurred in the last decade.
- Global temperatures 0.85°C above the 1951-1980 average.
- 1.1°C increase since Industrial Revolution (late 19th century).
- Green house gases account for ~ +2.5W/m^2 of warming.
- Fine particles (PM2.5) from combustion of fossil fuels scatter radiation back out into space, negative forcing ~ -1.1W/m^2 of cooling.

Without fine particles the global temperature rise would be even higher!!

Source: https://earthobservatory.nasa.gov/images/149321/2021-continued-earths-warming-trend?src=eoa-iotd

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Typical Particle Sizes

Type	Size(μm)
Smog	0.01-2
Oil smokes	0.03-1
Tobacco smoke	0.01-1
Black carbon	0.01-3
Clouds and fog	2-60
Fly ash	1-200
Cement dust	3-100
Plant spores	10-30
Bacteria	0.3-30
Human hair	30-200
Light scattering	0.5-5
Visible to eye	50 upwards
600dpi printing	42

Why Study Fine Particles?

Health implications

PM2.5 travel deep into the lungs, have direct access to the blood stream.

Absorb and scatter visible light

Fine particles are many times more efficient at scattering visible light than coarse particles. Public can see pollution!

Travel large distances

Fine particles stay in the atmosphere for days and weeks travel around the globe. Transported across countries.

Affect climate

Fine particles may have a negative climate forcing effect comparable to the positive forcing of greenhouse gases. Better understanding needed for climate modelling.

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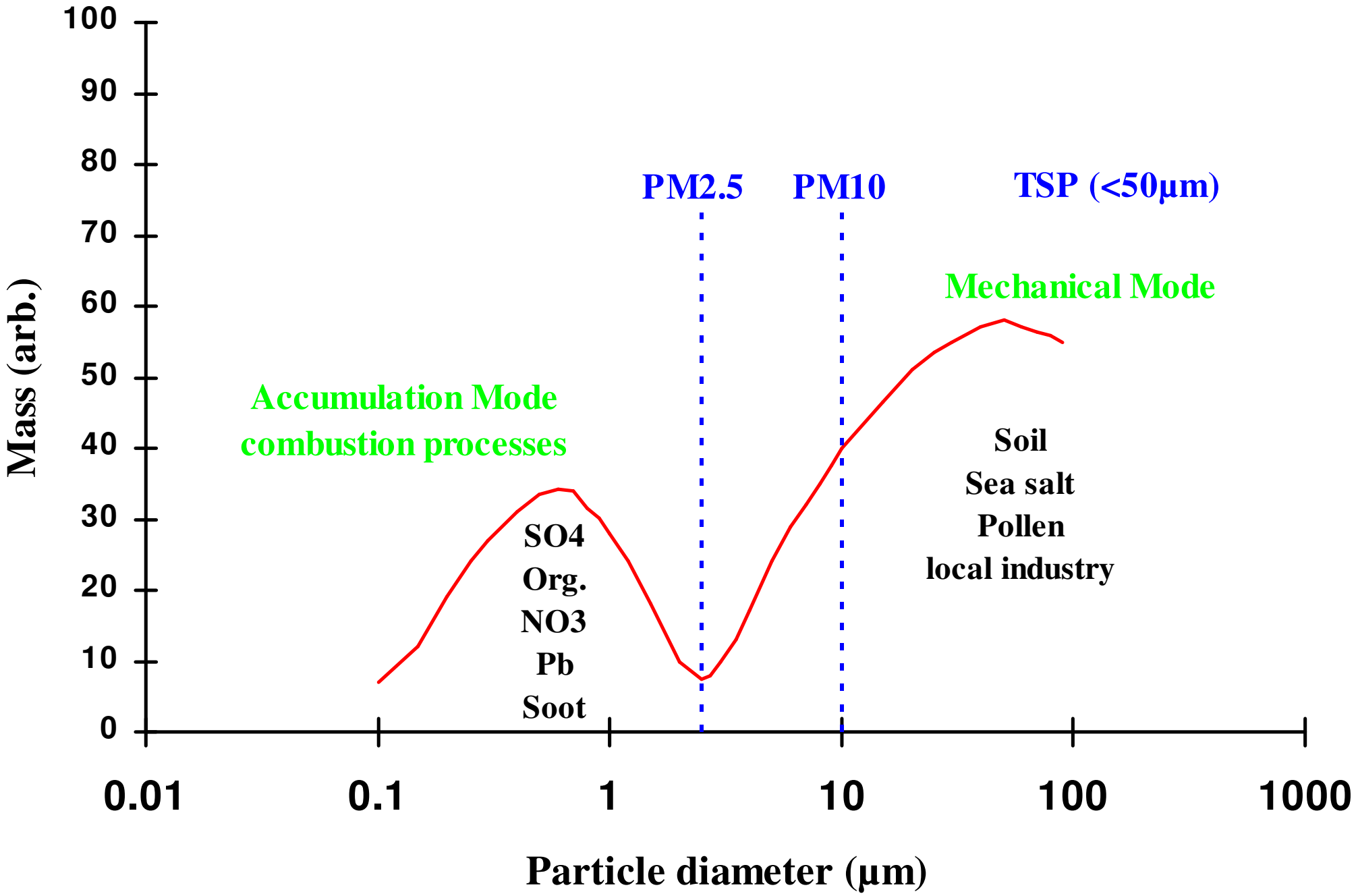
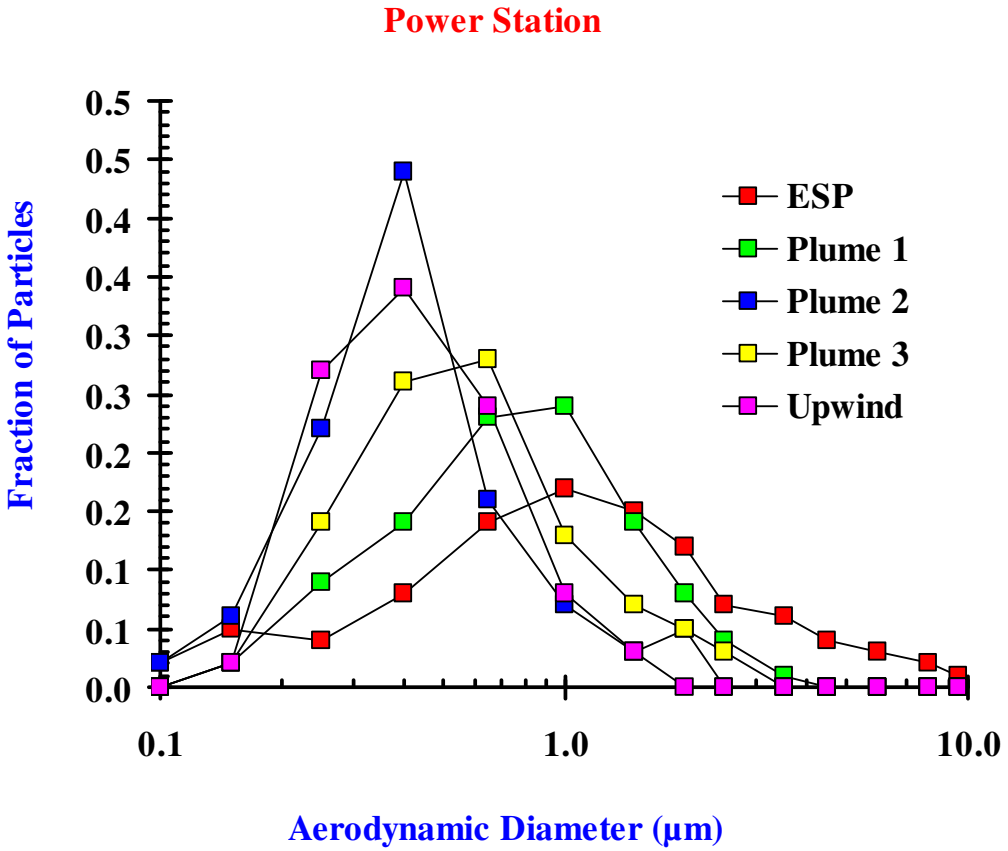
Mass of Particle vs Size

