Yale 耶鲁大学–南京信息工程大学大气环境中心
Yale-NUIST Center on Atmospheric Environment

Potentially Regional Sources of High Airborne Particulate Arsenic over the Subtropical Free Troposphere

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Working Experiences

2007/01-2016/12

- Research Center for Environmental Changes (RCEC), Academia Sinica, Postdoc and Assistant Research Scholar

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- Atmospheric Sciences Research Center (ASRC), SUNY at Albany, Postdoc

2017/01-2017/12

- Toxic and Chemical Substances Bureau, Senior Specialist

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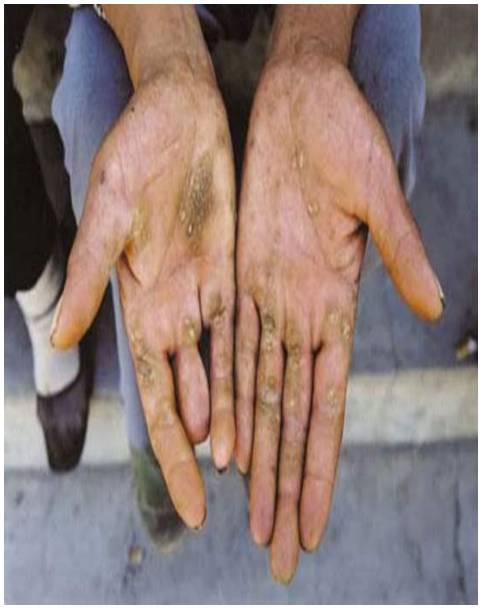
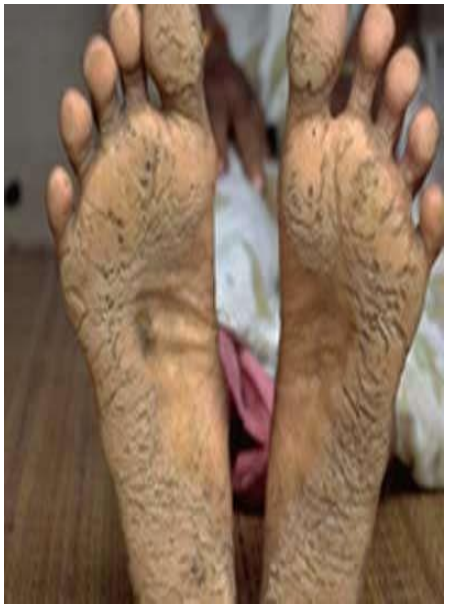
Research Interests

- Chemical Characteristics of Airborne PM.
- Stable Isotopes (carbon and lead) Related Issue in Atmosphere
- Source Identification of Ambient Particulate
- Stratosphere-troposphere Air Exchange and Its Impact on Atmospheric Chemistry.
- Mechanism and Formation of Airborne Nitrate

Introduction-Arsenic

- Arsenic (As), categorized into carcinogenic species by International Agency for Research on Cancer (IARC), has long been considered as a toxic element, having adverse effects on human health.





Where is As from?

- Atmospheric As is released from both natural and anth… bal emi…
- Volc… …les are … …ion and … …ntly anth…

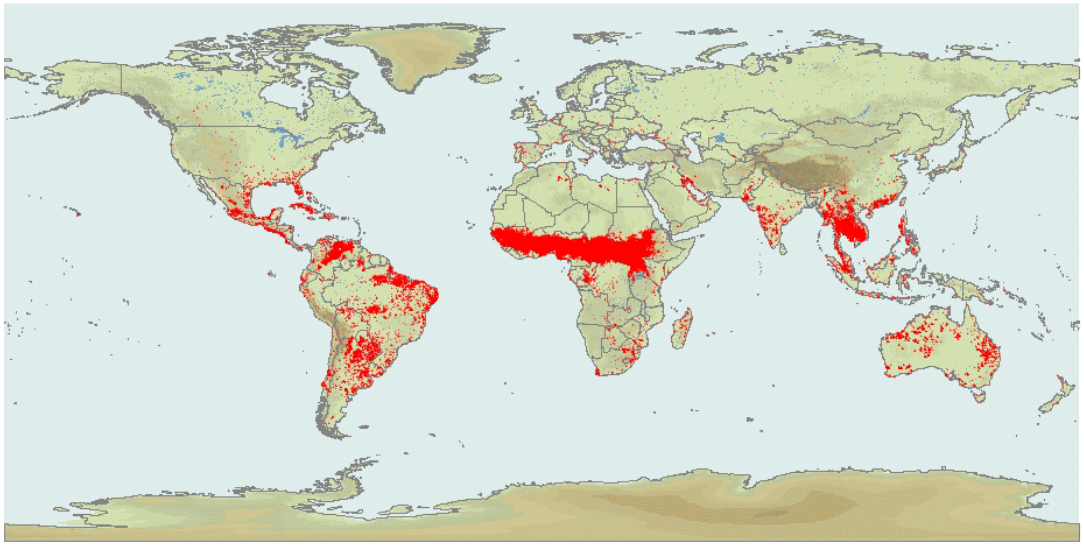
Trace metal	Anthropogenic source	Natural source	Total emission	Natural/total emissions*
As	19 (12–26)	12 (0.86–23)	31 (13–49)	0.39
Cd	7.6 (3.1–12)	1.3 (0.15–2.6)	8.9 (3.2–15)	0.15
Cr	30 (7.3–54)	44 (4.5–83)	74(12–137)	0.59
Cu	35 (20–51)	28 (2.3–54)	63 (22–105)	0.44
Hg	3.6 (0.91–6.2)	2.5 (0.10–4.9)	6.1 (1.0–11)	0.41
Mn	38 (11–66)	317 (52–582)	355 (63–648)	0.89
Mo	3.3 (0.79–5.4)	3.0 (0.14–5.8)	6.3 (0.93–11)	0.48
Ni	56 (24–87)	30 (3.0–57)	86 (27–144)	0.35
Pb	332 (289–376)	12 (0.97–23)	344 (290–399)	0.04
Sb	3.5 (1.5–5.5)	2.4 (0.07–4.7)	5.9 (1.6–10)	0.41
Se	6.3 (3.0–9.7)	9.3 (0.66–18)	16 (2.5–24)	0.58
V	86 (30–142)	28 (1.6–54)	114 (32–220)	0.25
Zn	132 (70–194)	45 (4.0–86)	177 (74–280)	0.34

All figures in units of 10^9 g yr^{-1}. Emissions from anthropogenic sources are from Nriagu and Pacyna[1]; these are median values, with the ranges in estimated emissions given in parentheses.

* Median values only.

Biomass Burning

- **Biomass burning** (BB) emissions associated with natural and human activated fires are the important sources of atmospheric particulate and trace gases.



