

TABLE OF CONTENTS

Summary	iii
Zusammenfassung	ix
Acknowledgments	xiii
Table of Contents	xv
List of frequently used abbreviations	xix
1 General introduction	1
1.1 The trace element selenium	2
1.2 The speciation of Se	2
1.2.1 Inorganic species	2
1.2.2 Organic species	3
1.3 Se and human health	4
1.4 Food sources of Se	5
1.5 Soil Se distribution	7
1.6 The global biogeochemical Se cycle.	8
1.7 Outline of thesis	12
1.8 References	15
2 Terrestrial selenium distribution in China is potentially linked to monsoonal climate	25
2.1 Abstract	26
2.2 Introduction	26
2.3 Methods.	28
2.3.1 Geochemical analyses	28

2.3.2	Fe K-edge XAS.	30
2.3.3	Se leaching tests	30
2.3.4	Statistical analysis of trace element and proxy data	31
2.3.5	Wavelet analysis	31
2.3.6	Principal component analysis (PCA)	31
2.4	Results.	32
2.4.1	Sources of Se and common trends with global benthic $\delta^{18}\text{O}$	32
2.4.2	Se linked to monsoon proxies	33
2.5	Discussion	34
2.6	Acknowledgements	38
2.7	Supplementary Information.	39
2.7.1	Supplementary discussion.	39
2.7.2	Supplementary figures	43
2.7.3	Supplementary tables	49
2.8	References	53
3	Increasing atmospheric Se deposition recorded in Chinese Loess during the Holocene	59
3.1	Abstract	60
3.2	Introduction	60
3.3	Study sites and methods.	62
3.3.1	The Chinese Loess Plateau.	62
3.3.2	The Mangshan and Luochuan sequences	63
3.3.3	Geochemical analyses	64
3.3.4	Enrichment factors.	67
3.3.5	Regression model.	68
3.3.6	X-ray absorption spectroscopy	68
3.4	Results.	68
3.4.1	Trends in trace element concentrations	68
3.4.2	Comparing trace element concentrations to pedogenic proxy data	70
3.5	Discussion	74
3.5.1	Predictions of Se, As and Pb trends	74

3.5.2	Enrichment in Se since MIS 3	76
3.5.3	A biogenic marine Se source?	77
3.6	Conclusions.	79
3.7	Acknowledgments	80
3.8	Supplementary information.	81
3.8.1	Supplementary figures	81
3.8.2	Supplementary tables	87
3.9	References	90
4	Marine primary productivity as a potential source of trace elements in rainfall	99
4.1	Abstract	100
4.2	Introduction	100
4.3	Methods and materials	102
4.3.1	Rainwater chemistry dataset from Plynlimon, UK	102
4.3.2	Origin of air masses.	103
4.3.3	Exposure of air masses to chlorophyll a.	104
4.3.4	Proxy of sea spray.	105
4.3.5	Exposure of air masses to anthropogenic emission proxies	105
4.3.6	Multivariate modeling	106
4.4	Results and Discussion.	108
4.4.1	Precipitation chemistry	108
4.4.2	Sensitivity analysis	109
4.4.3	Potential sources of rainwater constituents	111
4.4.4	Relationship between marine air exposed to chlorophyll a and precipitation chemistry	111
4.4.5	Primary productivity as a source of trace elements?	112
4.5	Conclusions.	116
4.6	Acknowledgements	117
4.7	Supplementary information.	118
4.7.1	Supplementary figures	118
4.7.2	Supplementary tables	123

4.8	References	124
5	Conclusions and Future Outlook	135
5.1	Linking Se concentrations to precipitation proxies in terrestrial sediments and soils. .	136
5.2	Influences of marine biological productivity on modern rainwater Se concentrations .	139
5.3	Implications and concluding remarks	141
5.4	References	142