

# Improving estimates of pollution outflow at Gosan using $^{222}\text{Rn}$

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The best understanding of climatic, ecological and health effects from increasing Southeast Asian emissions will likely be achieved by models coupled to detailed emission inventories and remote sensing data. To improve model accuracy and forecast horizons, careful evaluation against appropriate observations is essential. To minimize the chance of misleading comparisons, it is important to ensure ground-based reference observations are well matched with the model output, especially regarding fetch regions and scales of observation. To quantify upstream emissions based on ground-based observations it is necessary to: (i) understand the measurement “footprint”, (ii) identify observations most representative of air that has been in good contact with the surface over which it has travelled, and has not been significantly diluted by fronts or deep convection in trans-

it, (iii) ensure observations are representative of the whole boundary layer (BL), (iv) minimize the influence of local emissions, (v) characterize changes in mixing depth, and (vi) characterize evolving “background” concentrations.

Trace gas sampling at Gosan is conducted by the Korean Ministry of Environment, and meteorological observations by the Korean Meteorological Administration. CO measurements are made by NDIR absorption (detection limit 0.03 ppm and accuracy was 10%), and SO<sub>2</sub> by UV fluorescence (detection limit 0.5 ppb and an accuracy of 3.2%).<sup>[5]</sup> Direct, hourly radon observations have also been made at Gosan since 2001.<sup>[4]</sup> Radon is an unreactive poorly-soluble radioactive gas of terrestrial origin that is an ideal tracer of transport and mixing.<sup>[1,2,3]</sup> The detector is calibrated monthly, has a lower limit of de-

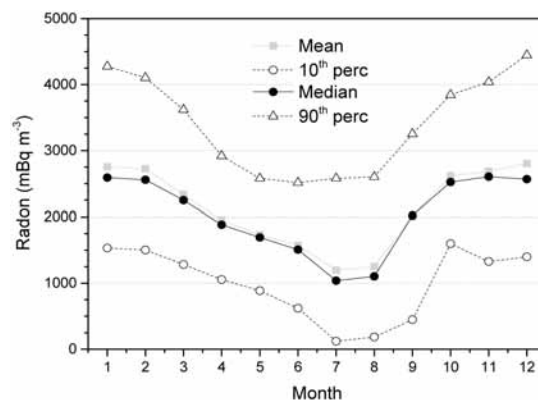
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tection of  $\sim 0.04 \text{ Bq m}^{-3}$ , and uncertainty of  $\sim 12\%$ . Jeju Island is sparsely populated, so the main Gosan pollution sources are: China (500 km west), Kyushu, Japan (200 km east), and mainland Korea (100 km north).

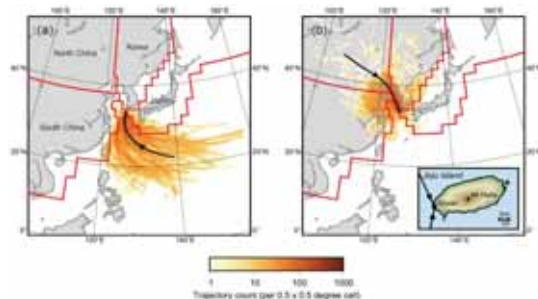
The seasonal radon cycle (Figure. 1) was characterized by a winter maximum and summer minimum, corresponding to extremes of terrestrial influence during the winter and summer monsoon periods (e.g. Figure. 2). Although sparsely populated, under certain conditions local emissions can contaminate Gosan observations. The diurnal radon cycle (Figure. 3a) is characterized by a morning maximum and afternoon minimum. In the afternoon, when the BL is well-mixed, the measurement footprint is large (representative of distant fetch regions). At night, radon accumulates until sunrise. Since radon has a terrestrial source, and the nearest alternative land is over 100 km away, the nocturnal radon accumulation must be a result of local influences. The diurnal CO cycle (Figure. 3b) is similar to radon. Therefore the observed nocturnal CO accumulation is also from local sources. By contrast, there are few large  $\text{SO}_2$  sources on Jeju, so at night, when the nocturnal inversion isolates surface observations from the influence of remote sources, a gradual ( $1.3\% \text{ h}^{-1}$  in summer) decline in  $\text{SO}_2$  is observed until sunrise (Figure 3c). Consequently, when using Gosan observations to characterize emissions from remote fetch regions, a 4-5 hour diurnal sampling window

near midday should be imposed.



**Figure 1**

10-year composite monthly mean and distribution of Gosan radon concentrations.