Long-range transported BB pollution

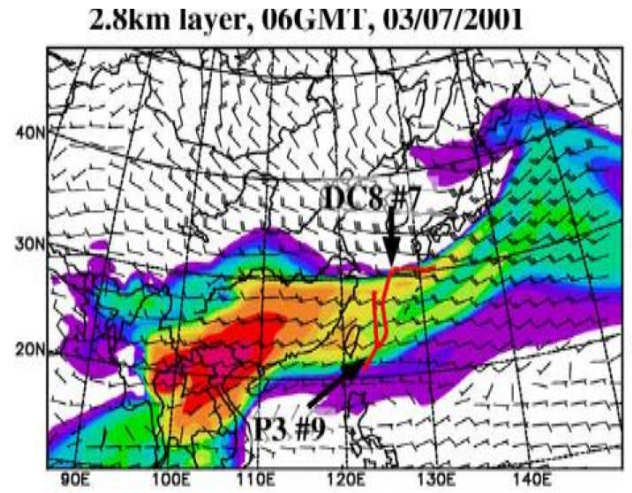
JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 108, NO. D21, 8824, doi:10.1029/2002JD003110, 2003

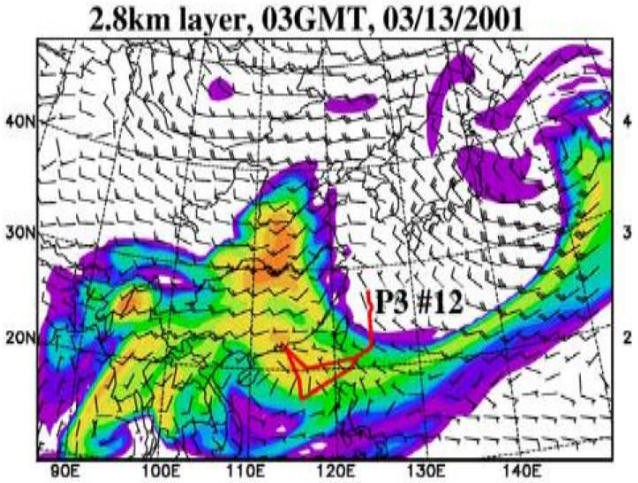
Influences of biomass burning during the Transport and Chemical Evolution Over the Pacific (TRACE-P) experiment identified by the regional chemical transport model

Youhua Tang,[1] Gregory R. Carmichael,[1] Jung-Hun Woo,[1] Narisara Thongboonchoo,[1] Gakuji Kurata,[2] Itsushi Uno,[3] David G. Streets,[4] Donald R. Blake,[5] Rodney J. Weber,[6] Robert W. Talbot,[7] Yutaka Kondo,[8] Hanwant B. Singh,[9] and Tao Wang[10]

Received 31 October 2002; revised 18 April 2003; accepted 25 April 2003; published 4 November 2003.

[1] Using a regional chemical transport model, STEM 2K1, and the emission inventory for the Transport and Chemical Evolution Over the Pacific (TRACE-P) period [*Woo et al.*, 2003 *Streets et al.*, 2003], we successfully simulated important features of the biomass burning (BB) CO outflow. Simulated results agree well with the TRACE-P aircraft measurements and Thailand surface observations. On the basis of sensitivity studies with and without biomass emissions, we identified nine flight segments that are affected by biomass plumes during the TRACE-P period and compared the characteristics of the BB air masses with the other air masses. The BB air masses emitted from Southeast Asia contain relatively high HCN ($\Delta HCN/\Delta CO \sim 0.0015$) and potassium ($\Delta K^+/\Delta CO \sim 0.0038$) but very low NO ($\Delta NO$ /



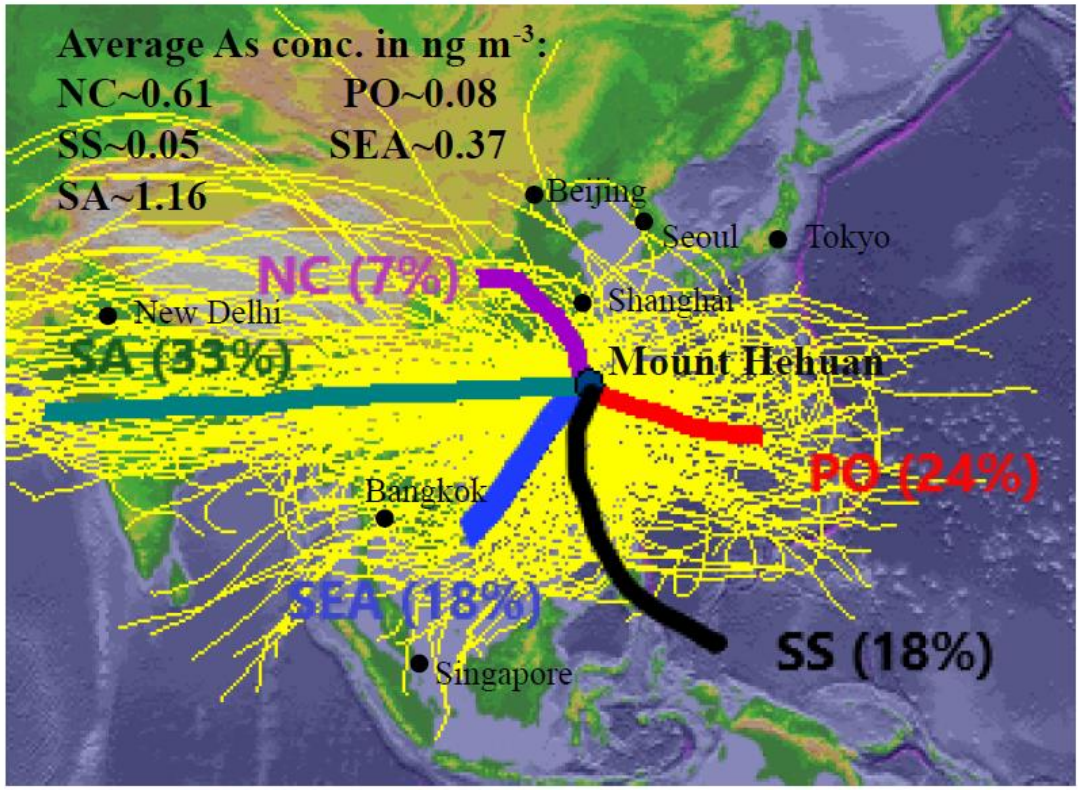


Experiment

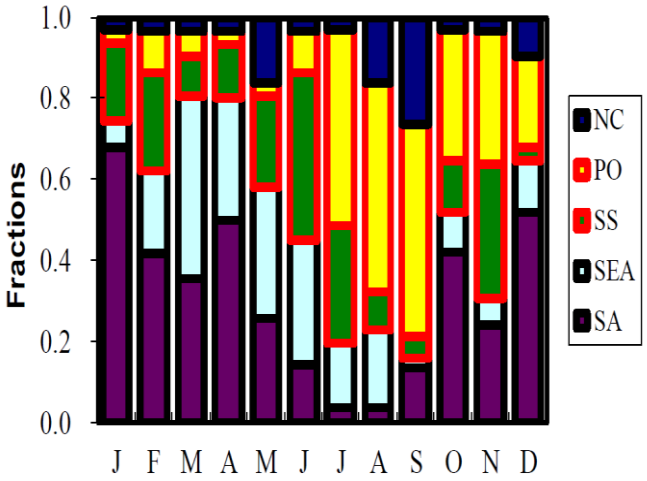
Our sampling site is located at Mount Hehuan (24.16N, 121.29E, 3001 m a.s.l.), Taiwan. The experimental campaign was conducted from September 2011 to September 2012.