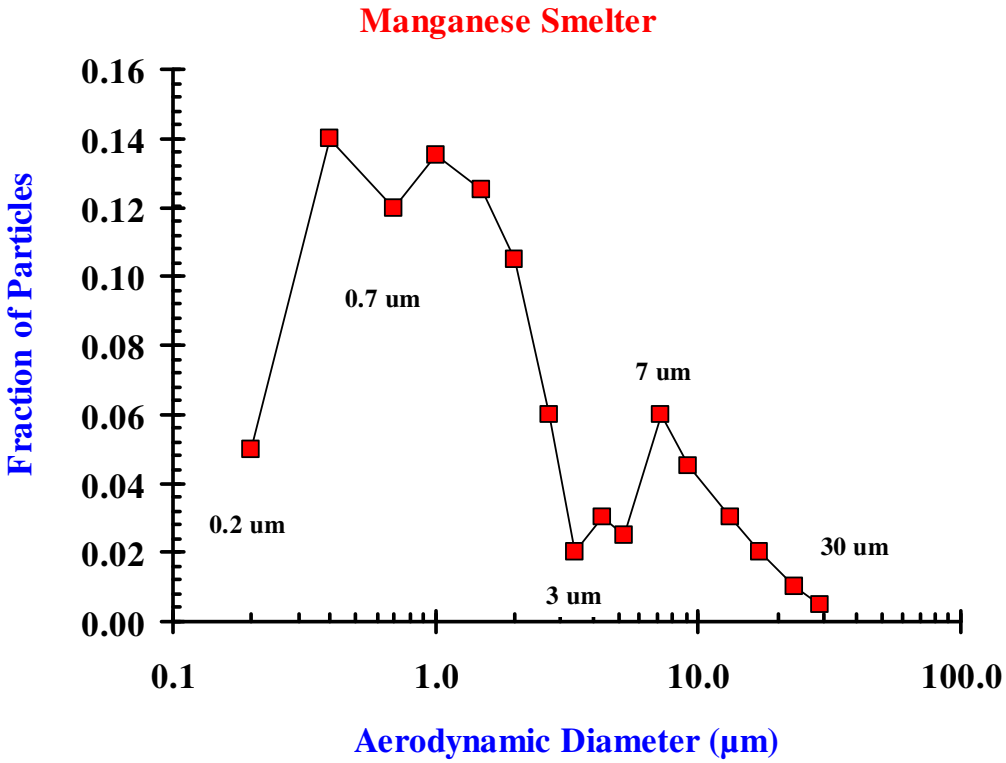


Measured Particle Sizes

Combustion



Combustion + mechanical



Fine and coarse components

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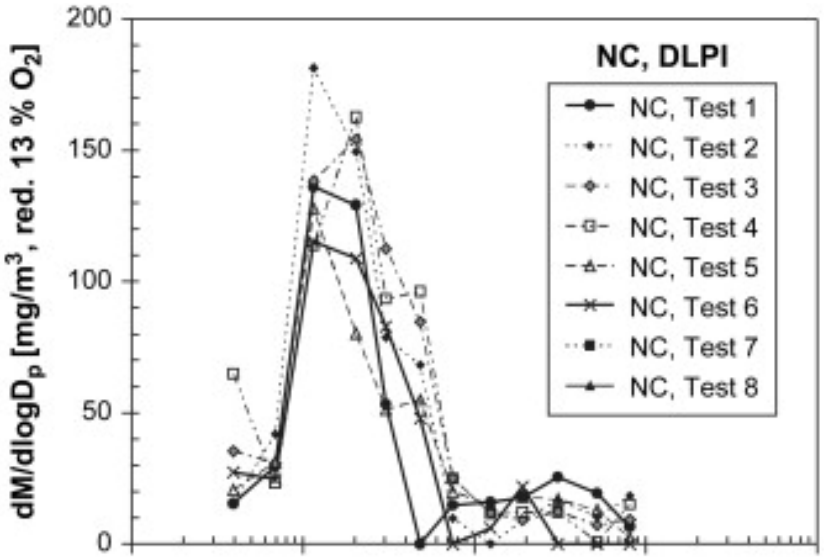
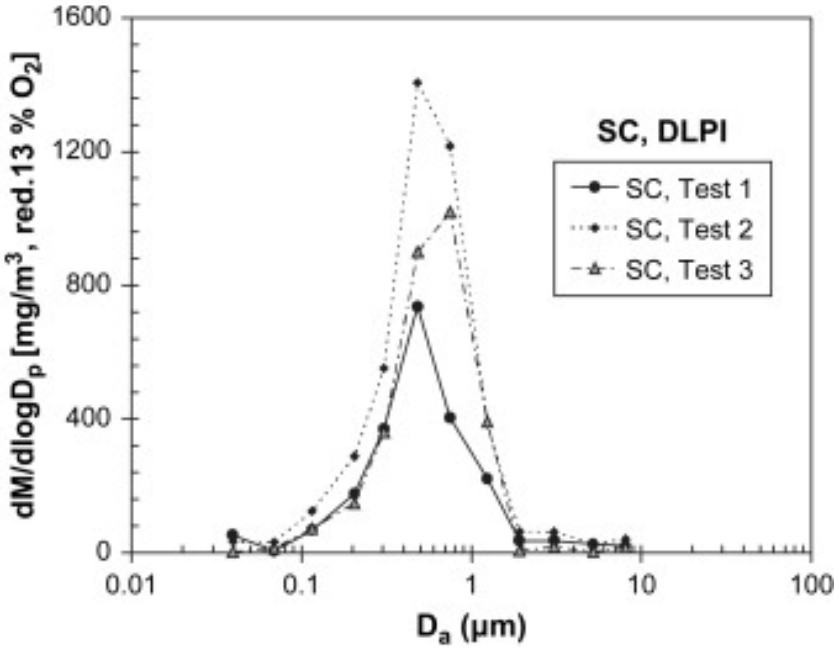
Fine Particles from Wood Combustion Heaters



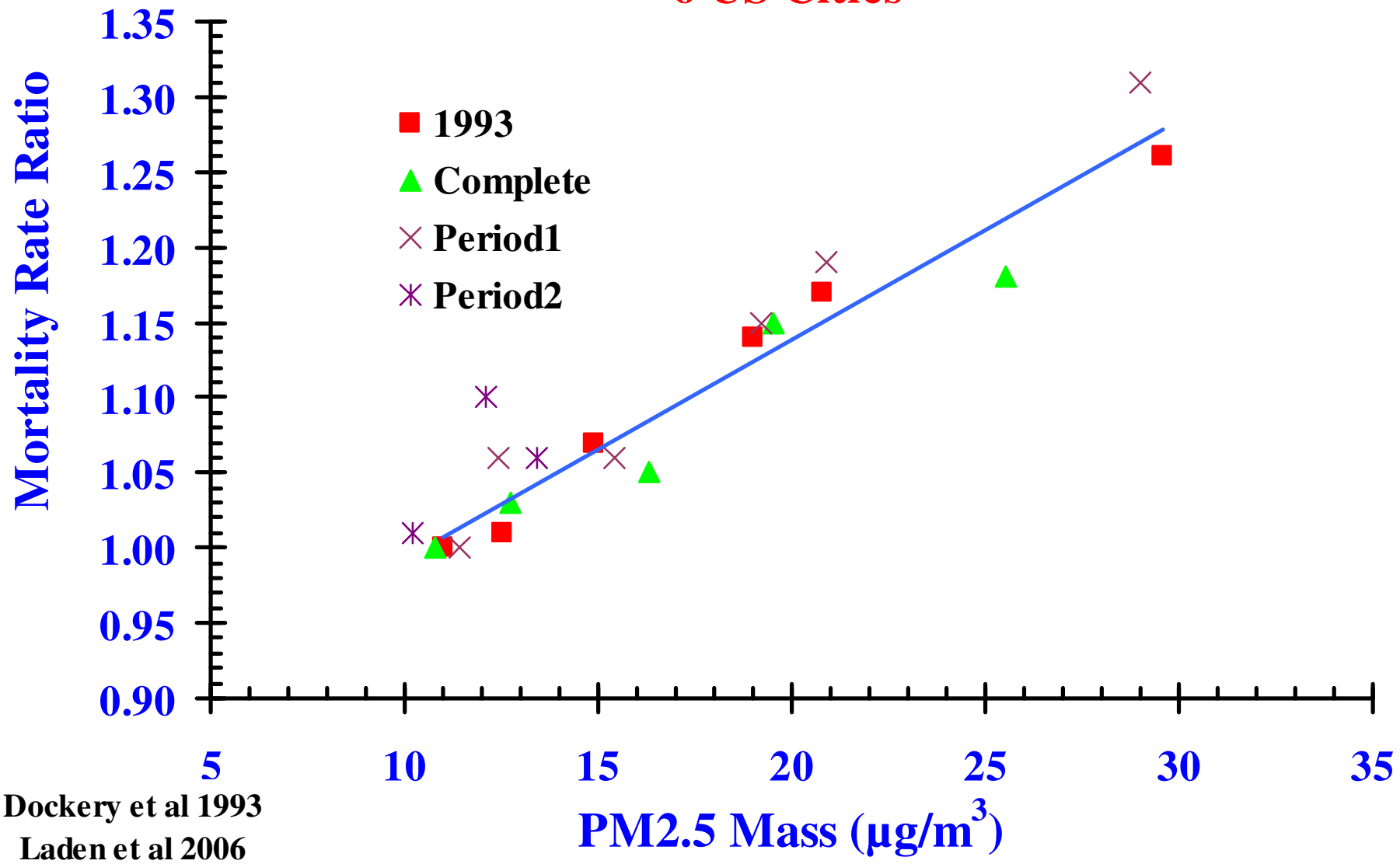
Normal wood combustion



Smoldering wood

After Tissari et al Atmospheric Environment 42 (2008) 7862–7873

Relative Mortality Rate Ratios
6 US Cities



Dockery et al 1993
Laden et al 2006

Appears to be no lower threshold for PM2.5 particles
What component of the mass is producing this death rate?

