

## Supplementary Material

Table S1. Summary of monthly wet deposition data in units of mm. Data over the periods shown in Table 1 for each site were used to obtain monthly-averaged deposition for rain and snow, where only days with recorded precipitation are included in the analysis. “n” represents the number of days in each month with either rain or snow.

	Mesa Verde				Chiricahua				Organ Pipe				Bryce Canyon				Gila Cliff				Bandelier																			
	Rain Ave	$\sigma$	n	Rain Ave	$\sigma$	n	Rain Ave	$\sigma$	n	Rain Ave	$\sigma$	n	Rain Ave	$\sigma$	n	Rain Ave	$\sigma$	n	Rain Ave	$\sigma$	n	Rain Ave	$\sigma$	n																
Jan	7.43	6.12	0.25	3.84	3.71	3.91	7.65	13.88	3.13	1.27	0.06	3.47	4.69	1.44	0.98	0.53	0.50	0.00	5.37	7.98	3.94	2.87	3.24	1.94	3.98	4.02	3.63													
Feb	4.23	3.03	0.75	3.80	3.92	3.45	4.15	5.56	3.50	2.54	1.80	0.13	3.86	4.24	2.75	4.45	3.29	0.81	0.00	4.30	5.03	5.88	2.94	3.26	2.19	3.01	3.39	3.00												
Mar	1.86	1.92	0.75	4.47	5.49	2.82	4.61	4.07	2.38	5.84	9.04	0.69	3.16	3.19	2.31	3.47	3.40	1.56	0.00	3.94	4.87	5.63	4.87	5.85	10.13	4.10	5.39	11.81												
Apr	3.60	6.07	1.50	2.38	2.26	1.91	8.08	10.49	1.38	5.14	6.24	0.56	3.65	4.40	2.25	3.94	4.97	2.19	0.00	4.06	6.24	6.12	2.06	4.06	6.24	3.88	0.00	5.37	7.98											
May	2.98	3.36	3.13	4.25	6.14	1.73	3.96	3.95	0.63	1.97	2.09	2.13	3.65	6.12	2.06	4.06	6.24	3.88	0.00	4.44	6.13	5.31	2.81	4.44	6.13	5.31	0.00	5.37	7.98											
Jun	3.65	4.73	2.75	4.06	6.83	3.91	0.76	0.36	0.88	3.70	3.43	2.25	5.37	7.86	2.81	4.44	6.13	5.31	0.00	3.94	4.39	3.06	2.83	2.87	1.25	3.72	5.10	2.19	0.00	5.37	7.98									
Jul	4.29	5.37	5.44	5.20	7.61	13.64	4.34	8.46	5.50	3.73	3.92	5.63	4.87	5.85	10.13	4.10	5.39	11.81	0.00	4.87	5.85	10.13	4.10	5.39	11.81	0.00	5.37	7.98												
Aug	4.12	5.33	8.00	5.98	8.74	15.09	9.00	11.83	5.25	3.84	3.96	5.50	5.16	5.91	11.81	5.68	7.62	10.38	0.00	4.86	5.43	0.44	5.63	7.75	1.94	1.27	1.51	0.56	0.00	5.37	7.98									
Sep	6.31	8.12	6.06	4.51	7.00	6.36	5.67	8.54	2.25	6.07	9.06	4.06	5.94	8.37	7.13	4.23	5.83	8.06	0.00	4.06	5.94	8.37	7.13	4.23	5.83	8.06	0.00	5.37	7.98											
Oct	4.86	5.02	4.69	4.32	6.82	5.36	6.94	8.87	1.63	7.95	9.76	3.13	6.87	8.51	4.25	6.36	7.19	4.94	0.00	4.86	5.43	0.44	5.63	7.75	1.94	1.27	1.51	0.56	0.00	5.37	7.98									
Nov	2.31	2.35	0.56	4.37	6.26	2.64	3.59	3.98	1.00	4.86	5.43	0.44	5.63	7.75	1.94	1.27	1.51	0.56	0.00	4.86	5.43	0.44	5.63	7.75	1.94	1.27	1.51	0.56	0.00	5.37	7.98									
Dec	4.64	7.93	0.25	4.05	4.00	3.00	7.78	9.89	2.63	0.00	3.44	4.50	0.69	3.81	3.91	0.25	0.00	3.81	3.91	0.25	0.00	3.44	4.50	0.69	3.81	3.91	0.25	0.00	5.37	7.98										
<u>Snow</u>																																								
Jan	5.25	6.04	4.94	17.00	7.59	1.55	0.00	5.37	7.98	3.94	2.87	3.24	1.94	3.98	4.02	3.63	0.00	5.37	7.98	3.94	2.87	3.24	1.94	3.98	4.02	3.63	0.00	5.37	7.98											
Feb	4.25	4.44	4.75	7.00	6.17	0.64	0.00	4.30	5.03	5.88	2.94	3.26	2.19	3.01	3.39	3.00	0.00	4.30	5.03	5.88	2.94	3.26	2.19	3.01	3.39	3.00	0.00	5.37	7.98											
Mar	3.82	2.81	3.13	2.00	2.03	0.18	0.00	3.94	4.39	3.06	2.83	2.87	1.25	3.72	5.10	2.19	0.00	3.94	4.39	3.06	2.83	2.87	1.25	3.72	5.10	2.19	0.00	5.37	7.98											
Apr	7.66	10.24	0.88	1.00	1.27	0.09	0.00	4.60	4.72	3.38	2.73	2.70	0.25	3.35	4.10	0.94	0.00	4.60	4.72	3.38	2.73	2.70	0.25	3.35	4.10	0.94	0.00	5.37	7.98											
May	2.29	0.06	0.00	0.00	0.00	0.00	0.00	1.27	0.59	0.50	0.00	0.00	0.00	0.00	0.00	0.00	0.00	1.27	0.59	0.50	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00						
Jun	0.00	0.00	0.00	0.00	0.00	0.00	0.00	1.27	0.06	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	1.27	0.06	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
Jul	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
Aug	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
Sep	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00					
Oct	6.79	6.24	0.44	0.00	0.00	0.00	0.00	6.21	4.90	0.56	0.00	5.79	5.78	0.31	0.00	5.79	5.78	0.31	0.00	5.79	5.78	0.31	0.00	5.79	5.78	0.31	0.00	5.79	5.78	0.31	0.00	5.79	5.78	0.31	0.00	5.79	5.78	0.31		
Nov	3.15	3.51	2.25	2.00	0.25	0.18	0.00	3.27	3.44	2.69	6.45	2.00	0.31	2.65	2.68	1.25	0.00	3.27	3.44	2.69	6.45	2.00	0.31	2.65	2.68	1.25	0.00	3.27	3.44	2.69	6.45	2.00	0.31	2.65	2.68	1.25	0.00	3.27	3.44	2.69
Dec	4.34	5.40	5.00	15.00	7.96	1.36	0.00	4.77	6.05	3.63	3.21	3.61	1.19	3.21	4.46	3.13	0.00	4.77	6.05	3.63	3.21	3.61	1.19	3.21	4.46	3.13	0.00	4.77	6.05	3.63	3.21	3.61	1.19	3.21	4.46	3.13	0.00	4.77	6.05	3.63

