

Yale



耶鲁大学-南京信息工程大学大气环境中心

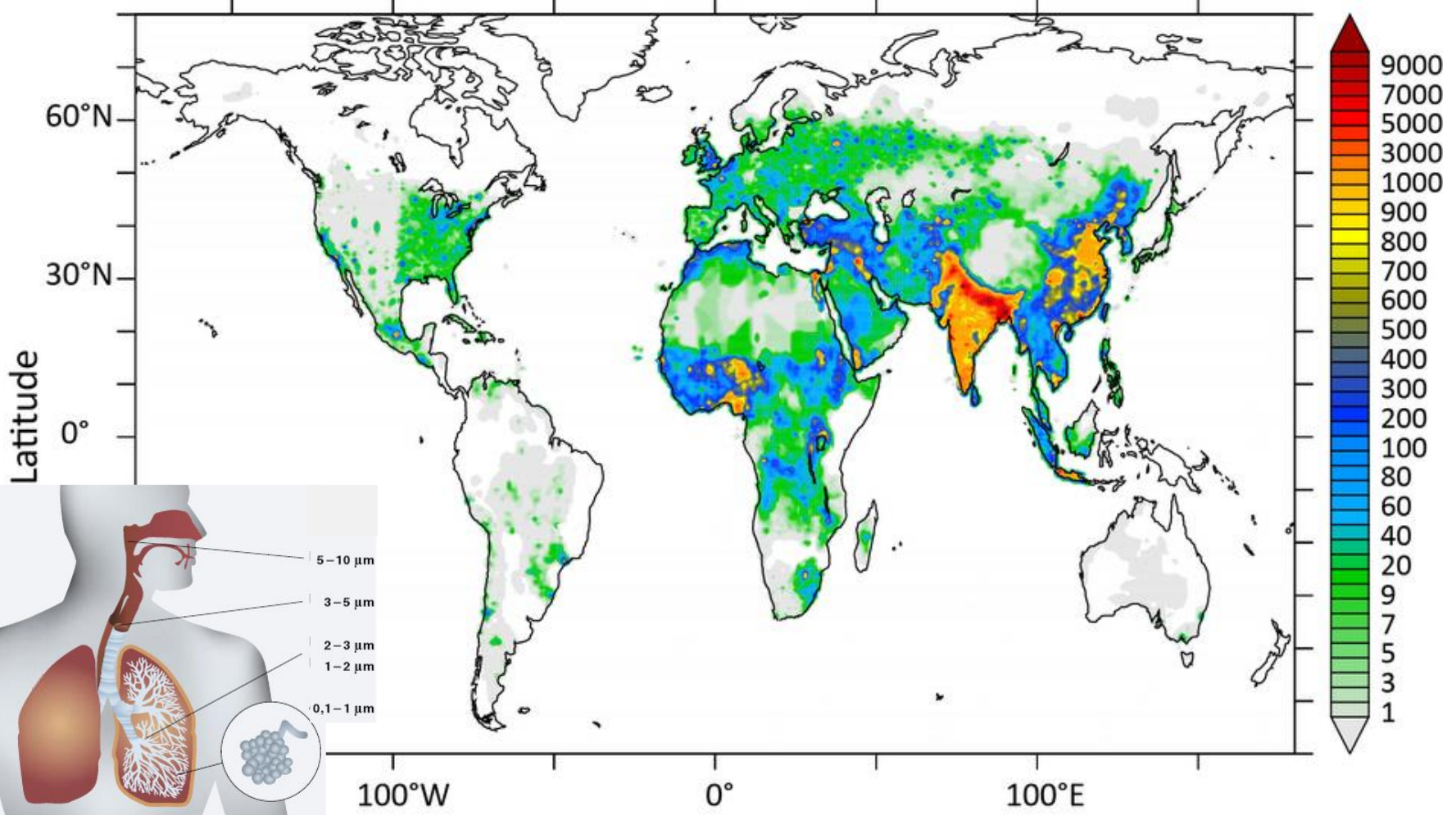
Yale-NUIST Center on Atmospheric Environment

基于同位素研究气溶胶的来源 和环境过程

章炎麟

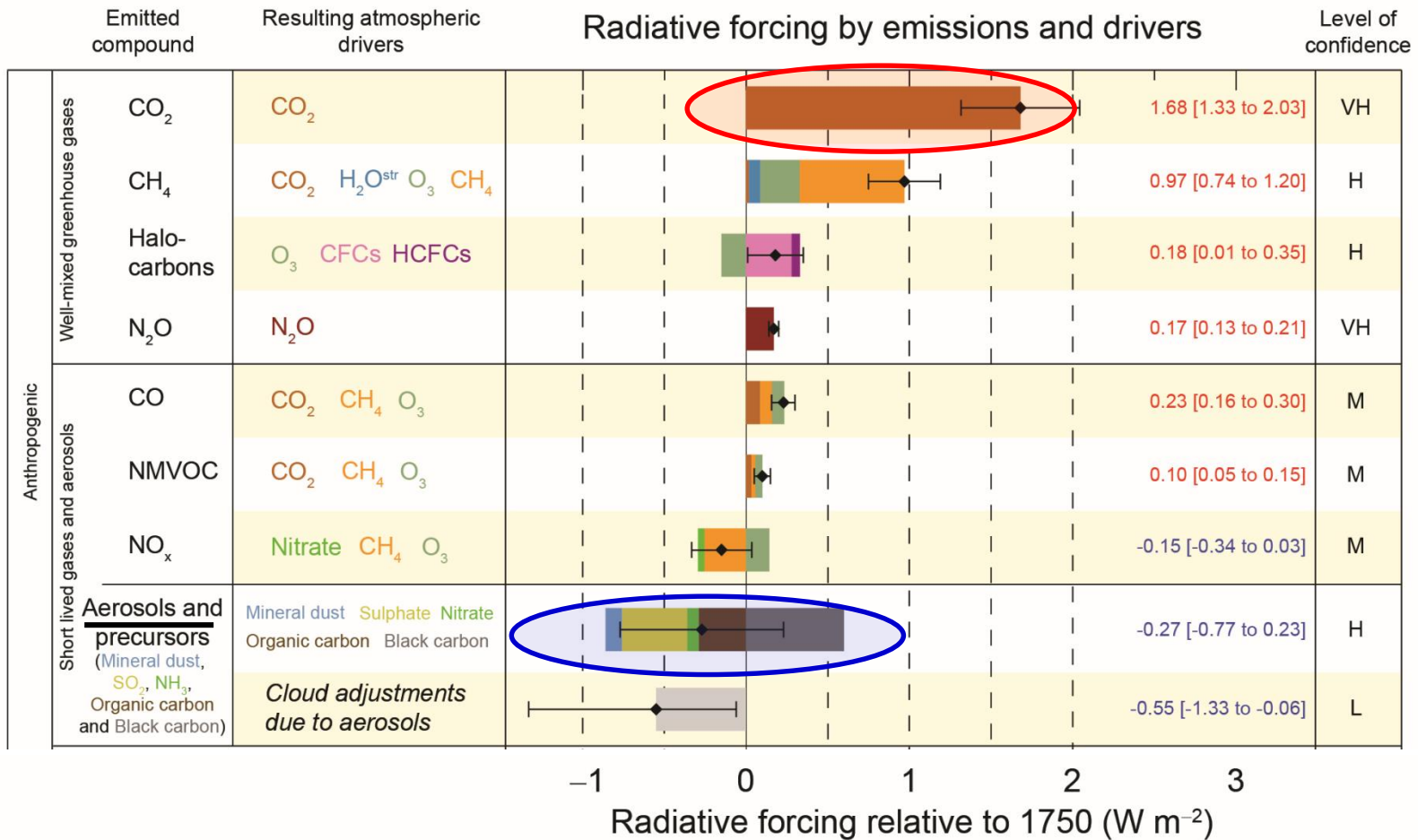
南京信息工程大学

室外空气污染造成全球每年330万人死亡



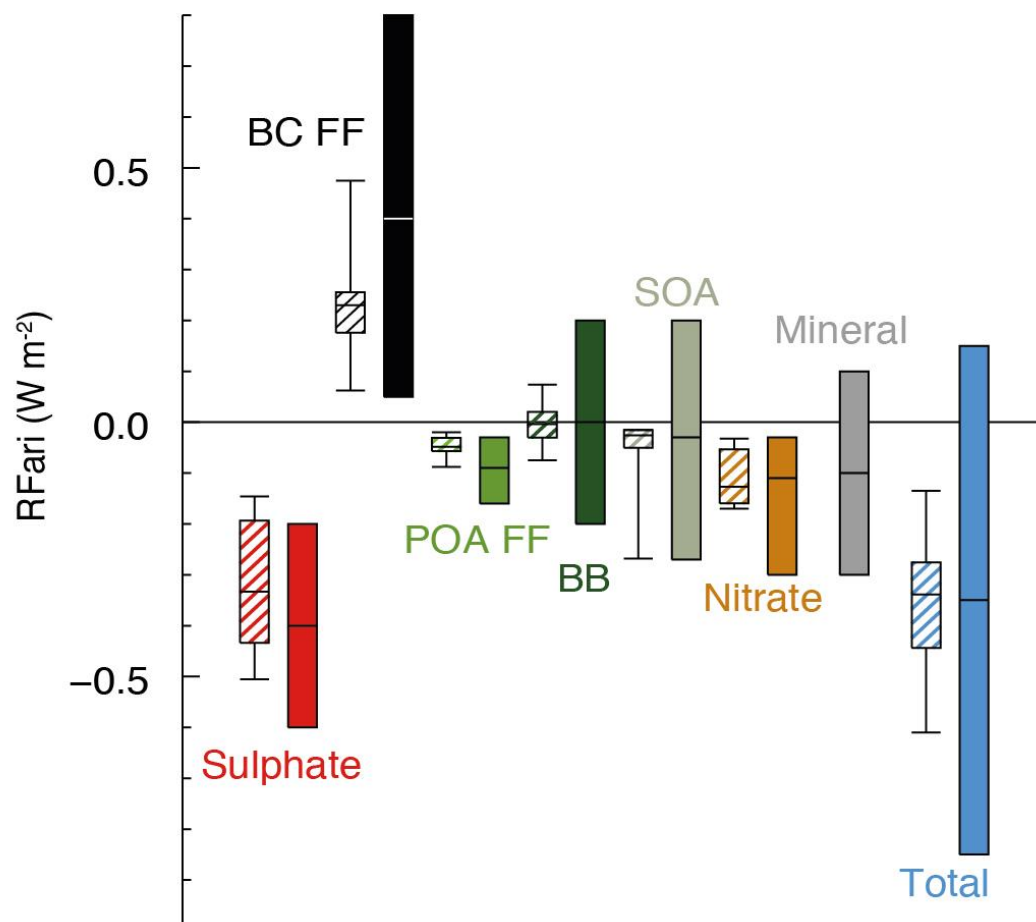
Mortality linked to outdoor air pollution in 2010. Lelieveld et al., **Nature**, 2015

