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Improving the representation of cross-boundary transport of anthropogenic pollution in Southeast Asia using Radon-222

***S.D. Chambers*[1], C-.H. Kang[2], A.G. Williams[1], J. Crawford[1], A. Griffiths1, K-.H. Kim[3] & W-.H. Kim[2]**

[1] ANSTO Institute for Environmental Research, Sydney, NSW, Australia

[2] Department of Chemistry, Jeju National University, Jeju 690–756, Korea

[3] Dept. of Civil & Environ. Engineering, Hanyang University, Seoul 133-791, Korea

7th Asia-Pacific GAW Workshop on Greenhouse Gases

October 22nd-23rd, 2015, Jeju, Republic of Korea.

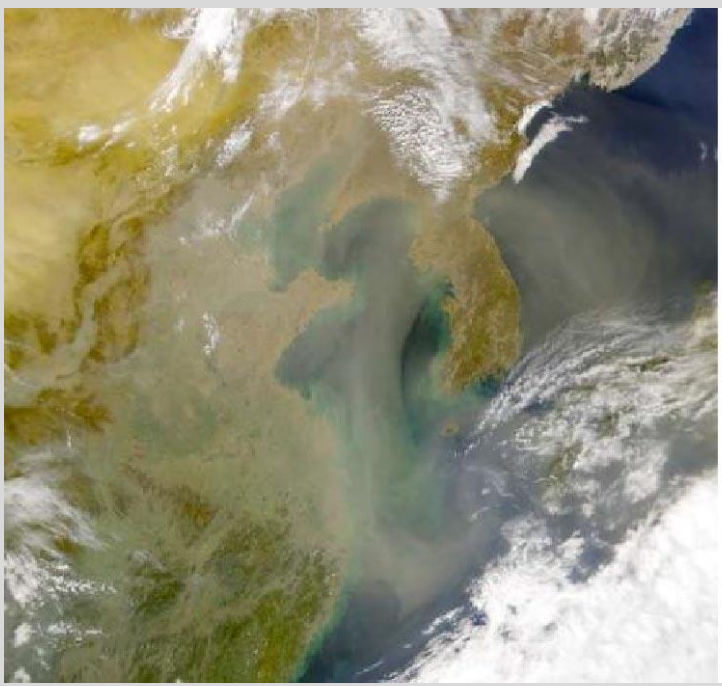
Background / Overview

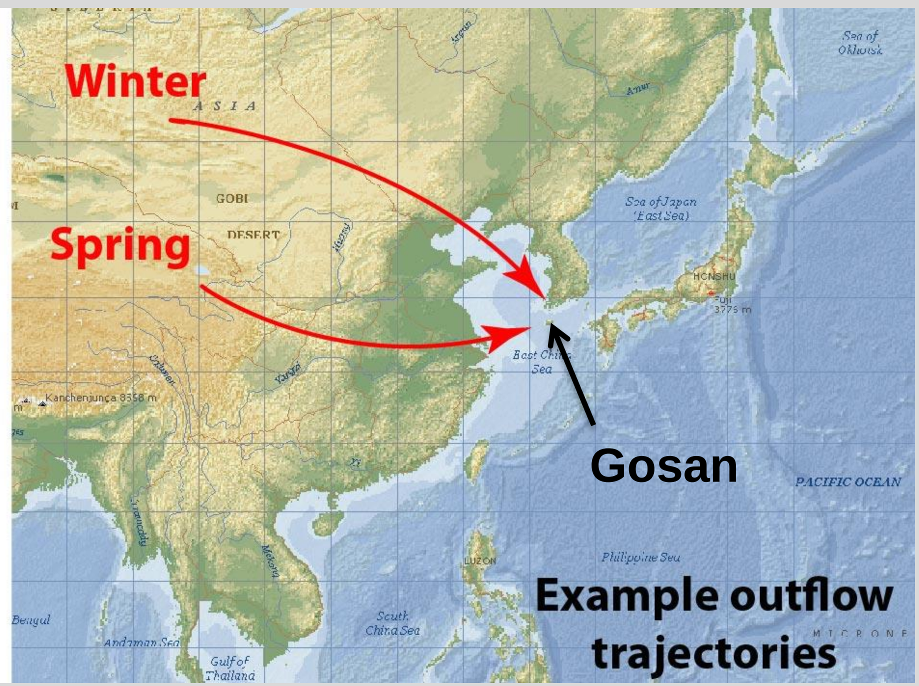
- Southeast Asia is presently contributing a large fraction of global anthropogenic emissions
- Due to the scale of the source regions and affected areas (& cost of detailed monitoring), we need to employ regional / global models to understand and estimate effects
- To improve model accuracy, and their ability to evaluate various emissions scenarios into the future, models need to be extensively evaluated & continually improved
- The number, quality, duration and temporal resolution of independent datasets for model evaluation (remotely-sensed and surface-based) also needs to be improved
- **Specifically**: surface-based measurements need to be suitably selected / processed to make them as representative as possible of intended fetch regions, and scaled appropriately to model resolution

Asian "outflow" and cross-boundary transport

- Natural and anthropogenic pollution leave continental Asia (a) at high-altitude (jet stream), and (b) low-altitude (in the MBL)
- Outflow in the MBL is tied to synoptic activity & monsoon circulation; not as far reaching as outflow in the jet-stream, but effects can extend beyond Japan
- Current emissions, and future changes, need to be accurately quantified; it is important to know exactly what our observations represent (w.r.t. scale & fetch)
- Gosan Station is ideally situated to observe MBL outflow events; can also be a good "background" site, since local emissions are low c.f. mainland Korea.

Easterly transport of natural and anthropogenic pollution from central and SE China to Korea and Japan



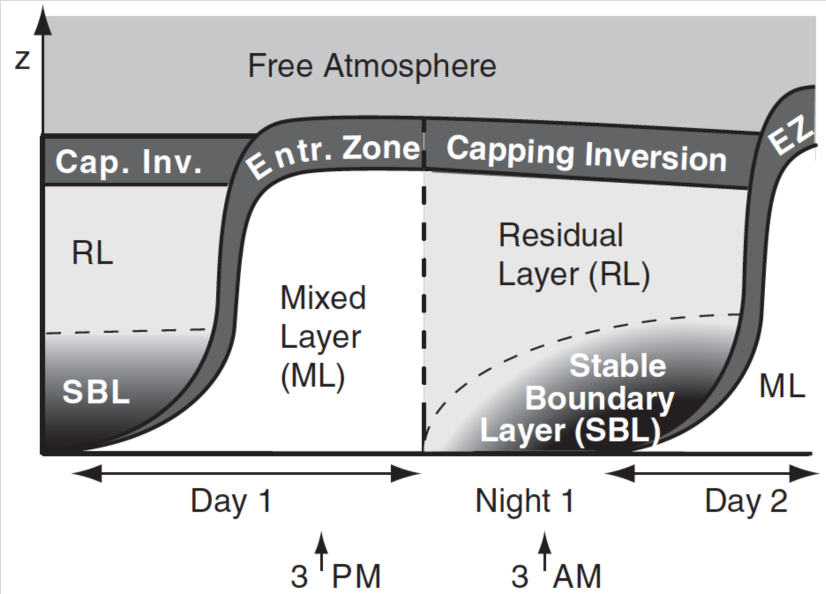


Factors affecting fetch representation

- Anthropogenic emissions are integrated by, and mixed within, the atmospheric boundary layer (i.e. from the surface to the synoptic inversion)
- The pollutant concentration of an air mass sampled "down-stream" depends on:
  (a) time in contact with surface sources (i.e. in the ABL)
  (b) ABL dilution processes during accumulation (detrainment, convection, fronts)
  (c) dilution *en route* to sampling location, and
  (d) uniformity of mixing & depth of the ABL at sampling location
- The influence of these effects can be assessed using a suitable tracer
- **Radon-222** is an unreactive, poorly-soluble, radioactive gas, with a short half-life and ~consistent (terrestrial) surface-based source
- These physical characteristics make radon an ideal tracer of transport and mixing
- For fetch regions with ~uniform radon flux, radon concentrations give qualitative information about air mass contact time with surface sources and dilution *en route* to the sampling location

Scale matching: spatial, vertical, temporal

- At most surface-based observation sites the ABL changes significantly over diurnal cycle
- During the day (when mixing is strong)
  (i) near-surface observations representative of whole ABL and large "footprint"
  (ii) pollution contributions from distant sources are **largest**, local contributions to observed pollution are **smallest**