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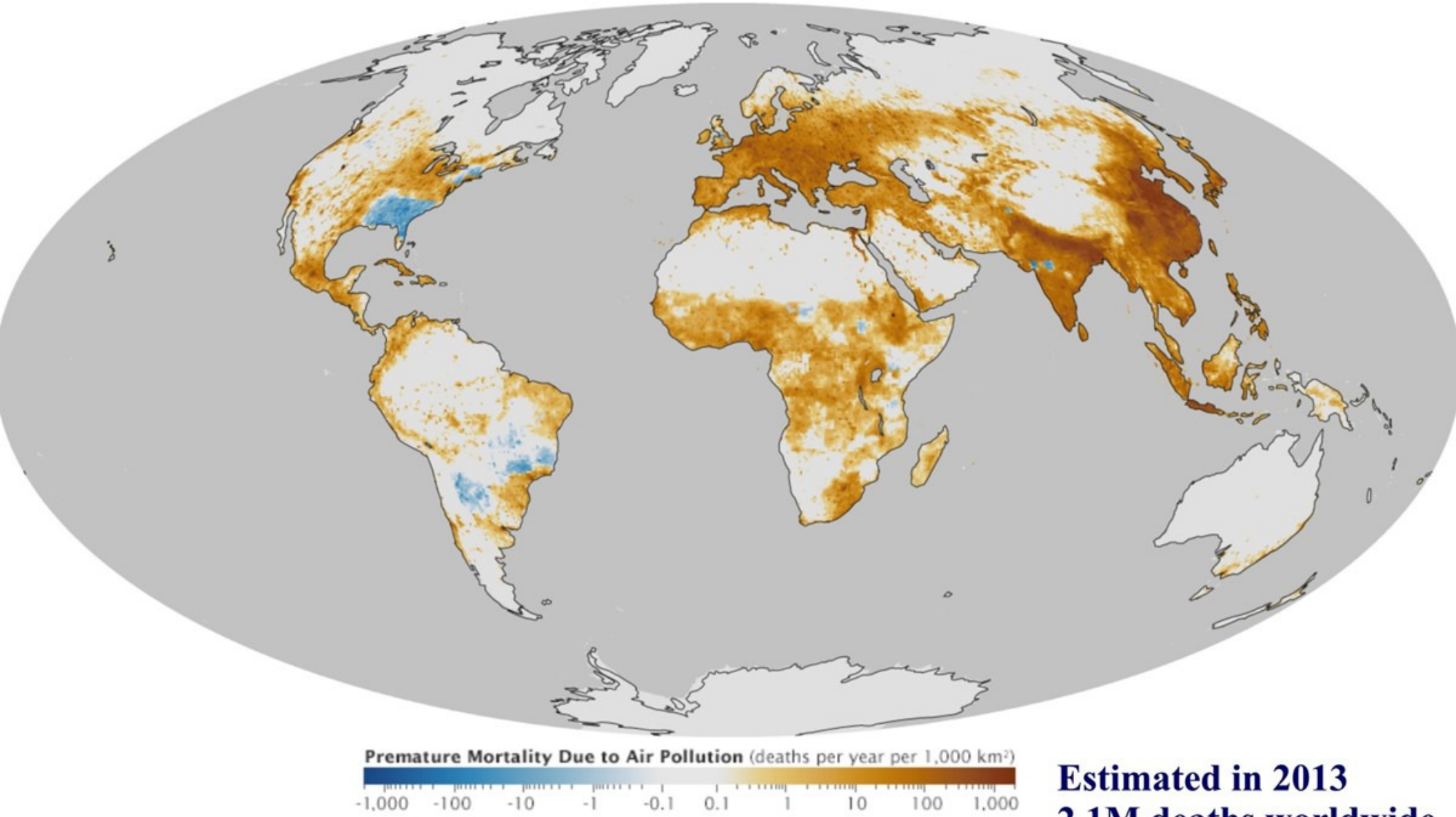
Premature Death Estimates Worldwide from PM2.5 Fine Particles



Source: NASA Earth Observatory site Jan 2013

Estimated in 2013
2.1M deaths worldwide from PM2.5

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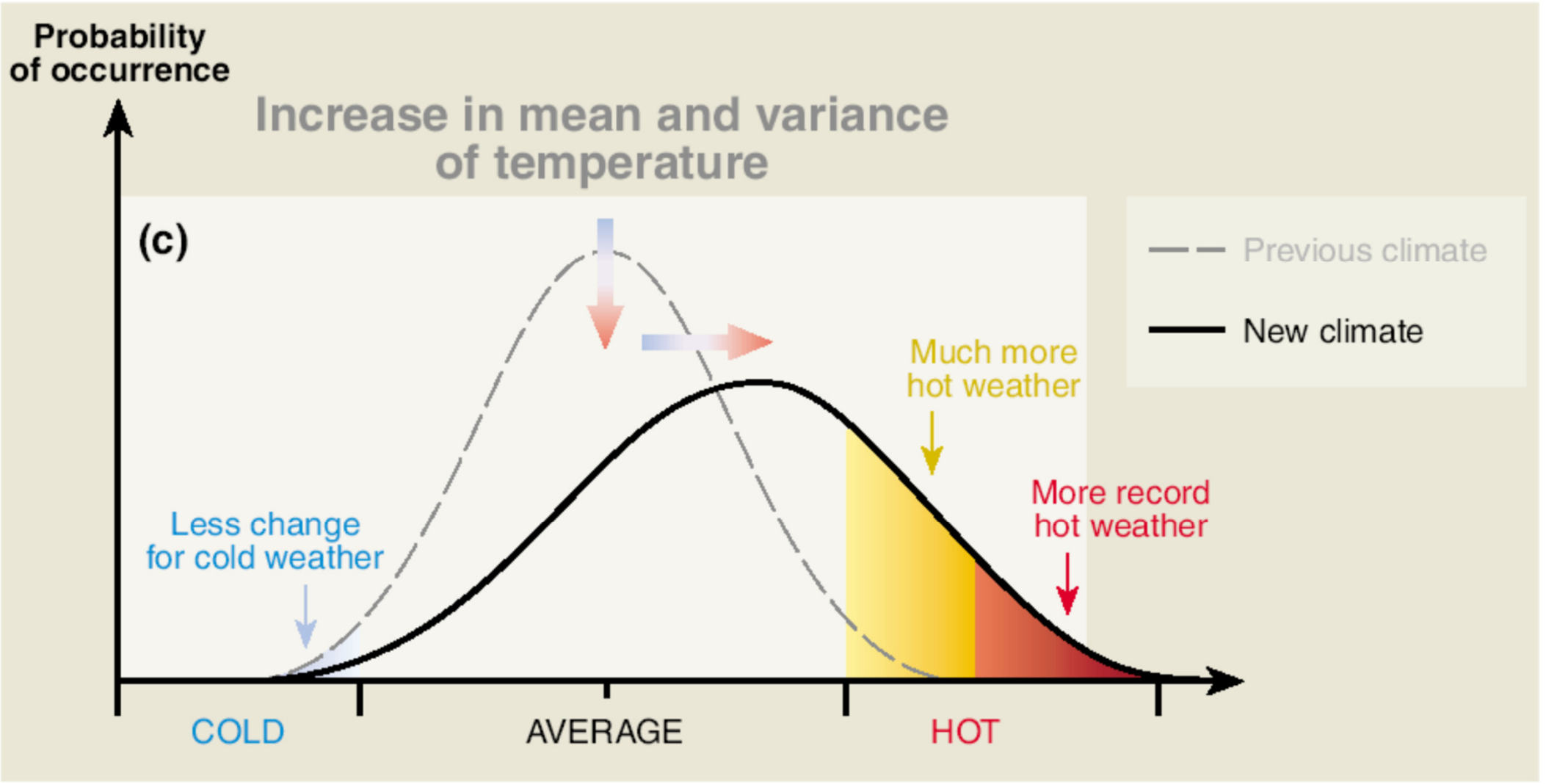

Increased Temps means more hot/ record weather

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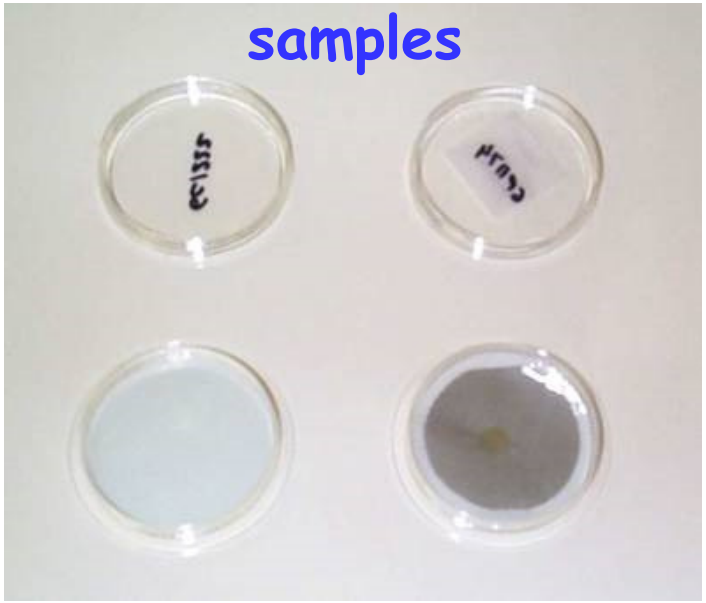
Fine Particle Pollution-what is it we are trying to measure?

- Average person inhales 30m^3 a day if not running marathons.
- For low to average levels of fine particle pollution at 10µg/m^3 this implies samples of only 300µg collected on filters.
- What should these filters be composed of so as not to interfere with this elemental analysis?
- How thick should these filters be compared with the deposited material?
- What techniques will measure a range of elements from hydrogen to lead on a filter in a few minutes with ng/m^3 sensitivity?
- If source apportionment techniques (PMF) split this 300µg into 6-10 sources, some sources may only have mass of 30µg!