Climate effects



Intergovernmental Panel on Climate Change (IPCC), 2014

辐射强迫因子



硝酸盐

硫酸盐

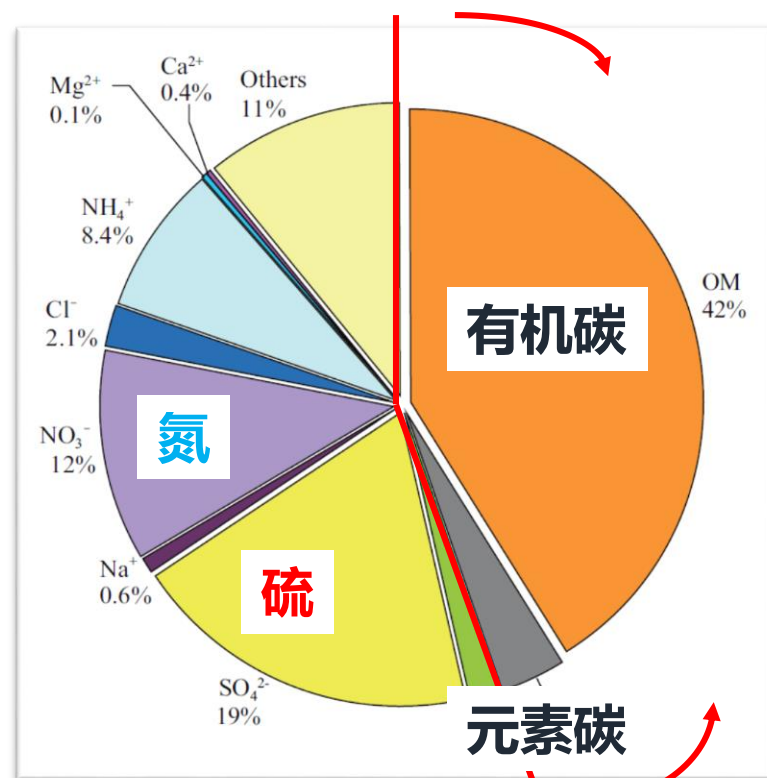
有机碳

黑碳/元素碳

IPCC报告

同位素在气溶胶领域的典型应用

硫、氮、碳



典型PM_{2.5}的化学组成

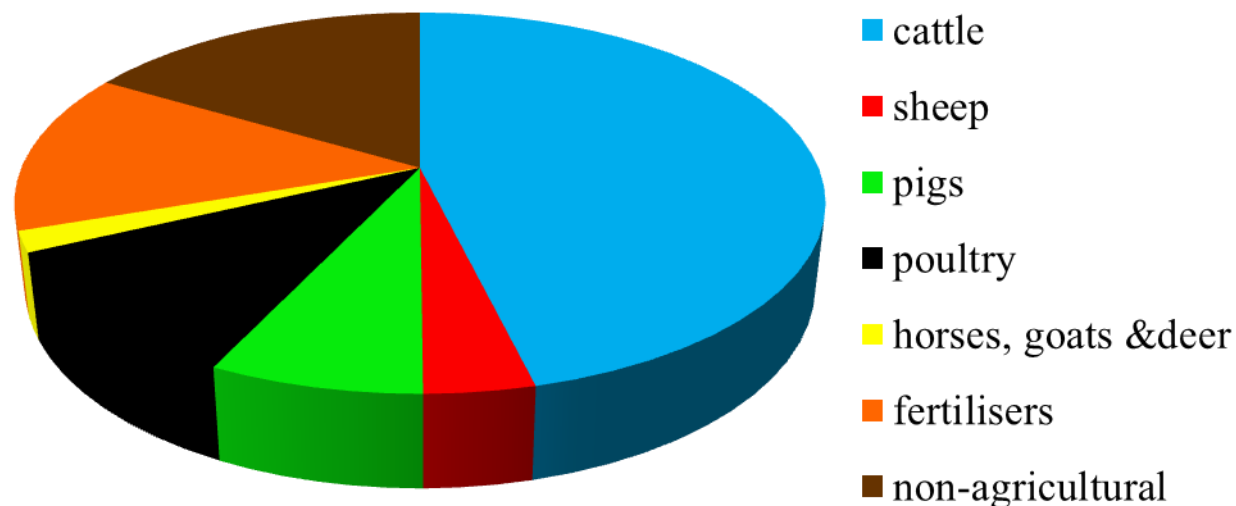
同位素：解析来源与过程的重要工具

报告内容

1. NH_x 的来源与过程
2. 碳气溶胶的来源与过程
3. 硝酸盐
4. 硫酸盐

问题背景

国内对非农业NH₃的研究方兴未艾



在英国，非农业源氨占到全国氨总排放量的**15%**

- 特点：** 1. 来源复杂； 2. 总量虽小，但却集中于城市区域； 3. 氨难以长程传输，易于在NO_x/SO₂-rich的城市生成PM

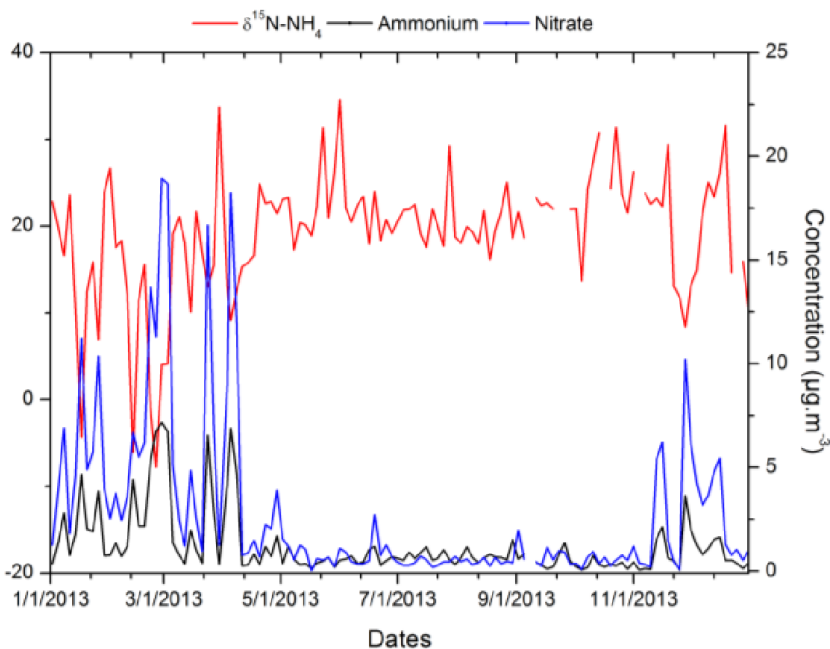
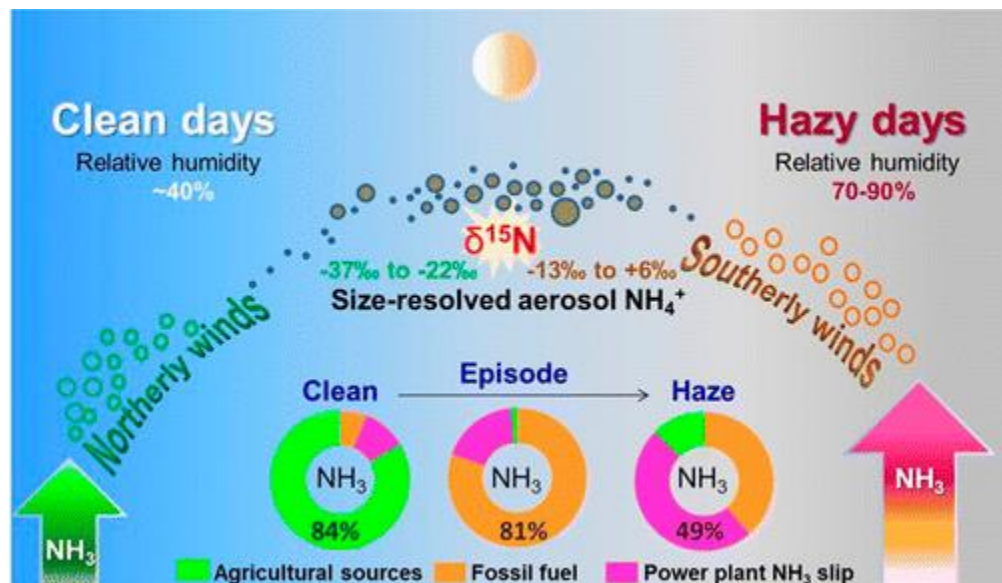


Figure 1. Temporal evolution of nitrate and ammonium concentrations, anti-correlated with $\delta^{15}\text{N}-\text{NH}_4$ in Grenoble (France).