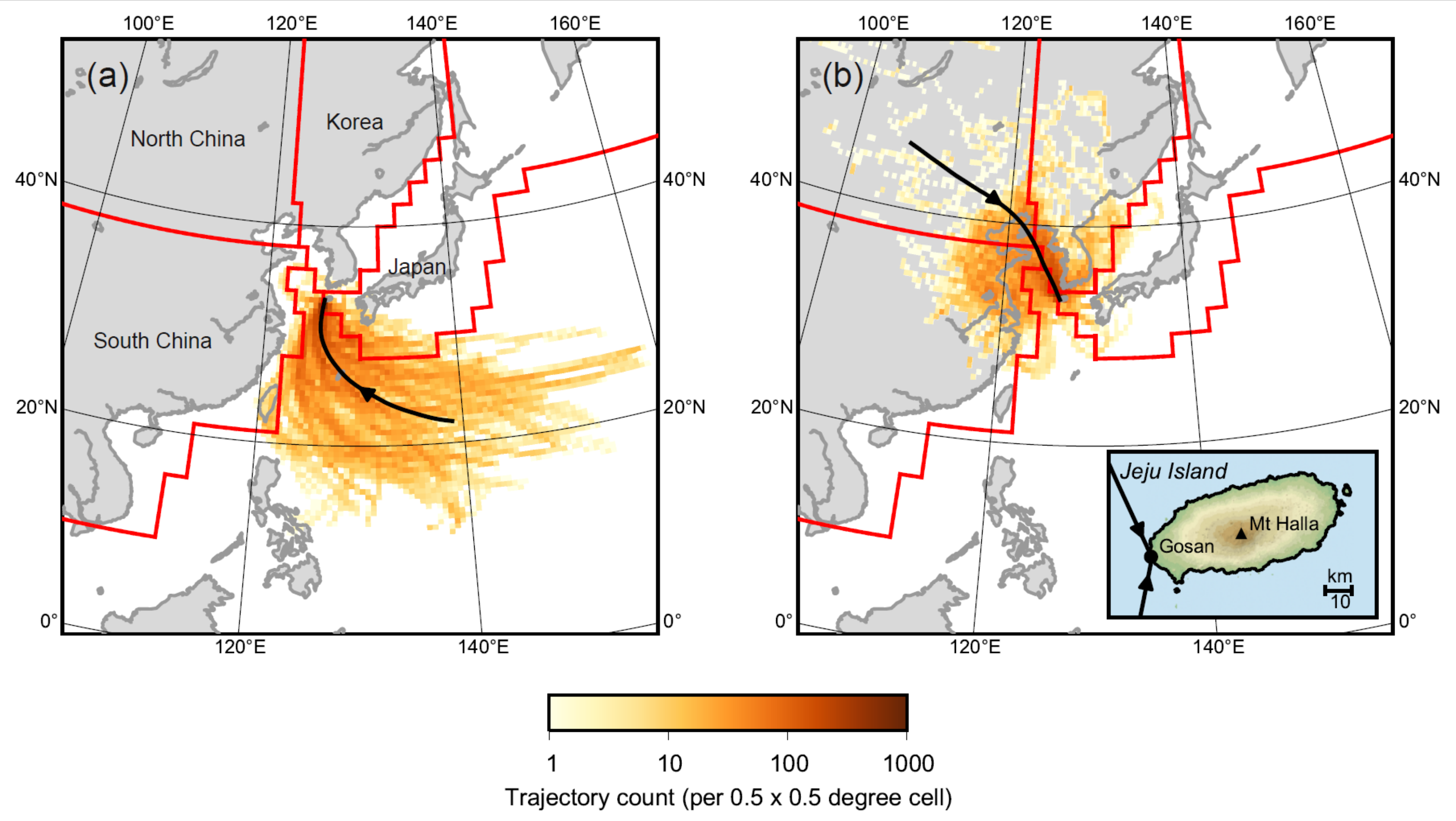


- At night the SBL inversion can isolate near-surface observations from the RL
  (i) vertical mixing is weak, likely not resolved by vertical model resolution
  (ii) near surface obs. represent small portion of the lower atmosphere (10s-100s m)
  (iii) spatial footprint of near-surface observations small (10s km)
  (iv) remote (advective) pollution contributions are **smallest**, local contributions to observed pollution are **largest**
- When a diurnal cycle exists, a diurnal sampling window should be used to provide the best scale match between observations and simulations

Gosan fetch seasonality & fetch regions

Summer monsoon (recent oceanic fetch)

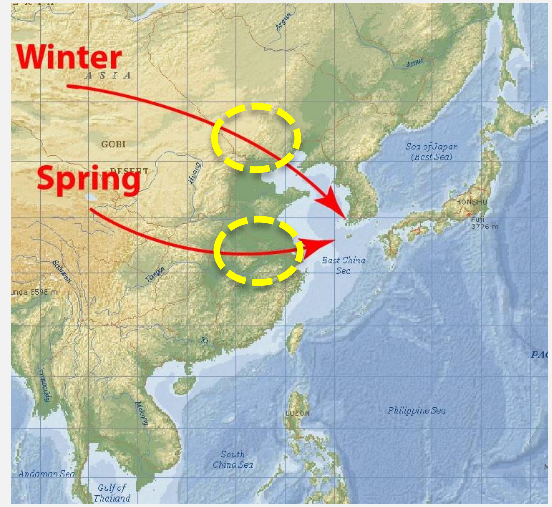
Winter monsoon (recent land fetch)

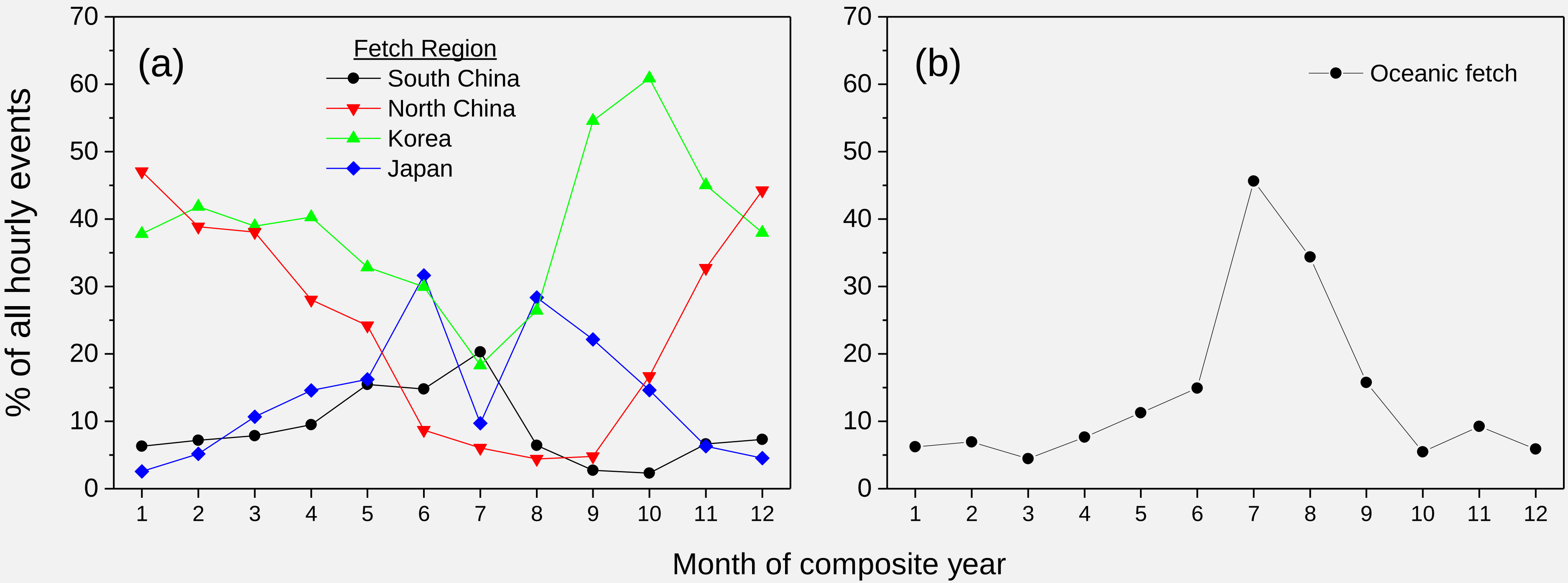


Transitional periods (mixed fetch conditions)

