ATMOSPHERIC DEPOSITION OF PARTICULATE POLLUTANTS TO THE YELLOW SEA

G. LAMMEL [1], Y.S. GHIM [2], R. LOHMANN [1,3,6], J.A.C. BROEKAERT[4]and H.W. GAO[5]

[1] Max Planck Institute for Meteorology, Bundesstr. 53, 20146 Hamburg, Germany, lammel@dkrz.de

[2] Korea Institute of Science and Technology, Air Resources Research Center, Seoul, Republic of Korea

[3] University of Lancaster, Environmental Chemistry Dept., UK

[4] University of Hamburg, Institute for Inorganic and Applied Chemistry, Hamburg, Germany

[5] Ocean University of China, Institute for the Marine Environment, Qingdao, China

[6] now at: University of Bremen, Research Center for Ocean Margins, Germany

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INTRODUCTION

Due to rapid development (including industrialisation and mobilisation) atmospheric pollution is a severe and frequent phenomenon in eastern China. Accordingly, pollutant inputs into the adjacent seas is a concern. We report on comparative aerosol elemental composition measurements at a coastal urban and a remote island site of the Yellow Sea.

METHODS

The sites chosen were Qingdao, eastern China, Shandong province (36°05'N, 120°20'E), and Gosan on Jeju Island, Korea (33°24'N, 126°00'E). Simultaneous sampling took place 9-21 June 2003. Particulate matter was collected by high-volume sampling on quartz filters without an upper size cut-off. For analyses ICP-MS was used upon acid digestion (Sciex Elan 5000, HP 4500) except for Ca, Mg, Fe, and Al from Gosan samples which were analysed by ICP (Leeman PS1000). Transports were analysed based on analysed back-trajectory calculations for the two sites (HYSPLIT model; Draxler & Rolph, 2003). Transport occurred between the two sites during two episodes of 1-2 days each.

RESULTS

A comparison with the results of earlier measurements in the area (Liu et al., 2002) indicates a trend of increasing pollution in Qingdao in recent years: V, Cu, Zn and Pb seem to have doubled or tripled since 1995/96. Ni, Pb, V, Cu and Zn were higher enriched in aerosol (relative to the composition of the earth's crust) in 2003, the latter three very significantly, by one order of magnitude. The concentrations at Qingdao in 2003 are comparable to observations in urban areas of present-day India and of Germany in 1981/82 i.e., before abatement measures were put in place.
Concentration levels were higher at the coastal site by a factor of 2-8 for Mg, Ti, Ca, Co, Mn, Ba and Pb and by a factor of 10-25 for Al, Ca, V, Fe, Cu, Zn and Sr. Crustal enrichment factors at Gosam were similar for most elements and indicate local sources of Ti and Cd.

For the episodes of transport between the sites, dry deposition velocities (cm s^{-1}) and depositional fluxes F ($\mu g\ m^{-2}\ d^{-1}$) to the Yellow Sea are derived as:

$$v_{dep\ E} = -\ln EF_{transport\ E} * h_{transport} / \Delta t + v_{dep\ Al}$$

$$F = v_{dep\ E} * (c_{Qingdao} - c_{Gosan}) / \ln(c_{Gosan}/c_{Qingdao})$$

with enrichment factor $EF_{transport} = (E_{Gosan}/Al_{Gosan})/(E_{Qingdao}/Al_{Qingdao})$, concentrations at the two sites $c_{Qingdao}$ and c_{Gosan}, mixing depth of the marine boundary layer (we adopt 500-1000 m) $h_{transport}$ and transport time Δt = 24-54 h. For derivation of values of $v_{dep\ E}$ and F assumptions on the Al mass size distribution (based on earlier observations in the Yellow Sea; Gao et al., 1997) and $h_{Gosan} \approx h_{Qingdao}$ were made.
The atmospheric input of metals into the Yellow Sea matches with levels observed in the North Sea in the early 1990s (Schulz, 1993) for most elements, but exceeds these levels by up to one order of magnitude for Cu (27 and 22 µg m^{-2} d^{-1}) and Pb (116 and 89 µg m^{-2} d^{-1} during the two episodes of transport, each).