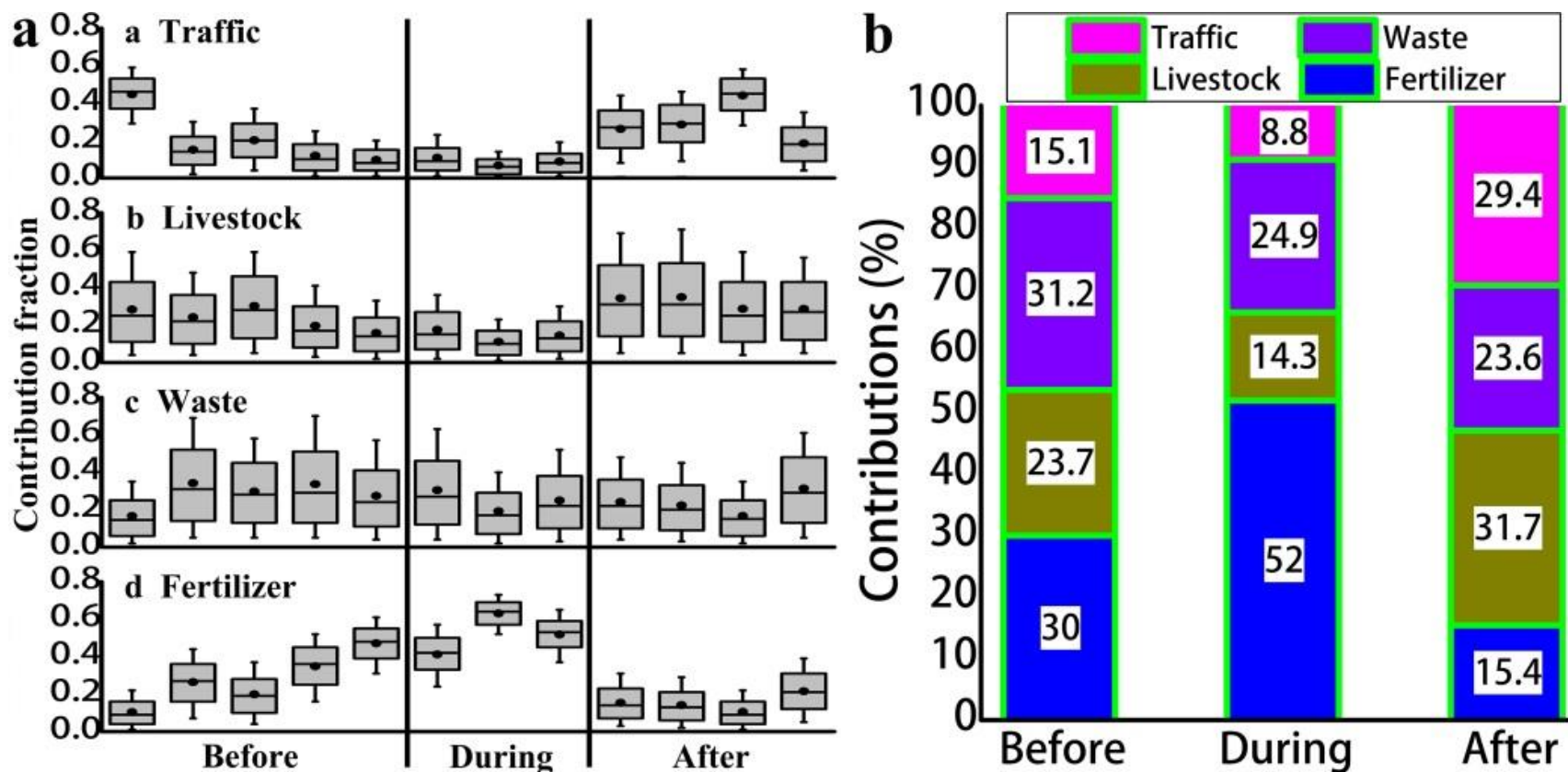


Pan et al., 2016

法国：高的 NH_4^+ ，低的同位素，与农业活动有关

中国：高的 NH_4^+ ，高的同位素，与化石燃料燃烧有关

将氮的同位素源谱应用于北京APEC前中后环境NH₃的源解析

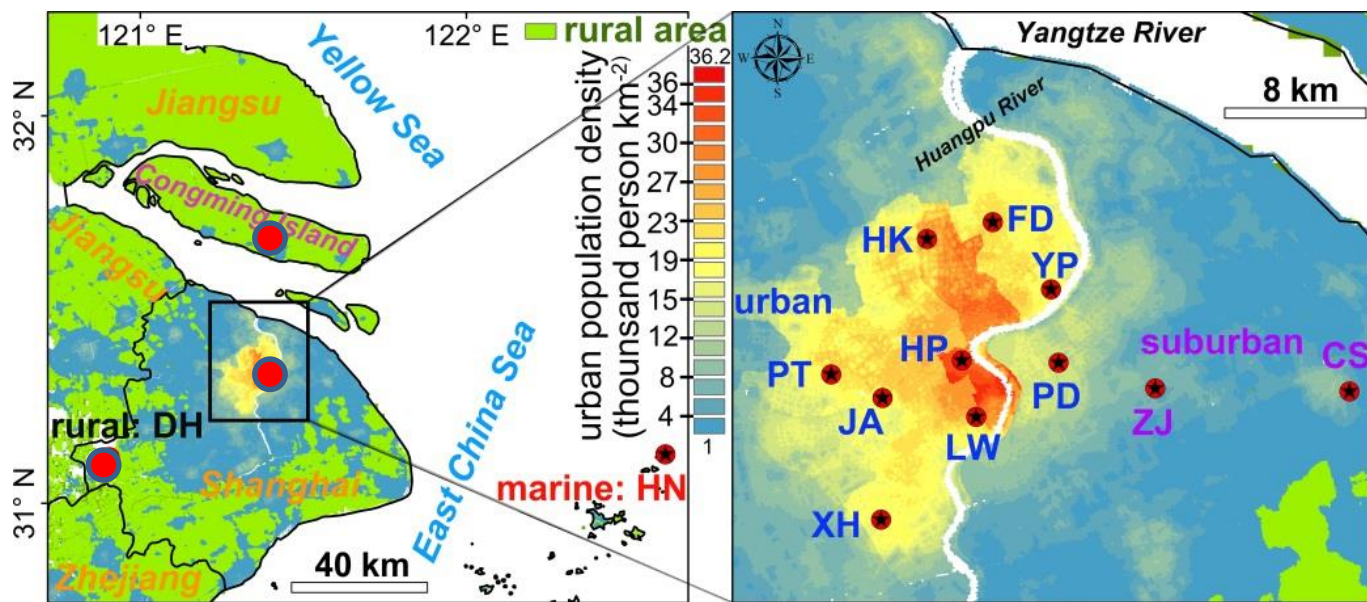


研究方法

上海NH₃监测网络

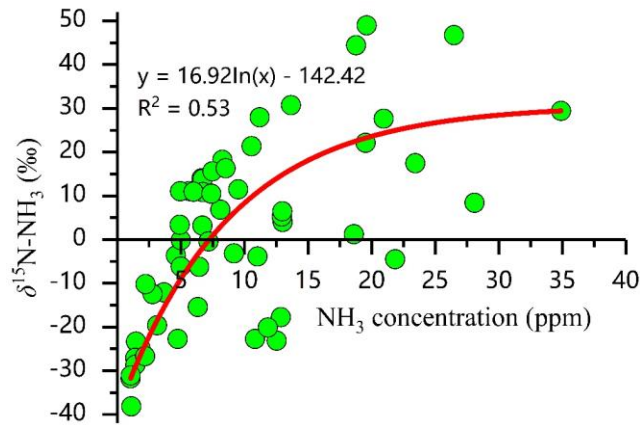
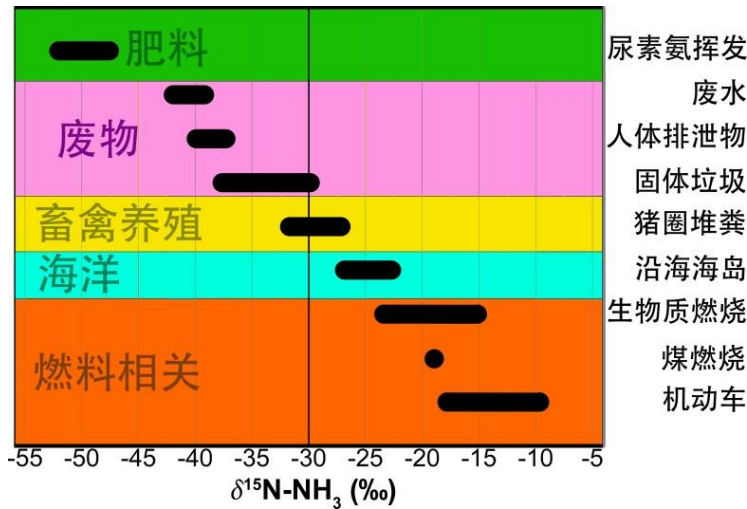
- 时间** 跨越农村、城市、背景（东平公园）条带的3大超级站
- 空间** 覆盖上海全市的10个密集Ogawa + ALPHA被动监测网络
- 维度** 同时包含化学组分和同位素分析在内的全方位监测体系

2014/2015夏季

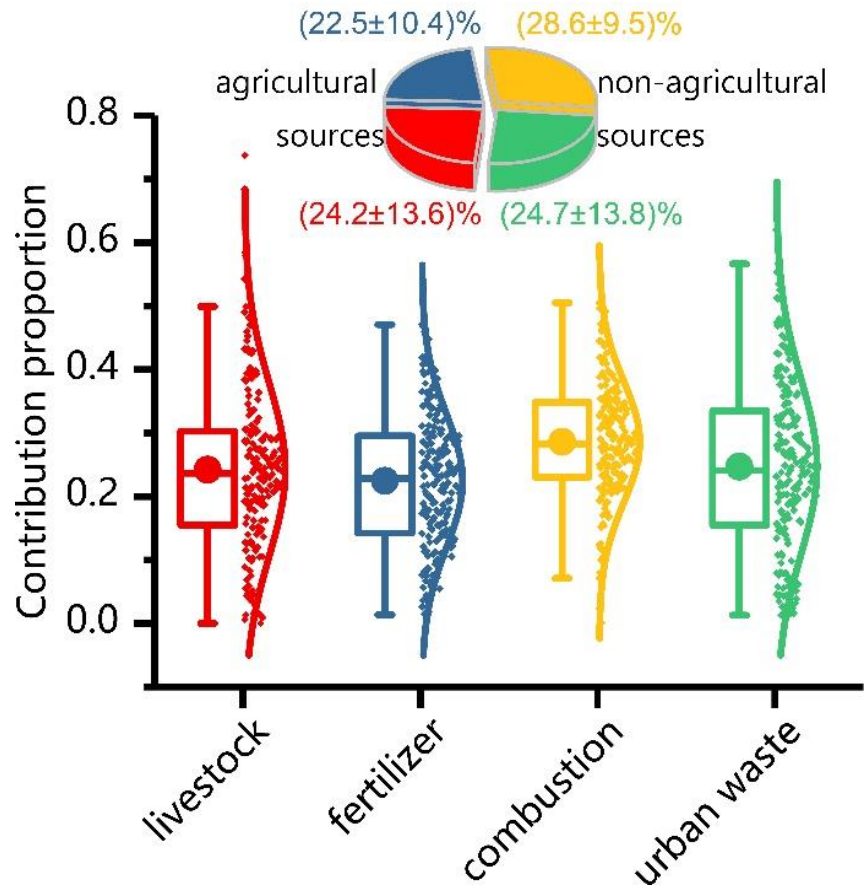


分析讨论-同位素

上海城市环境氨的源解析



机动车源谱



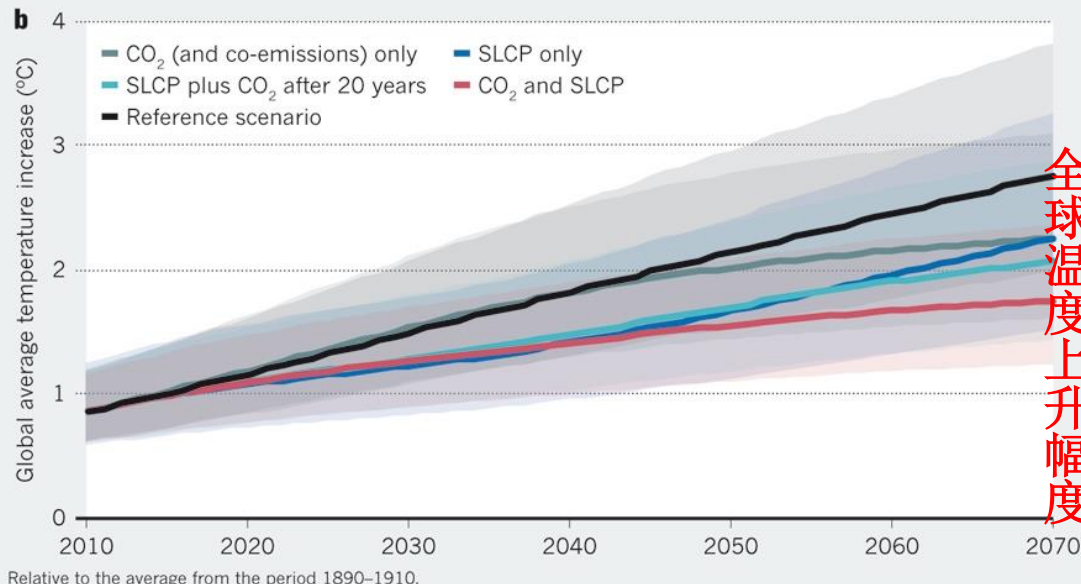
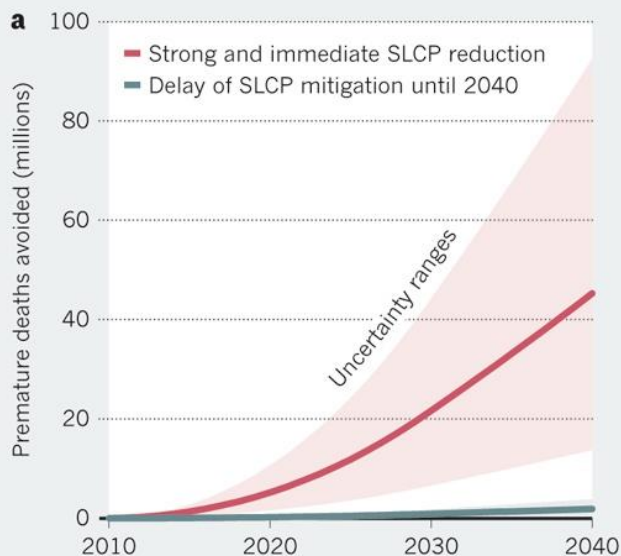
source: Chang, et al., unpublished.

空气和气候污染物“双赢”的减排策略

CO₂和SLCPs（如黑碳）的同时减排

CLEAN AIR

More than 40 million deaths from respiratory and cardiovascular diseases could be prevented by 2030 by halving the concentration of short-lived climate-forcing pollutants (SLCPs) in the atmosphere immediately (a). Joint approaches to mitigating SLCPs and carbon dioxide are more effective than separate measures in limiting global average temperature rise⁴ (b).