- Daily TSP samples were collected using a high-volume TSP sampler.
- TSP samples were digested with HNO3 and HF; analyzed elements through ICP-MS. Also, TSP samples were extracted with Milli-Q water and determined ions by IC.
- Hourly CO and O3 were monitored using commercial instruments.
- Backward trajectory was computed using HSPLIT model.
- WRF-Chem model was also used to simulate BB events.
- Fire spots were obtained from MODIS.

Backward Trajectory Analysis

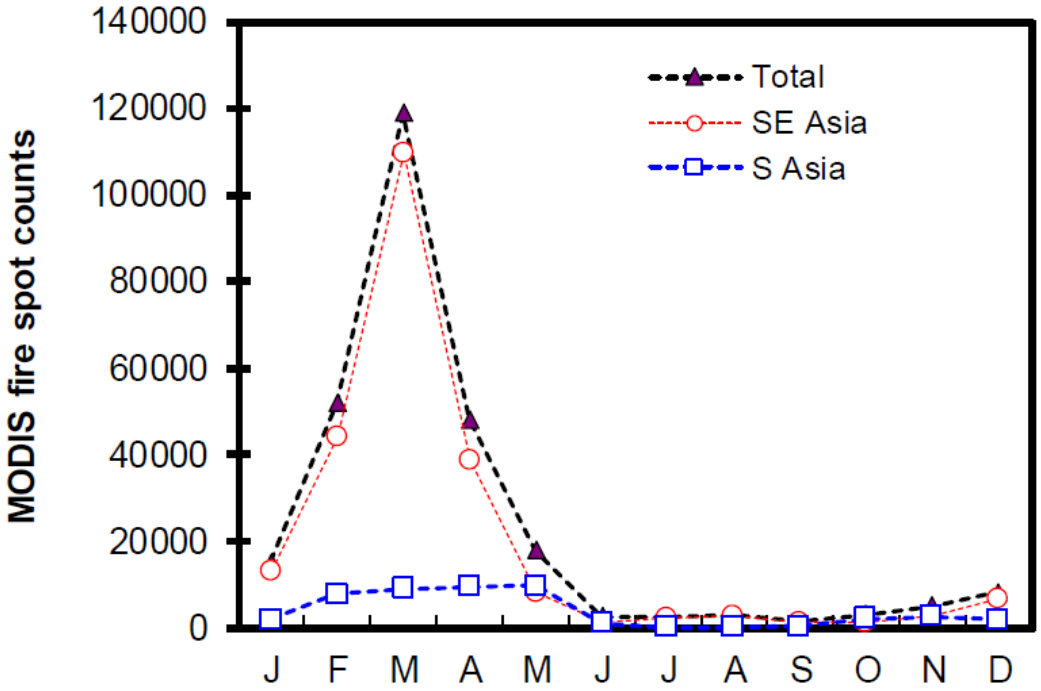


- Five-day backward trajectory arriving at 3000 m were computed once per day at 12:00 (local time).
- Four additional trajectories were generated of which starting location changed ± 0.5˚.



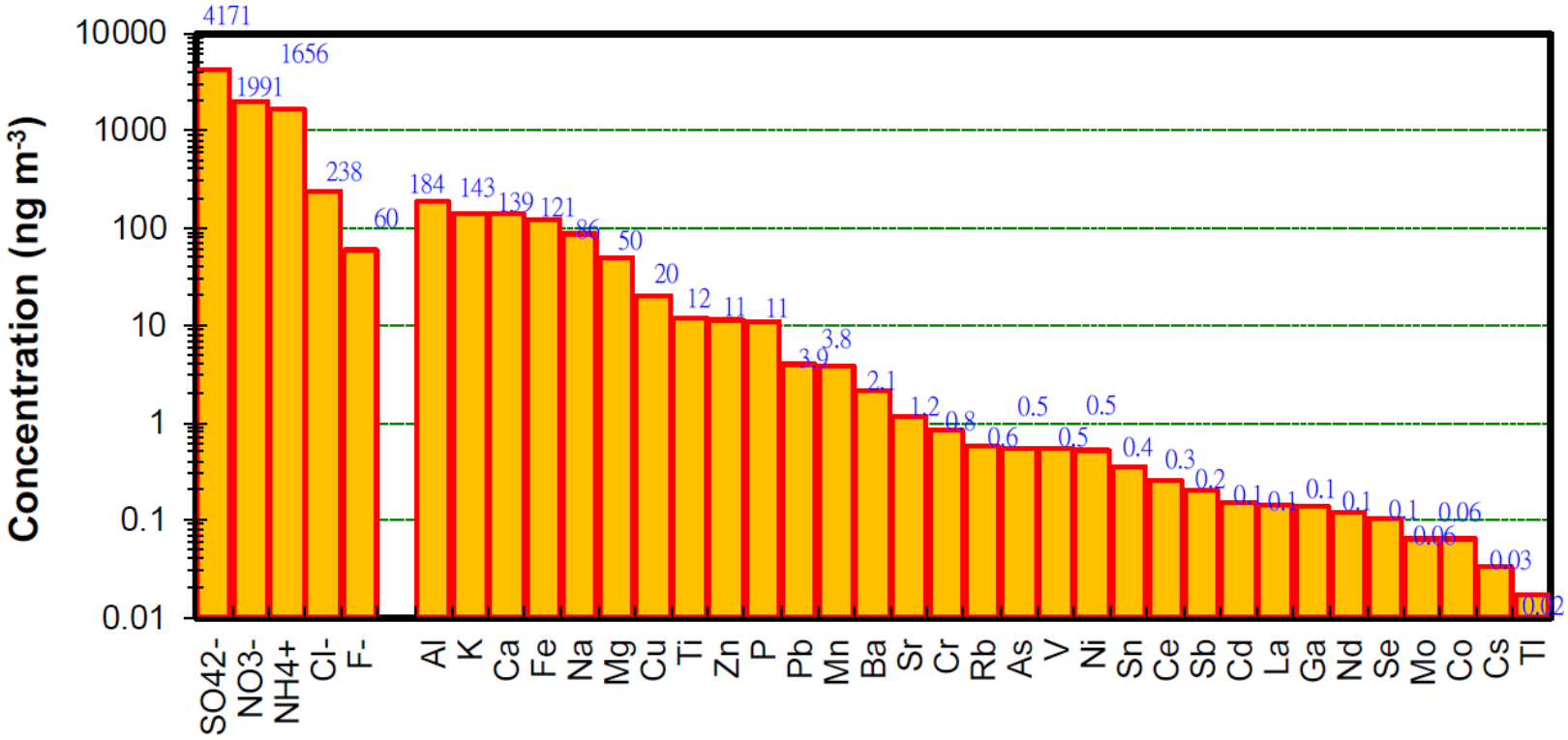
- SA was the predominant air parcel, followed by PO, SS, SEA and NC.
- SA, NC and SEA air masses were categorized into continental air whereas PO and SS were divided into marine air.

