# The Progress of Accelerator Mass Spectrometry and their Applications in China

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#### 1. Introduction

AMS- Leading technique for the detection of long-lived nuclides

1. Ultra high sensitivity:  $10^{-15}$ --10<sup>-16</sup> (~65Ka <sup>14</sup>C)

2. Small sample size: mg-µg

- 3. High throughput: ~3000 samples/yr
- 4. Extreme valuable for Archaeological dating, Paleo-environmental & Biomedical studies

#### Challenging Issue for a precise measurement

Interferences need to be eliminated as much as possible in every step of the measuring process:

- Sample collection and preparation
- Ion beam generation
- Beam transport and handling
- Gas stripping
- Data acquisition and processing

#### High quality AMS facility performances:

- Extremely low machine background
- Very low isotopic fractionation
- High beam transmission efficiency
- High resolution beam analysis
- High long term operation stability

# Among ~ 60 AMS facilities around the world, 5 are in China.

CIAE	HI-13MV Tandem	<sup>10</sup> Be, <sup>26</sup> Al, <sup>36</sup> Cl, <sup>41</sup> Ca, <sup>79</sup> Se, <sup>129</sup> I
PKU	EN 6MV Tandem	<sup>10</sup> Be, <sup>14</sup> C, <sup>26</sup> Al
PKU	0.6 MV Tandem	<sup>14</sup> C
XI'AN	3 MV	<sup>10</sup> Be, <sup>14</sup> C, <sup>26</sup> Al,
CENTER	Tandem	$^{41}$ Ca, $^{129}$ I
SINR	Cyclotron	<sup>14</sup> C

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#### 2. Typical AMS Facilities in China

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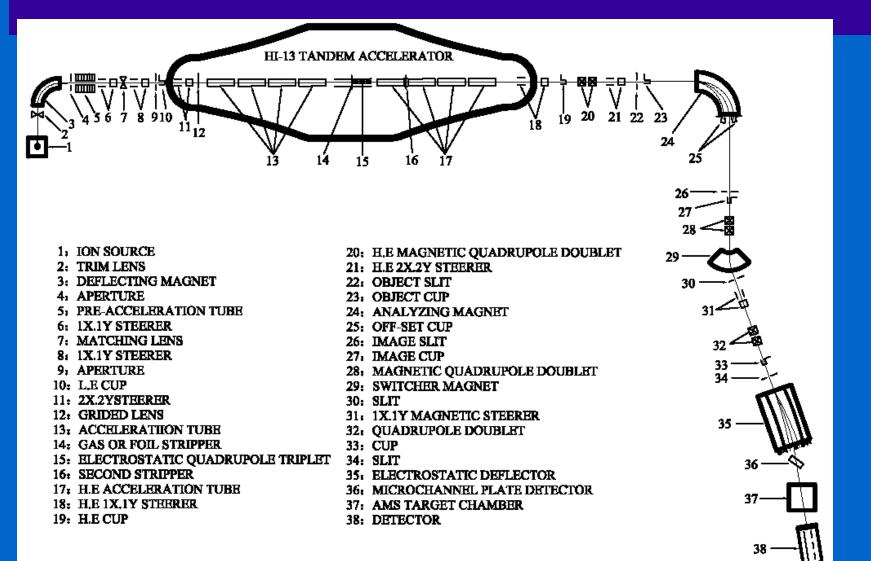


#### HI-13 Tandem Accelerator

#### China Institute of Atomic Energy (CIAE)

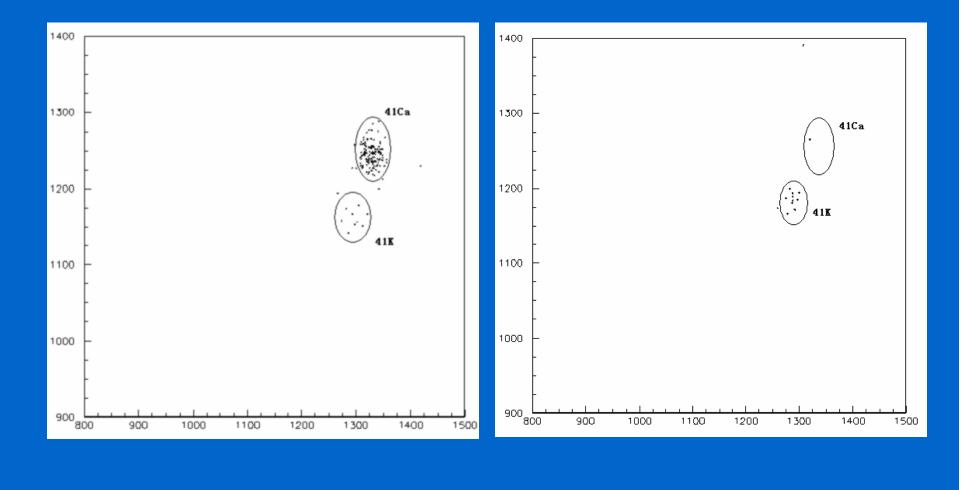


#### The layout of CIAE's HI-13 MV AMS system



### Two-dimensional spectra ( $\Delta$ E1- $\Delta$ E2)of <sup>41</sup>Ca standard(10<sup>-11</sup>) sample and blank sample