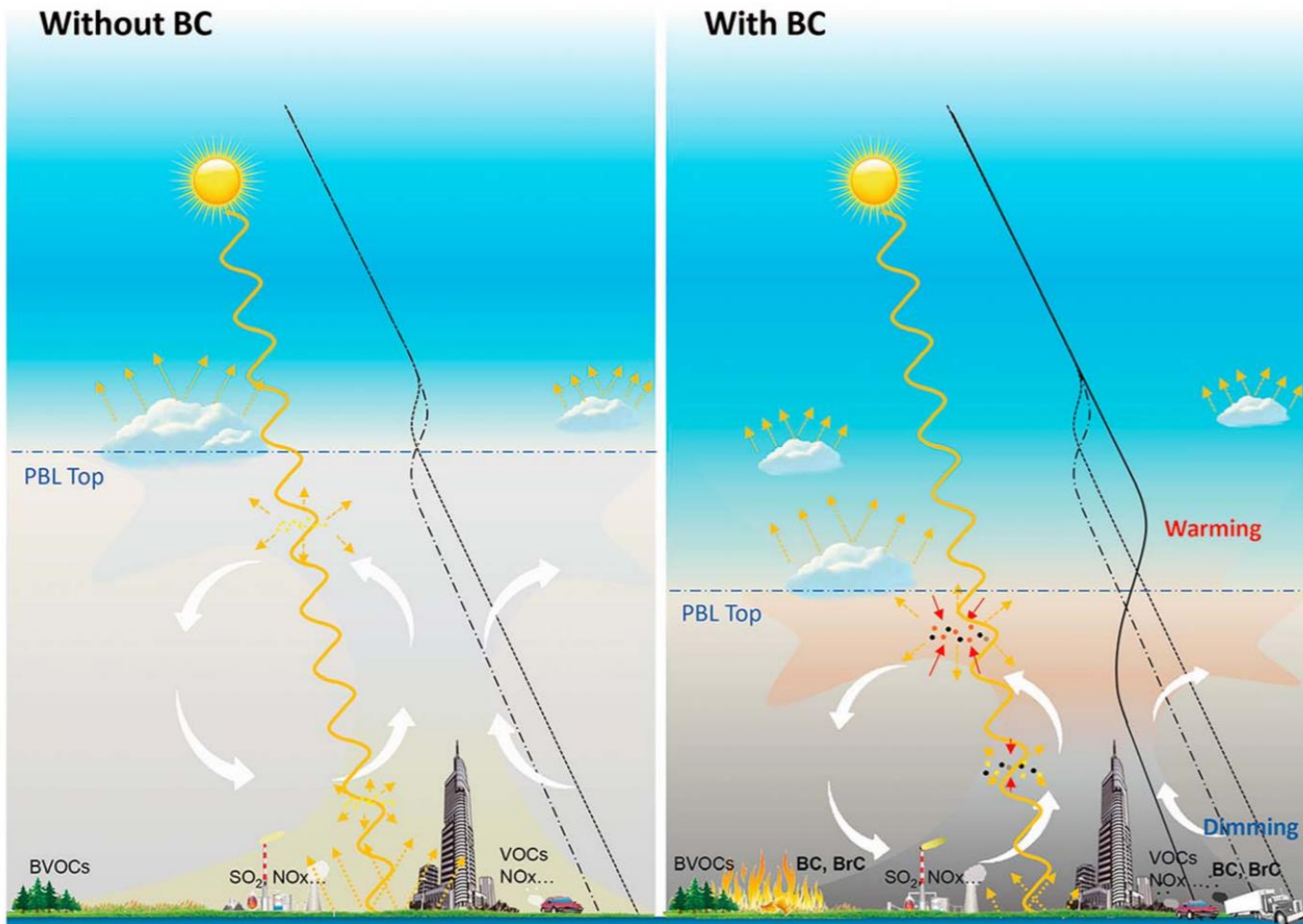
过早死

全球温度上升幅度

From nature

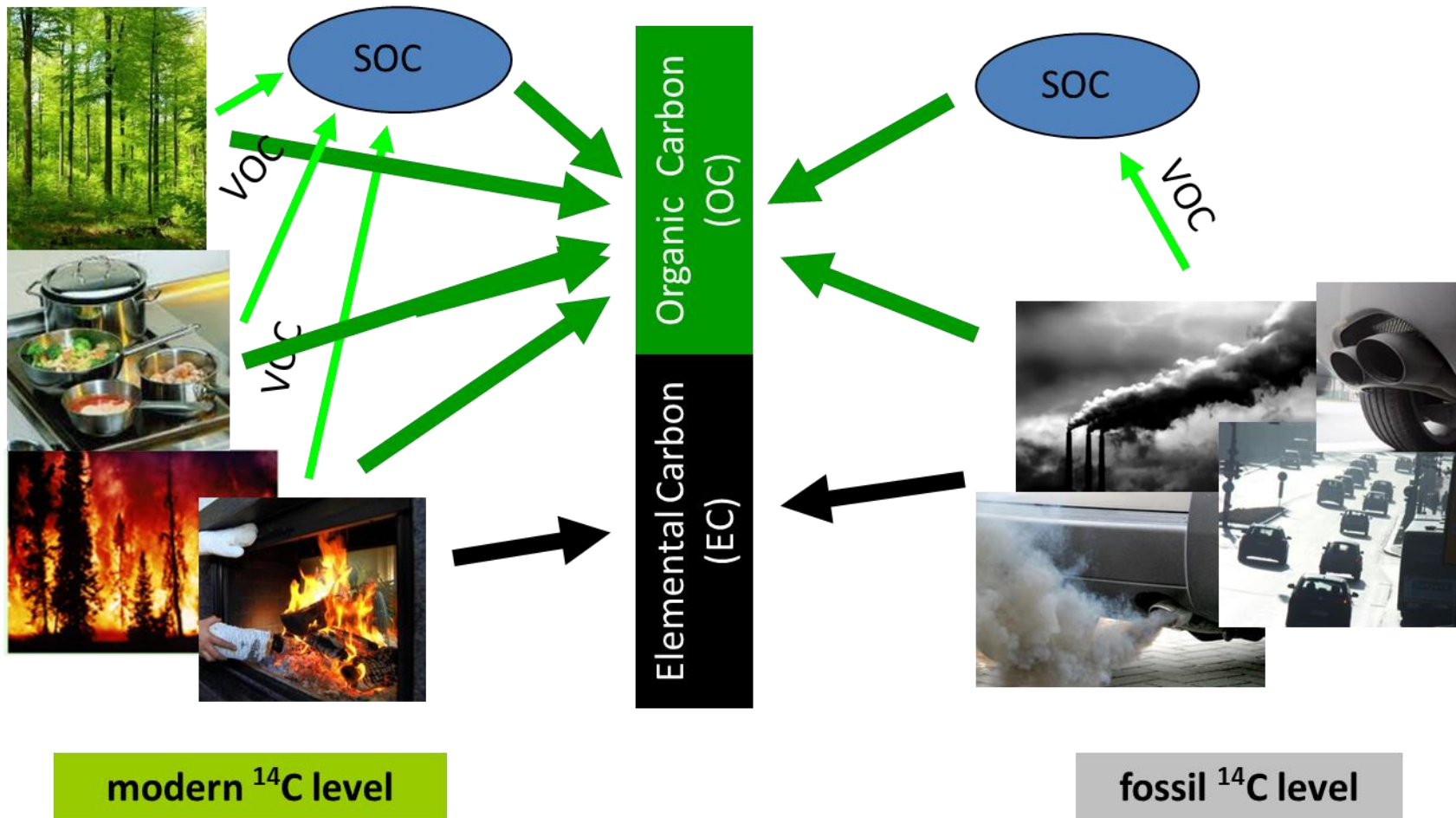
黑碳的减排是关键，效应最直接！

黑碳的穹顶效应(dome effect)



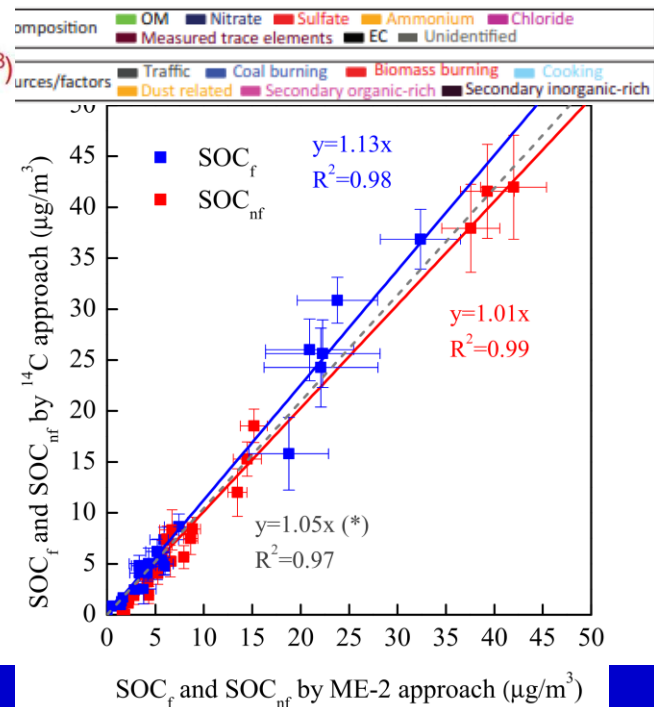
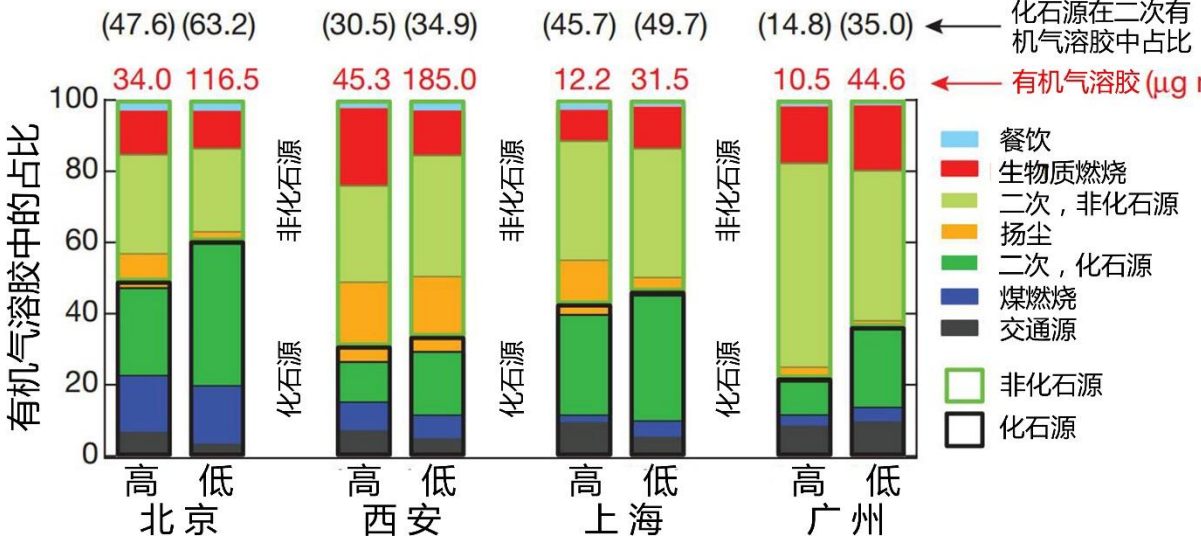
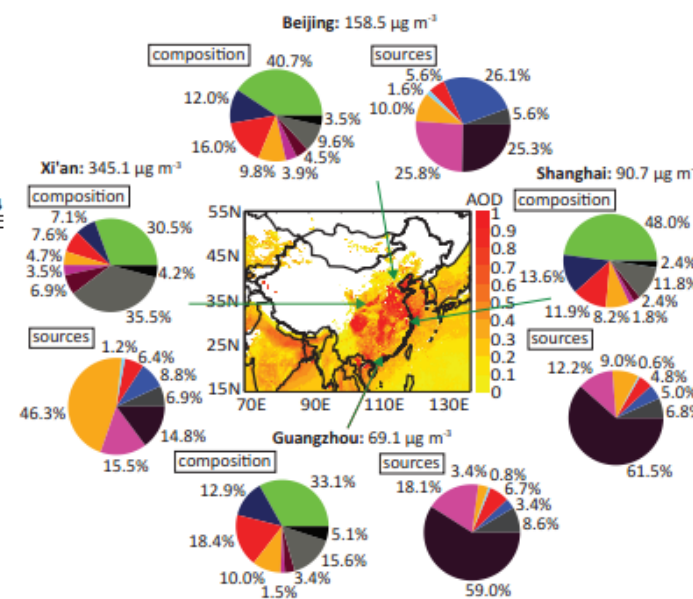
Ding et al., 2016

细颗粒物中的 ^{14}C

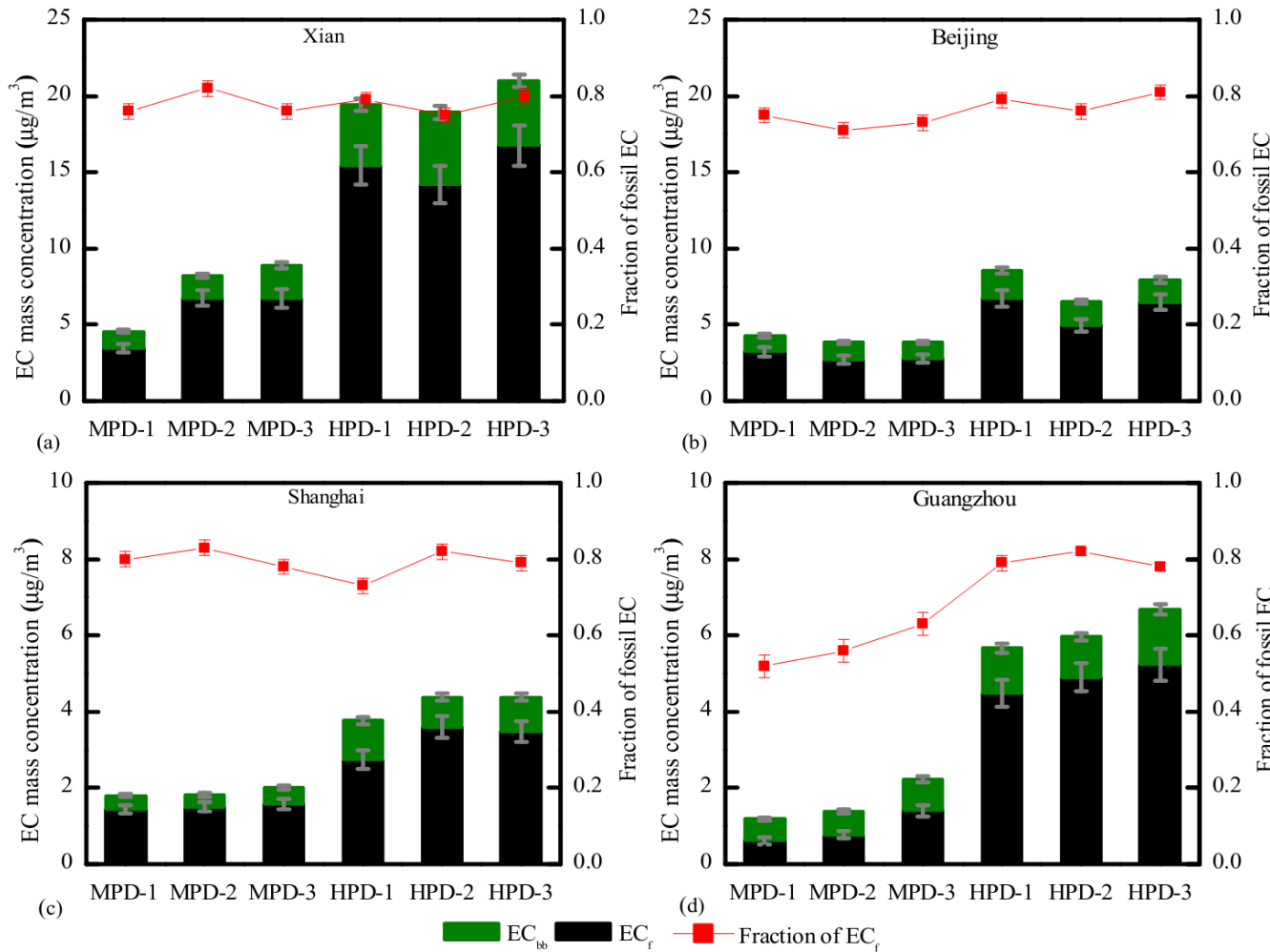


High secondary aerosol contribution to particulate pollution during haze events in China