PM2.5 & PM10 samples



IBA and XRF are standard analysis methods that can meet most of these requirements.

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ASP-Aerosol Sampling Program at ANSTO

PM2.5 Cyclone unit with its microprocessor at Lucas Heights

Exposed stretched Teflon filter, specifically design for IBA analysis

Filters exposed from midnight to midnight each Sunday and Wednesday. Ideal for IBA analysis, typically 300µg of sample on a 250µg/cm^2 thick filter. Can analysis for 30 elements at ng/m^3 levels, non destructively, in a few minutes of running.

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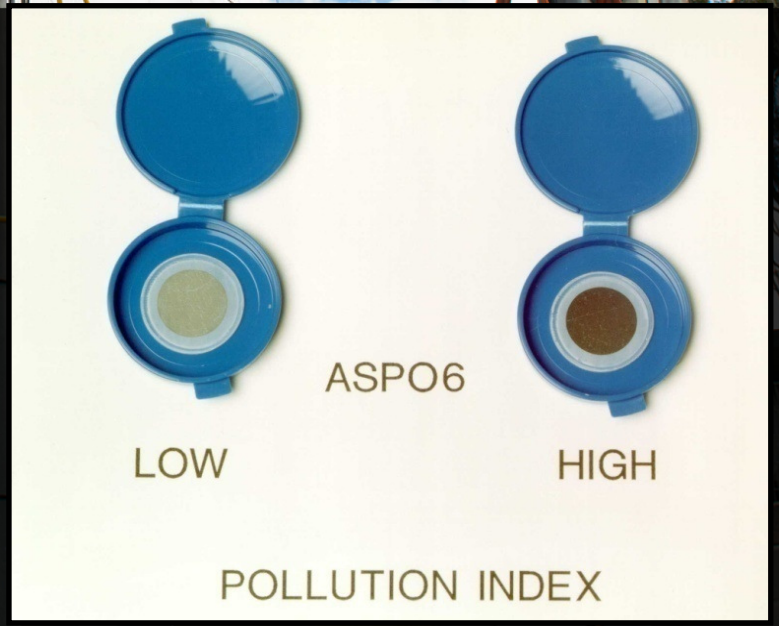
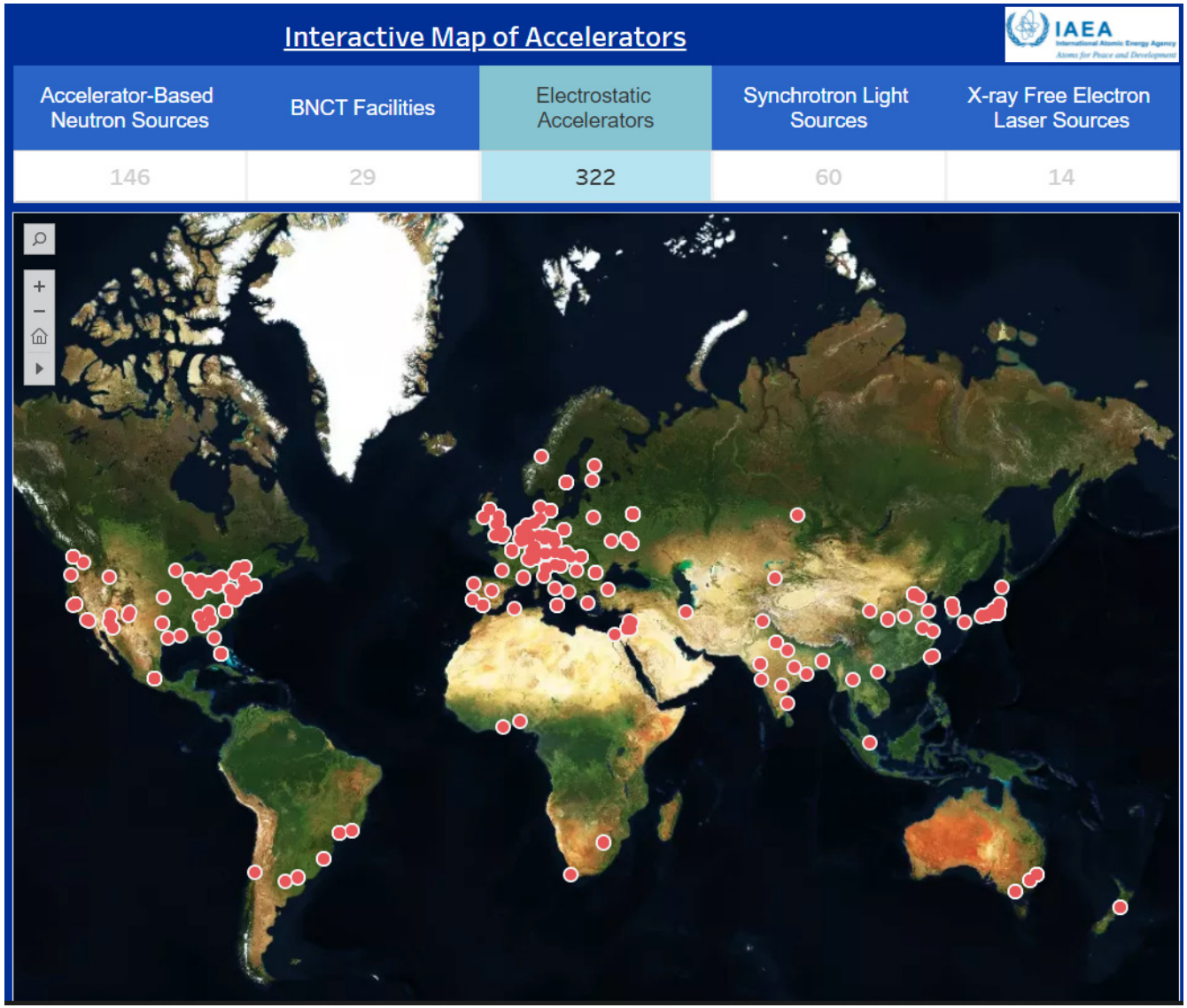
Fine Particle Aerosol Sampling Program - ASP

Source fingerprinting

Soil	Al	Si	Ca	Ti	Fe
Sea spray	Na	Mg	S	Cl	Br
Coal burning	H	C	S	As	Se



Accelerators Globally



http://nucleus.iaea.org/sites/accelerators/Pages/default.aspx

The full range of accelerator facilities and capabilities at ANSTO puts us in the top five megavolt ion facilities globally.