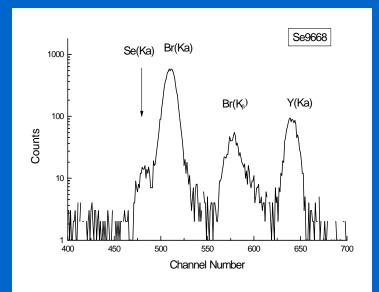
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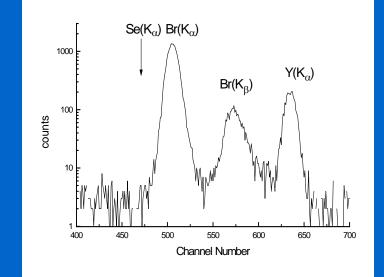


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# Projectile X-ray method for the identification of <sup>79</sup>Se

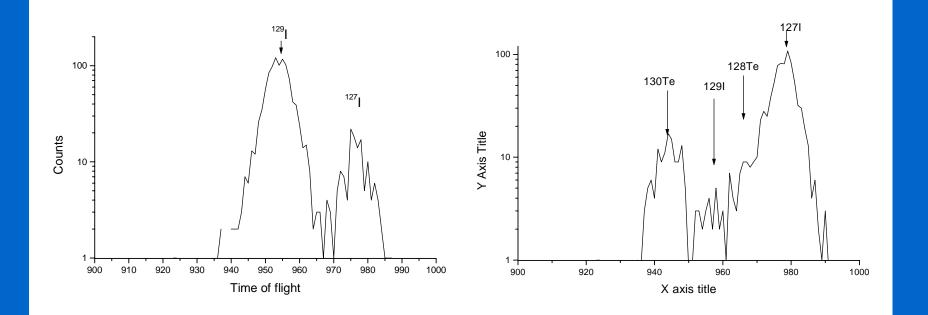
 According to different characteristic Kα ray energy, the <sup>79</sup>Se and <sup>79</sup>Br are identified.





# TOF method for isotope identification of <sup>129</sup>I

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### For the measurements of heavy elements (Pu, U isotopes), Mass resolution(M/ $\Delta$ M) increased from 90 to 400.



The upgraded injection system

#### PKUAMS at the Institute of Heavy Ion Physics, Peking University



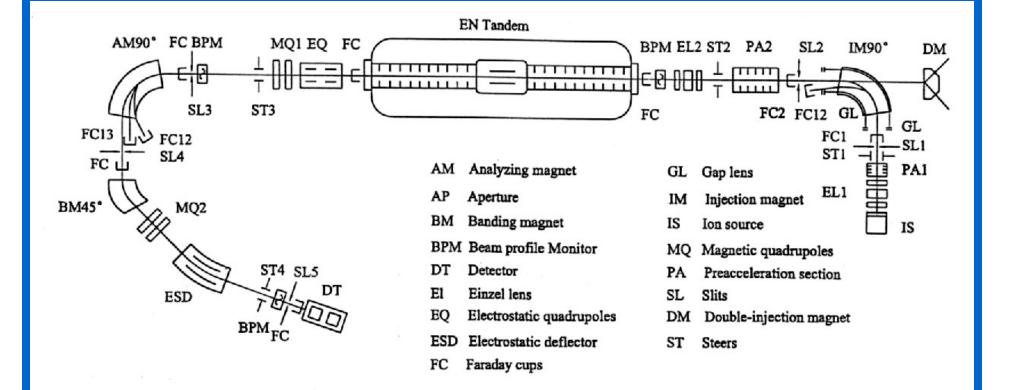


#### HVEC 6MV EN Tandem Accelerator

#### Upgraded Features of PKUAMS for the Xia-Shang- Zhou Chronology Project

- Precision of measurement : 0.4-0.5% for  $^{14}C$
- Machine background : <sup>14</sup>C ~ 1×10<sup>-15</sup>; <sup>10</sup>Be <6×10<sup>-15</sup>
- Beam transport efficiency: ~ 30%
- Serial sample calibration with Bayesian method developed
- Quality control for precise dating established