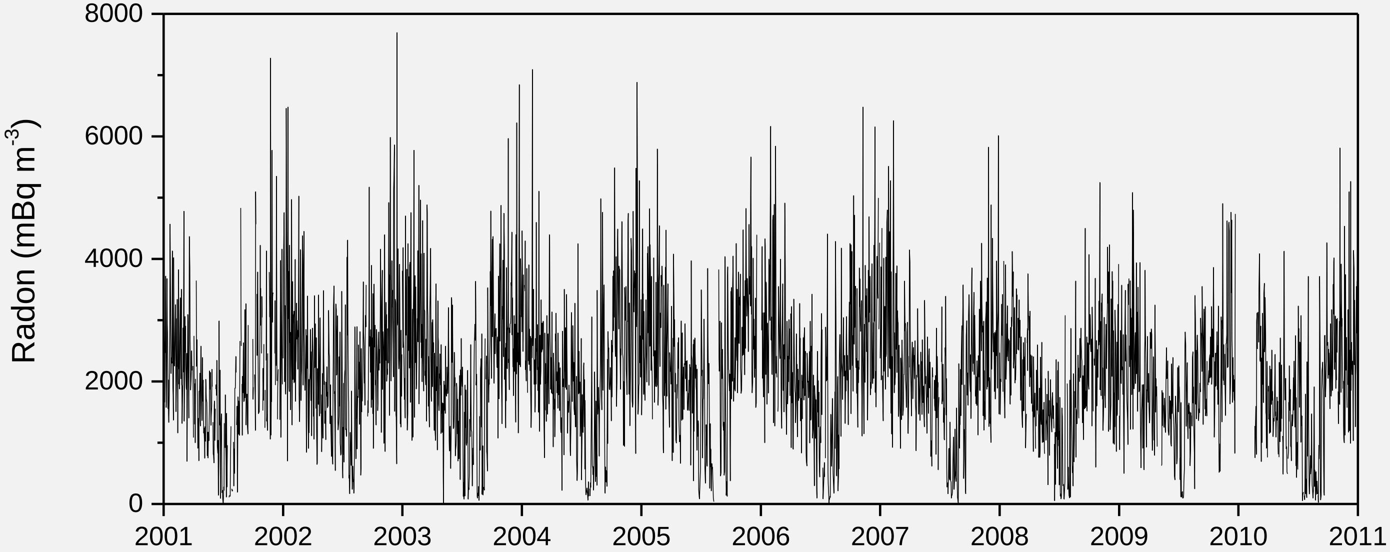
Gosan: monthly fetch-region breakdown

- *PC version of NOAA HYSPLIT v4.0, starting height 200 m*
- *5-day back trajectories, every hour, every day, 10-years*
- *Fetch region categorised by mean location of air mass over last 24 hours of land contact*
- *Strong seasonality noted in influence on Gosan air masses from the different regions*

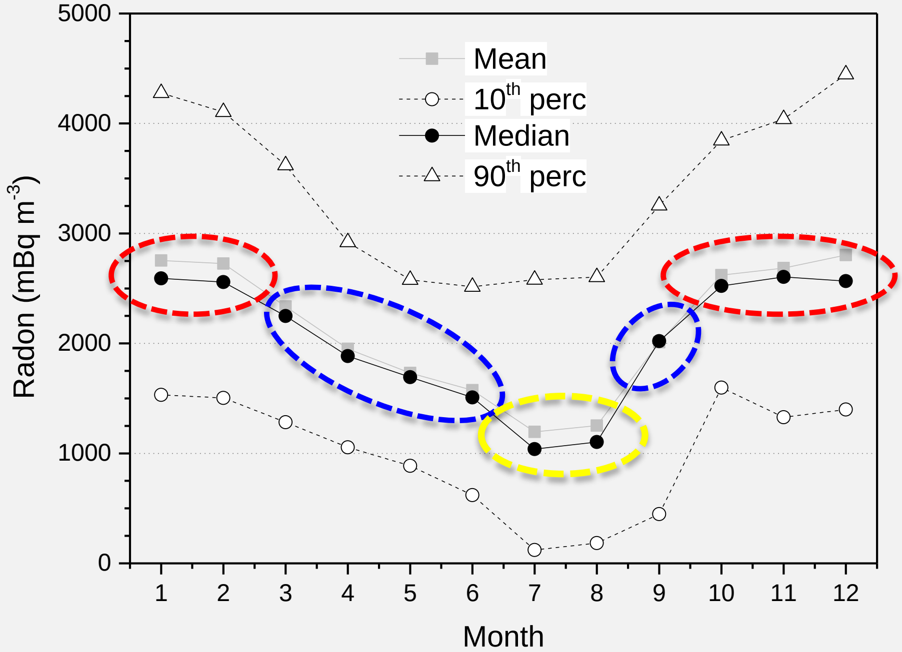




Gosan radon observations

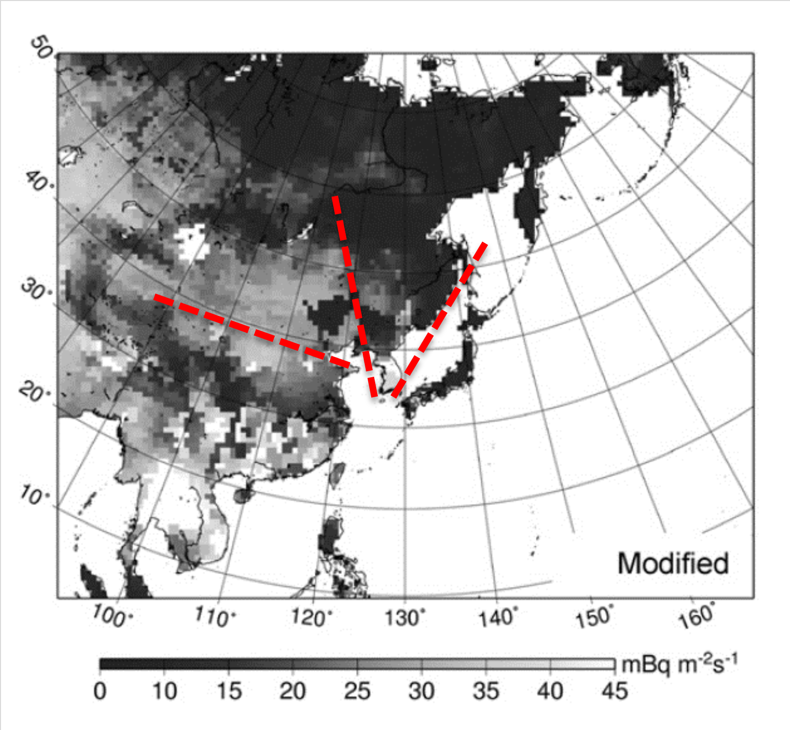


Hourly radon observations at Gosan Station: 2001 - 2011

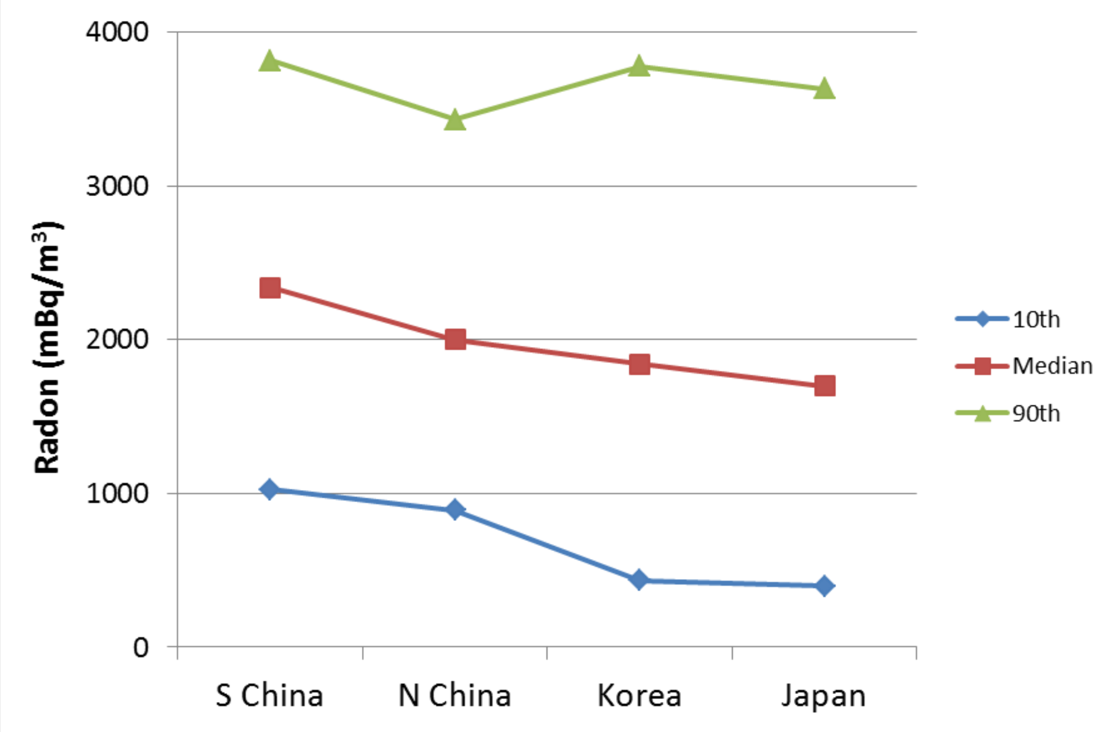


- *Large terrestrial / oceanic fetch contrast*
- *Seasonal cycle characterised by winter maximum and summer minimum*
- *Oct – Feb most consistent monsoonal "outflow" from continental Asia*
- *Jul – Aug most consistent oceanic fetch ("cleanest" air masses)*
- *Mar – Jun and Sep are "transitional" months (mixed fetch)*