Ru-Jin Huang^{1,2*}, Yanlin Zhang^{3,4}, Carlo Bozzetti¹, Kin-Fai Ho⁵, Jun-Ji Cao², Yongming Han², Kaspar R. Daellenbach¹, Jay G. Slowik¹, Stephen M. Platt¹, Francesco Canonaco¹, Peter Zotter¹, Robert Wolf¹, Simone M. Pieber¹, Emily A. Brunts¹, Monica Crippa^{1,†}, Giancarlo Ciarelli¹, Andrea Piazzalunga⁶, Margit Schwikowski^{3,4}, Gülcin Abbaszade⁷, Jürgen Schnelle-Kreis⁷, Ralf Zimmermann^{7,8}, Zhisheng An², Sönke Szidat³, Urs Baltensperger¹, Imad El Haddad^{1*} & André S. H. Prévôt¹



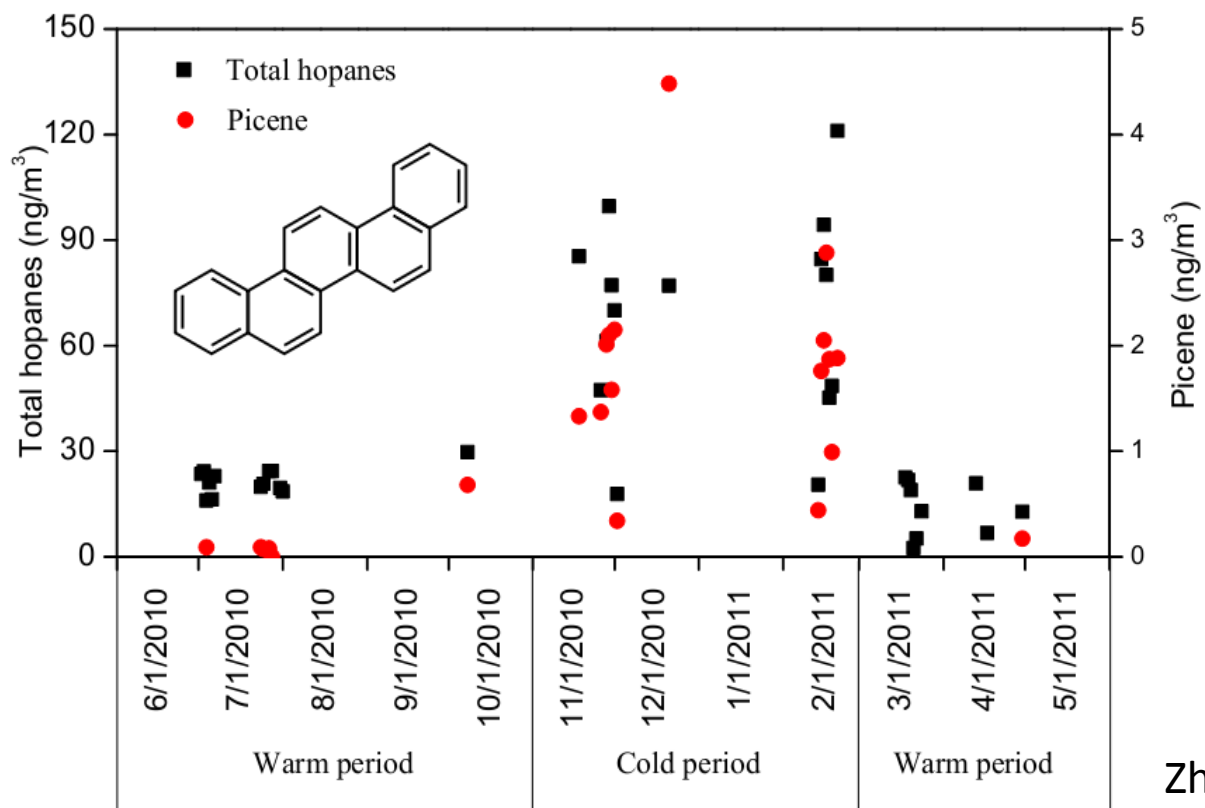
化石源 vs. 非化石源



MPD:
Moderately Polluted Days
HPD:
Heavily Polluted Days

如何进一步区分两类化石源：机动车/燃煤

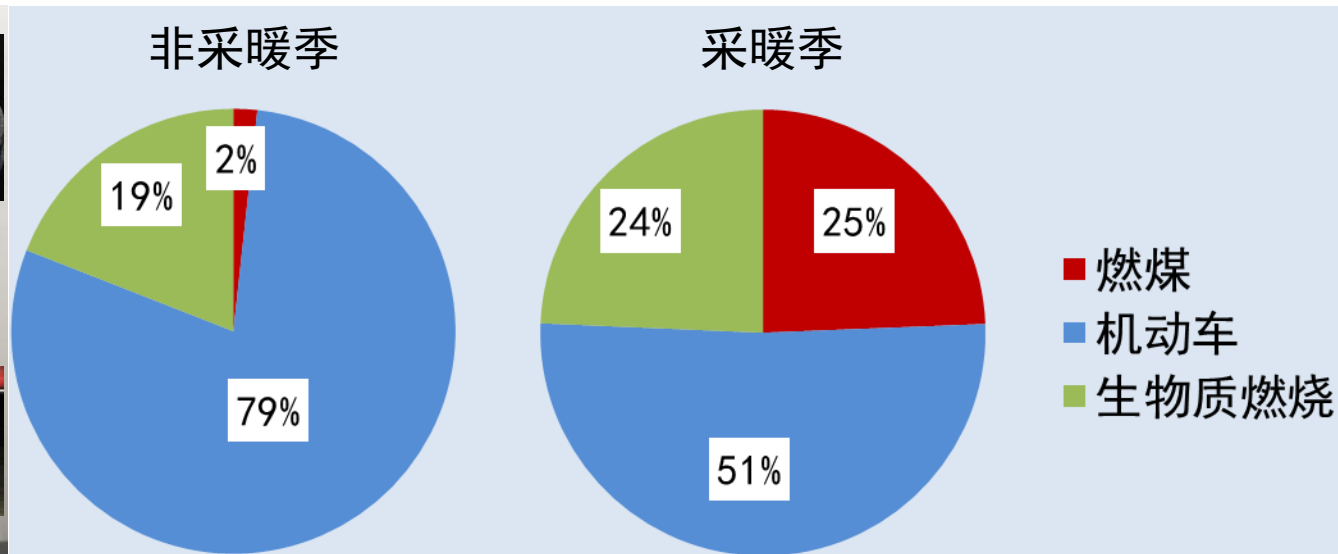
燃煤燃烧的生物标志物：picene (苝)