#### The Layout of the PKU-AMS



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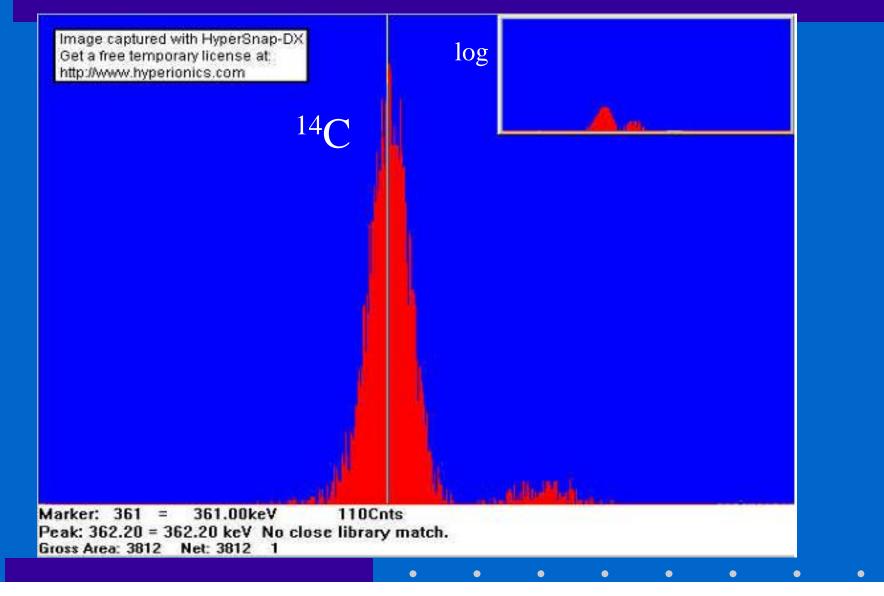
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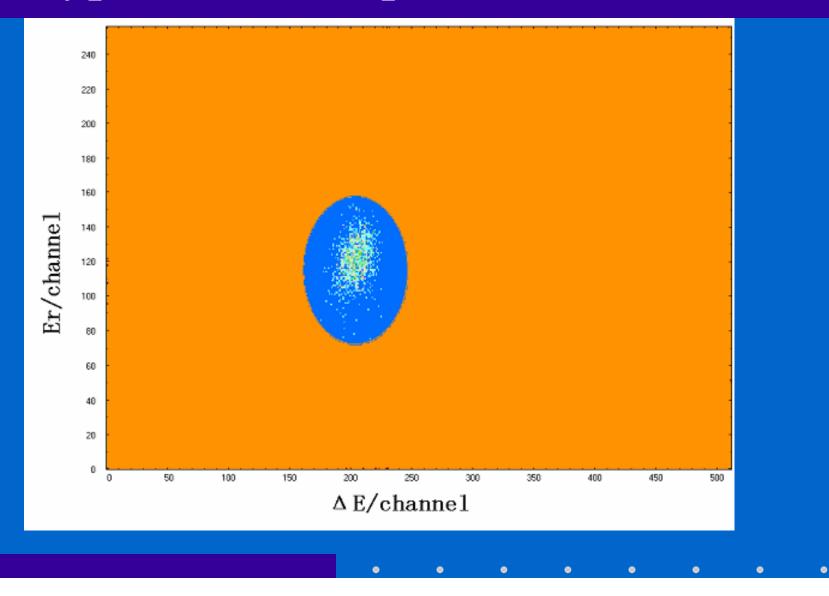
#### Computer Control System



#### Typical Spectrum of <sup>14</sup> C after upgrading



#### Typical $\Delta E$ --E Spectrum of <sup>10</sup>Be

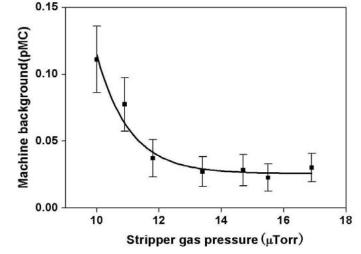


#### Modern Compact Facility for PKU-AMS



#### Machine Performance

 $\diamond$  Accuracy : 0.3 %, ~25 yrs  $\diamond$  Background: <4X10<sup>-16</sup>, ~65 Ka 🔶 Source output: 60-80 μA Transport efficiency : ~42% 0.00 10 Dating Capability : >3000/yr Standard sample comparison : OXI, OXII, ANU, & IAEA standard sample Comparison inter Labs : UC-Irvine, ANU, IAEA



#### The Sputtering Ion Source with 40 targets



►  $I_{inj}$ =50-80 µA <sup>12</sup>C<sup>-</sup> with high beam quality and stability