Ele-ment	Episode #1			Episode #2		
	$c_{Gosan}/c_{Qingdao}$ ($EF_{transport}$)	v_{dep}	F	$c_{Gosan}/c_{Qingdao}$ ($EF_{transport}$)	v_{dep}	F
Mg	0.16 (3.7)	1.12 ± 0.75	139 ± 86	0.30 (3.1)	1.36 ± 0.50	181 ± 50
Al	0.046	1.9 [a)]	1550	0.090	1.9 [a)]	1115
Ca	0.090 (2.0)	1.49 ± 0.40	521 ± 117	0.072 (0.75)	2.04 ± 0.13	886 ± 35
Ti	0.35 (7.8)	0.68 ± 1.16	75 ± 155	0.39 (4.0)	1.12 ± 0.62	118 ± 48
Mn	0.081 (1.6)	1.57 ± 0.22	19 ± 3	0.29 (3.2)	1.17 ± 0.34	18 ± 5
Fe	0.016 (0.20)	2.04 ± 0.28	1830 ± 440	0.089 (0.87)	1.95 ± 0.03	1230 ± 26
Co	0.037 (0.80)	1.95 ± 0.07	0.60 ± 0.03	< 0.12 (0.25)	0.85 ± 0.40	0.12 ± 0.06
Cu	0.039 (0.90)	1.92 ± 0.04	27 ± 1	0.16 (1.5)	1.65 ± 0.12	22 ± 1
Zn	0.015 (0.28)	2.19 ± 0.40	256 ± 53	0.13 (1.1)	1.83 ± 0.03	186 ± 3
Sr				0.048±0.05 (0.58±0.05)	2.13 ± 0.09	8.9 ± 0.6
Cd	0.25 (5.0)	0.77 ± 0.73	0.21 ± 0.20	0.29 (3.9)	1.04 ± 0.39	0.78 ± 0.29
Ba	< 0.03 (< 5.4)	0.72 ± 0.75	61 ± 65	0.66 (7.1)	0.69 ± 0.65	23 ± 20
Pb	0.039 (0.64)	2.22 ± 0.20	116 ± 10	0.26 (2.7)	1.26 ± 0.28	89 ± 20

[a)] adopted from Gao et al., 1997

Table 1. Concentration ratios, $c_{Gosan}/c_{Qingdao}$, aerosol transport enrichment factors, $EF_{transport} = (E/Al)_{aerosol\_Gosan}/(E/Al)_{aerosol\_Qingdao}$ (in brackets), deposition velocities, v_{dep} (cm s^{-1}), and dry depositional fluxes, F (µg m^{-2} d^{-1}), during transport episodes #1 and #2 between Qingdao and Gosan. Ranges given according to mixing depth ($h_{transport}$ = 750 ± 250 m) and Δt uncertainties.

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REFERENCES

Draxler, R.R., & Rolph, G.D. (2003). HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model, NOAA Air Resources Laboratory, Silver Spring, USA, *internet*: http://www.arl.noaa.gov/ready/hysplit4.html.

Gao, Y., Arimoto, R., Duce, R.A., Zhang, X.Y., Zhang, G.Y., An, Z.S., Chen, L.Q., Zhou, M.Y., & Gu, D.Y. (1997). Temporal and spatial distributions of dust and its deposition to the China Sea, *Tellus* 49B, 172-189.

Liu, C. L.; Zhang, J.; & Shen, Z. B. (2002). Spatial and temporal variability of trace metals in aerosol from the desert region of China and the Yellow Sea, J. Geophys. Res. 107, 4215, doi:10.1029/2001JD000635.

Schulz, M. (1993): Räumliche und zeitliche Verteilung atmosphärischer Einträge von Spurenelementen in die Nordsee, *PhD thesis*, *Schriftenreihe Angewandte Analytik* No. 17, Hamburg, Germany, 191 pp.