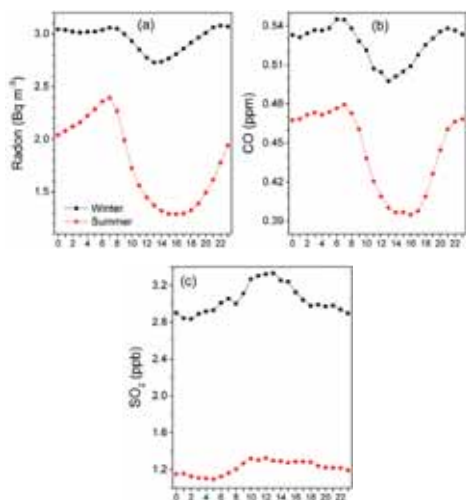
**Figure 2**

Trajectory density plots for Gosan fetch during the summer and winter monsoons.

A corollary of these observations is that Gosan aerosol samples integrated over 24-hour periods may contain a “local source” bias. Furthermore, ground-based observations reported when the BL is well-developed are better suited to model evaluation, since model spatial or vertical resolution may be insufficient to resolve the nocturnal boundary layer or island effects.

The dominant terrestrial fetch regions contributing to anthropogenic pollution observed at Gosan are: South China, North China,

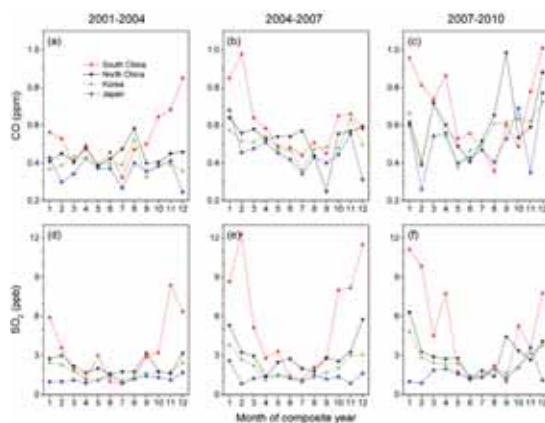
Korea and Japan (Figure. 2). Fetch regions were assigned hourly based on the mean air mass location over its most recent 24 hours of land contact, as indicated by 5-day NOAA Hysplit v4.0 back trajectories. Radon's physical characteristics ensure that an air mass' radon concentration will be closely linked to terrestrial influence over the past 2-3 weeks. For a given fetch, the higher an air mass' radon concentration, the longer it has spent in contact with surface sources, or the less dilution it has been subjected to, and the more likely observations are to be representative of surface-based emissions over that fetch. Here, "dilution" is understood to be a result of either (i) tropospheric injection, during fronts or other severe weather events, or (ii) deep convection, which can vent emissions from the BL<sup>[6, 7]</sup>.



**Figure 3**

Summer and winter diurnal cycles of (a) radon, (b) CO and (c) SO<sub>2</sub>, at Gosan.

To select observations most representative of emissions from each fetch region, we (i) selected only observations within the 5-hour diurnal sampling window to exclude local influences and mixing effects, (ii) retained only those air masses with radon greater than the monthly median value for each fetch region (assuming that low radon events are poorly representative of surface-based sources due to dilution or limited interaction with the corresponding BL).

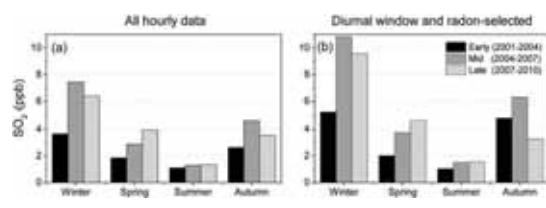


**Figure 4**

Monthly-mean CO and SO<sub>2</sub> for each fetch region across the decade, based on measurements within the diurnal sampling window and upper 50% of radon concentrations.

Mean monthly CO and SO<sub>2</sub> values by fetch region based on the sampling method proposed here are summarized in Fig. 4. Across the decade, results have been grouped in three 4-year composites: 2001-2004; 2004-2007 and 2007-2010. In general, emissions increased across the decade, were highest from South China and lowest from Japan.

Figure. 5 compares seasonal mean SO<sub>2</sub> across the decade from South China based on all hourly data, and the procedure described here. Our findings indicate that unless local influences are minimized, and representative air masses sought, emissions from South China are likely to be substantially underestimated (less so in the case of the other fetch regions). Furthermore, unless appropriate care is taken to ensure observations are representative of their intended fetch regions, and to better match vertical and horizontal scales of observations to model grid cell dimensions, results of subsequent evaluations could be misleading.



**Figure 5**

Seasonal mean SO<sub>2</sub> across the decade from South China based on (a) all hourly data, and (b) the sampling method proposed in this study.

## References

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