#### The Sequential Injection System

Biased vacuum
 chamber for ~10 Hz
 cycling sequential
 injection

◆ <sup>12</sup> C <sup>-</sup>	0.3 ms
13C-	1.0 ms
<sup>14</sup> C-	100 ms

Fractionation &
 beam loading effect
 minimized



#### Special Recirculation Gas Stripper

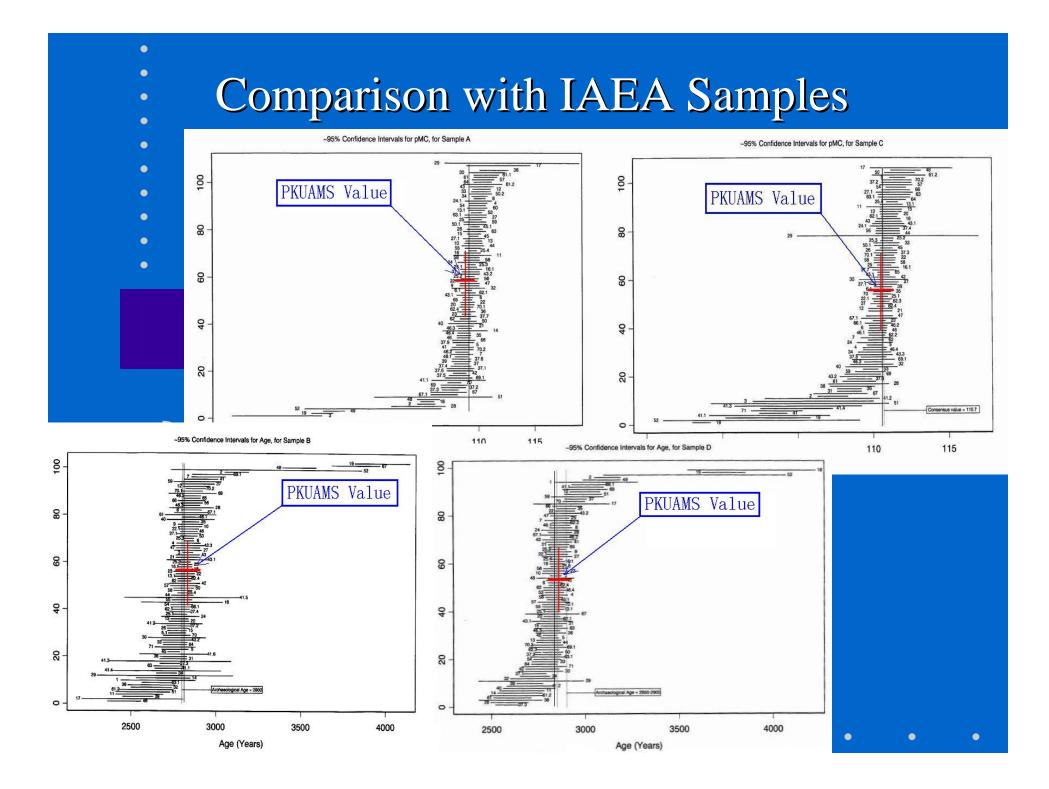
 Stripping channel \$ 8X450 mm
 Pressure inside the stripping channel 17-19X10<sup>-3</sup> Tor.
 Two molecular pumps for recycling stripping gas



#### Detector & Data Acquisition

♦ An Au Surface **Barrier** Detector with a lifetime about 1500 hrs  $\diamond$ Clean <sup>14</sup>C spectrum can be obtained at 200 counts/sec for modern carbon sample







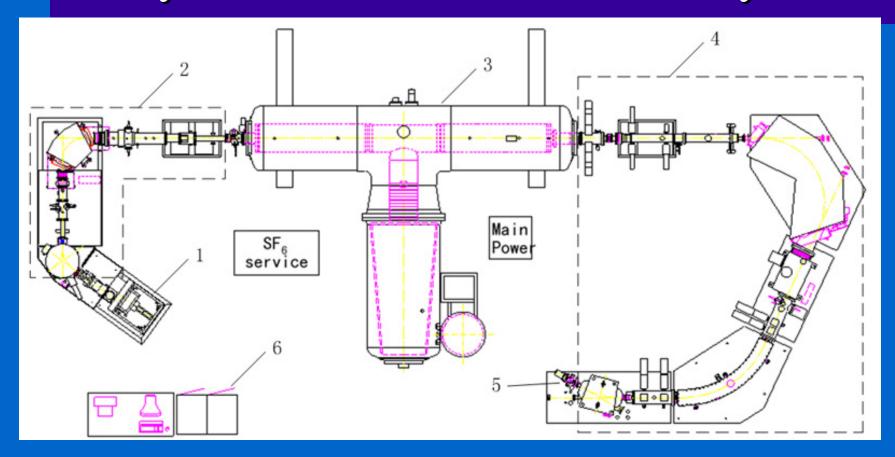
Jointly established by Institute of Earth Environment, CAS and Xi'an Jiaotong University in 2004

#### Xi'an 3 MV Multi-element AMS

(10Be, 14C, 26AI and 129I)



#### Layout of the Xi'an AMS Facility



1. Ion source2. Injector3. HVEC 3MV Tandem4. High energy analyzing system5. Detector6. Control system

#### **Special features**

Ion source for both solid and potencially CO<sub>2</sub> samples
Sequential injection at 100Hz cycling frequencies
Low injection energy with Q-snout
Accelerator tube with combined magnetic & electrostatic suppression results in low X radiation level
Flat Top mass independent transmission





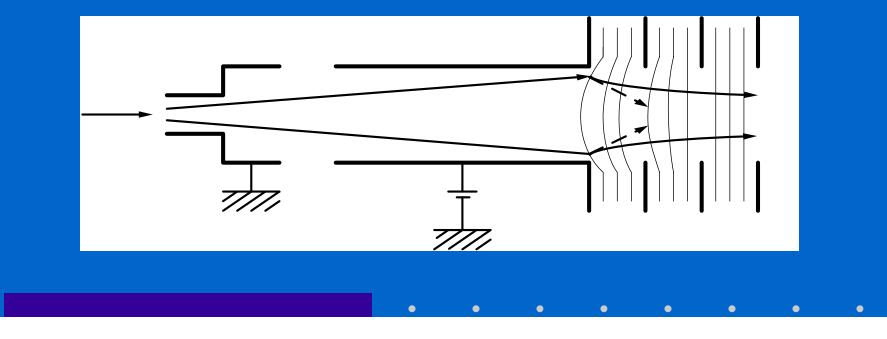
Negative ion source with 50 sample holders

