LONG RANGE TRANSPORT OF FINE PARTICULATE SOIL, SULPHATE AND BLACK CARBON ACROSS THE EAST ASIAN REGION AND BEYOND DURING 2001-03.

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INTRODUCTION

A more comprehensive knowledge of aerosols and fine particles in the atmosphere and their effects on the earth's radiation balance is critical to a better understanding of climate change and global warming [1]. Tropospheric fine particles, which may be anthropogenic or natural, have the largest uncertainty but play a significant role in climate models [2,3]. The key components of these fine particles, like sulfates, black carbon and windblown soils [1,2,4], need to be better quantified on a global, regional and seasonal scale.

The Australian Nuclear Science and Technology Organisation (ANSTO) has been using accelerator based ion beam analysis (IBA) techniques [5-8] to characterise fine particulate matter at five sites related to the large international Aerosol Characterisation Project in Asia, ACE-Asia [2-4]. These IBA techniques provide detailed information on over 20 different chemical species from hydrogen to lead [5-6]. This enabled key species, important in visibility studies, human health of urban populations [9] and climate change models, like soil, sulphates and black carbon [10] to be quantitatively determined from the total fine mass loadings across a region covering Vietnam, Korea, China, Japan and Philippines [11].

This paper reports quantitatively on seasonal variations in and the transport of several of these key chemical components of fine particulate matter across thousands of kilometers of land and sea in and around the east and southeastern Asian region. This is the longest continuous record of fine particle sampling data covering over 2.8 Mkm^2 of the Asian region collected to date. It covers four separate winter and summer seasons across the region.

Through the use of appropriate back trajectory methods and emissions models on a large regional scale [11-14], we show the origins of these very large fine particulate mass events, measured at certain times of each year, to be the industrial regions of eastern China and the desert regions of northern China.

SAMPLING LOCATIONS AND PROTOCOLS

Fine particle ($PM_{2.5}$) cyclone units and $PM_{2.5}$/ PM_{10} stacked filter sampling units were located at selected sites at Manila in Philippines (14°39'N, 121°04'E), Hong Kong (22°13'N, 114°15'E), Cheju Island South Korea (33°17'N, 126°10'E), Hanoi Vietnam (21°02'N, 105°51'E) and Sado Island off west coast of Japan (38°08'N, 138°17'E). These site locations are shown as large triangles and stars in the maps of Figs 3 to 6 discussed further below. Generally sampling occurred twice a week for 24 hours from midnight to midnight. The cyclone system sampling at 22L/min and the stacked filter system at 16 L/min. Sampling was designed to commence in January 2001 and run continuously till June 2004. However, the

Cheju Island and Hanoi sites did not commence until March 2001 and Sado Island commenced even later in September 2001. More detailed descriptions of sampling sites and regimes can be found elsewhere [3], Manila and Hanoi were the more urban sites of the five. Here we report on the first three years of sampling to December 2003 for the $PM_{2.5}$ data only as this was most relevant for the long-range transport issues discussed here.

RESULTS AND DISCUSSIONS

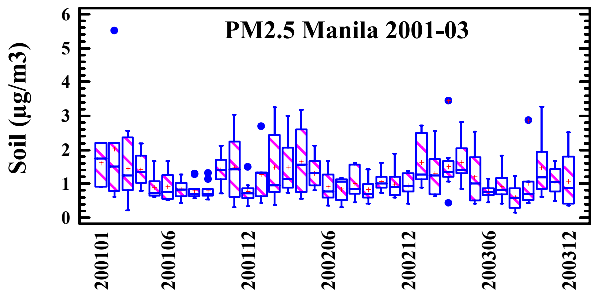
The Asian regions covered experience distinct well documented seasonal differences [2,4,11] with total fine and coarse particulate matter concentrations generally being large in colder winter months and smaller in summer or monsoonal rainy months. These have been discussed qualitatively by Huebert et al [2] and quantitatively by Cohen et al [3]. The characterisation of these total fine mass measurements by IBA methods allows partitioning of the total mass into the more interesting quantities for this study namely, soil, sulfate and black carbon (BC). Table 1 gives 3-year averages for these quantities for the five sampling sites.