Zhang et al., ES&T, 2015

城市黑碳气溶胶来源

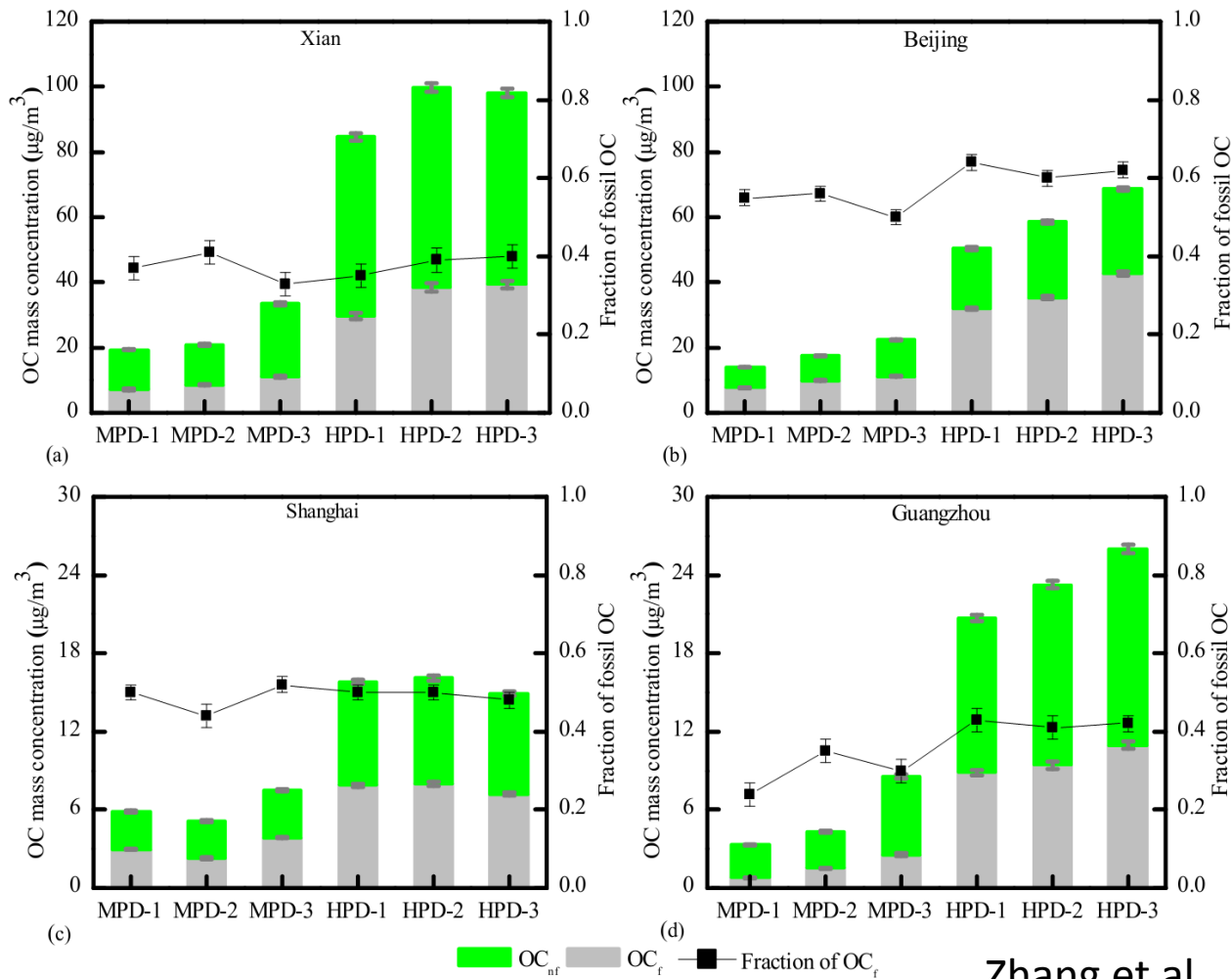
北京



方法：
放射性碳、有机分子标志物（hopanes和picene等）

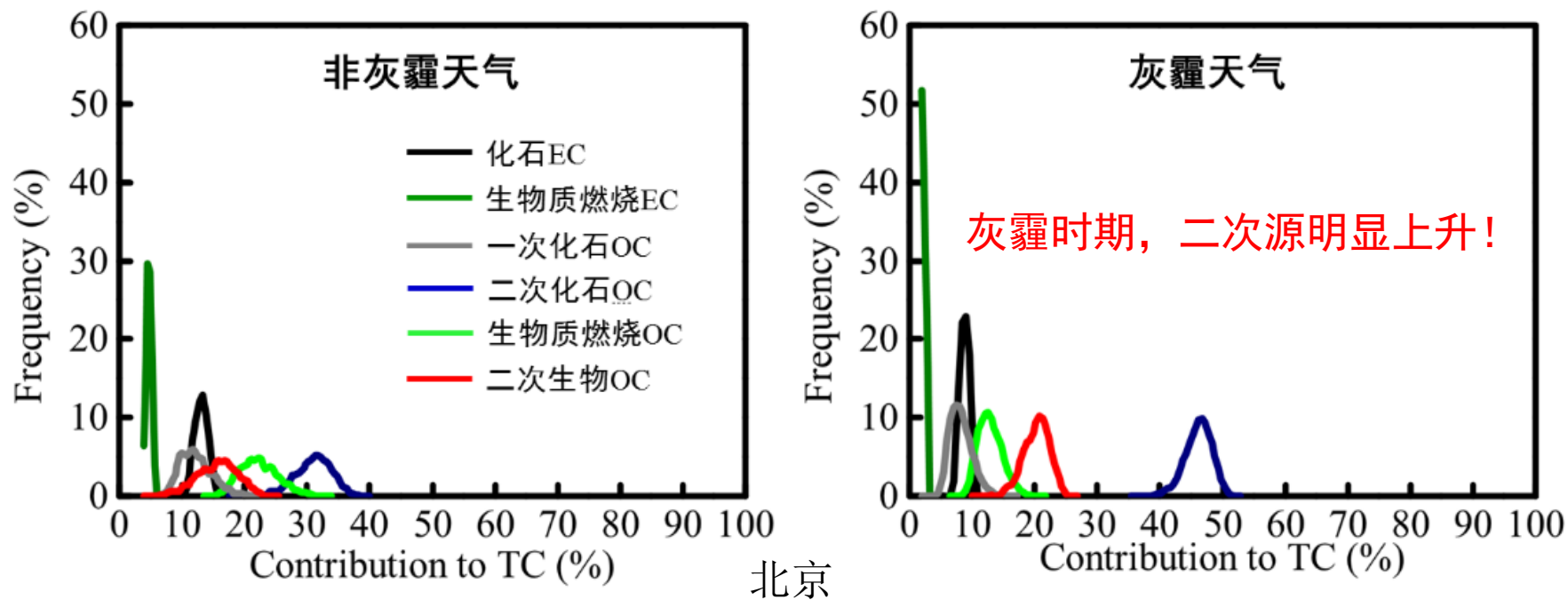
机动车是北京黑碳最主要来源；在采暖季燃煤贡献迅速增加，而生物质燃烧也稍有增长。

化石源 vs. 非化石源



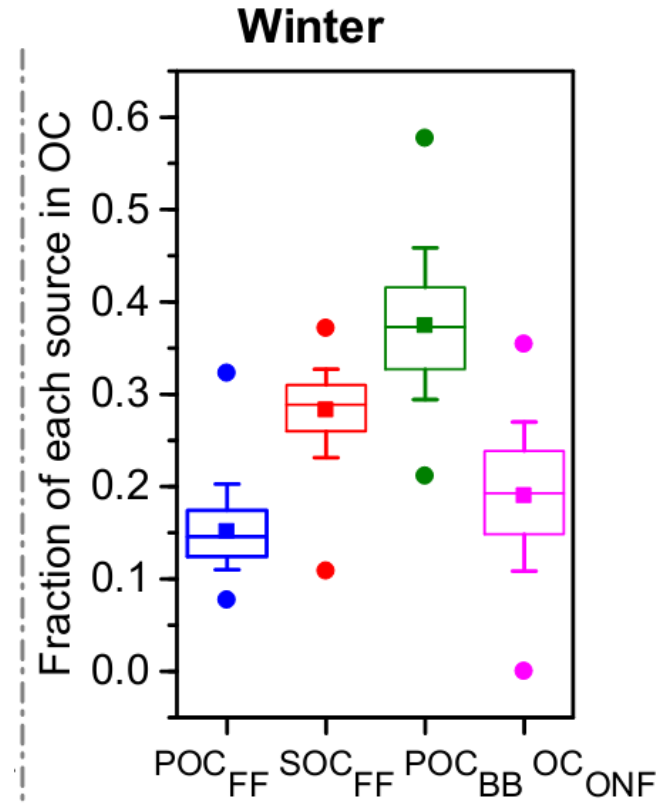
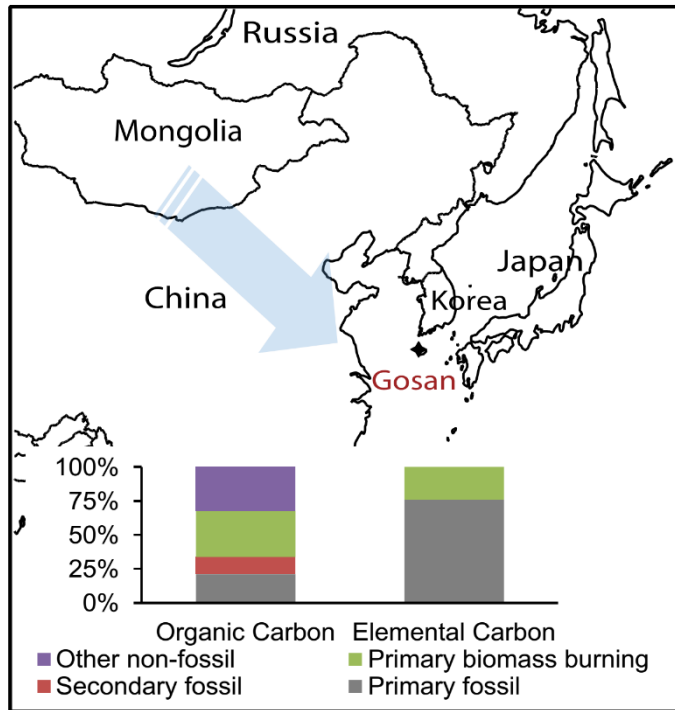
Zhang et al., ACP, 2015

一次源与二次源对含碳颗粒物的贡献



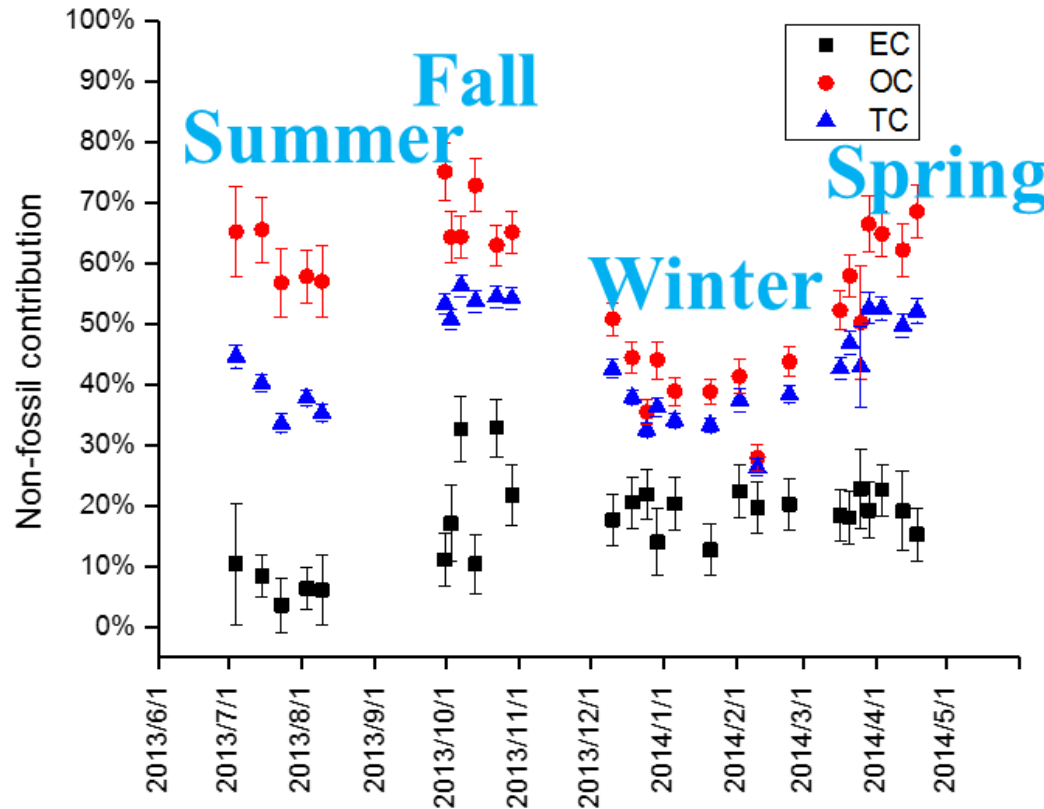
①揭示了二次有机气溶胶对我国重霾污染PM_{2.5}污染的重要贡献; ②提供了PM_{2.5}来源解析的新思路与新方法; ③提出了我国应加强控制气溶胶前体物排放的建议。相关研究成果发表在 (*Nature*, 514, 218-222, 2014) 和 (*Atmos. Chem. Phys.*, 15, 1299-1312, 2015)