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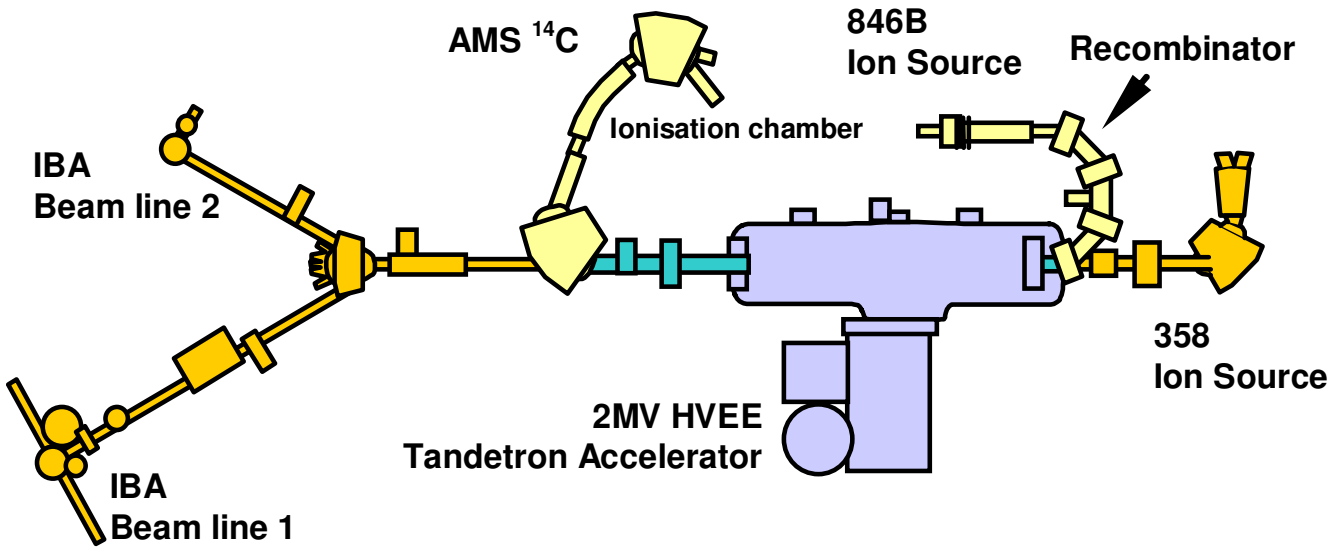
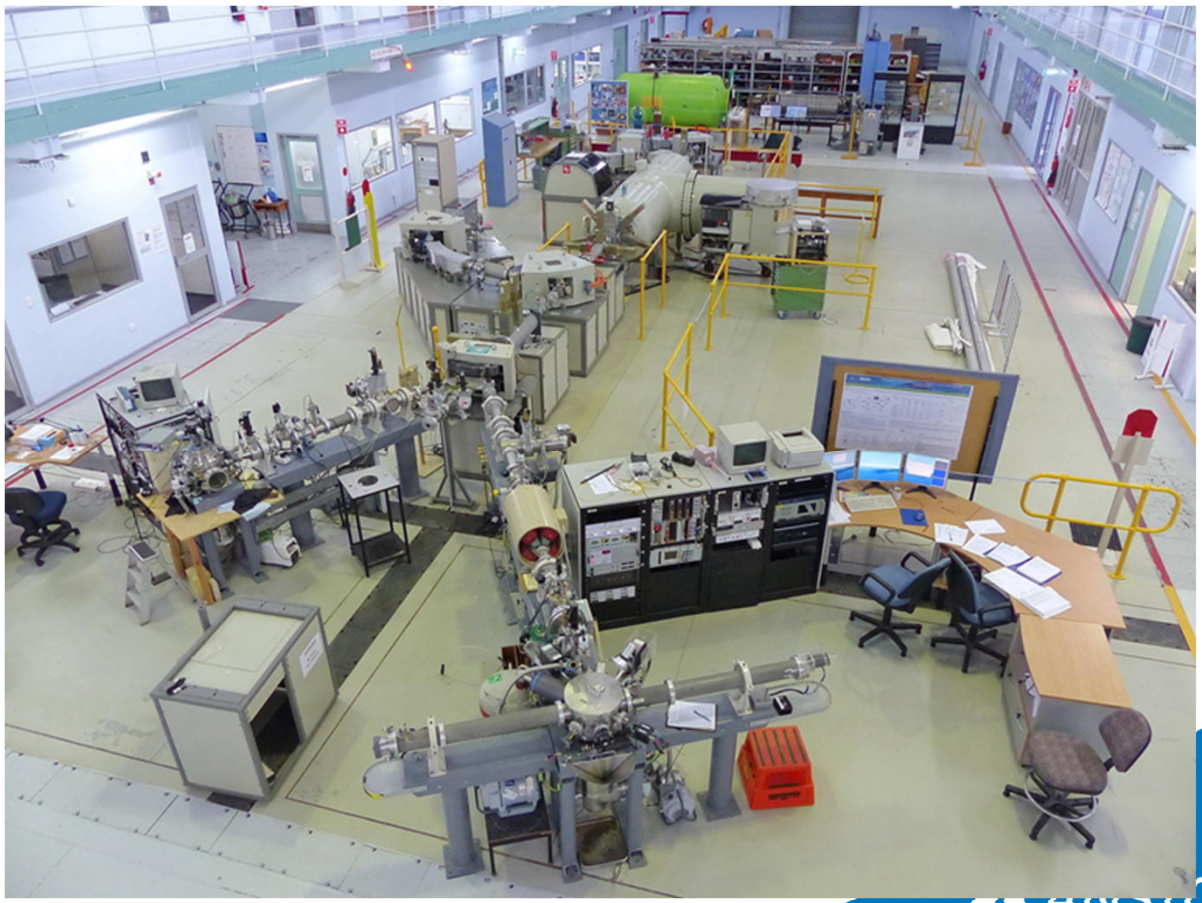
STAR Accelerator (2MV)



- 2MV HVE Tandem
- 2 Duoplasmatron ion sources for H, He
- 1 Sputter ion source for heavy ions C, ….
- 3 High energy beamlines; radiocarbon, materials characterisation, ion beam techniques.



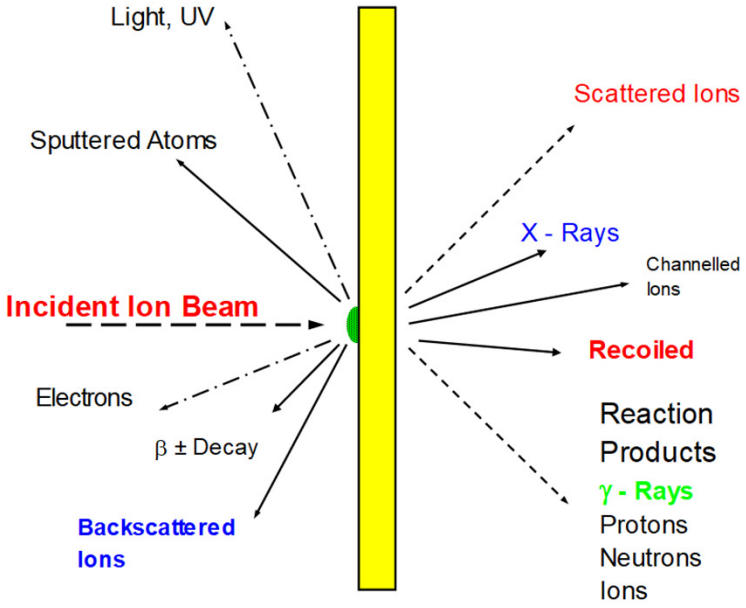
Fine Particle PM2.5 IBA Characterisation

Typical fine mass filters for 24 hour collection weighs ~300µg.

Filters analysed using nA beams of MeV protons and IBA techniques give over 30 different elemental and chemical species.

Analysis does not destroy the sample.

Exposed stretched Teflon filter, optimised for IBA analysis.



ANSTO 2MV STAR accelerator

PIXE – Interactions with electrons, characteristic keV x-rays from Al to U
PIGE – Interactions with nucleus, gamma rays for light elements (Mg, Al, F, Na..)
RBS – Rutherford Backscattering for C, N, O ..
PESA – Particle Elastic Scattering analysis for total H content.

These 4 techniques can be run simultaneously in a few minutes and provide data from H to U with sensitivities of (µg/g) on sample sizes as small as pg.

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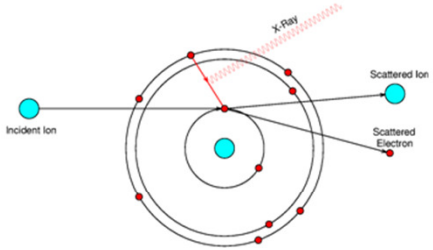
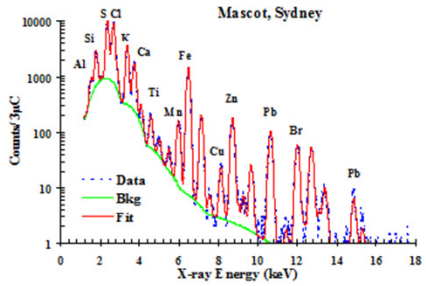
IBA Techniques

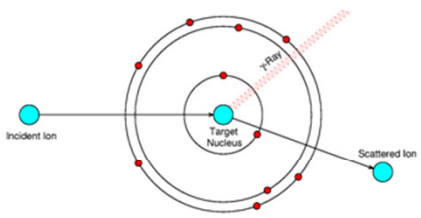
PIXE

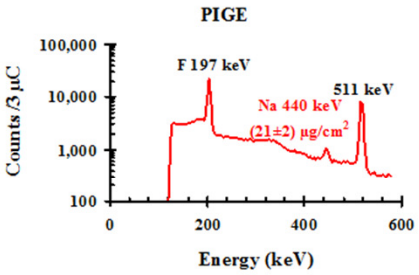




Al to Pb (X-rays)

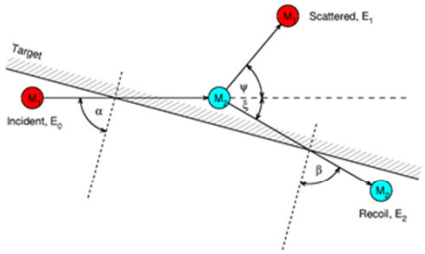
PIGE

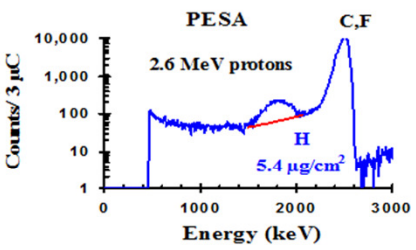




F, Na (γ-rays)

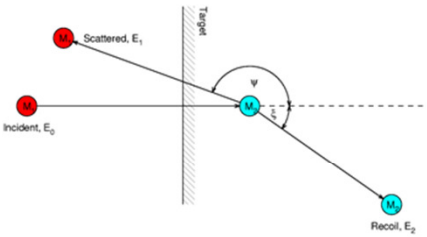
PESA

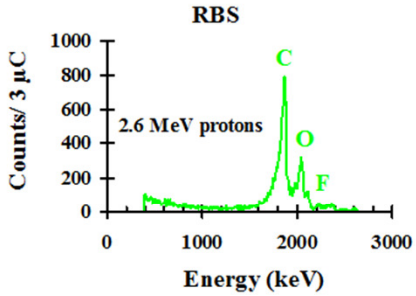




H (forward scatter)

RBS





C, N, O (back scatter)

⇩**multi-elemental (23 elements)**

H, C, N, O, Na, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Br, & Pb

Techniques can be run simultaneously

- **Are non destructive**
- **High sensitivity – ppm levels in a few minutes**
- **Small samples – nanograms (10^{-9}g)**