$PM_{2.5}$ ($\mu g/m^3$)	Manila (272 points)	Hong Kong (276 points)	Cheju Is. (218 points)	Hanoi (273 points)	Sado Is. (190 points)
Mass	43±17(99)	27±17(122)	18±13(93)	50±33(222)	12±9(61)
Soil	1.16±0.7(5.5)	1.8±2.2(17)	1.6±2.6(22)	4.8±4.1(28)	0.83±1.5(13)
Sulfate	4.74±2.7(15)	8.9±4.9(30)	5.4±3.1(19)	9.5±6.5(39)	3.9±3.0(23)
BC	12.8±4.8(26)	2.0±1.0(6.8)	1.3±1.0(7.9)	4.2±1.7(11)	0.86±0.5(2.9)

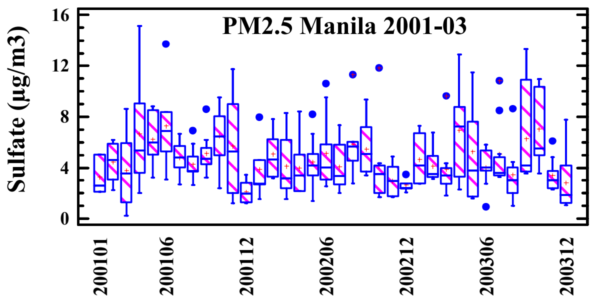
Table 1. Three year averages and 1σ standard deviations fine masses ($PM_{2.5}$) for soil, sulfate and black carbon. The values given in brackets are the 24-hour maxima for the period.

During the study period all sites expect the Sado Island site in Japan exceeded the current US EPA $PM_{2.5}$ health goals of 15 $\mu g/m^3$ annual average and 65 $\mu g/m^3$ 24-hour maxima. Soil, sulfates (as ammonium sulfate) and black carbon represented between 44% and 59% of the total fine ($PM_{2.5}$) mass, the other major component being organic matter, measured but not discussed here. Manila and Hanoi had relatively higher sulfate and black carbon due to their stronger local urban influences.

Monthly box and whisker plots for the five sampling sites for soil and sulfate ions only are shown in Fig. 1 (a) to (j). The dots represent significant outlier events. Black carbon plots were somewhat similar to the sulfate ion plots and due to space limitations could not be shown here (see also Ref. [3]).



(a)



(f)

