P2.5 INFLUENCE OF TURBULENT MIXING AND AIR CIRCULATION IN THE LOWER ATMOSPHERE ON FETCH AREAS OF SELECTED WMO ATMOSPHERE WATCH BASELINE AIR POLLUTION STATIONS

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1. INTRODUCTION

The World Meteorological Organisation (WMO) established the Global Atmosphere Watch (GAW) Programme in 1989. The scientific goals of GAW relate to investigating the role of atmospheric chemistry in global climate change, and include: understanding the complex mechanisms with respect to natural and anthropogenic atmospheric change; and improving the understanding of interactions between the atmosphere, ocean, and biosphere.

At present there are 24 “global” stations in the GAW network, tasked with the mission to make reliable, comprehensive observations of the chemical composition and selected physical characteristics of the atmosphere on global and regional scales. With a view to characterising long term (decadal +) trends in the baseline composition of the global atmosphere, global GAW stations are situated in remote locations and considered representative of large geographic fetch areas. The network of global stations is supplemented by numerous “regional” and “contributing” stations worldwide.

When interpreting results from these stations it is important first to consider how seasonally changing fetch regions and local mixing affect long-term observations at each site, and how comparable observations are between stations in the network in a global context.

The combined influence of terrestrial source distribution, species’ entrainment into air parcels analysed at the stations, and the parcels’ transport in the lower atmosphere, result in station-specific geographic areas (fetch regions) which are over- or under-represented in station records. Hence, interpreting species’ variability requires, at least, that extremes of fetch areas be independently delineated and their temporal variability understood and documented. To these ends, the aim of this study is to compare and contrast seasonal changes in local mixing and regional-scale air mass fetch between three GAW stations (two global and one regional) using hourly radon concentration data collected between Jan-2002 and Dec-2006.

Mauna Loa Observatory (MLO; Hawaii), and Cape Grim Baseline Air Pollution Station, (CGBAPS; Tasmania, Australia), are well established global GAW stations, and Gosan Station (Jeju Island, South Korea) is a regional GAW station that is playing an increasingly important role in the investigation of natural and anthropogenic pollution emissions from East Asia.

2. METHODS

Irrespective of whether it is natural or anthropogenic, most atmospheric pollution has terrestrial sources. Due to the transient nature and often complex source-sink mechanisms of many aerosols and other atmospheric constituents, we chose to use the passive tracer radon-222 – an unambiguous indicator of terrestrial influence on an air mass – to investigate fetch regions and local mixing at each site.

Radon is a naturally occurring, radioactive, noble gas, with a low solubility in water. It is emitted from terrestrial surfaces at an approximately constant rate on diurnal timescales and the emissions are assumed to be uniform on local to regional scales. Terrestrial emissions of radon are at least two orders of magnitude higher than oceanic emissions, enabling independent differentiation between air parcels predominantly influenced by land or water. The half-life of radon (3.8-days) is comparable to the lifetimes of many short-lived atmospheric species and is optimum for local mixing studies, since it is long compared with typical turbulent timescales (≤1-hour), but short enough to constrain the concentration of radon in the free troposphere to be 1-3 orders of magnitude lower than near surface values. The combination of these properties makes radon an excellent tracer for atmospheric transport and mixing studies.

Atmospheric radon concentration is measured at each site using 1500 L dual flow loop, two filter detectors (Zahorowski et al., 2005); although at CGBAPS a 5000 L detector is also in operation. These kinds of detectors are designed for multi-year deployment and provide continuous hourly radon concentration data. They have a 45 minute response time with a lower limit of detection of 10 and 40 mBq m^3 for the 5000 L and 1500 L models, respectively.

We used seasonal plots of diurnal composite radon concentration at each site to investigate the influence of local mixing and topography on routine measurements. In addition we used radon concentrations in conjunction with back-trajectory analyses to characterise seasonal changes in fetch region for each site instead of wind sector analysis, since the direction of an air mass' final approach to a site was often not representative of its long-term fetch.

10-day back trajectories were generated hourly for each site using the PC version of NOAA Air Resources Laboratory's (ARL) HYbrid Single-Particle Lagrangian Integrated Trajectory model HYSPLIT [version 4.6; Draxler and Hess, 1998] and a Visual Basic batch processing tool. The resulting collection of trajectories was incorporated into a database along with the calibrated hourly radon concentrations. Groups of trajectories delineating fetch regions corresponding to extremes of terrestrial influence were then selected by season based on corresponding radon concentrations ($\geq 90^{th}$ percentile: high events; $\leq 10^{th}$ percentile: low events).