Table S2. Correlation matrix (r values) for rain water constituent concentrations measured at the sites shown in Table 1. Values are only shown when statistically significant (95%) with a two-tailed Student's T-Test.

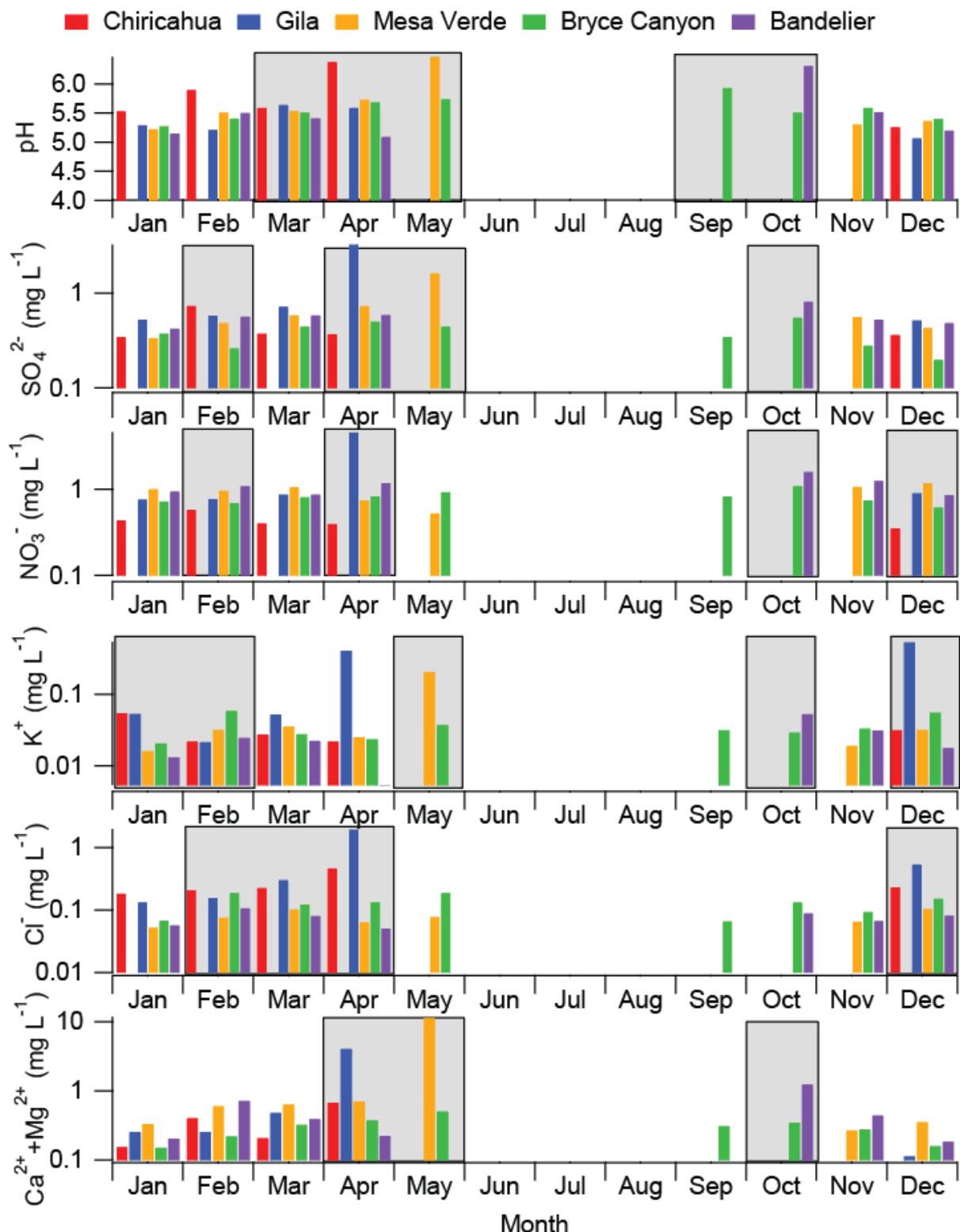
Bandelier Rain (n = 248)										Bryce Canyon Rain (n = 129)									
	Ca	Mg	K	Na	NH <sub>4</sub>	NO <sub>3</sub>	Cl	SO <sub>4</sub>	pH		Ca	Mg	K	Na	NH <sub>4</sub>	NO <sub>3</sub>	Cl	SO <sub>4</sub>	pH
Ca	1.00										1.00								
Mg	0.93	1.00									0.92	1.00							
K	0.48	0.56	1.00								0.59	0.66	1.00						
Na	0.69	0.74	0.33	1.00							0.66	0.71	0.64	1.00					
NH <sub>4</sub>	0.46	0.43	0.32	0.26	1.00						0.65	0.60	0.34	0.36	1.00				
NO <sub>3</sub>	0.77	0.73	0.43	0.51	0.68	1.00					0.83	0.81	0.48	0.55	0.83	1.00			
Cl	0.77	0.84	0.42	0.89	0.38	0.67	1.00				0.77	0.78	0.61	0.88	0.52	0.73	1.00		
SO <sub>4</sub>	0.76	0.73	0.43	0.64	0.61	0.78	0.71	1.00			0.79	0.80	0.60	0.73	0.66	0.83	0.82	1.00	
pH	0.46	0.42	0.21	0.27	0.25	--	0.19	--	1.00	0.39	0.38	--	--	0.34	--	--	--	1.00	
Chiricahua Rain (n = 206)										Gila Rain (n = 294)									