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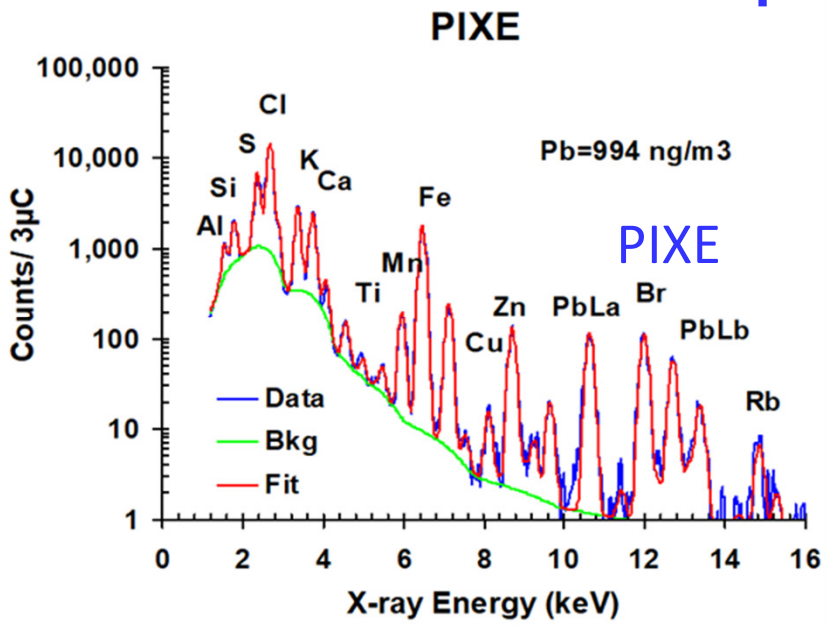
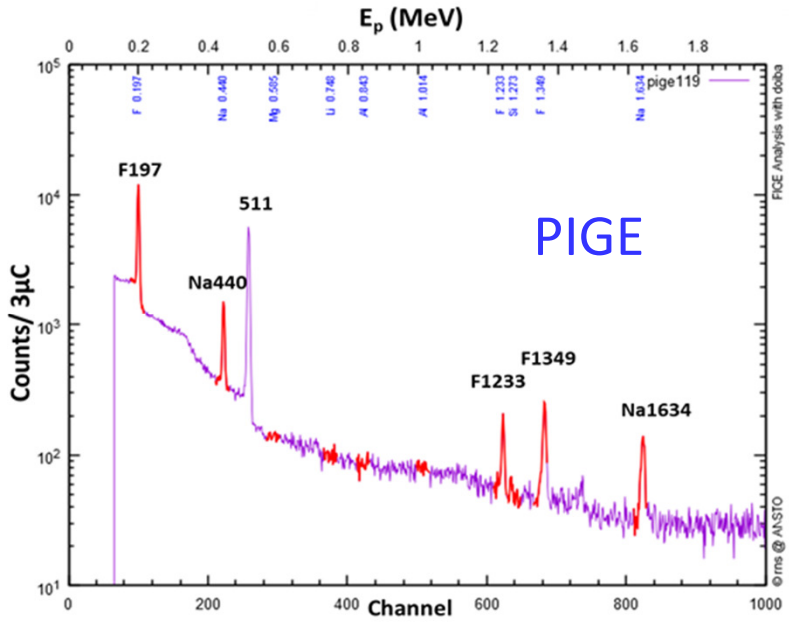
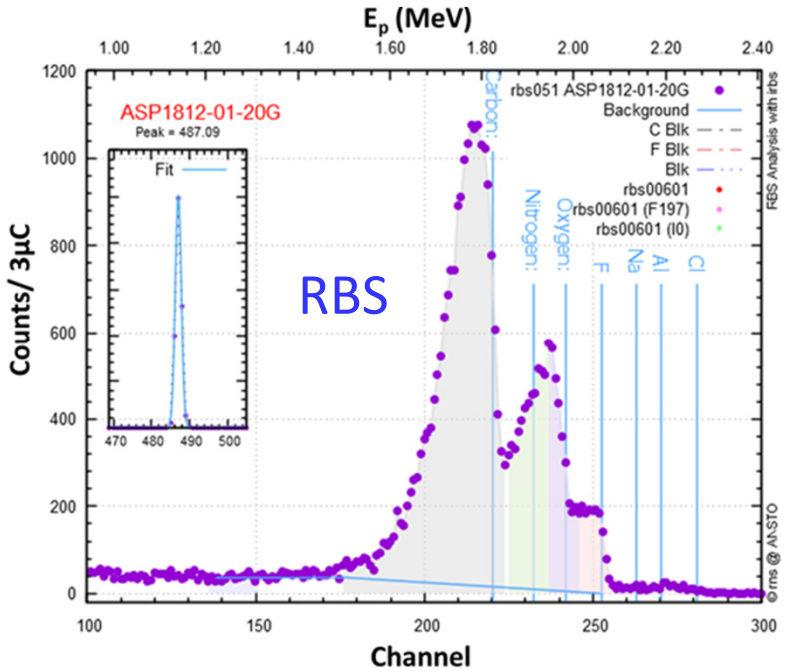
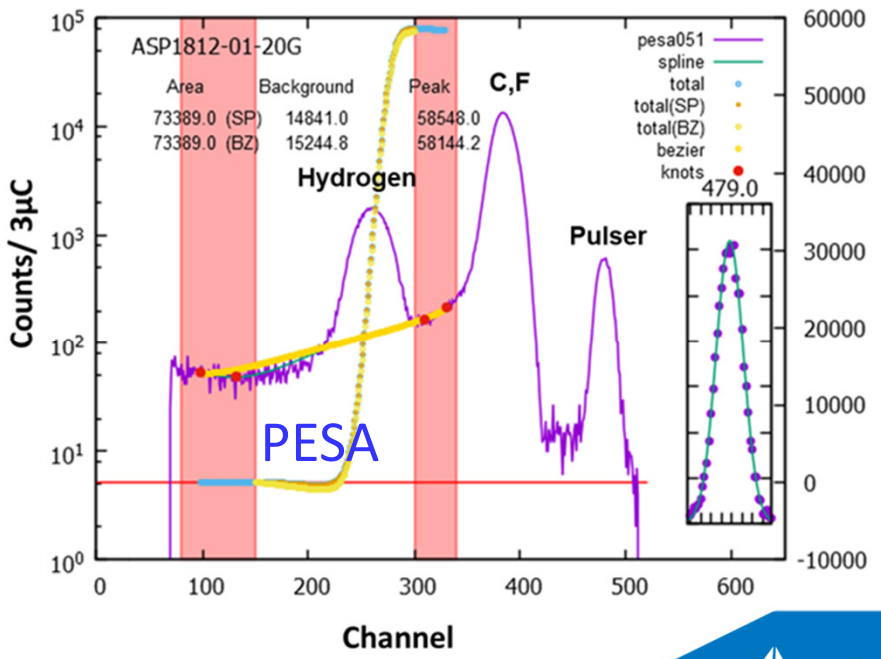
IBA Spectra – iBAT Analysis Code

2.5 MeV protons, 10nA for 5mins









Measure >30 elements from H to U, run 100 samples a day, non-destructively

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Modern trends combine:

- IBA composition
- Source fingerprinting and apportionment
- Back trajectory wind speed and direction.

Positive Matrix Factorisation (PMF)

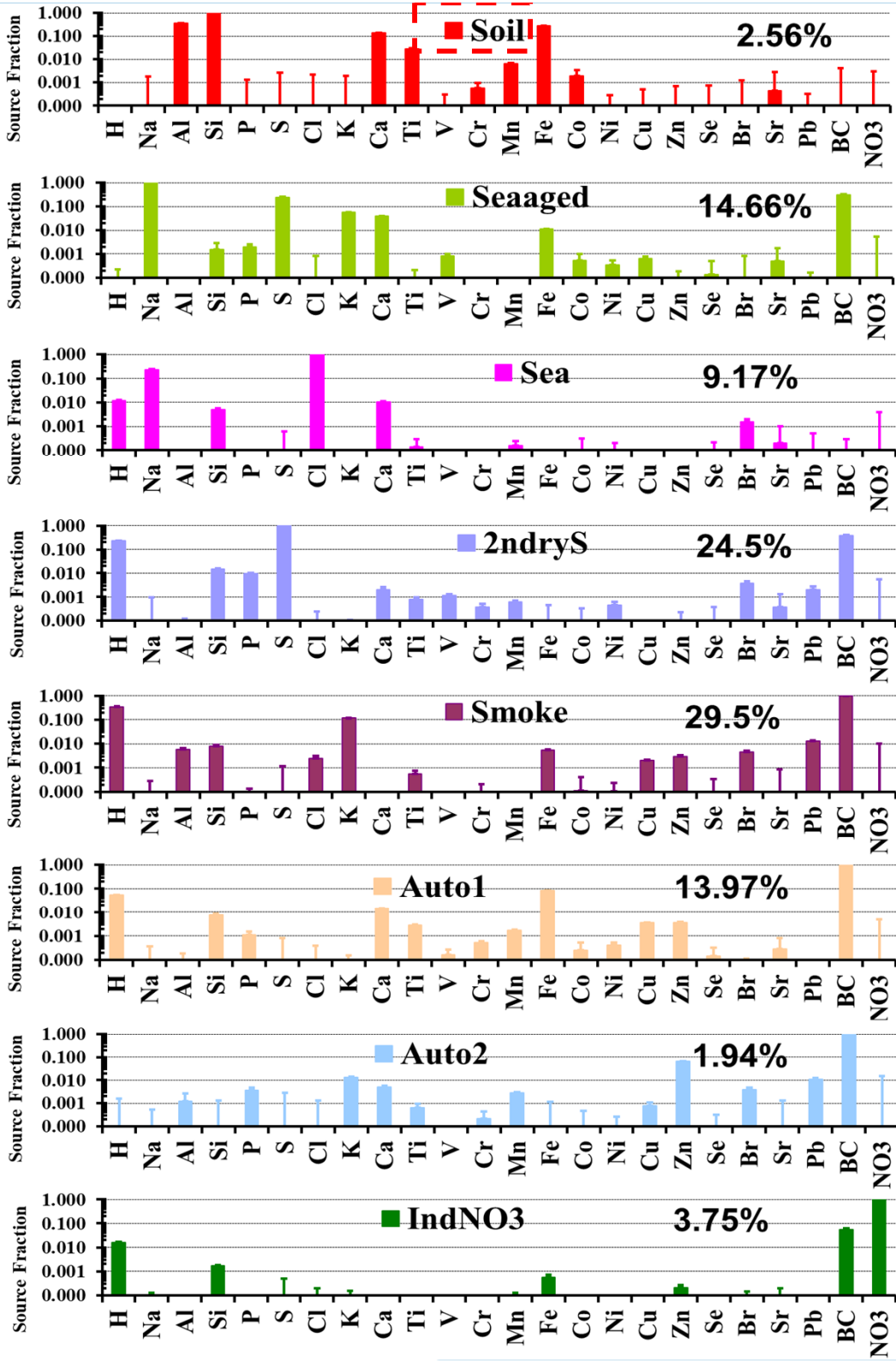
$$M_{i,j} = \sum_{k=1}^{p} F_{k,j} * G_{i,k} + E_{i,j}$$