3. RESULTS AND DISCUSSION

3.1 Cape Grim Baseline Air Pollution Station

Located at 90m above sea level (asl) on an exposed bluff on the northwest coast of Tasmania, and more than 30 km from the nearest significant township, Cape Grim is an ideal site for observing long term changes in the baseline global atmosphere. South of Tasmania the Southern Ocean offers uninterrupted oceanic fetch for air mass transit times in excess of two weeks. The site has three main fetch regions, which are broadly categorised into the following wind direction sectors: oceanic, $190 \leq \theta < 280$; mainland Australia, $280 \leq \theta < 90$; Tasmania, $90 \leq \theta < 190$). Since changes in air mass origin between these fetch regions are driven synoptically (timescale 1-2 weeks), as opposed to changes in regional or seasonal circulation patterns, the oceanic fetch region is well represented in measurements throughout all months of the year.

On an annual average basis almost half of all air masses arriving at Cape Grim have experienced long-term oceanic fetch and would thus be suitable for characterising baseline atmospheric conditions for the Southern Hemisphere.

The frequent occurrence of oceanic fetch conditions at Cape Grim has a strongly stabilising effect on the depth of the atmospheric boundary layer. Consequently, only a weak concentration / dilution signal for atmospheric constituents was found as a result in diurnal changes in mixing depth. A composite plot based on all hourly observations indicated a weak diurnal signal (amplitude 0.06 Bq m^{-3}) in the CGBAPS radon record. This signal was characterised by a broad morning maximum and an afternoon minimum. Repeating the analysis using observations made only during times of oceanic fetch, when baseline monitoring occurs, the amplitude of the diurnal cycle became negligible (0.01 Bq m^{-3}). Therefore baseline sampling at CGBAPS during oceanic fetch conditions can be performed independently of the time of day.

Due to the nature of synoptic systems that pass Cape Grim, after a wind direction change to the oceanic sector radon concentrations typically decrease for the first 12-24 hours to a relatively stable value representative of the baseline atmosphere. Usually, after 4-5 days in the oceanic sector, radon concentrations then begin to rise once more preceding a change in fetch sector. Consequently, air masses which persist in the oceanic sector for 2-4 days (inclusive) are referred to as "deep" oceanic events and are most suited to baseline sampling.

The 10-day fetch region for deep oceanic events covers a large portion of the southern Indian Ocean, extending 8000-10,000 km WSW of Cape Grim (Figure 1). Very little seasonal variability was observed in the shape or extent of this oceanic footprint for deep oceanic events.

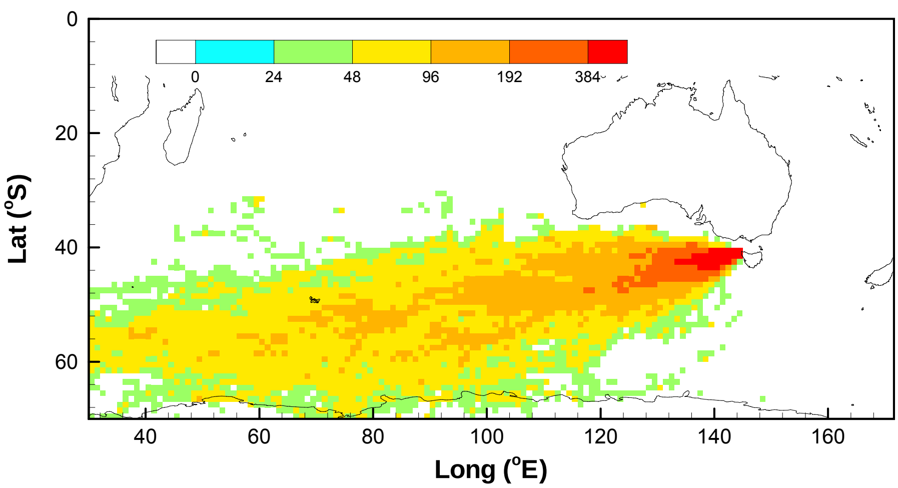

Figure 1: 1°x1° resolution back trajectory density function for CGBAPS deep oceanic events.