Contact time with surface and dilution



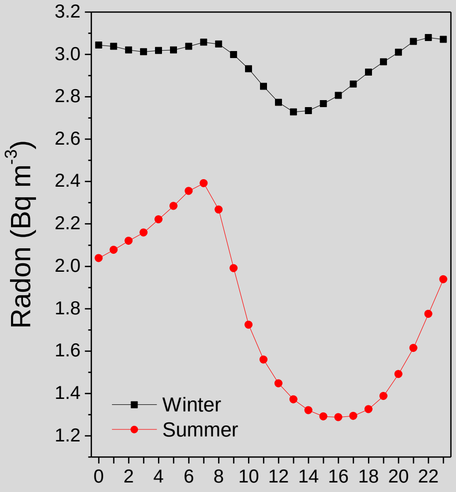
Asian radon flux map (Hirao et al., 2010)

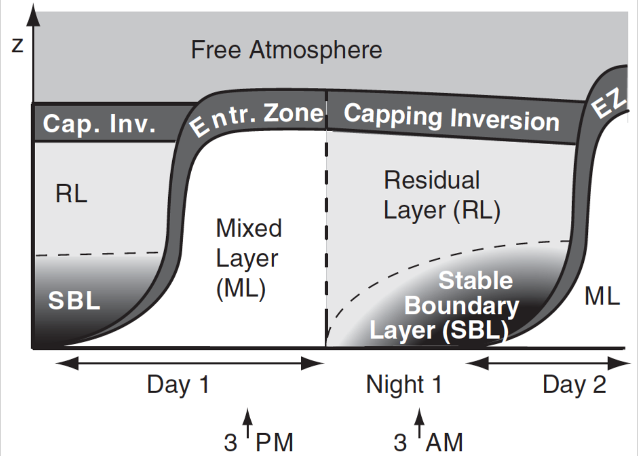


Radon distribution by fetch region

- *Despite variability in radon source function, when air masses have spent a long time over land their concentrations are fairly consistent (90th perc)*
- *Many factors can result in low observed radon (altitude, dilution, etc.)*
- ***Selection Criteria**: air masses with **Rn > median** value have (i) had most consistent contact with land-based sources, and (ii) experienced least dilution en route to Gosan*
- *These air masses are **most representative** of surface-based sources in the fetch regions*

Matching scales: horizontal and vertical





- Gosan diurnal cycle of radon is small (strong oceanic influence)
- Diurnal Rn cycle characterised by afternoon minima and morning maximum
- **Midday**: ABL well mixed (i) surface obs. representative of whole ABL depth, and (ii) large measurement footprint contributes to observations (advection)
- **Night**: accumulation of radon in **shallow surface layer (NBL)**
  Rn (terrestrial), Jeju (island), nearest land > 100km
  → nocturnal accumulation MUST be a **local effect** (reduced measurement footprint)
- Large-scale models may not resolve island-scale (horizontal) or NBL-scale (vertical)
- Model evaluation datasets need to be selected accordingly
- Diurnal CO similar to radon, Diurnal SO_2 180° out of phase

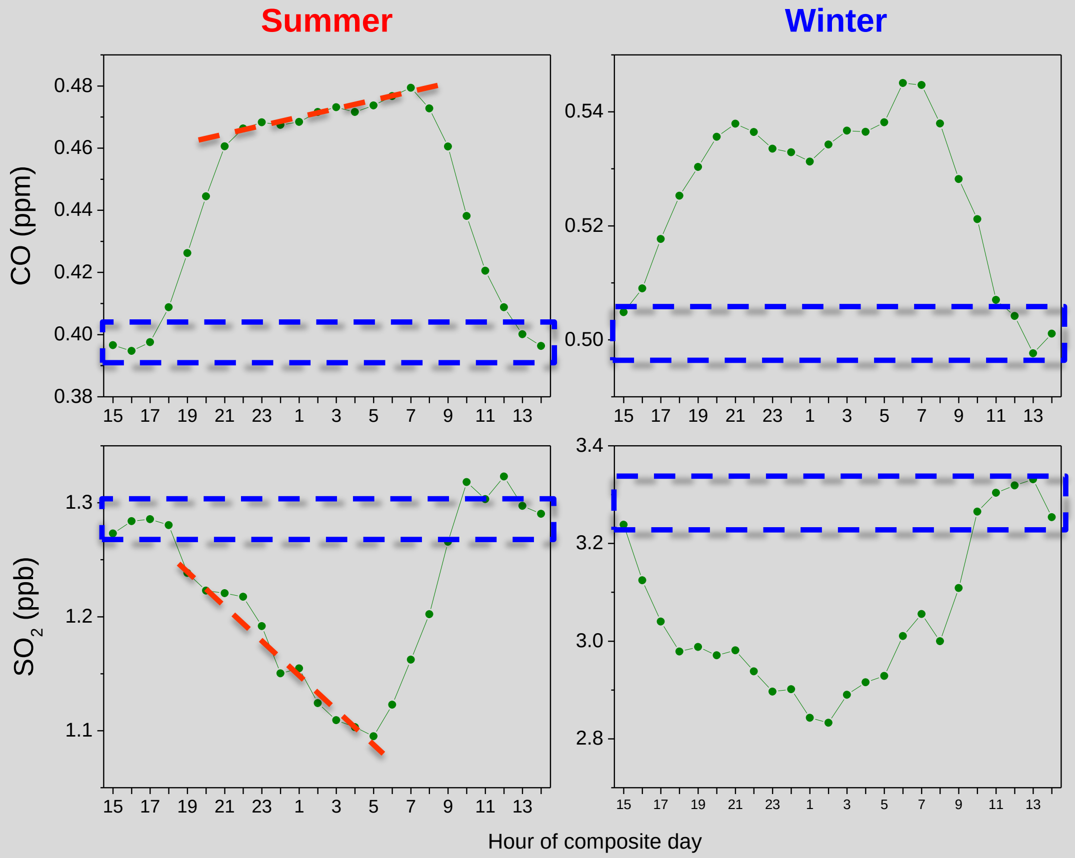
Matching scales: continued (time shifted)

Nocturnal accumulation of LOCAL CO under stable inversion

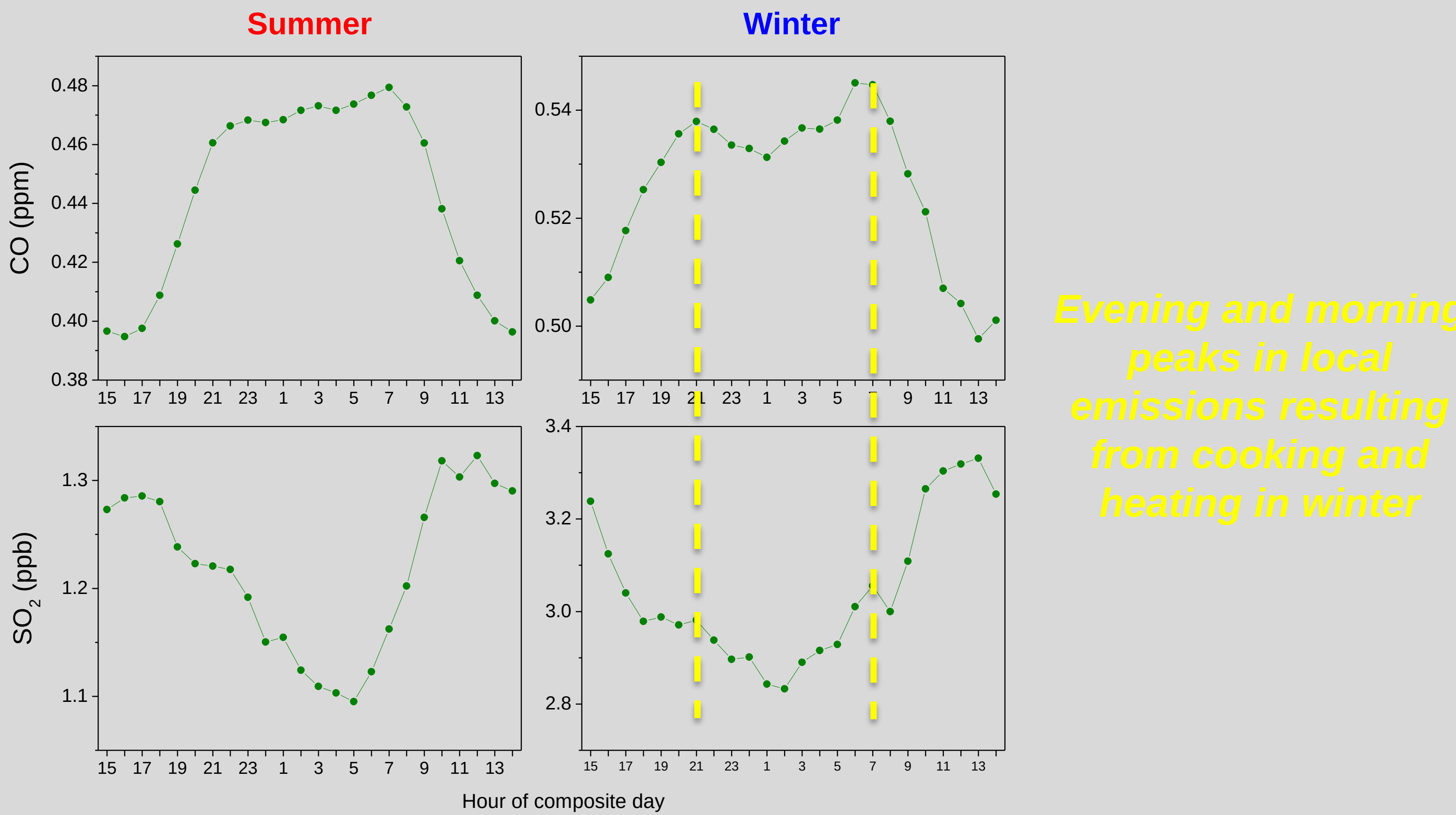
CO from distant sources in the well-mixed ABL