- 54° electrostatic spherical analyzer (R=470mm)
- 90° bouncer magnet (R=400mm)
- Fast sequential injection at 100 Hz, <sup>14</sup>C 9.5 ms, <sup>12</sup>C & <sup>13</sup>C 100 µs

#### Low injection energy and Q-snout

Low Injection energy 35 keV Using small electromagnetic components

Q-snout: solving the overfocusing on ions while entering into the accelerator with low energy



#### **Results of the Acceptance Tests**

<sup>10</sup>Be、<sup>14</sup>C、<sup>26</sup>AI、<sup>129</sup>I (2006.7)

		Sensitivity ( Tested )	Statistic Precision ( Tested )
<sup>14</sup> C (Graphite)	<sup>14</sup> C/ <sup>12</sup> C	3.0×10 <sup>-16</sup>	0.18%
	<sup>13</sup> C/ <sup>12</sup> C		0.070%
<sup>10</sup> Be	<sup>10</sup> Be/ <sup>9</sup> Be	3.6×10 <sup>-15</sup>	1.40%
<sup>26</sup> A1	<sup>26</sup> Al/ <sup>27</sup> Al	2.3×10 <sup>-15</sup>	0.38% (3.0MV)
<sup>129</sup> I	<sup>129</sup> I/ <sup>127</sup> I	2.0×10 <sup>-14</sup>	1.75%

High <sup>14</sup>C precision reached by fast sequential injection
 High <sup>10</sup>Be detection sensitivity at 3MV AMS

#### FUTURE POSSIBLE EARTH SCIENCES RESEARCH

High resolution study on climate & cosmic events
Establishing Chinese climatic history
Studies on Chinese civilization origin
Dating of groundwater
Control and preventability of environmental pollution
Physiognomy evolvement of Tibetan Plateau
Geodynamics

**3. Typical Applications** 

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#### **Archaeological Studies : Chronicle Dating**



Si Ma-qian 145 BC-90 BC

Shi JI, written by Si Ma-qian, is the most authoritative book on the chronicle of ancient Chinese history starting from 841 BC. It also gives all the King's names and their genealogy since Xia. However, Chinese chronology has not been well established so far.

For the date of the conquest of Shang by King Wu of Zhou, at least 44 different solutions have been offered spreading over a span of 112 years.

Dynasties	Dates
Xia	? BC - ? BC
Shang	? BC - ? BC
Western Zhou	? BC – 771 BC
Eastern Zhou	770 BC – 252 BC

The Xia-Shang-Zhou Chronology Project integrates Archaeological studies, Astro-chronological studies & Radiocarbon dating etc. to establish a chronicle frame for Xia, to Western Zhou on the base of scientific evidence

Nine series of samples, including 7 sites, one tomb series and a special series of oracle bone samples, were studied with AMS <sup>14</sup>C dating

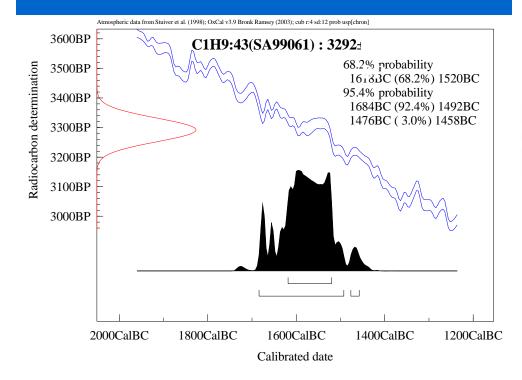
The inscriptions on oracle bones recorded the King's name, activities, war, sacrificial offerings and important astronomical phenomena.

Oracle bones from Yinxu site of Shang Dynasty

# Calibration of serial samples with Bayesian method

 $\succ$  Tree rings used to convert <sup>14</sup>C age into calendar years

#### Calibration of serial samples using Bayesian method to reduce uncertainty



B	Coundary Begin		
Г	Phase Early W. Zhou - I		
T	014-88QJ7H147 90.9%	where the second	-
t	006-86QJ4M6266 103.1%		
t	007-86QJ4M6081 90.9%		1 1 1 1
Г	Phase Early W. Zhou - II		1 I I I I
1	008-86QJ4M6306 67.1%	min i	-
F	017-86QJ7H78 87.4%		<u> </u>
t	016-80QIH109(3) 98.9%		
Г	Phase Middle W. Zhou - I		
╞	009-86QJ4M6411 87.4%		· · · · · ·
┢	019-840J7H23 102 9%		+ + + +
+	018-82QIVH410 114.4%		
с г	Phase Middle W. Zhou - II		1 1 1
	021-82QIVH402:1 109.8%	- Maria	
╞	020-82QIVH402 111.5%		
	Phase Late W. Zhou - I		+
+	010-86QI2M5215 151.8%		·····
+	022-82QIIIH326 132.0%		
	Phase Late W. Zhou - II		+ + + + + + +
-	011-860I2M5217 146.9%		· · · · · · · · ·
P	Coundary End		
D	oundary Ena		