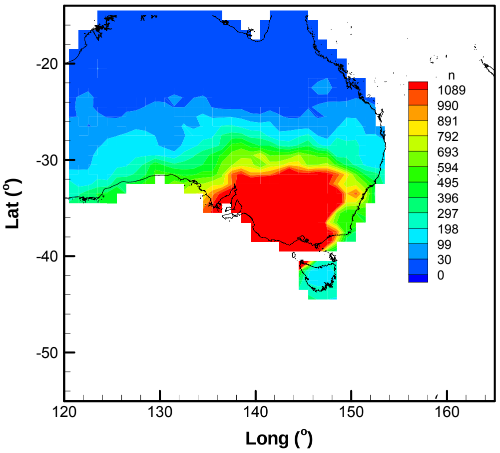

Figure 2: 1°x1° resolution back trajectory density function of CGBAPS Mainland sector observations.

Non-oceanic events, mainland Australian events in particular, can be used to characterise anthropogenic emissions and to estimate emissions inventories of climatically active gases from southern Australia (Figure 2). Consequently the size and seasonal evolution of the CGBAPS terrestrial footprint is of particular interest. The portion of mainland Australia represented by mainland events is limited primarily to the Victorian region (Figure 2). A strong seasonal variability in the CGBAPS terrestrial footprint was observed (Figure 3), such that only from May through August of each year do mainland events spend sufficient time over land to be considered representative of regional terrestrial emissions.

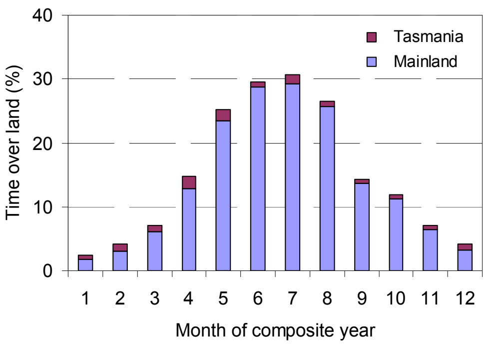

Figure 3: Monthly mean fraction of 10-day trajectories corresponding to Mainland sector events that were in contact with land.

3.2 Mauna Loa Observatory

Mauna Loa Observatory is situated on the northern slope of the Mauna Loa volcano, Hawaii, in the central eastern Pacific Basin. At an elevation of 3,394 m asl it is not subjected to the effects of changing atmospheric mixing depths in the same sense as most low lying stations, and provides an opportunity for direct sampling of air from the northern hemisphere's lower troposphere. On the windward side of Hawaii the undisturbed oceanic fetch extends 6000-8000 km. At face value the remote location of this site, as well as minimal influence of vegetation and human activity, provide an ideal setting for monitoring constituents of the global atmosphere that are implicated in climate change. However, despite the setting, the suitability of MLO as a platform to observe long-term changes in the baseline global atmosphere is strongly time dependent.

The composite diurnal MLO radon concentration exhibits a pronounced diurnal signal characterised by a broad early morning minimum and sharp mid-afternoon maximum (Figure 4). While the amplitude of this signal is small (0.08 Bq m^{-3}) it is almost 180° out of phase with diurnal signals of atmospheric constituents at lower elevation sites. The nature of this signal is attributable to local topographic effects, specifically, the anabatic and katabatic winds on the slopes of the Mauna Loa volcano. During the day, anabatic winds (flowing up the face of the volcano) contaminate MLO observations with radon and other trace species emitted locally from the island. At night katabatic flow dominates. Cool air flows down the face of the volcano, drawing tropospheric air to the site. Only under these conditions (ie. during the nocturnal sampling window shown in Figure 4), are observations representative of tropospheric conditions.

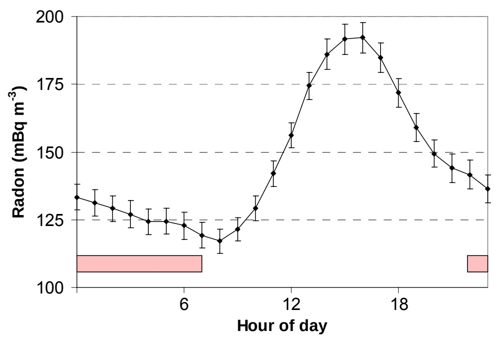

Figure 4: Diurnal composite radon concentration at MLO. Hashed bar indicates nocturnal tropospheric sampling window.