SO_2 from distant sources in the well-mixed ABL

Nocturnal removal of SO_2 (~1% h^{-1}) in the stable boundary layer

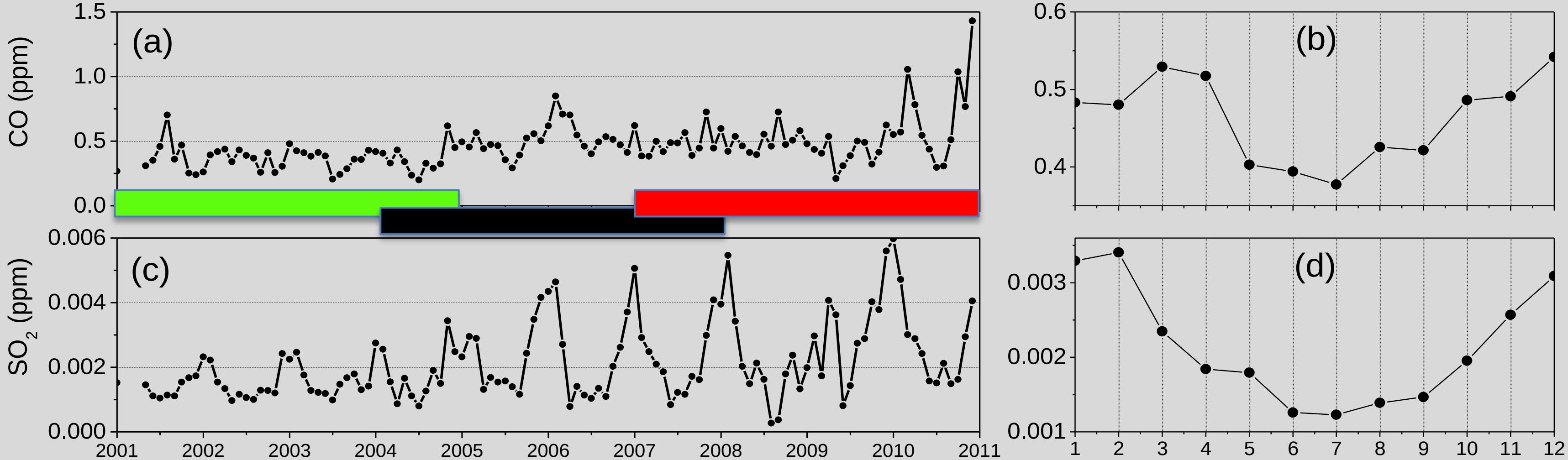


Matching scales: continued (time shifted)



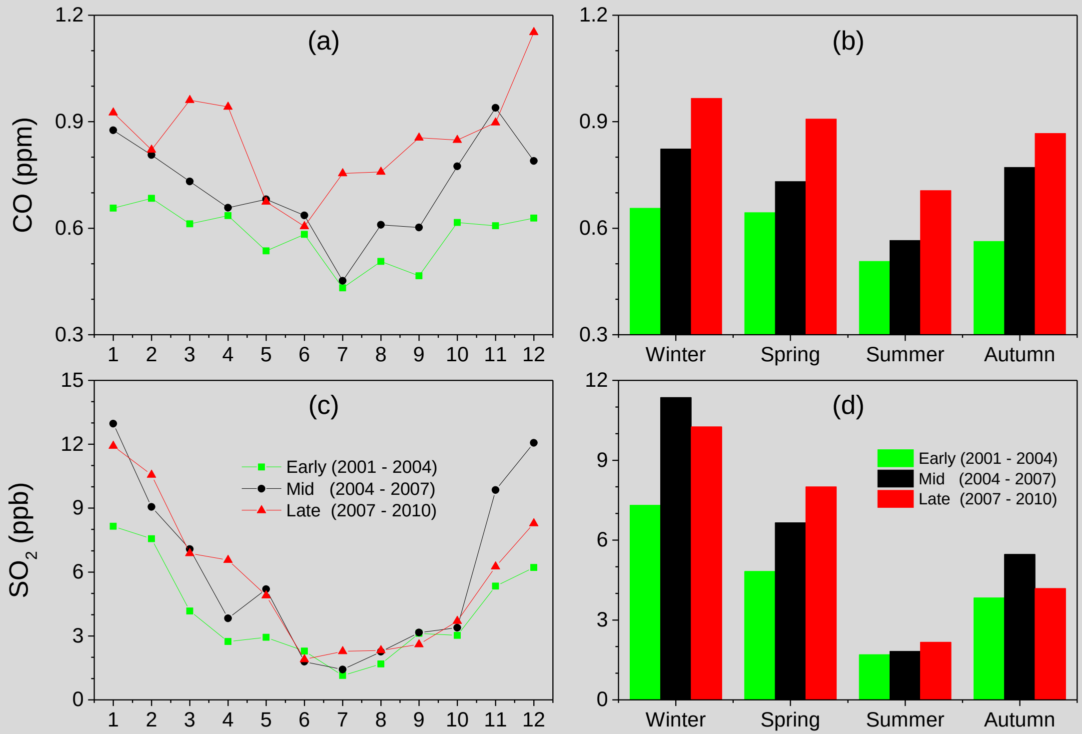
- Not all hourly Gosan observations are equally representative of remote fetch regions across the diurnal cycle
- Local emissions can significantly influence Gosan observations at night (potentially problematic for daily-integrated observations)
- **Selection Criteria**: A 4-6 hour diurnal sampling window centred on the well-mixed ABL period would minimise local influences and best represent remote fetch regions