# Results via synthesis of various means

Dynasties	Dates
Xia	ca. 2070 BC – ca. 1600 BC
Shang	ca. 1600 BC – 1046 BC
Western Zhou	1046 BC – 771 BC

Chronological frame of Xia, Shang & Western Zhou

## Studies on Neolithic culture in North China Donghulin Site discovered & excavated





Tombs, ash-pits, fireplaces and other vestiges along with chipped stone implements, microliths, polished stone tools and potteries were found at Donghulin site



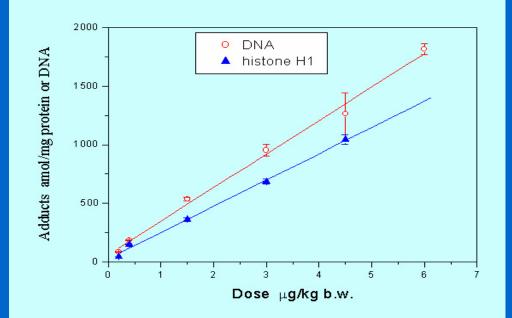
#### Studies on Neolithic culture in North China

- More than 20 charcoal and bone samples were collected from this site and dated on PKUAMS.
- The results show that the Donghulin Man lived from 9000BC to 7000BC, the early Neolithic period



#### **Bio-science studies using PKUAMS**

Measurement on<sup>14</sup>C-labled nicotine-DNA adducts



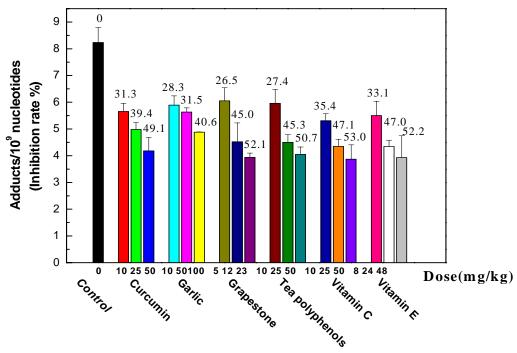
Recently, the inhibitory effects of curcumin, garlic,squeeze, grape-seed extract, tea polyphenols, vitamin C and vitamin E on nicotine-DNA adduction have been investigated.

#### Inhibition Effects of Adductions by Dietary Constituents

•Amongst the six agents, grape-seed extract showed the strongest inhibition to the DNA adduct formation

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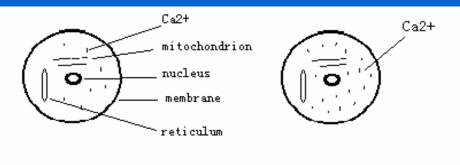
•The reduction rate reached about 50% for all agents except garlic squeeze (40%)



#### **Biomedical applications at CIAE**

• Using <sup>26</sup>Al as a tracer to study pneumoconiosis

• Using <sup>41</sup>Ca as a tracer to investigate the origin of increased free Ca<sup>2+</sup> in cells when exposed to carcinogenic substance.



(a) a normal cell

(b) a cell exposed to C.S

10<sup>7</sup> Ca atoms may go into each cell when the cells are exposing to carcinogenic substance 1 hour

# CONCLUSIONS

• AMS technology has been well developed in China

• Each of the 4 Tandem based AMS performed successfully to meet their needs in the fields of Archaeology, Earth Science, Environmental Sciences and Biomedical sciences and etc.

• All of the facilities will be further upgraded according to the requirements of new applications

# Thank you!

#### The Campus of Peking University

#### Demands of Xia-Shang- Zhou Chronology Project

- To define the chronological frame from the start of Xia dynasty up to 841 B.C. by precise <sup>14</sup>C dating on serial samples from various historical sites and oracle bones
- Precision better than  $\pm 30$  years
- Counts of <sup>14</sup>C per measurement 100 300k
- High long-term operation stability