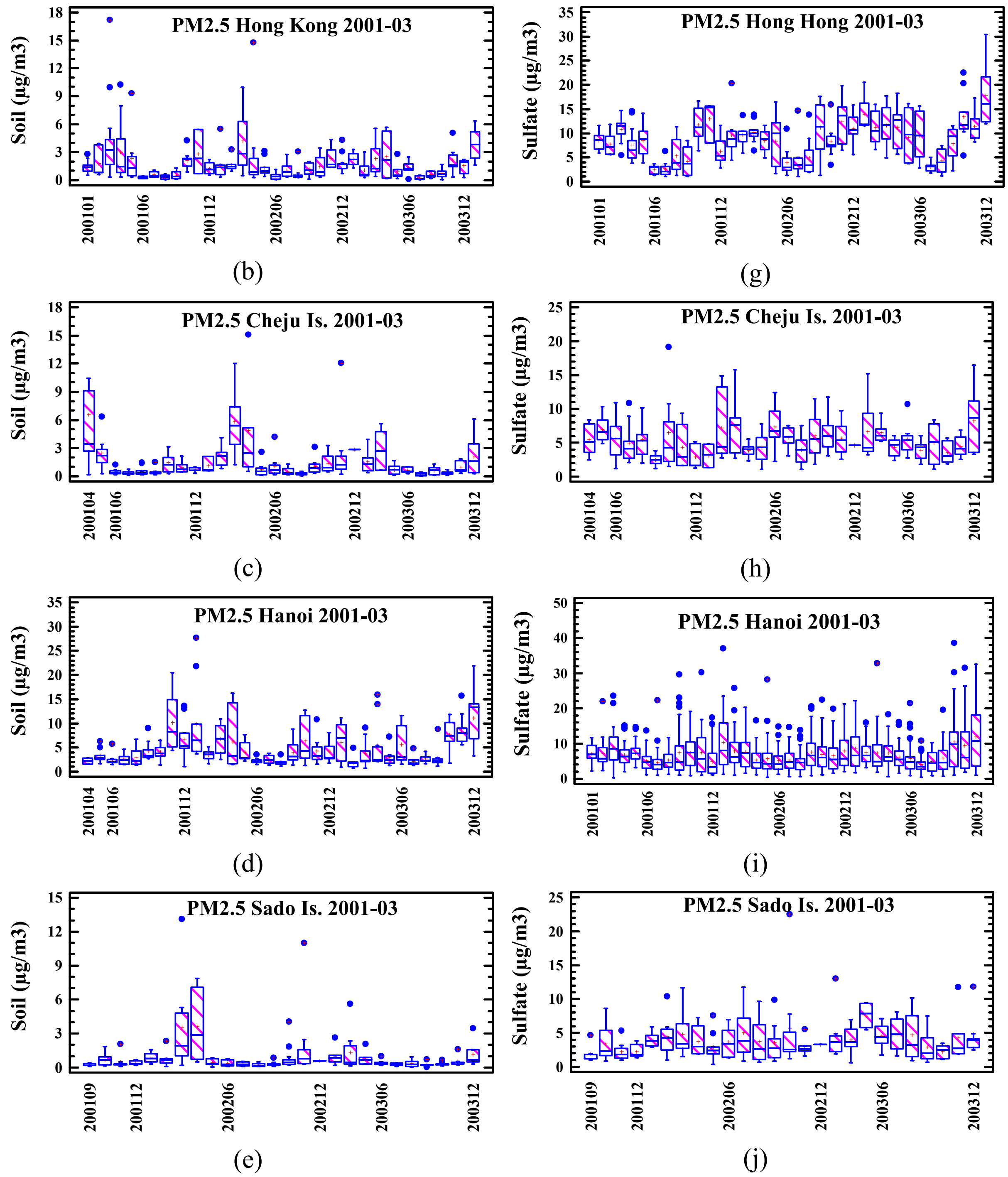


Fig. 1. Monthly box and whisker plots for $PM_{2.5}$ soil (a) to (e) and sulfate ions (f) to (j) at 5 sites for the period 2001-03. The dots represent outlier events.

Cheju and Sado Islands showed significant soil events, 3 to 10 times background, particular during the spring months of February to April each year. These are the well-documented Asian Dust Events (ADEs) [2,4,11]. Fig. 2 shows the hourly wind event rose for the Hong Kong site for the period January to April 2002, demonstrating that the prevailing winds are predominantly from the north to east sector at this time of the year for the region. Later in the year, during summer, the prevailing winds switch 180º and come more from the south and west sector. This seasonal pattern is typical for most of our sampling region.

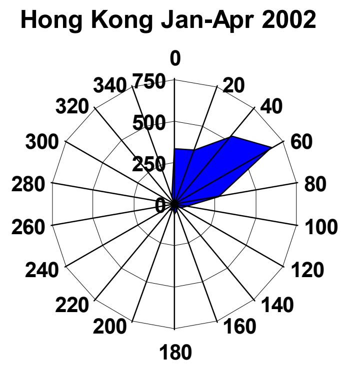


Fig. 2. Hourly wind event rose for the Hong Kong site for period January to April 2002.

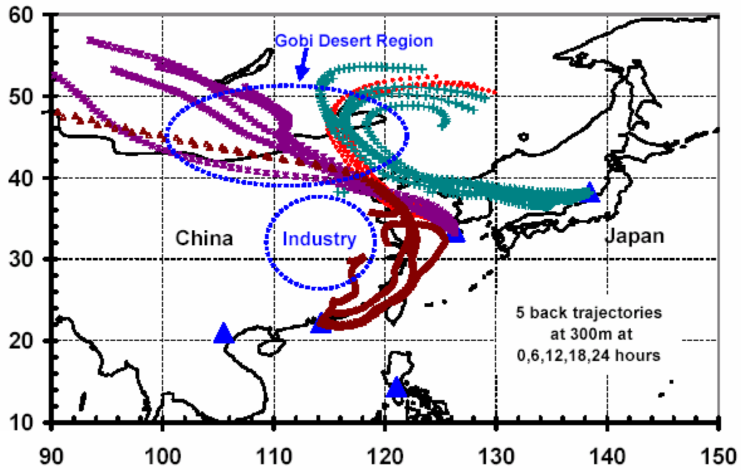


Fig. 3. 10 day back trajectories for Hong Kong, Cheju and Sado Islands for the period 10-17 April 2002.

Fig. 3 shows 10 day back trajectories plots for the Hong Kong, Cheju and Sado Island sites for the period 10-17 April 2002. The trajectories were calculated every 6 hours starting at 300m above each site [12,13]. They show major soil transport routes (covering 30 degrees of longitude and latitude) from the Gobi and Taklamakan desert regions of China, near and west of the Mongolian border. Some trajectories are over 5,000km long and the Gobi desert regions are over 3,000km west of our Sado Island site.

Similar back trajectories have been calculated for the extreme sulfate events at Hanoi, Hong Kong and Manila during 22-26 October 2003 [see dots in Figs. 1(f), (g) and (i)]. The 10 day back trajectories for these 3 sites are shown in Fig. 4. Sulfate ions were transported hundreds of kilometres from the central and eastern industrial regions of China into Hong Kong and Hanoi during this period. It also shows long range transport of air masses into Manila from southern Japan, Korea and eastern China in the Beijing region. These trajectories extend back into the Gobi desert regions of northern China, however if wind velocities in the region are too low insufficient soil is pushed up into the atmosphere to be transported the great distances required to reach Hanoi, Hong Kong and Manila.