Anthropogenic emissions: 2001-2011



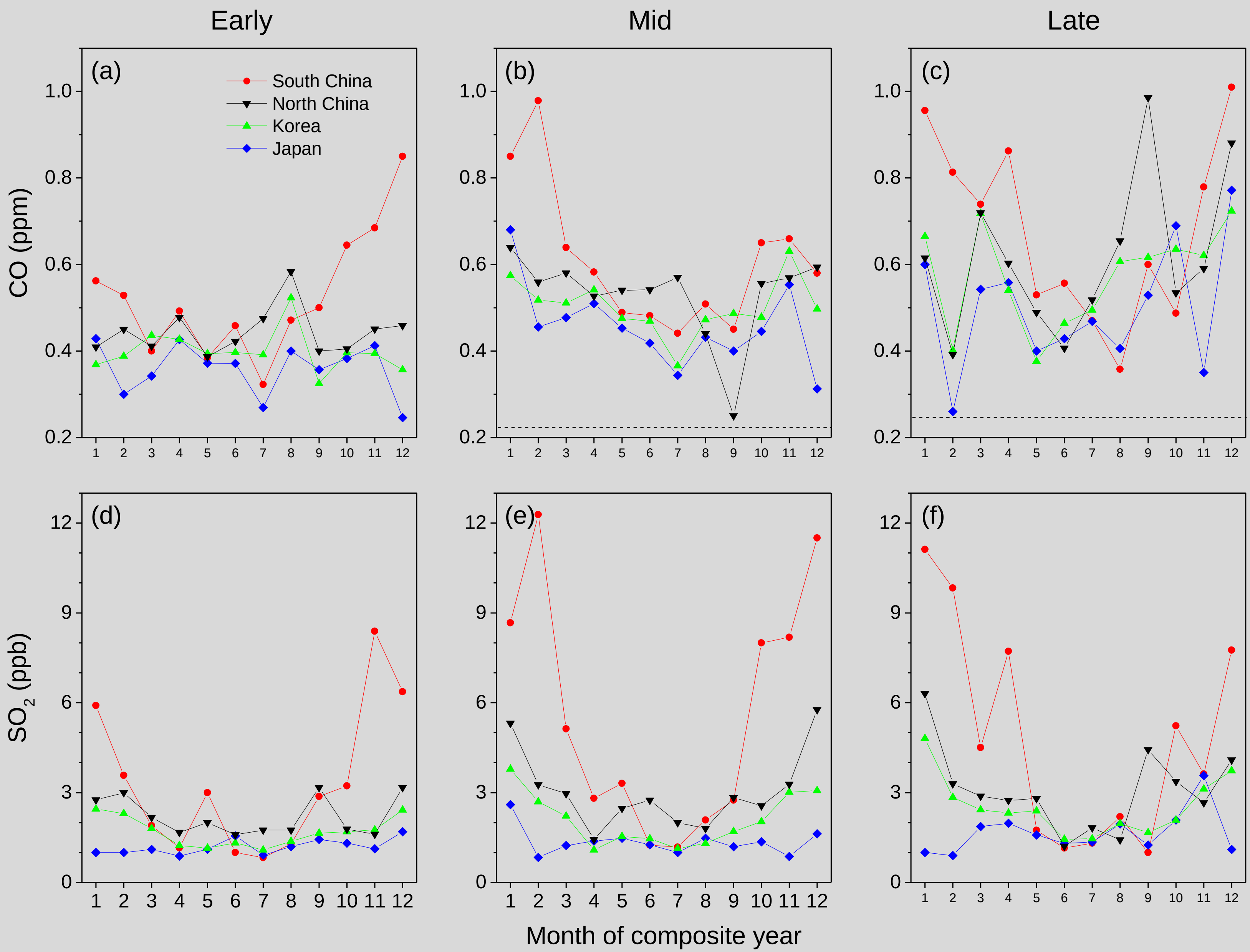
Increase over decade

Strong seasonal cycle



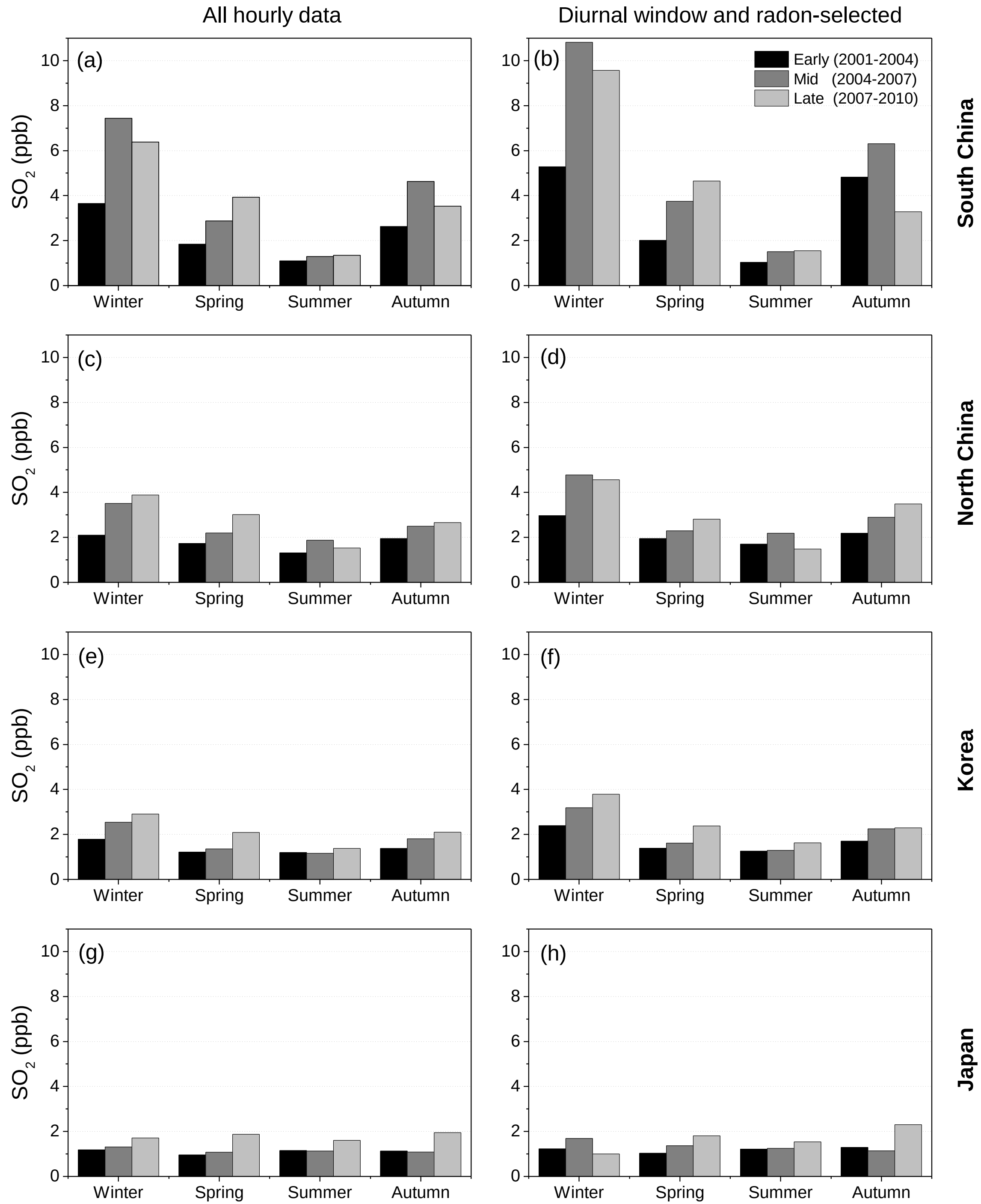
- *4-year composite plots*
- *Seasonal cycle of pollutants closely correlated to recent time-over-land*
- *CO: monotonic temporal increase, all seasons*
- *SO_2: recent winter reduction (various mitigation measures reported ≥ 2006)*
- *SO_2 changes in summer less pronounced (recent land fetch Jeju / Japan – fewer strong SO_2 sources)*

Emissions fetch breakdown



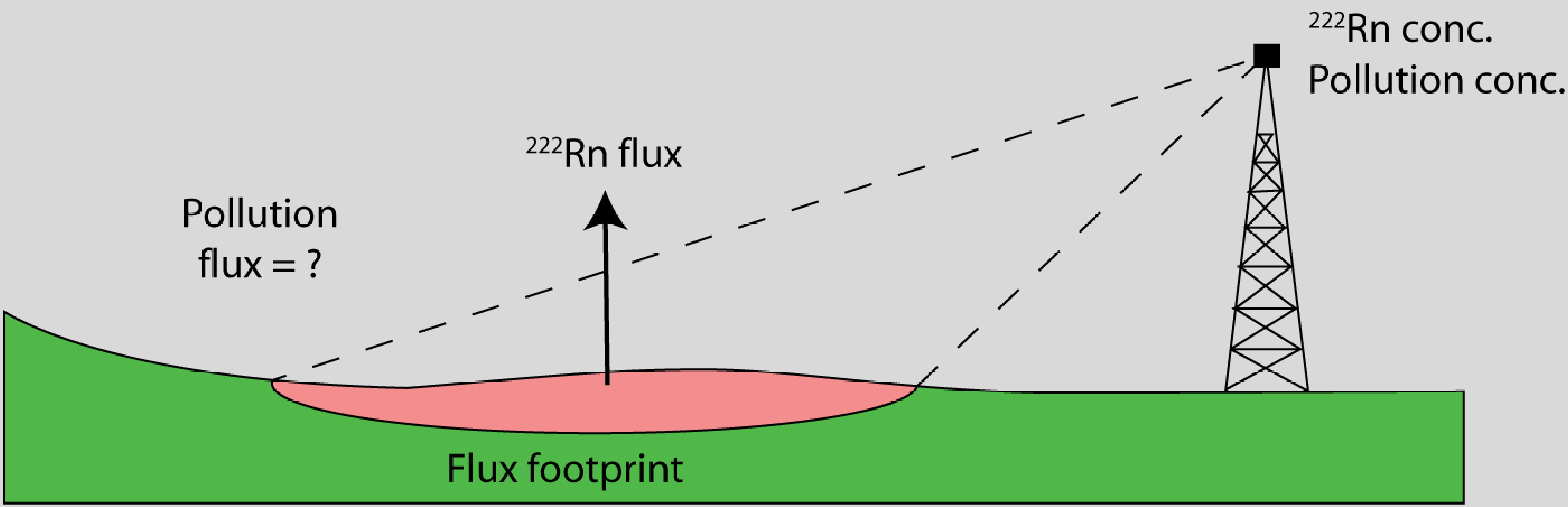
Importance of representative observations