In addition to the diurnal restrictions on tropospheric sampling at MLO, air mass fetch analysis indicated large seasonal changes in the footprint of sampled tropospheric air (Figure 5 a,b). In the northern hemisphere summer fetch regions were constrained almost entirely within the Pacific Basin. Under these conditions, and within the nocturnal sampling window, observations would be representative of northern hemisphere baseline concentrations. However, at other times of the year – particularly during the winter/spring transition and mid autumn when the tropospheric jet stream is positioned near 30ºN – fetch regions for tropospheric air can reach deep into continental Asia. When assisted by the jet stream, air with a strong terrestrial signature (that has been lifted by cold fronts or deep convective processes over Asia), can reach MLO in less than a week. Under these conditions, even during the suggested nocturnal sampling window, measurements may not be representative of global baseline values. An inspection of the corresponding radon concentrations would be required to determine the degree of terrestrial influence. Furthermore, depending on the position of the jet stream, fetch regions within continental Asia can shift from the south (dominated by anthropogenic pollution sources) to central Asia (dominated by natural sources of aerosols).

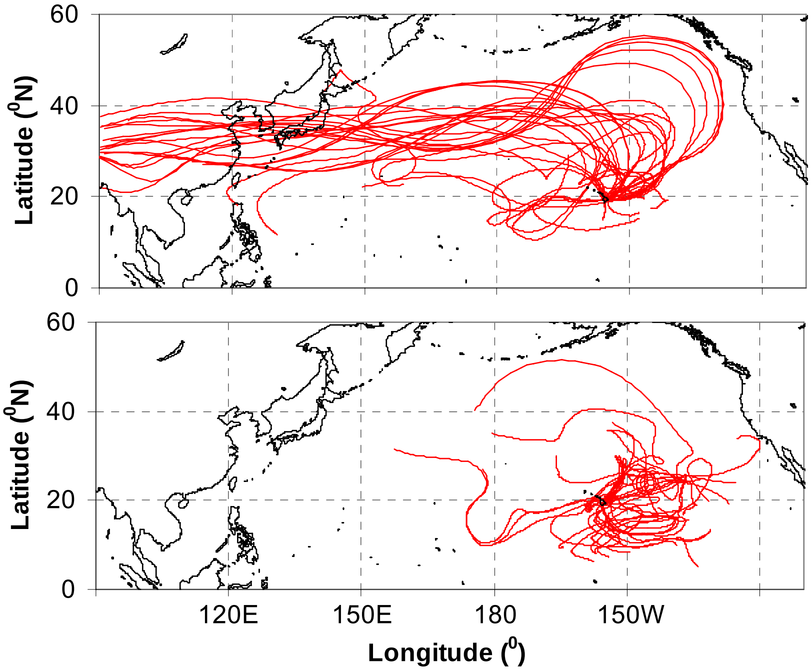

Figure 5: Example of typical Asian continental and Pacific Basin fetch regions for MLO tropospheric samples.

3.3 Gosan Station

The Gosan Station radon detector is situated on a bluff at 50 m asl on the west coast of Jeju Island, South Korea. The Chinese mainland lies approximately 500 km to the west across the Yellow Sea, the Korean Peninsula approximately 100 km to the north, and Japan approximately 300 km to the east. Beyond Jeju Island the only open oceanic fetch to this site is from the south to southeast across the East China Sea into the Pacific Ocean. The site lies near the latitudinal centre of observed winter-monsoon pollution outflow events from continental Asia and was a focal point for a large proportion of the international research effort during the first Intensive Operation Period of the Asian Aerosol Characterisation Experiment (ACE-Asia; Huebert et al., 2003).

A marked diurnal signal (amplitude 0.85 Bq m^{-3}) is evident in the composite annual radon record for this site. This signal is characterised by a brief morning maximum and a broad afternoon minimum, consistent with a significant terrestrial influence on the local atmospheric boundary layer. When considered by season (Figure 6) the range in amplitude of the diurnal signal is from 0.44 Bq m^{-3} in winter (when the most recent fetch is over the Yellow Sea) to 1.39 Bq m^{-3} in summer (when the most recent fetch of Pacific Ocean air masses is terrestrial, 20 to 60 km across Jeju Island).

Gosan Station is a regional (not global), GAW network site. Based on the magnitude and nature of the diurnal cycle at Gosan, measurements in the early to mid-afternoon (when atmospheric boundary layer depths are maximised) would be most representative of regional atmospheric conditions of the prevailing fetch region.

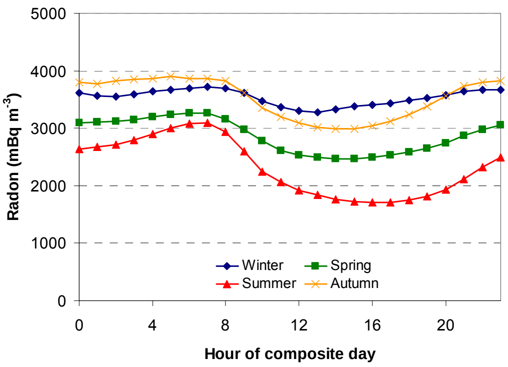

Figure 6: Diurnal composite plots by season at Gosan Station.