MODIS fire spots

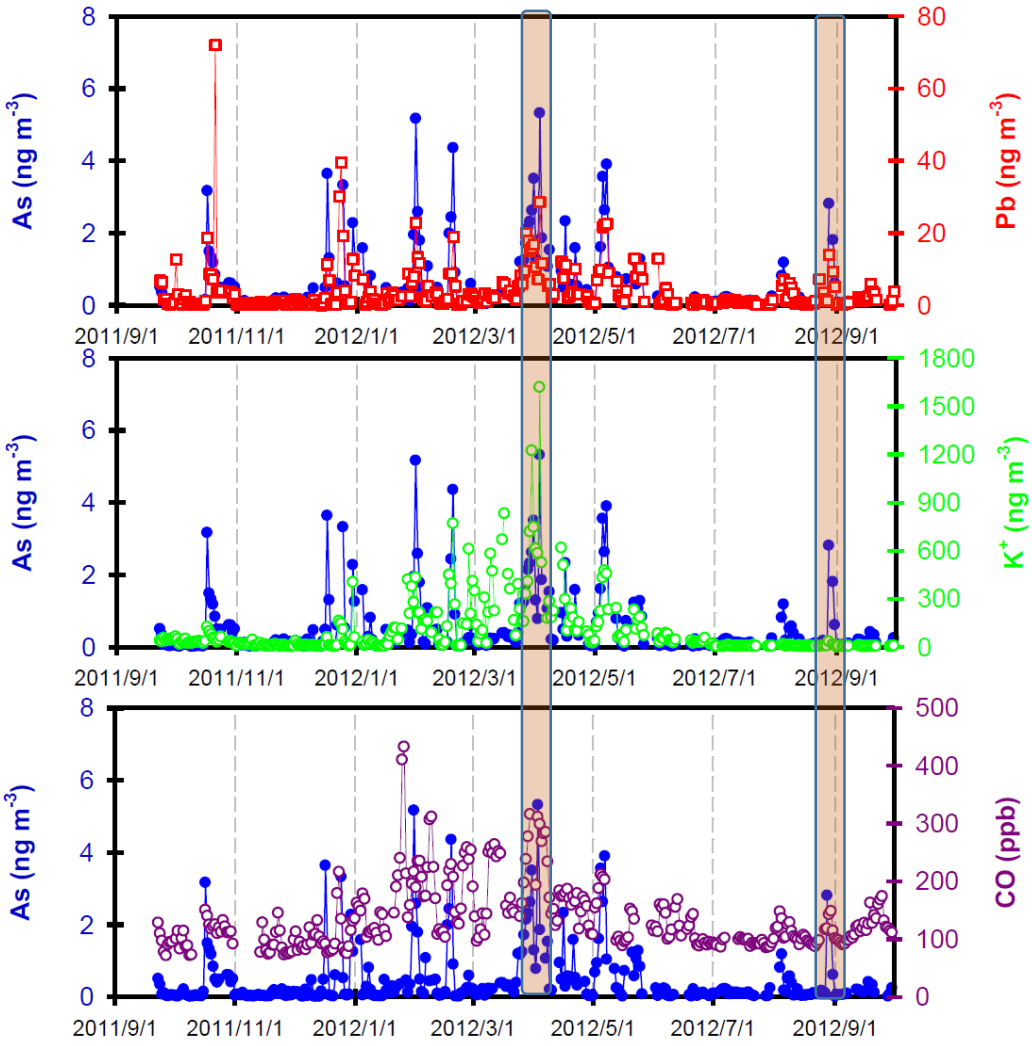


- In SE Asia, the BB activities showed strong seasonality with a maxima in March.
- In terms of S Asia, the fire spots accounted 20% for those in SE Asia.