- *Without appropriate data selection: emissions from South China substantially underestimated*
- *Emissions from North China and Korea slightly underestimated*
- *Emissions from Japan sometimes overestimated*
- *Significant implications for regional inventory analyses and model evaluations*



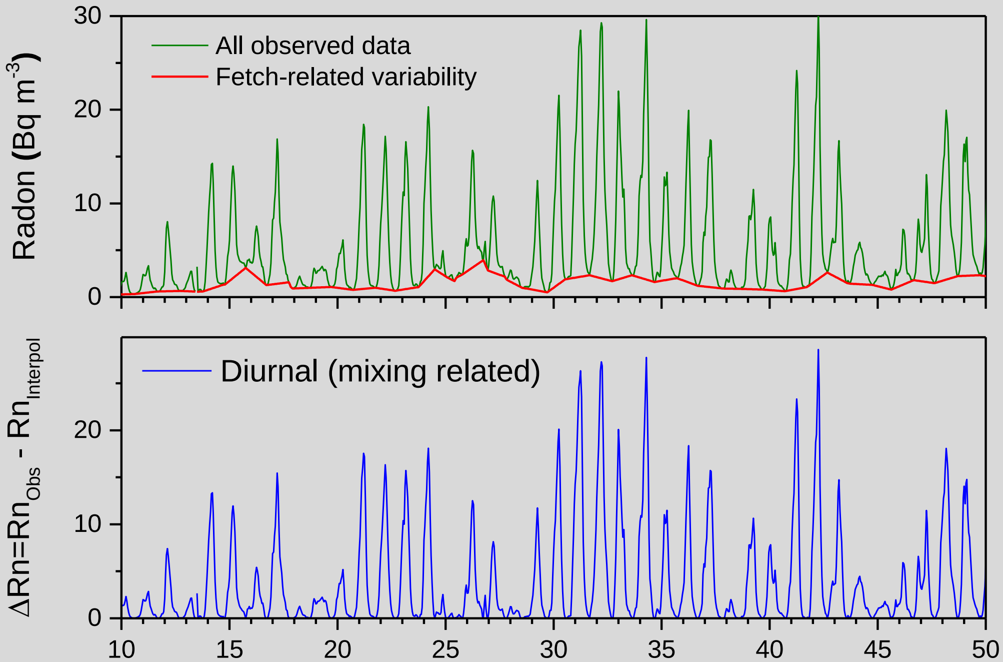
Other applications for hourly atmospheric radon observations at inland or coastal sites

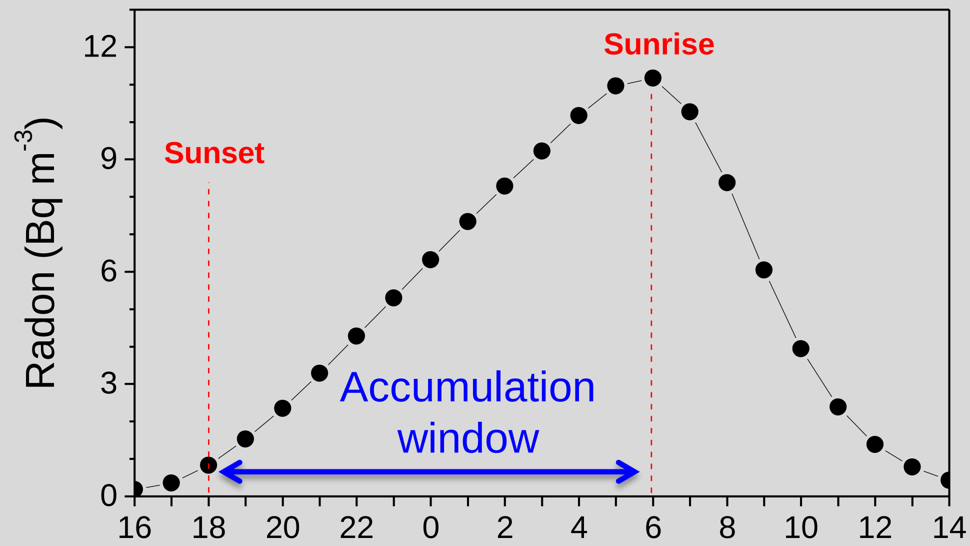
Regional emissions inventories (radon-calibrated flux technique)

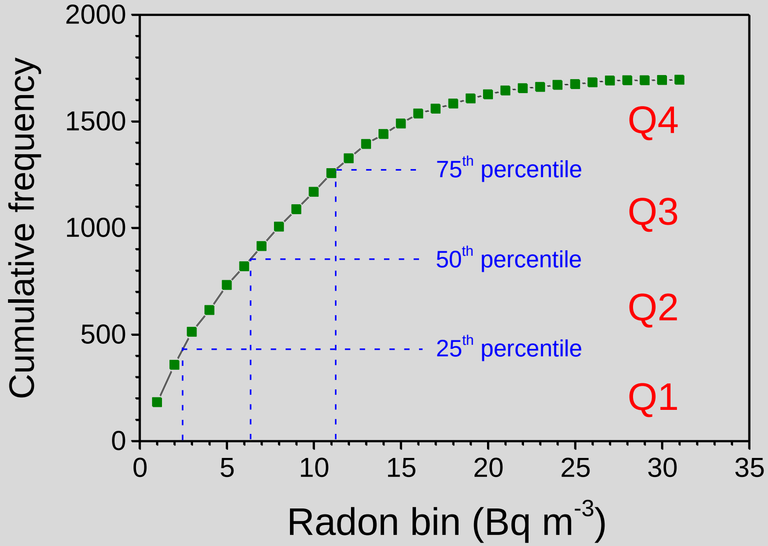


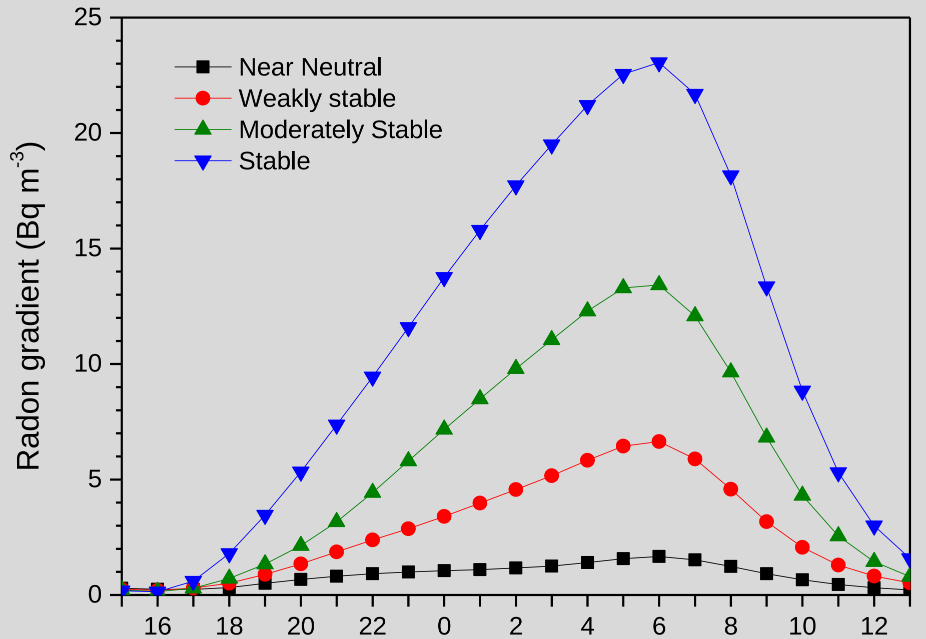
$$\frac{[^{222}Rn]}{^{222}Rn_{FLUX}} = \frac{[Pollution]}{Pollution_{FLUX}}$$