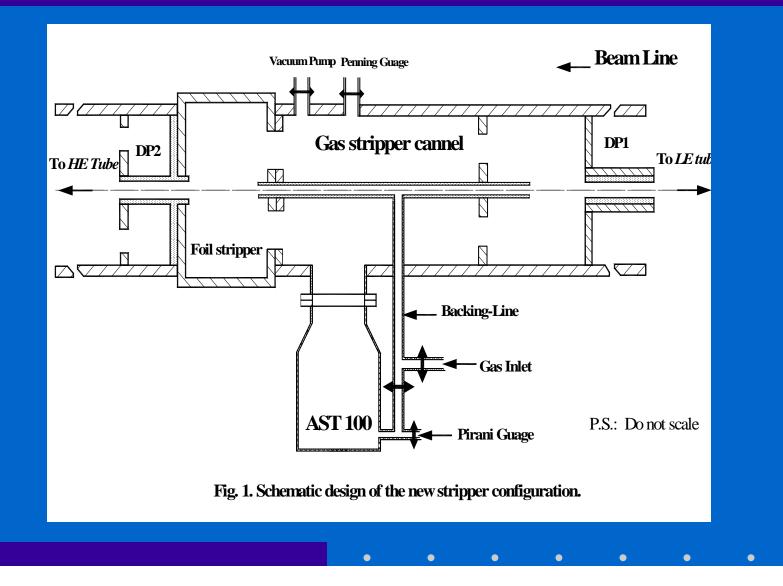
# Quality control for precise Dating

- Machine error analysis and reduction
- Standardized beam tuning procedure
- Well-designed measuring procedure and data manipulation program
- Statistical testing
- Multiple targets with *t* or *F* test
- Multi-standards (OX-I, OX-II, ANU, IAEA...]
- Inter-comparison between Laboratories

# Upgraded components of PKU-AMS

- High output ion source with 40 sample-holders
- Fast cycling sequential injection system
- High efficiency, flat-topping, mass independent beam transport
- Installation of recycling stripping gas
- Highly stabilized power sources
- Computer controlled operation & data acquisition

#### Recirculation Stripper for EN Tandem



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# The Conquest of Shang by King Wu

> A profile was found in Fengxi site (near Xi'an) with the remains from Pre-Zhou to the late Zhou > Fengxi site was dated. It was the home of the Zhou people before the establishment of Western Zhou ▶ King Wen of Zhou (周文王) moved his capital to Fengxi, and KingWu (周武王), son of King Wen, conquered Shang from Fengxi

# The Conquest of Shang by King Wu

- AMS results: the Conquest occurred mostly during 1060 BC – 1000 BC
- LS results: the Conquest occurred mostly during 1050 BC – 1010 BC

Considering the dating results of Yin site and oracle bones from late Shang as well as Liulihe site and Tianma-Qucun site from early Western Zhou, the most possible intervals was 1050 BC – 1020 BC

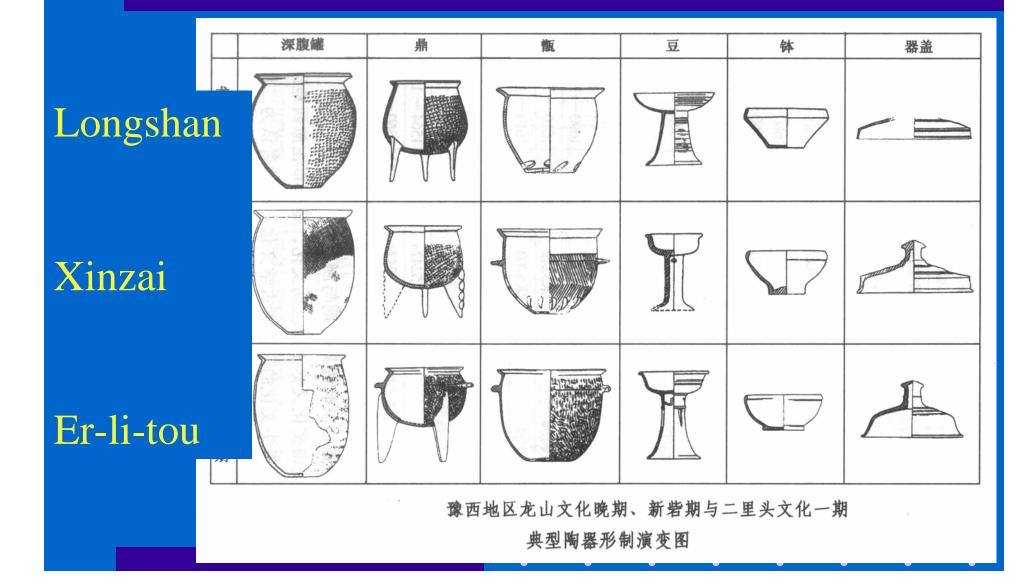
# Xinzhai site – A Study on Xia



1000 m<sup>2</sup> Cultural
 remains in Xinzhai site
 contain late Longshan
 and early Er-li-tou.

• Bayesian method is used again for the calibration of the serial samples from 3 sites

#### Evolution of the form of Potteries



#### Unearthed pottery from Xinzhai site



#### Late Longshan phase



#### Unearthed pottery from Xinzhai site



#### Xinzhai Phase



#### **Xinzhai Phase**

It is believed Xia started from late Longshan and finished in Erlitou culture

There was a gap between late Longshan and Erlitou culture

Xinzhai culture filled this gap

Boundary Start	
Phase Late Longshan	
002-1 H123_103.8%	
R_Combine 014 H126 107.4%	
008 H122 106.7%	
007-2 H120 101.9%	
R_Combine 001 H119 101.4%	
Boundary 1-2	
- Phase Early Xinzhai	
<i>R_Combine 006 T1(6)C 109.0%</i>	
<i>R_Combine 012 H116 101.8%</i>	
005-2 H112 100.6%	
<i>R_Combine 019 H115 112.2%</i>	
028 H61(6) 113.9%	
Boundary 2-3	
- Phase Late Xinzhai	
<i>R_Combine 018 H40 80.3%</i>	
<i>R_Combine 017 H26 113.3%</i>	
009 H76 125.8%	
<i>R_Combine 010 H48 118.3%</i>	
<i>R_Combine 013 H45 112.6%</i>	
<i>R_Combine 016 H29(1) 119.2%</i>	
021-2 H66 120.7%	
020-2 H30 79.5%	
Boundary End	