Chemical Species in TSP

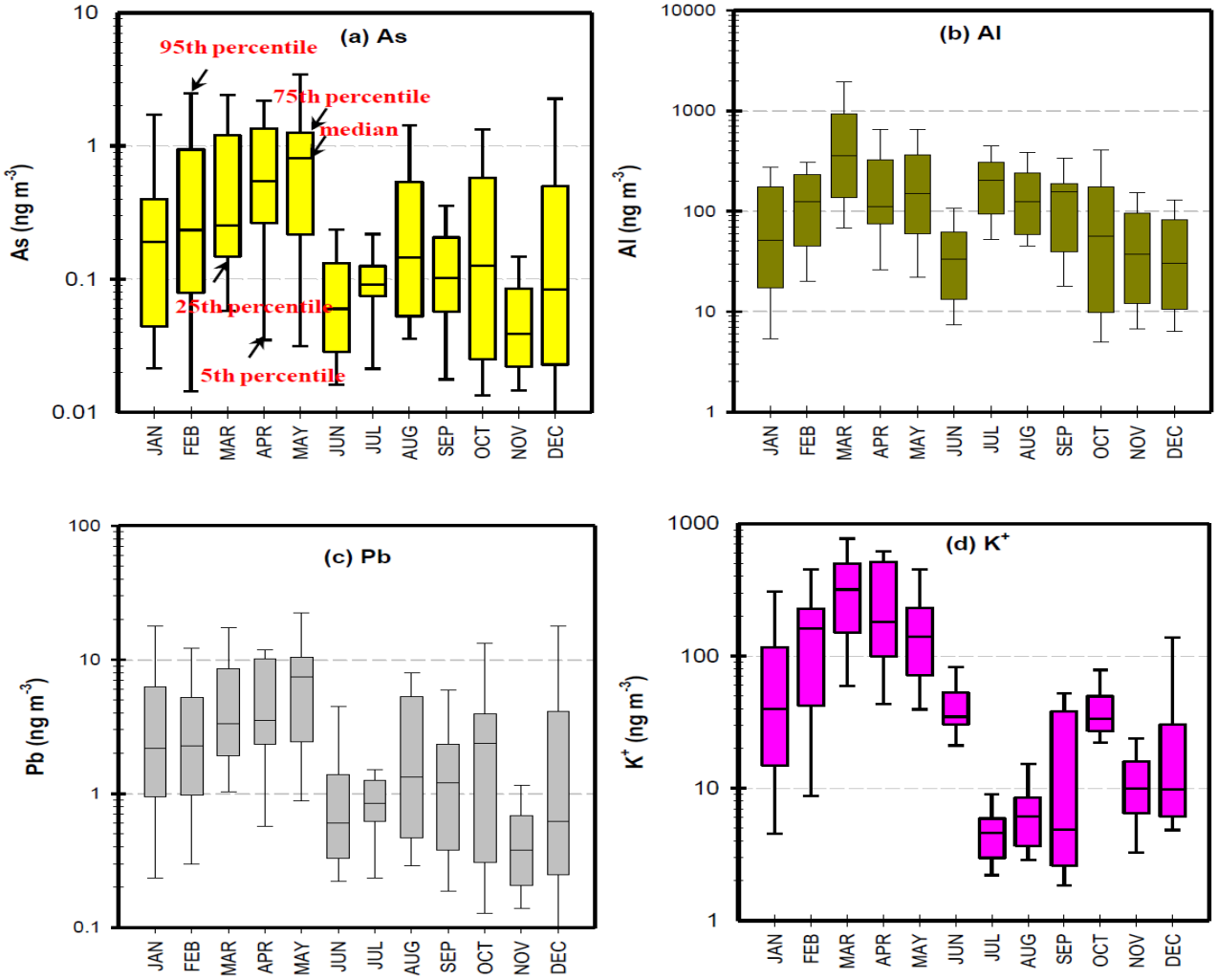


Daily As concentrations at Mount. Huhuan

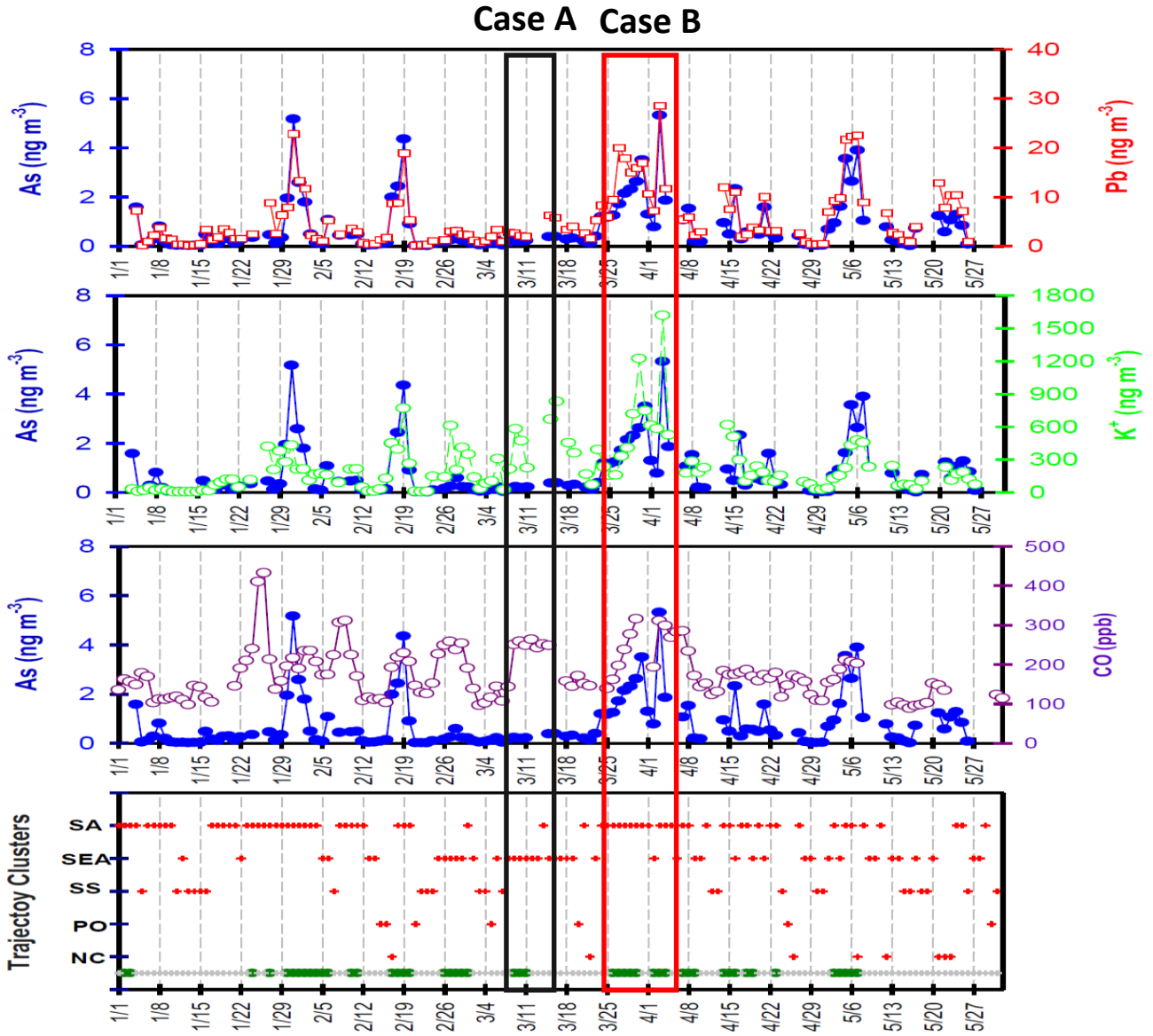


- On average, As concentration at Mount Hehuan was 0.5 ng/m^3.
- Lower As concentration (0.1 ng/m^3) in the summer was in agreement with that at Mauna Loa.