Effect of nocturnal stability on pollution

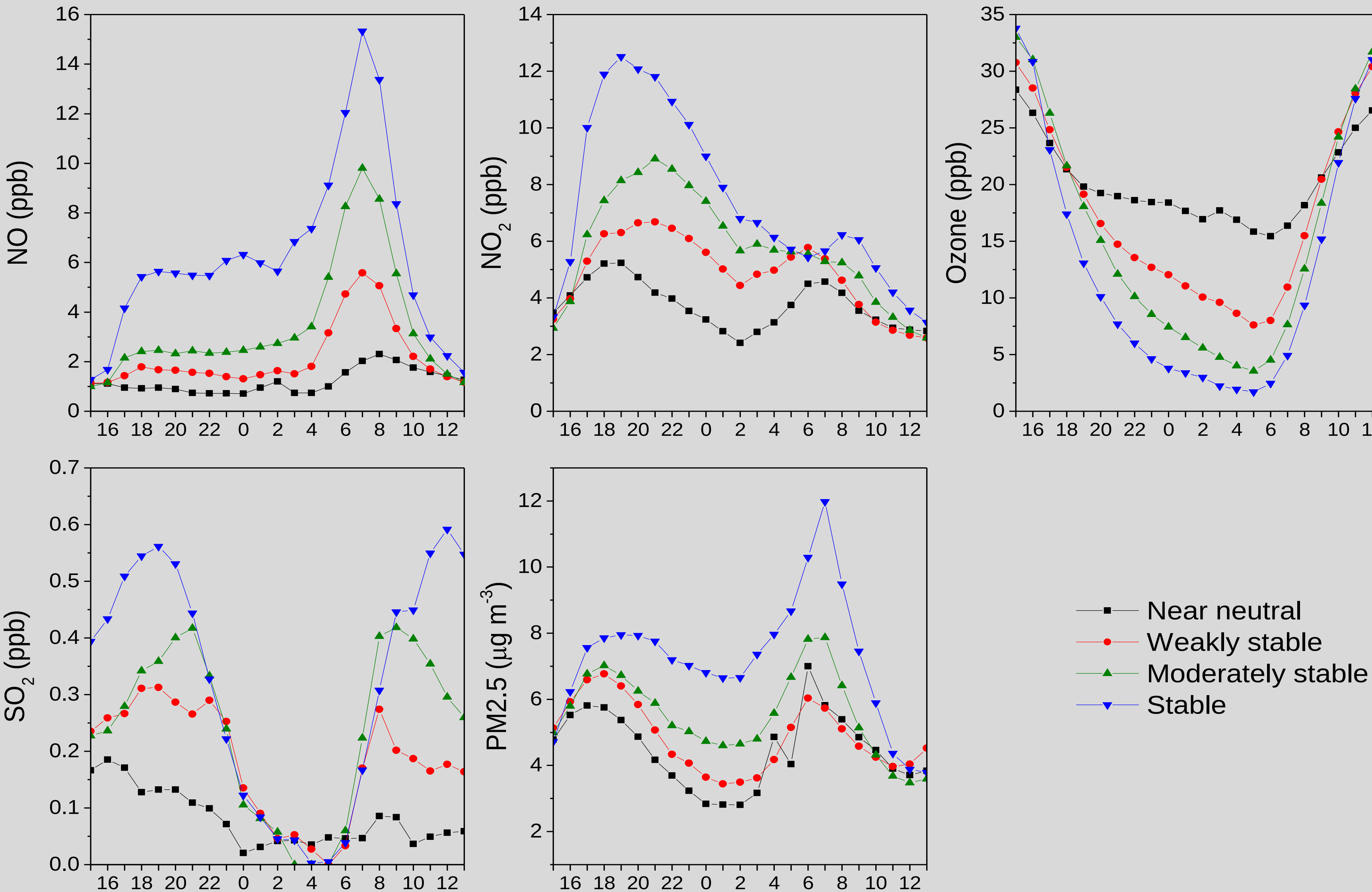






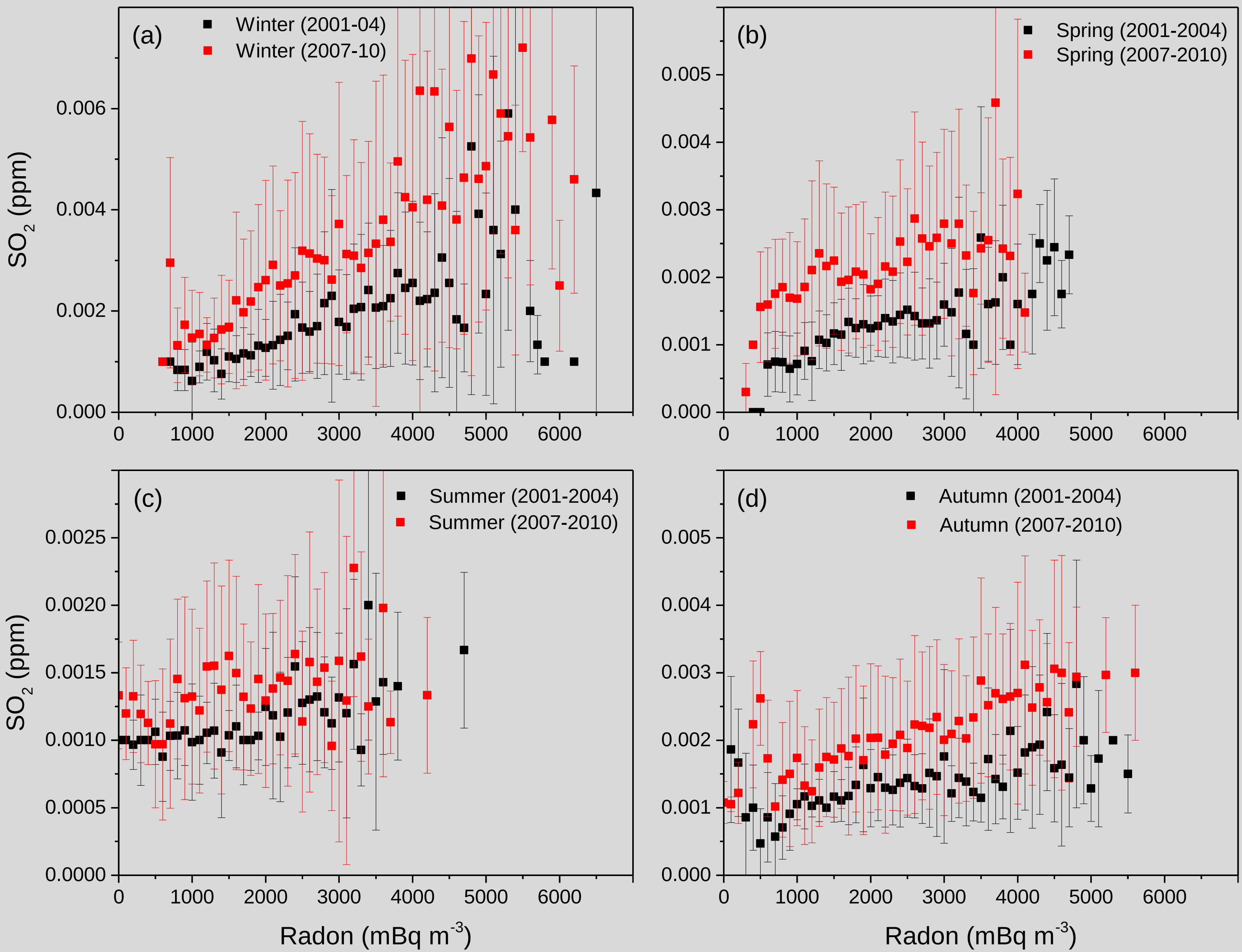


Stability effects on pollution concentration



Completely independent of site meteorology

Identifying "background" pollutant concentration



Conclusions

- Radon can be used to improve representation of near-surface sources in distant fetch regions
- A diurnal sampling window for obs. at island sites can help to match the temporal and spatial scale to simulations
- Not accounting for these effects may lead to significant errors in emission estimates
- Radon can also be used for (i) urban pollution studies, (ii) background characterisation, (iii) regional inventory assessment, and (iv) pollution model evaluation studies.

Thank You

Chambers, S.D., C-.H. Kang, A.G. Williams, J. Crawford, A.D. Griffiths, K-.H. Kim and W-.H. Kim. **Improving the representation of cross-boundary transport of anthropogenic pollution in Southeast Asia using Radon-222**. Aerosol and Air Quality Research, in press, October 2015,

Crawford, J., Chambers, S.D., C-.H. Kang, Griffiths, A. and W-.H. Kim. **Analysis of a decade of Asian outflow of PM10 and TSP to Gosan, Korea; also incorporating Radon-222.** *Aerosol and Air Quality Research*, 6, 529-539, 2015.

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