济州岛有机碳来源



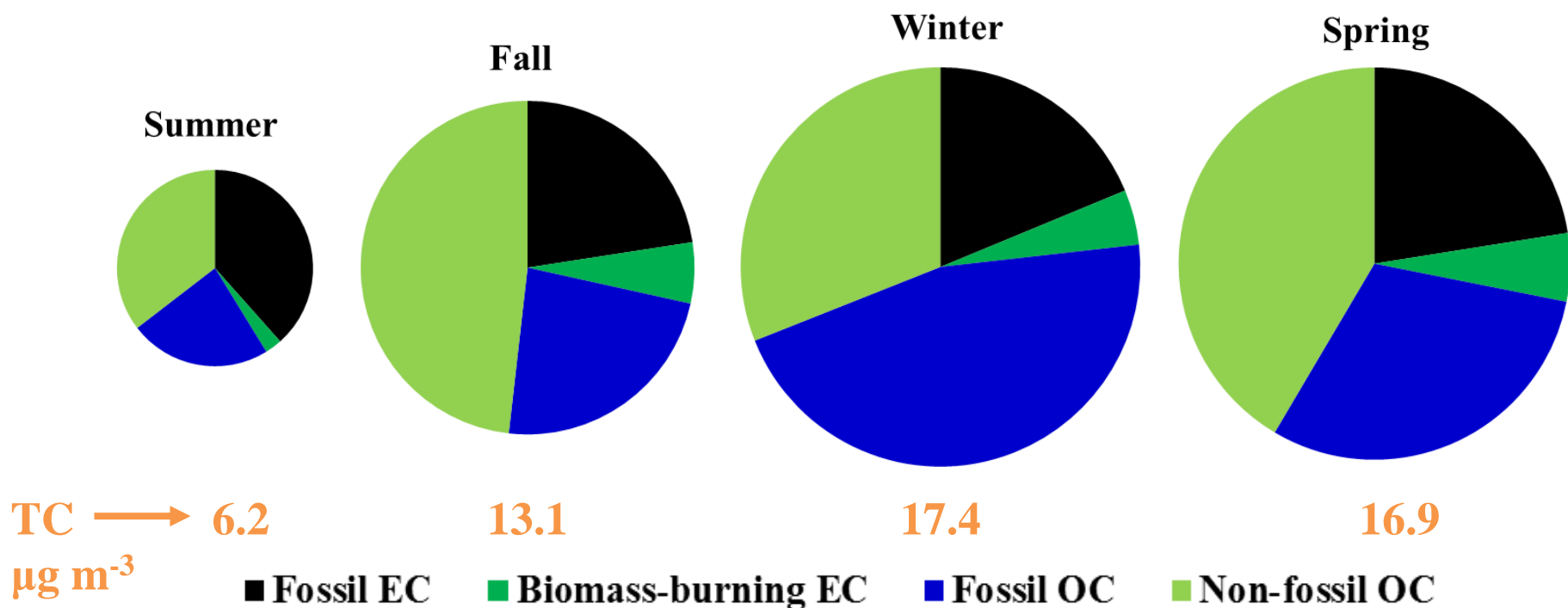
Zhang et al., ES&T, 2016

^{14}C results

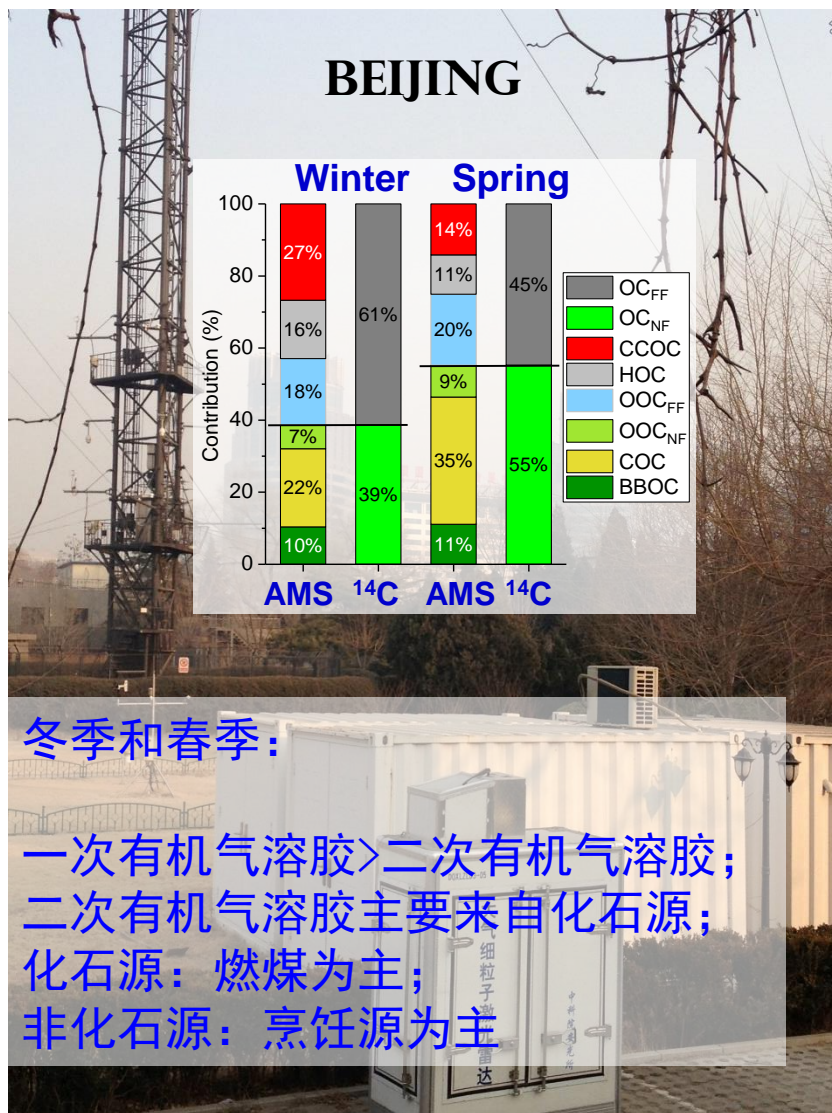


- I. OC and TC: similar source and variations
- II. Non-Fossil OC: 28-75% (55%)
- III. EC: highest fossil in summer

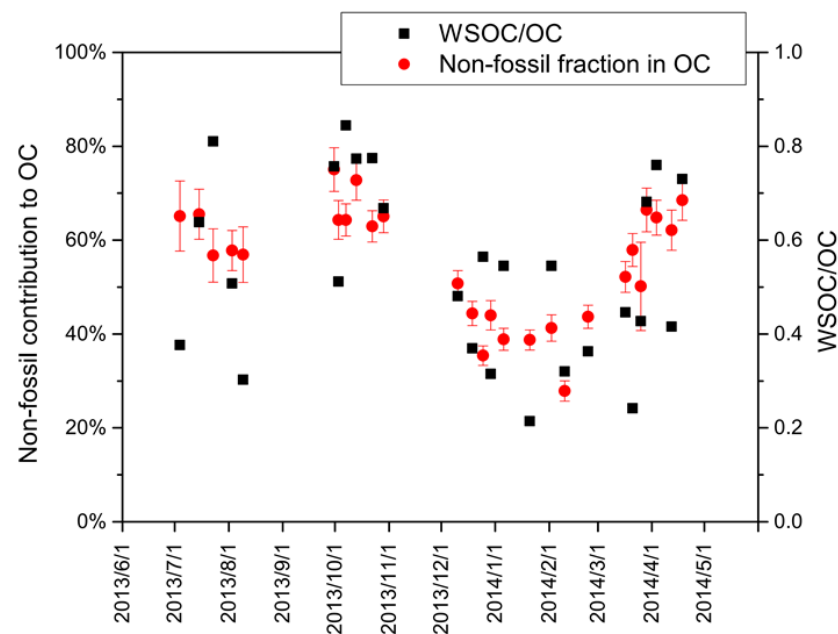
PM1 有机碳和黑碳的化石源/非化石源贡献



- ❑ Non-fossil OC was a dominant contributor (~60%) in summer, fall and spring.
- ❑ Fossil OC became the most important contributor (~60%) in winter.
- ❑ EC was dominated by fossil sources (>80%) in all seasons.
- ❑ Biomass burning only contributed to ~7% of EC in summer, but increased to ~20% in other seasons.



北京PM1有机气溶胶来源

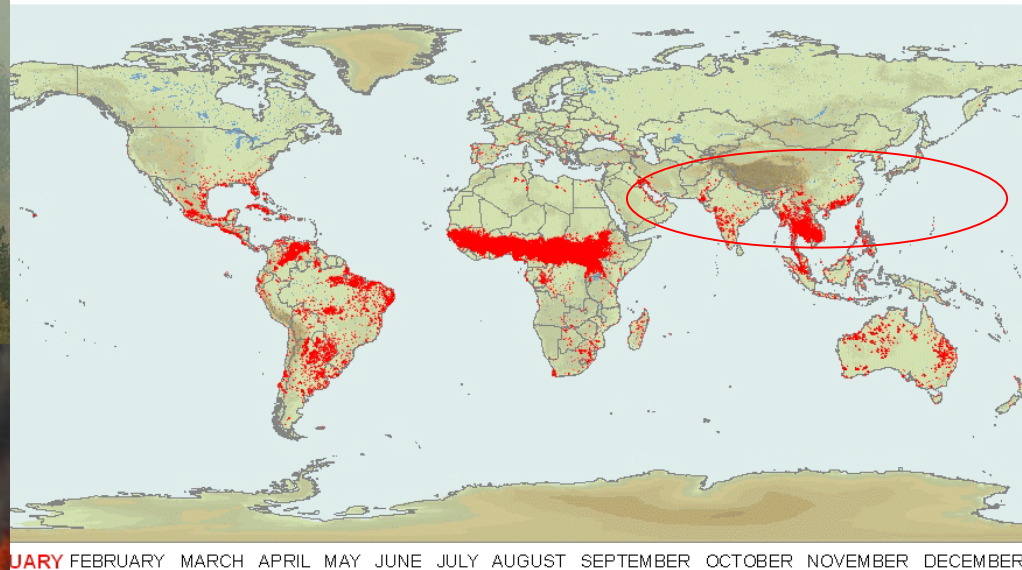


生物源主要和生物质燃烧和二次有关

An important contributor: open biomass burning

Biomass (including residential biofuels) burning is an important contributor to air pollution in Asia (Streets et al., 2003).

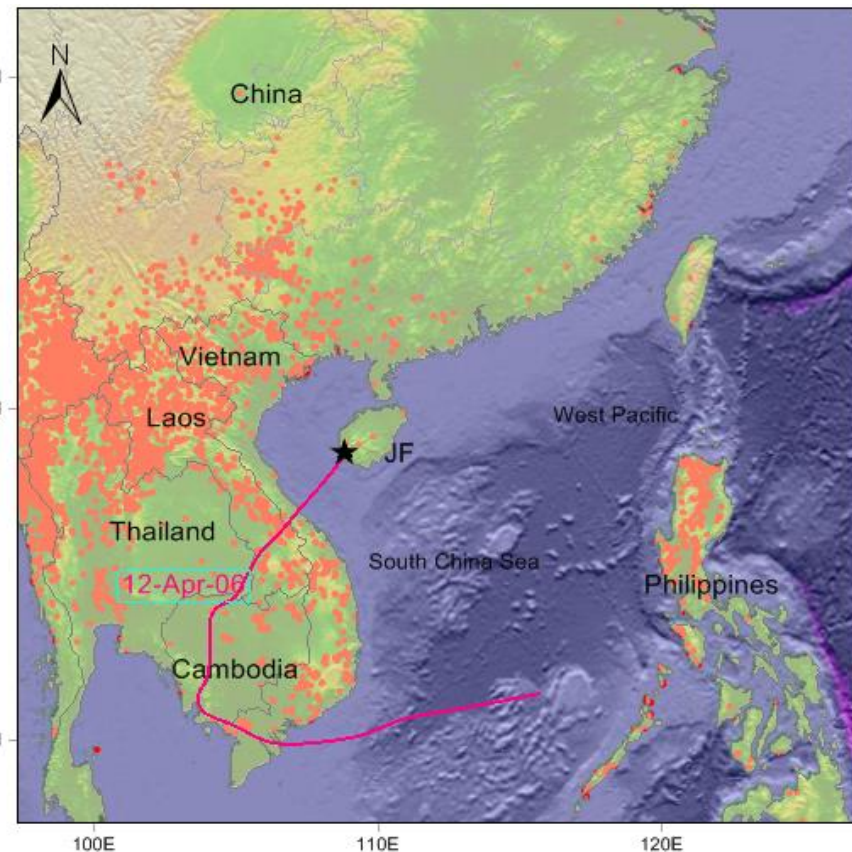
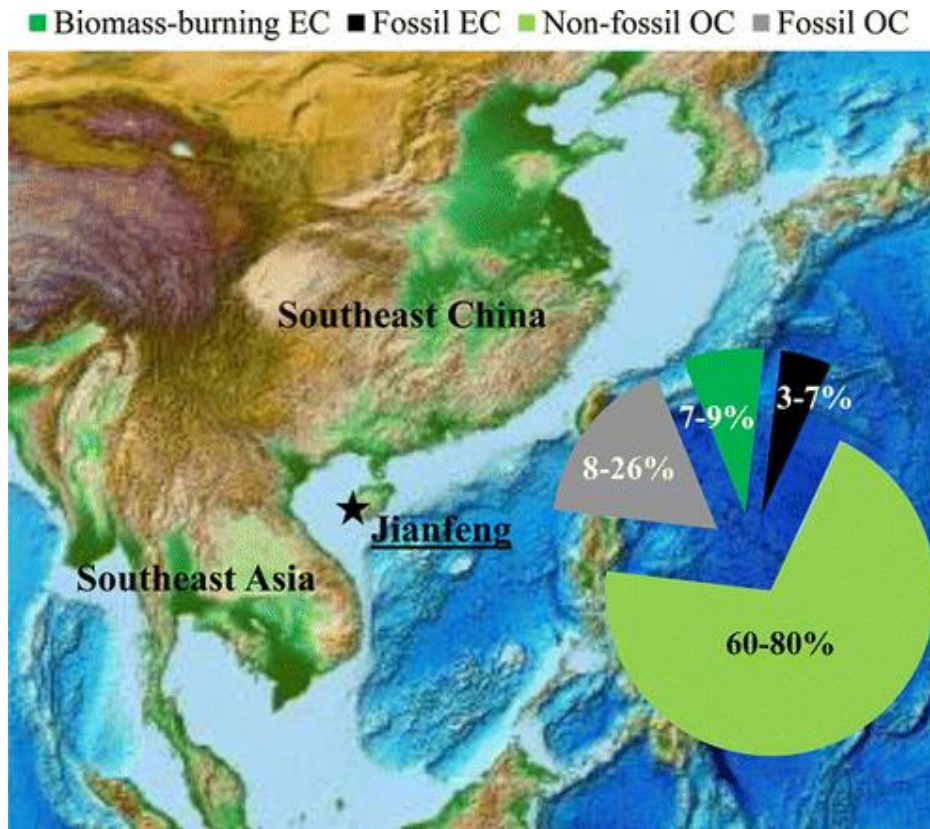
MODIS Rapid Response Fire Detections for 2005



Zhang et al., 2014

Can open biomass-burning activities influence on the air quality of this background site?