Seasonality of Chemical species

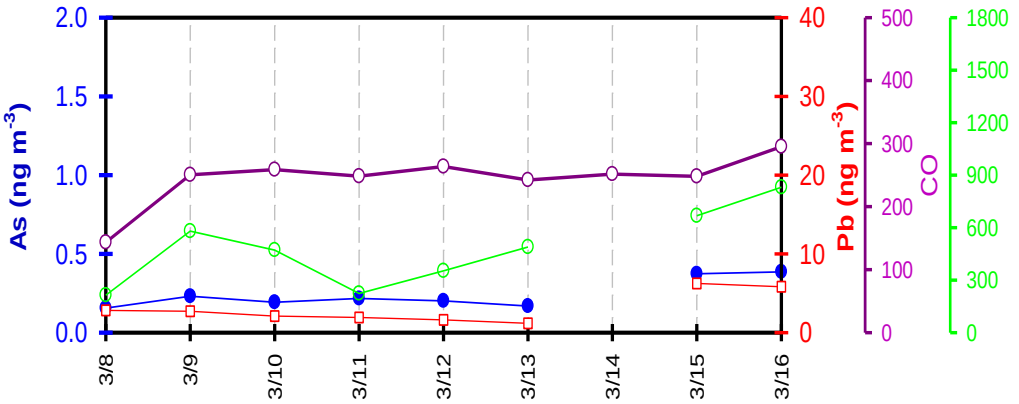


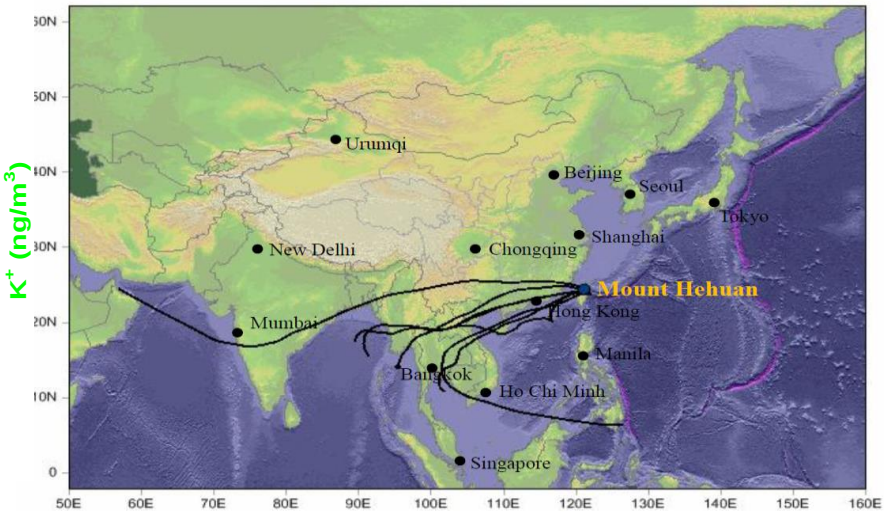
Daily As concentrations during the BB seasons

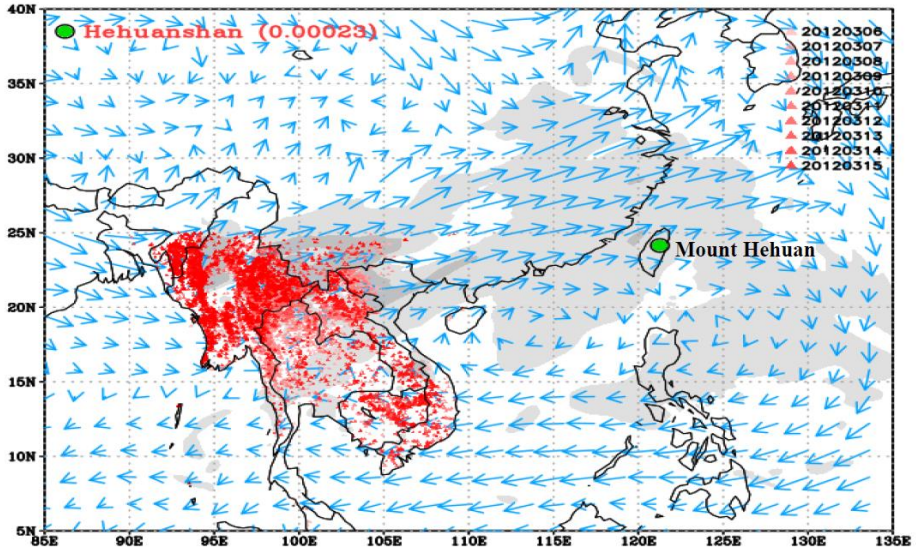


- We choose K^+ and CO concentrations of 109 ng/m^3 and 160 ppb as criterions for identifying the suspected BB events.

Case A (3/8-3/16)