From mid-August, through winter, to mid-May, air mass fetch to Gosan Station is primarily off the Asian continent. For the remainder of the year fetch is primarily oceanic (Figure 7). Only for a short time (4-6 weeks of each year in summer) is the station fetch almost exclusively from the Pacific Ocean via the East China Sea. This is the sole window of opportunity for measurements at Gosan Station to be representative of near-baseline conditions for the northern hemisphere. For the months when fetch is primarily from the Asian continent the regional signal observed at Gosan Station is of mixed origins, representing the Korean Peninsular, northeast China, Mongolia and Siberia. Briefly (for 2-3 weeks in spring) the site's fetch region shifts to the west (eastern and central China).

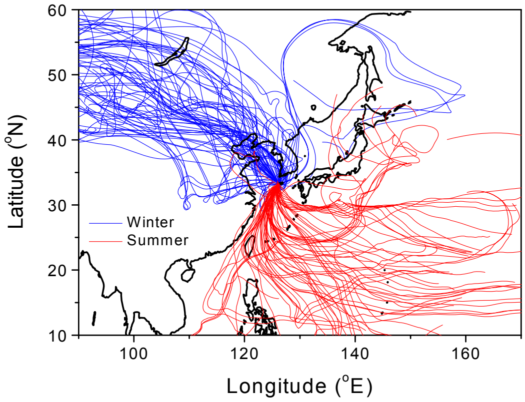

Figure 7: Examples of Gosan Station fetch regions for winter terrestrial fetch (blue) and summer oceanic fetch (red).

4. CONCLUSIONS

The Cape Grim Baseline Air Pollution Station is an ideal global GAW station. There are minimal anthropogenic sources of pollution nearby and almost half of the hourly observations each year represent conditions over an extensive unperturbed fetch of the Southern Ocean that shows little seasonal variability in extent or location. Events occurring within deep oceanic conditions can be sampled independently of time of day, and deep oceanic conditions are statistically well represented throughout all months of the year.

The Mauna Loa Observatory is ideally situated to sample directly from the lower troposphere of the northern hemisphere. However, anabatic winds associated with the local steep topography limit sampling of unperturbed tropospheric air to an 8-hour temporal window of each day. Furthermore, for more than half of each year the fetch of tropospheric air masses reaches well into continental Asian. This is of particular concern to baseline sampling efforts during the winter/spring transition, and in mid-autumn, when the combination of frontal uplift / deep convection over Asia and the tropospheric jet stream, transport air with a strong terrestrial signature to MLO in less than a week. Observations of northern hemisphere baseline conditions are best made at MLO during the northern hemisphere summer within the nocturnal sampling window. Or other times of the year when radon observations indicate minimal terrestrial influence.

The Gosan Station exhibits the greatest terrestrial influence of the three sites in this study. Consequently, observations at this site would be most representative of the regional atmosphere when mixing depth is maximised (early to mid afternoon). There is a narrow window of opportunity each summer for quasi-baseline sampling at this site. For the remainder of the year fetch regions for this site are in continental Asia, primarily northeast China, and around the Mongolia-Siberian border. Unfortunately, terrestrial fetch regions for this site lie close to, or beyond, the upper latitude threshold of regions thought to contribute significantly to Asian pollution outflow events across the Pacific Basin.

5. REFERENCES

Draxler, R. R. and Hess, G. D. 1998. An overview of the HYSPLIT 4 modelling system for trajectories, dispersion and deposition. Australian Meteorological Magazine, 47, 295-308.

Huebert, B. J., Bates, T. Russell, P. B., Shi, G., Kim, Y. J., Kawamura, K., et al. 2003. An overview of ACE-Asia: strategies for quantifying the relationships between Asian aerosols and their climatic impacts. J. Geophys. Res., 108 (D23), 8633-8633.

Zahorowski, W., S. Chambers, T. Wang, C.-H. Kang, I. Uno, S. Poon, S.-N. Oh, S. Werczynski, J. Kim and A. Henderson-Sellers. Radon-222 in boundary layer and free tropospheric continental outflow events at three ACE-Asia sites. Tellus, 57B, 124-140, 2005.