Biomass burning in South Asia



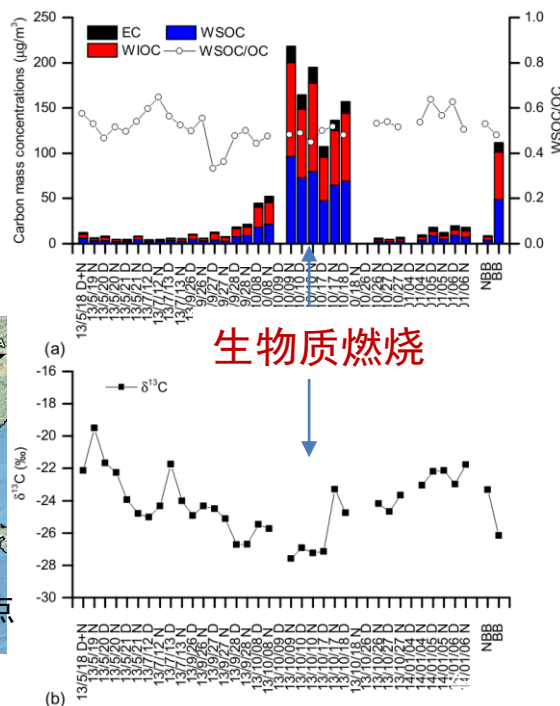
Zhang et al., 2014

生物质燃烧： ^{13}C 证据

2013年秋季
实地照片



三江平原沼泽湿地生态试验站



- ✓ 发现生物质燃烧严重影响东北三江平原地区的空气质量；其主要生物质燃烧类型为C3植物；
- ✓ 启示：生物质燃烧(土地利用方式的变化可能)严重影响该地区含碳物质的地球化学循环过程

$\delta^{13}\text{C}$: 大气化学过程

↓ Depletion in heavier isotope ^{13}C

↑ Enrichment in heavier isotope ^{13}C

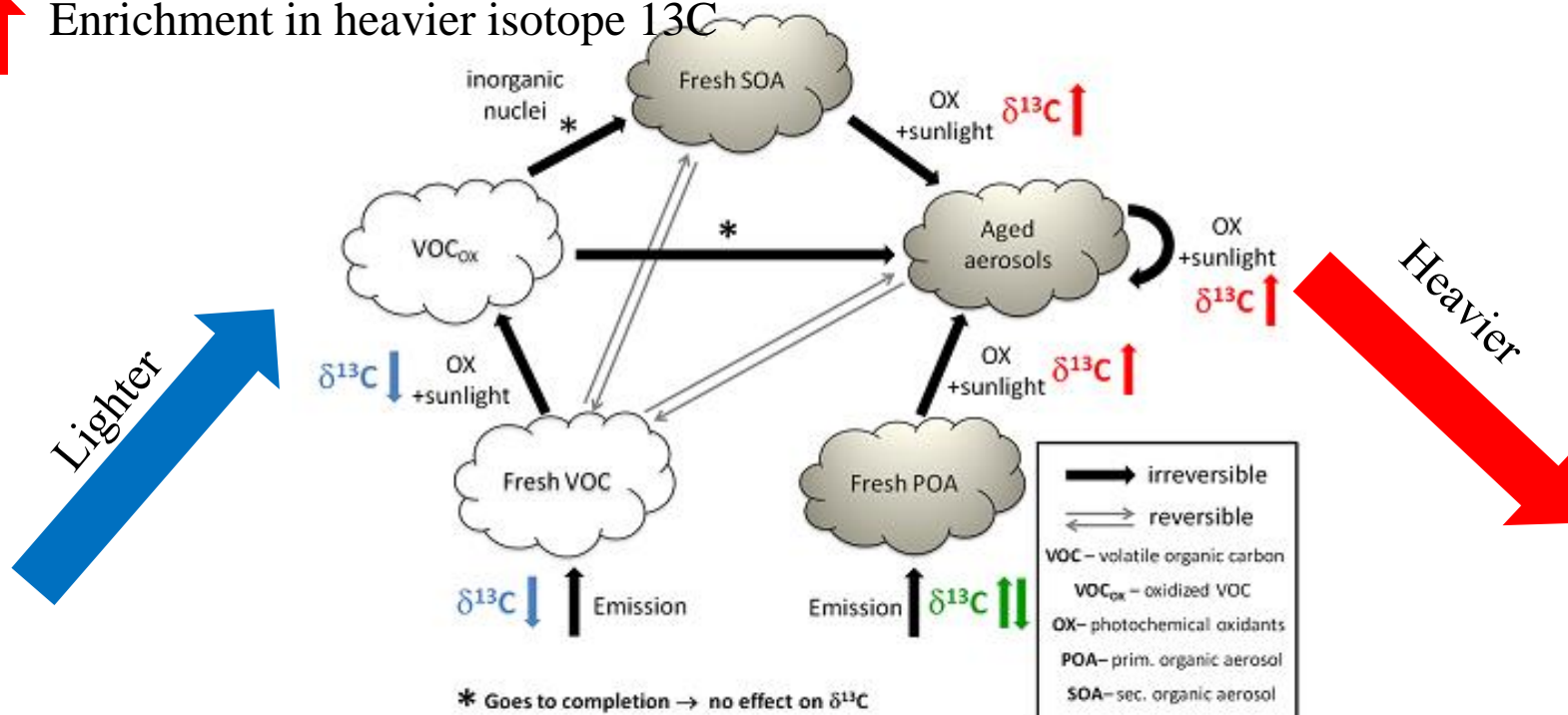
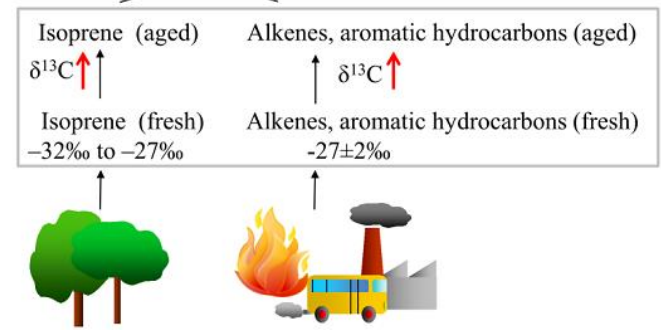
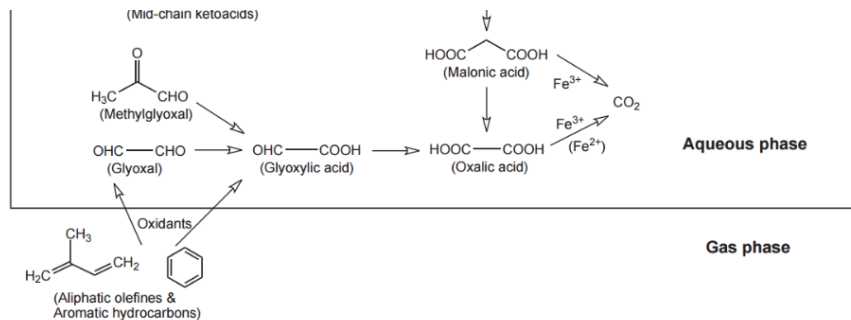
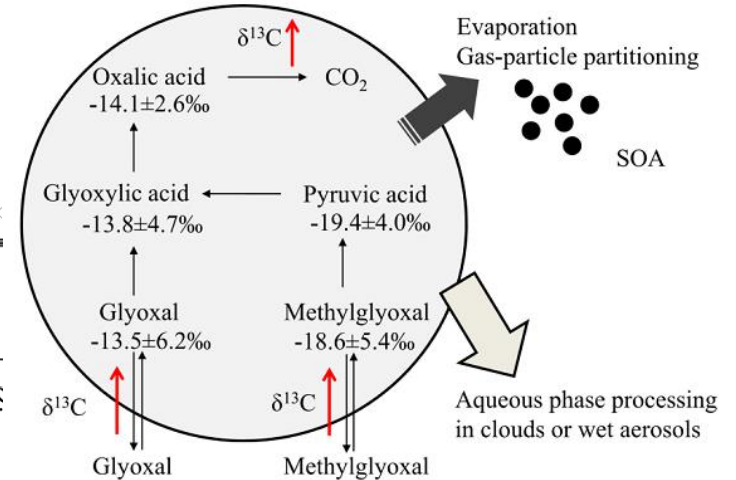
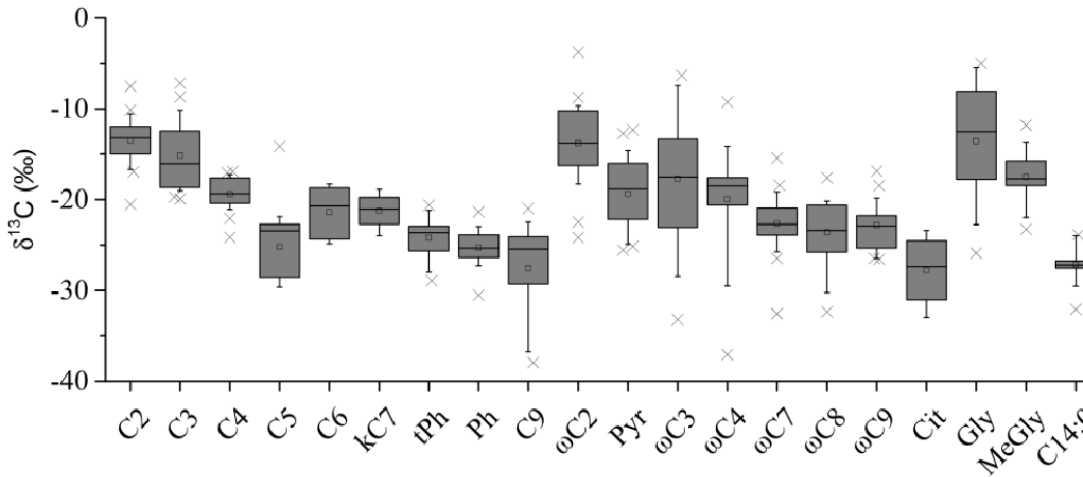


Fig: A simplistic illustration

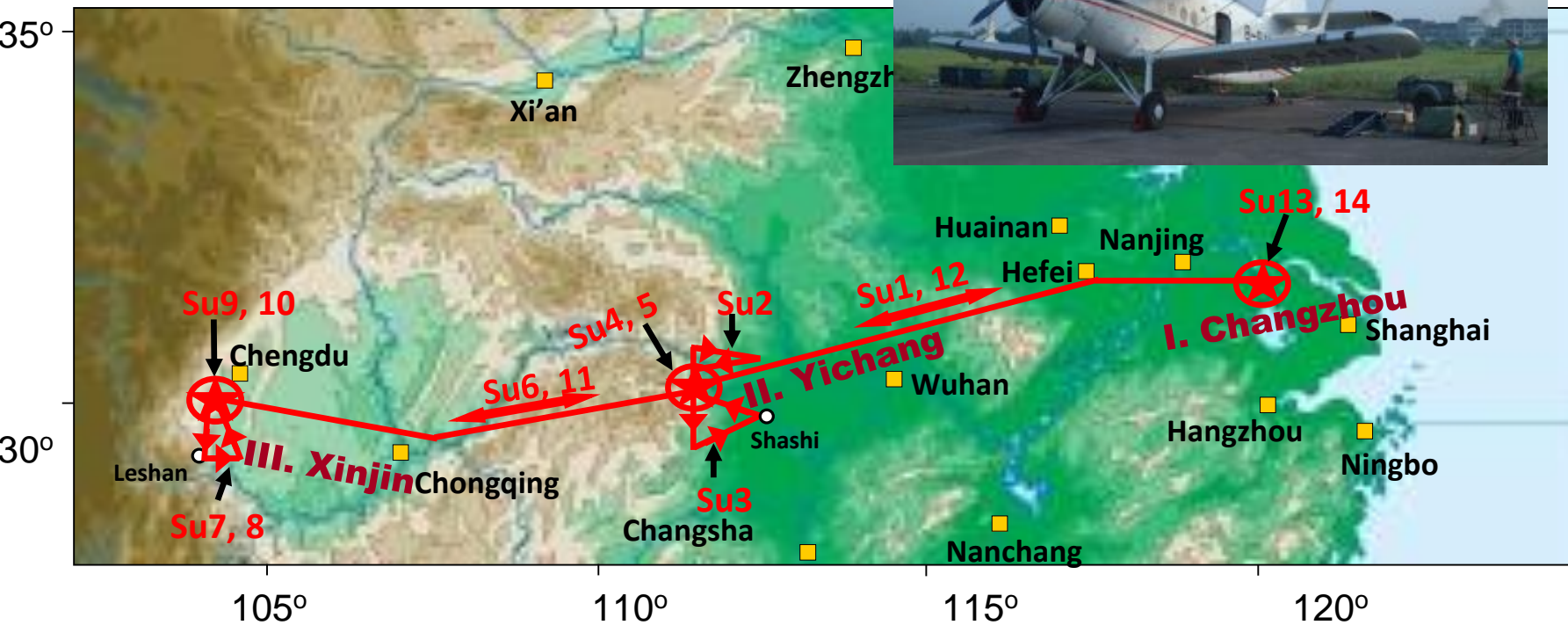
有机气溶胶老化: ^{13}C 证据



Zhang et al, JGR. 2016

有机气溶胶航测

Summer 2003



(zhang et al., ACP, 2016)

结论与展望

- 含氮和含碳气溶胶的同位素与气溶胶排放源以及气溶胶的大气化学过程（气粒分配、气溶胶老化等）。
- 厘清与大气化学过程有关的同位素分馏过程是进行稳定同位素溯源研究的前提。
- 稳定同位素分析也是研究污染物的大气化学过程的有效手段。
- 大气化学研究迫切需要大气科学、环境科学、地球化学、同位素生态学等多学科交叉。



Stable Isotope Lab @NUIST

Gas-Bench

GC

Con-flo IV

Precon

MAT 253

EA

同位素大气化学分会场研讨会
(17日早上三楼钟山厅)

欢迎晚上参观大气环境中心和大气成分稳定同位素实验室

章炎麟, dryanlinzhang@outlook.com, 18151606893

欢迎与我们联系, 一起开展同位素大气化学合作研究