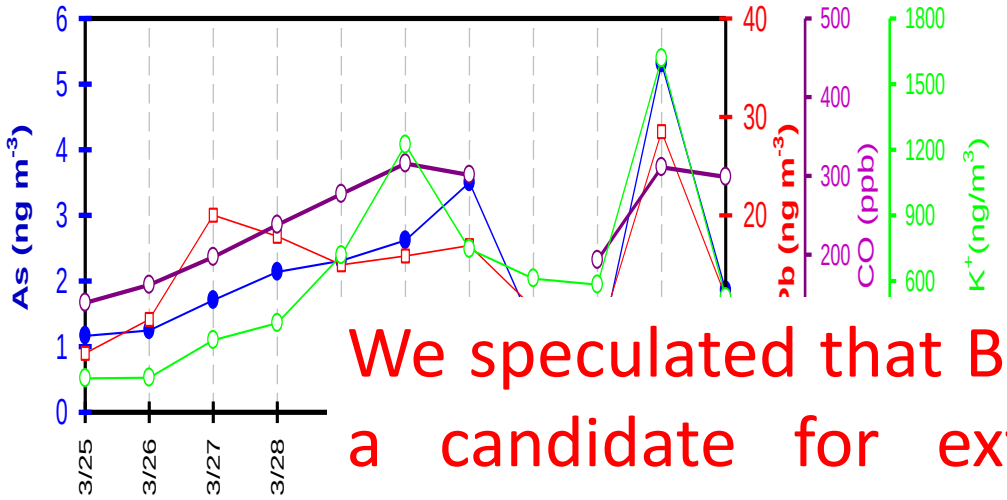


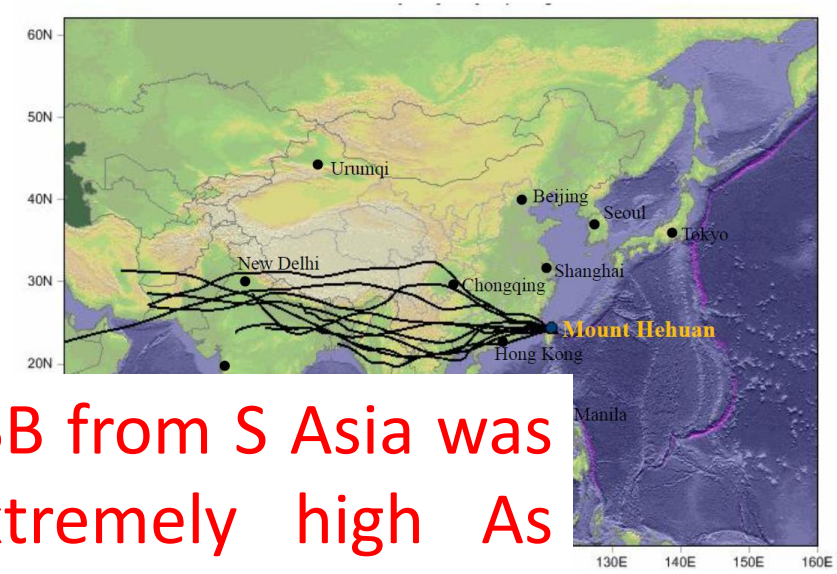




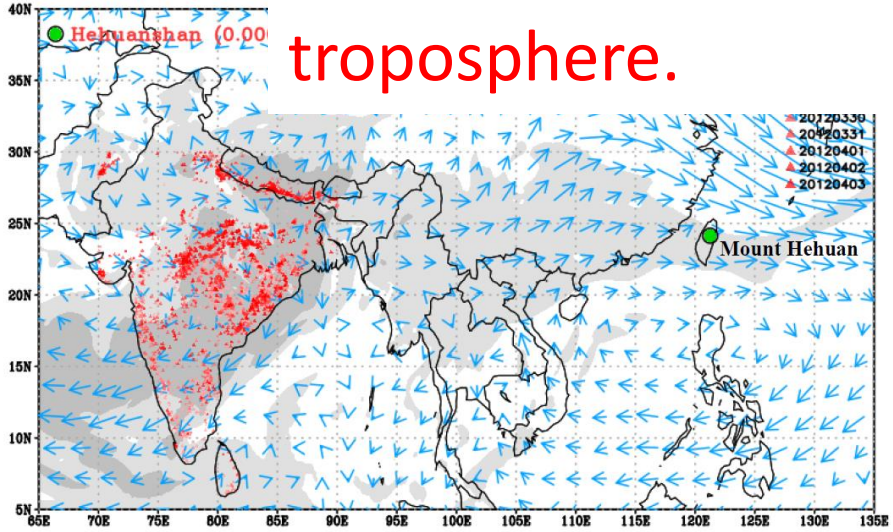
- In this case, air parcels mainly originated from Indo-China Peninsula, crossing over southern China and then arriving at Taiwan.
- No enhancements of As were found in this case.

Case B (3/25-4/3)





We speculated that BB from S Asia was a candidate for extremely high As concentrations over the subtropical troposphere.



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er northern
part of Indo-China Peninsula and southern China before subsiding at Taiwan.

- Significant increases of As concentrations were observed in this case.

Correlation between As and K^+

Typical types and formation mechanisms of haze in an Eastern Asia megacity, Shanghai

K. Huang[1,2], G. Zhuang[1], Y. Lin[1], J. S. Fu[2], Q. Wang[1], T. Liu[1], R. Zhang[1], Y. Jiang[1], C. Deng[1], Q. Fu[3], N. C. Hsu[4], and B. Cao[1]

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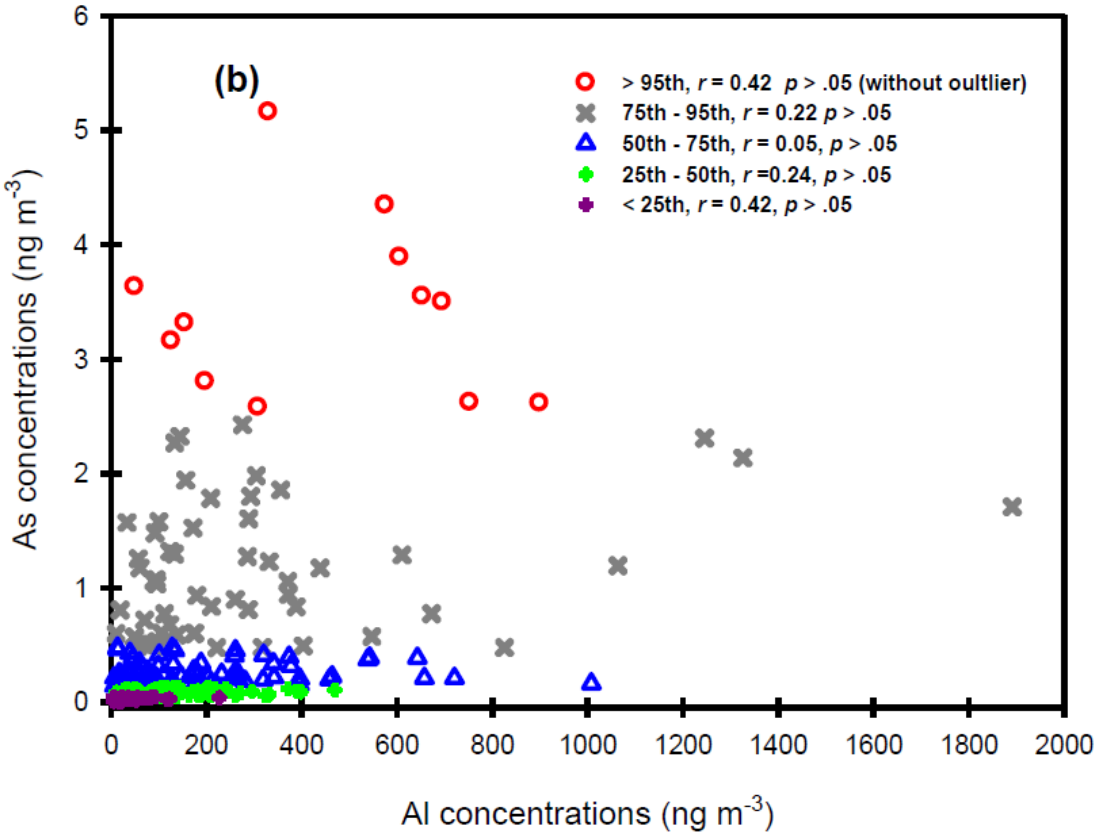
Revised: 6 November 2011 – Accepted: 16 November 2011 – Published: 2 January 2012

Abstract. An intensive aerosol and gases campaign was performed at Shanghai in the Yangtze River Delta region over Eastern China from late March to early June 2009. This study provided a complementary picture of typical haze types and the formation mechanisms in megacities over China by using a synergy of ground-based monitoring, satellite and between CO and $PM_{2.5}$ corroborated that organic aerosol dominated aerosol chemistry during biomass burning, and the high concentration and enrichment degree of arsenic (As) could be also partly derived from biomass burning. Aerosol optical profile observed by lidar demonstrated that aerosol was mainly constrained below the boundary layer and com-

A good correlation of As and K^+ was found in the BB events. Again, the compelling evidence supports the hypothesis.

Correlation between As and Al



Wind-eros[illegible]n particles

Why S Asia?