M_{ij} is a daily mass matrix, F_{kj} are p fingerprints using the j measured elements and G_{ik} is the daily contributions matrix for each of the p fingerprints. E_{ij} is error term to be minimized.

Opposite is the *p=8* fingerprint fit to 2,302 sampling days between January 1998 and February 2022. These 8 fingerprints divide the daily PM2.5 mass (Av.=8.1±5.2 µg/m^3) into 8 separate and distinct sources.

The *Soil* fingerprint was driven by Al, Si, Ca, Ti and Fe. The (Al/Si) ratio was typical of aluminosilicates.
The *Sea* fingerprint was driven by Na and Cl in the correct ratio and traces of Si, Ca and Br.
The *Smoke* fingerprint was driven by H reflecting the organic component, K indicative of biomass burning and BC for the soot content. Other traces such as Cl, Zn, and Br were typical of biomass burning.

http://www.helsinki.fi/~paatero/PMF/pmf2.zip



HYSPLIT Hourly Back Trajectories for Soil at Liverpool for 2001-22

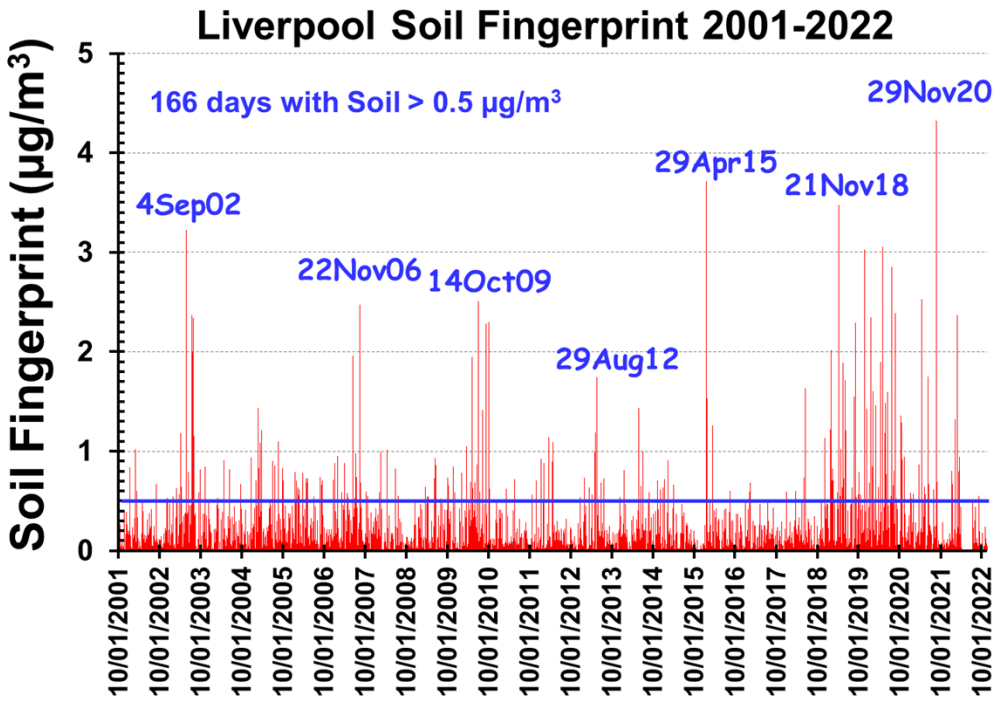
Desert dust storm in Birdsville, QLD, Australia.
27 January 2006.

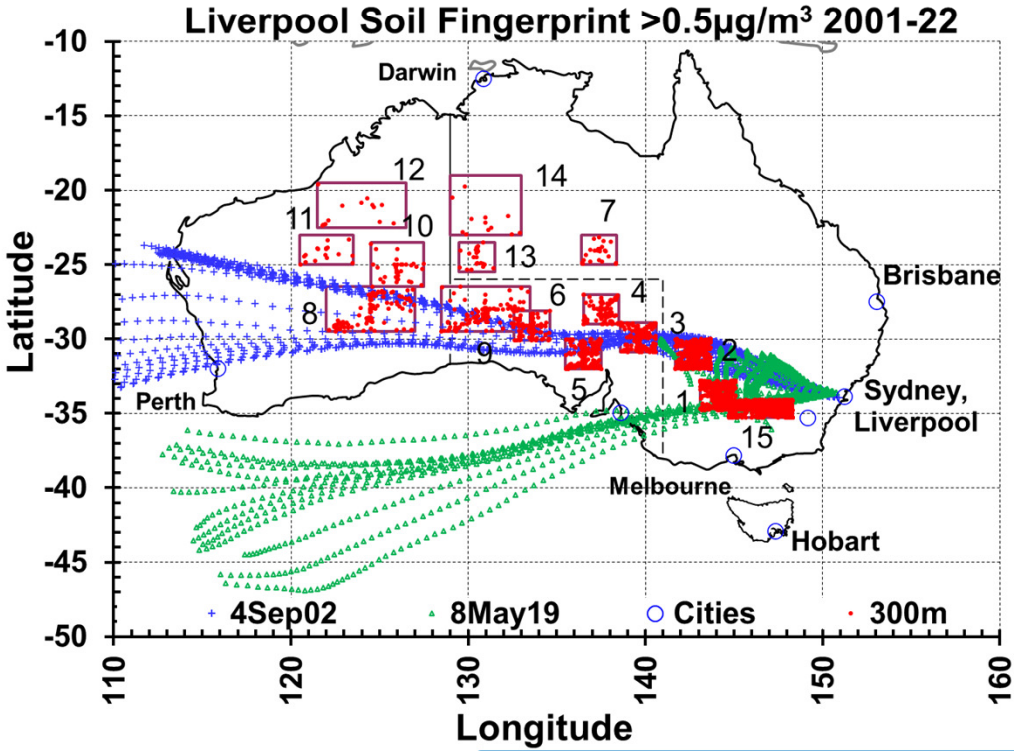
300m Desert	%Soil	Desert	%Soil
15Riverina	41.2	6LakeEyreNorth	2.1
1LakeMungo	25.6	10Gibson	0.7
2LakeWindaunka	11.8	7SimpsonDesert	0.5
4OlympicDam	5.2	13GrtSandyE	0.5
3EastFlinders	4.6	11LitSandy	0.4
9GreatVicE	2.4	12GrtSandyW	0.3
8GreatVicW	2.2	14Tanami	0.3
5EmuFieldsSalt	2.2		

In past 22 years 41% of the dust measured in the Sydney basin (>0.5µg/m^3) at Liverpool had trajectories through the agricultural region, Riverina Box15.

The desert regions (boxes 1-14) tended to contribute only on occasional extreme Soil days.

HYSPLIT: http://ready.arl.noaa.gov/HYSPLIT.php





Accelerator Mass Spectrometry (AMS) - Isotopic Dating

Measurements out to 10 half-lifes

^{14}C

5,730yrs
$1;10^{-12}$

100yrs — **60,000**yrs

Climate and archaeology

$$^{14}_{7}N + n \rightarrow {}^{14}_{6}C + p$$

stratosphere by thermal neutrons

^{36}Cl

301,300yrs
$1;7\ 10^{-13}$

30,000yrs — **3,000,000**yrs

Ground water management and geological exploration

Neutrons on ^{35}Cl, proton spallation of ^{39}K and ^{40}Ca.

^{10}Be

1,390,000yrs
Trace

Cosmic ray interactions with ^{14}N, ^{16}O.
Spallation of ^{16}O, ^{27}Al, ^{28}Si and ^{56}Fe.