Calendar date

#### Preliminary Results of Xinzhai Site

The transition time from Longshan to Xinzhai was about 1840BC – 1820BC

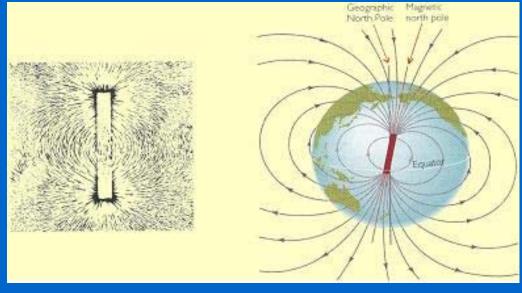
The end age of Xinzhai (~ 1720BC) is about the same with the beginning of Er-li-tou

It is not clear if Longshan culture belongs to Xia, but most archaeologists in China believe Xinzhai culture belongs to Xia, therefore the beginning of Xia dynasty was at least before 1820BC

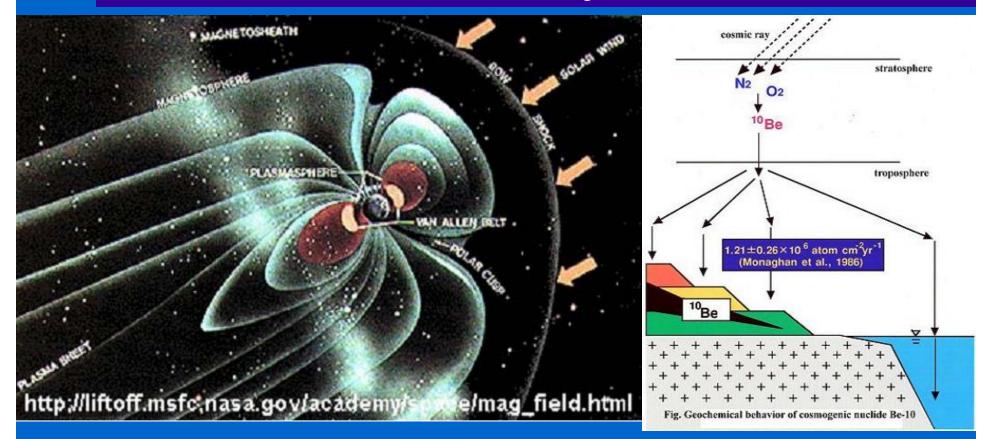
#### Earth Science Studies with <sup>10</sup>Be

• The Matuyama-Brunhes polarity reversal which happened 0.78 Ma ago was recorded in loess L8 in Chinese loess sequence which corresponds to a glacial period.

• The same polarity reversal boundary by the marine record was in an interglacial period



# Geomagnetic field shielding of cosmic rays



<sup>10</sup>Be is a useful isotopic tracer for studying the formation of Aeolian deposits and Quaternary climate evolution

#### Matuyama-Brunhes polarity reversal

- One explanation is that the loess record of the Matuyama-Brunhes boundary (MBB) is displaced downwards by over 100 cm
- To determine the true position of the MBB in loess, <sup>10</sup>Be concentration in loess from Louchuan, Shanxi Province is being dated, as the increase of the <sup>10</sup>Be production rate due to the reduction of geomagnetic field intensity during the polarity reversal is significant.

#### Matuyama-Brunhes polarity reversal

PKUAMS was tuned to the best condition for separating <sup>10</sup>Be from <sup>10</sup>B with a gas-filled chamber. An inter-comparison with Uppsala University was in good agreement

➤ More than 80 loess samples have been measured and the  ${}^{10}$ Be/ ${}^{9}$ Be ratios are found in the range of  $10^{-11}$ - $10^{-12}$ , the results will be compared & processed further. Earth's magnetic field was reversed many times in the past. The latest reversal occurred at 780 thousand years ago, the Brunhus-Matuyama reversal. These geomagnetic polarity reversals are recorded in different types of geological sediments, e.g. deep-sea sediments and loess (the wind-blown dust widely diustributed in northern China,). However, the Brunhus-Matuyama reversal boundary (MBB) is found in different positions corresponding to glacial and interglacial times in loess and marine sediments respectively. Someone suggested that this apparent disparity between ocean and land records is due to the delayed lock-in of reversal signals in loess. One way to test their hypothesis is to measure the Be-10 concentration in loess profiles. As the cosmogenic Be-10 production is expected to increase at the time of geomagnetic reversal due to the reduction of geomagnetic field intensity, a Be-10 peak may be detected and it should point to the true position of the reversal. This is one of the examples of our AMS application at PKU.