ARTICLES
PUBLISHED ONLINE: 15 NOVEMBER 2009 | DOI: 10.1038/NGEO685
nature geoscience

Anthropogenic influences on groundwater arsenic concentrations in Bangladesh

nature geoscience
ARTICLES
PUBLISHED ONLINE: 13 DECEMBER 2009 | DOI: 10.1038/NGEO723

Arsenic release from paddy soils during monsoon flooding

nature geoscience
PROGRESS ARTICLE
PUBLISHED ONLINE: 17 JANUARY 2010 | DOI: 10.1038/NGEO750

Vulnerability of deep groundwater in the Bengal Aquifer System to contamination by arsenic

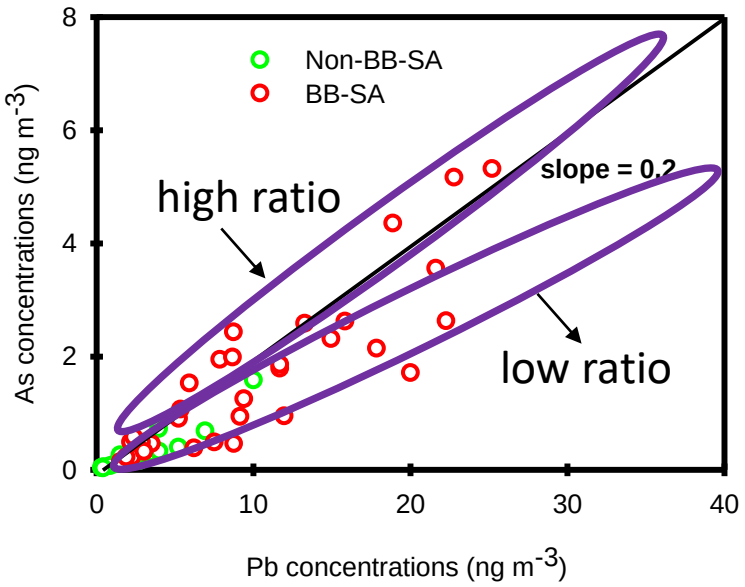
W. G. Burgess[1]*, M. A. Hoque[1], H. A. Michael[2], C. I. Voss[3], G. N. Breit[4] and K. M. Ahmed[5]

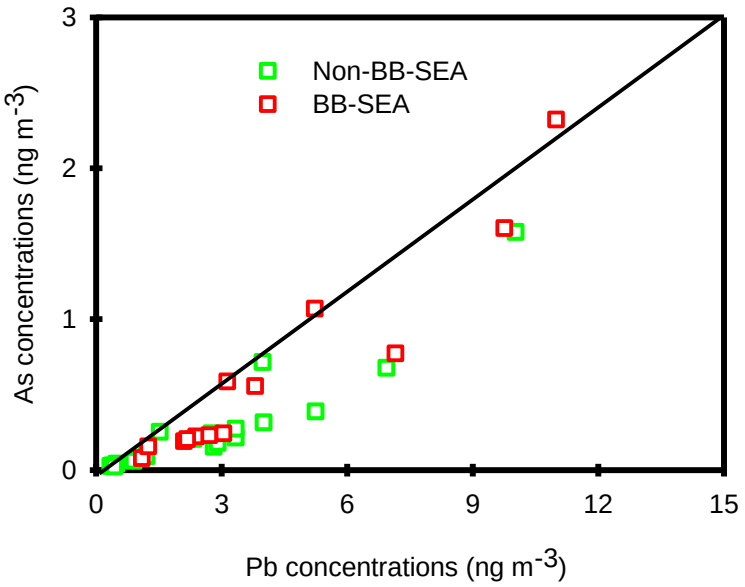
Lead Arsenate

- Lead arsenate **(LA, $[Pb_5OH(AsO4)_3]$)** was the most extensively used of the arsenical insecticide in the world. Although LA was officially banned as insecticide in 1990s in many developed country, but has not been banned in India nowadays.

- **LA has a As/Pb ratio of 0.22.**

As vs. Pb





Different emissions in northern India exhibited distinct ratios of As/Pb:

- Lead smelting: 0.002
- Natural soil, paved and unpaved roads: 0.02- 0.13
- Coal-combustion in stove: 0.0016
- Coal-combustion in power plant: 0.0026
- **Lead arsenate: 0.22**

As concentrations between non-BB and BB events in SA and SEA air clusters

Categories	As ($ng\ m^{-3}$)	Pb ($ng\ m^{-3}$)	K^+ ($ng\ m^{-3}$)	CO (ppb)
SA air cluster				
Non-BB				
Max	3.5	16.9	831	432
Min	0.05	0.6	15	102
Mean	0.6	4.5	207	188
Std.	0.7	3.8	173	86
BB				
Max	5.3	28.5	1617	316
Min	0.13	1.6	71	156
Mean	1.6	10.2	404	217
Std.	1.4	7.3	336	42
Differences[1]	1.0	5.7	197	29
SEA air cluster				
Non-BB				
Max	1.6	10.0	452	282
Min	0.02	0.3	4	95
Mean	0.4	2.9	151	148
Std.	0.4	2.5	141	45
BB				
Max	2.3	11.0	609	259
Min	0.08	1.1	139	170
Mean	0.6	4.2	328	212
Std.	0.7	3.2	178	39
Differences	0.2	1.3	177	64

$\Delta K^+/\Delta CO \sim 0.0043$; $\Delta AS/\Delta CO \sim 10^{-5}$

$\Delta K^+/\Delta CO \sim 0.0018$; $\Delta AS/\Delta CO \sim 10^{-6}$

1. Difference for each species are calculated by the mean values in BB and non-BB events.

- The contribution of BB over S Asia was roughly estimated to be 1.0 ng/m3 which accounted 63% for total As concentrations in the springtime.
- We roughly estimated that the emission rate of As from S Asian BB activities was 17 Mg/year, which was much lower than that (~18.8 Gg/year) of globally anthropogenic As emissions.

Conclusions

- Previously, high As concentrations over free troposphere in Northern Pacific region was always considered as contribution of industrial emissions (Perry et al., 1990; Wai et al., 2016). From our study, we propose a new concept for a potential source of high As over the subtropical free troposphere, that is, BB activities over S Asia might be an important source of airborne arsenic

Thank you for your attention

Scatter plots of As vs. K^+ and As vs. Al in different As bins

Motivation

- To investigate the influence of Asian continental outflow on atmospheric chemistry over subtropical free troposphere, aerosol samples and gaseous pollutants (CO and O_3) were continuously collected and monitored at Mount. Hehuan, Taiwan from September 2012 to September 2013.
- Arsenic (As), has long been considered as a toxic element, having adverse effects on human. Extremely high As concentrations were observed at Mount Huhuan during the springtime.
- ***What are the potential sources of extremely high As?***