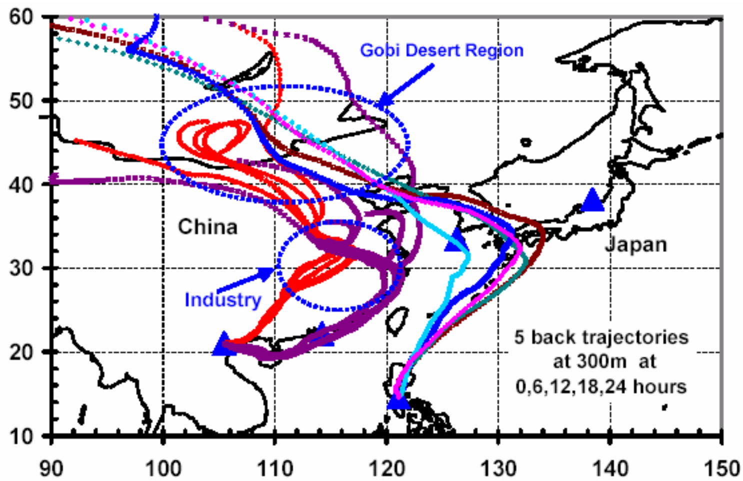


Fig. 4. 10 day back trajectories out of Hanoi and Manila for the period 22-26 October 2003.

Further evidence of this long range transport of air masses and hence both natural and anthropogenic fine particle pollution plumes can be obtained from the emissions modelling at Naval Research Labs [14], shown in Figs. 5 and 6 for soil and sulfate respectively. The 4 stars represent the sampling sites of Hanoi, Hong Kong, Cheju and Sado Islands, the Manila site was off the bottom of these maps and is not shown.

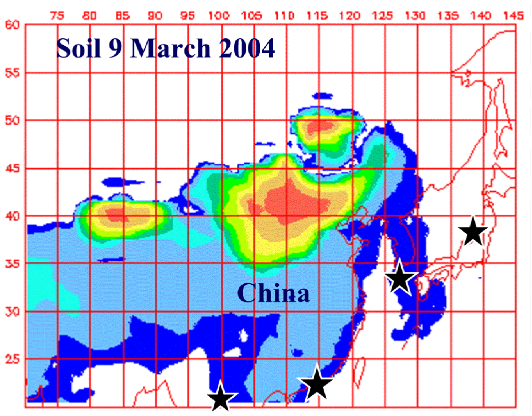


Fig. 5. Model soil emissions [14] from the east Asia region for 9 March 2004.

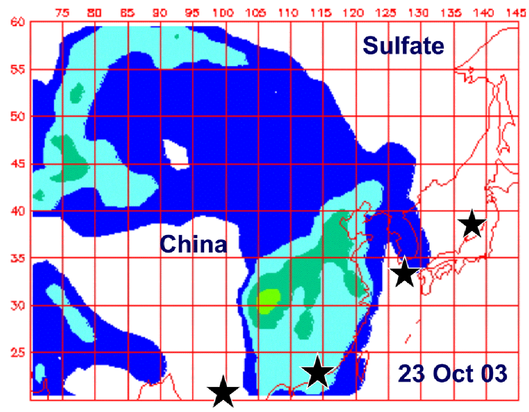


Fig. 6. Model sulfate emissions [14] from the east Asia region for 23 October 2003.

These models can be run at 6 hourly intervals for any day of the year. Fig. 5 shows the start of a major Asian Dust Event in March 2004. The Taklamakan (40°N, 85°E) and Gobi desert (43°N, 110°E) soil source regions are clearly visible. The corresponding sulfate source regions, for the October 2003 event shown in Fig. 6, are more diffuse but cover the central and east coast industrial regions of China (25°-40°N, 105°-125°E).

SUMMARY

ANSTO has collected and characterised data on fine particles in the eastern and southeastern Asia regions at five sites covering an area of more than 2.8Mkm^2 and a period of three years from 2001 to 2003 inclusive. This is a unique data set. Seasonal variations in key components of soil and sulfates have been presented. Significant outlier events have been related to long range soil and sulfate transport across national borders and oceans covering thousands of kilometres. Clearly at certain times of each year there are significant anthropogenic (sulfate and black carbon) and natural (soil) fine particle events affecting several countries. Data collected to date show annual average $PM_{2.5}$ mass concentrations in excess of current US EPA fine particulate health goals at some sites.

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