500,000yrs — **14,000,000**yrs

Soil erosion and geomorphology

- Megavolt accelerator systems can accelerate most sotopes in the periodic table. Individual atom counting.
- Can measure isotopic ratios to $1:10^{15}$ with a precision of 0.5%.
- Match the isotope half-life to the timescale required.

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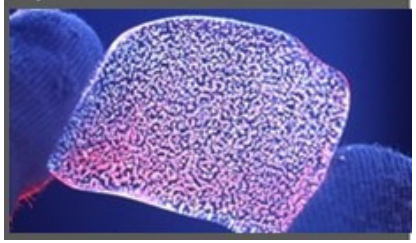
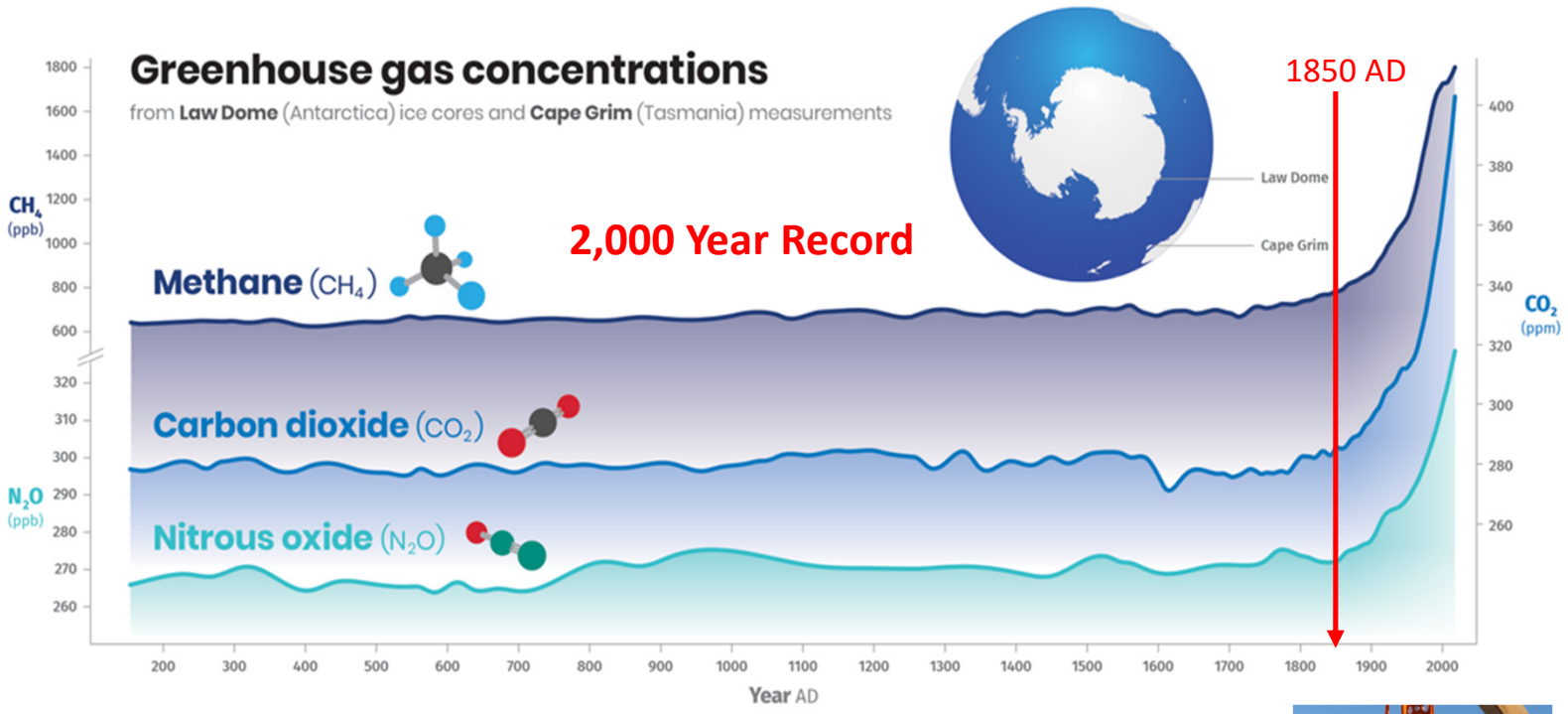
Trapped CO_2 in Antarctic Ice Cores

The three principal greenhouse gases methane, carbon dioxide and nitrous oxide increased dramatically over the industrial period.





In last 2,000 years;

CH_4 – increase ~ 700 ppb to ~1,800 ppb (x25 stronger GHG than CO_2).

CO_2 – increase ~ 280 ppm to ~ 410 ppm.

N_2O – increase ~ 270 ppb to ~ 330 ppb.

Major increases occurred since 1850 AD.

Current levels unprecedented in the last 800,000 years.

- Falling snow which compacts with time traps and preserves gases.
- Air samples as old as 800,000 years have been recovered from Antarctica.
- 1Tonne of ice core contains ~20µg of carbon – ^{14}C AMS works with µg samples.
- ^{14}C-AMS good at distinguishing between modern carbon (livestock, landfill) and ancient carbon (fossil fuels, permafrost).

Accelerator based ^{14}C Measurements

Extraction of solid C from gaseous CO_2 requires microgram sample preparation capabilities, with no cross- contamination.

Microgram carbon samples are pressed into cathodes for insertion into the accelerator ion source.

Megavolt machines accelerate the isotopes through torturous paths from the ion source to the detector.

Only a given isotope with the required (ME/q^2) reaches the detector.

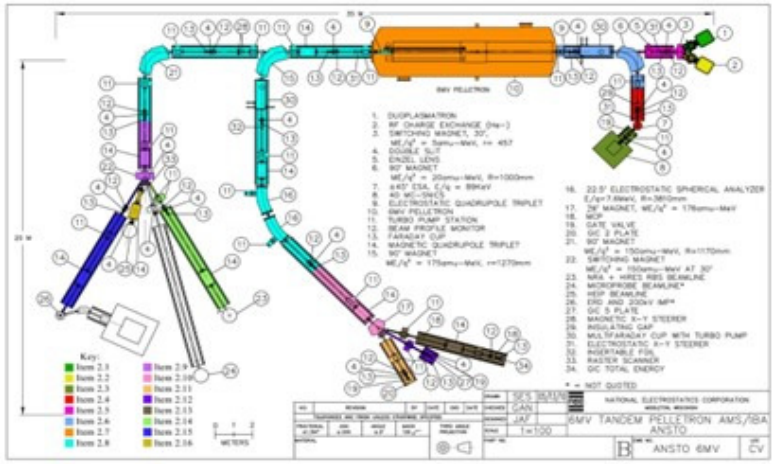
Isotopic selectivity can be as good as $1:10^{15}$.

Measure 10 half-lifes, precision ±0.5%.

A micro-furnace system

Cathode and ion source cathode wheel

High energy beamlines for a 6MV Tandem Accelerator



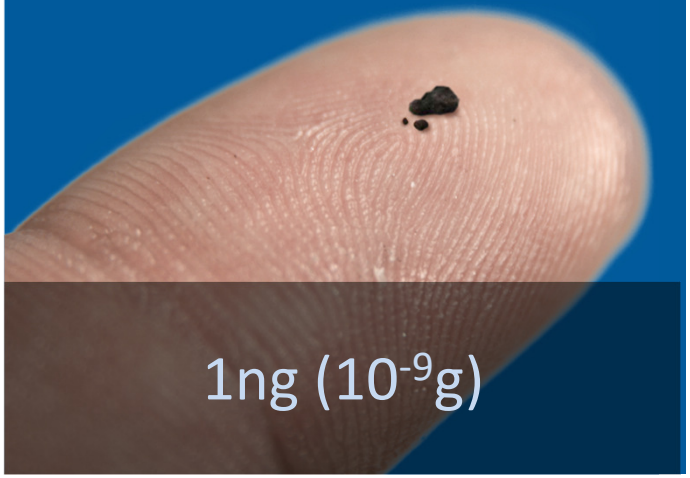
Layout for a 6MV IBA and AMS Tandem Accelerator

Why use accelerators?

Very sensitive

1 in 10^{23} atoms

Very small samples



1ng (10^{-9}g)

Very short processing time

Minutes to hours
not days to weeks

Summary

- The 4 simultaneously obtained IBA spectra of PIXE, PIGE, RBS and PESA are ideal to analyse most key elements (H to Pb) of concern for fine particle (PM2.5) air pollution studies. IBA is a non-destructive technique.
- Typical IBA sensitives are around 1-10 ng/m^3 of air sampled after 3µC runs (10nA for 6 mins) which is more than adequate when total mass concentrations are ~ 10 µg/m^3.
- PMF source apportionment splits the PM2.5 mass into 7-9 different source fingerprints including, windblown soil, secondary sulfates, sea spray, automobiles, smoke from biomass burning and industrial emissions.
- PMF fingerprints couples with back trajectory wind speeds and directions identify the source fingerprint locations and help to better understand long range transport of air pollution.
- AMS techniques using selected isotopes such as ^{14}C, ^{36}Cl and ^{10}Be provide a very broad range of dating methods applicable to environmental and climate change studies.
- AMS can detect isotopic ratios to $1/10^{15}$ with precisions down to 0.5% on samples as small as 10 µg.

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Thank you for your attention

Questions?

Info: http://www.ansto.gov.au/ASP

http://www.ansto.gov.au/IBA

http://www.ansto.gov.au/ASPdatabases

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