	Ca	Mg	K	Na	NH <sub>4</sub>	NO <sub>3</sub>	Cl	SO <sub>4</sub>	pH		Ca	Mg	K	Na	NH <sub>4</sub>	NO <sub>3</sub>	Cl	SO <sub>4</sub>	pH
Ca	1.00										1.00								
Mg	0.80	1.00									0.87	1.00							
K	0.59	0.64	1.00								0.45	0.56	1.00						
Na	0.50	0.90	0.50	1.00							0.66	0.74	0.55	1.00					
NH <sub>4</sub>	0.43	0.29	0.28	--	1.00						0.39	0.44	0.25	0.25	1.00				
NO <sub>3</sub>	0.68	0.53	0.41	0.27	0.79	1.00					0.64	0.66	0.36	0.34	0.75	1.00			
Cl	0.46	0.88	0.53	0.99	--	0.24	1.00				0.56	0.76	0.74	0.88	0.29	0.41	1.00		
SO <sub>4</sub>	0.67	0.57	0.51	0.40	0.72	0.72	0.37	1.00			0.64	0.69	0.40	0.46	0.67	0.82	0.51	1.00	
pH	0.41	0.28	0.22	0.17	0.15	--	--	--	1.00	0.38	0.29	0.16	0.28	--	-0.14	0.19	-0.19	1.00	
Mesa Verde Rain (n = 188)																			
	Ca	Mg	K	Na	NH <sub>4</sub>	NO <sub>3</sub>	Cl	SO <sub>4</sub>	pH										
Ca	1.00																		
Mg	0.83	1.00																	
K	0.74	0.75	1.00																
Na	0.67	0.72	0.64	1.00															
NH <sub>4</sub>	0.19	0.46	0.38	0.22	1.00														
NO <sub>3</sub>	0.33	0.65	0.48	0.46	0.79	1.00													
Cl	0.46	0.67	0.61	0.88	0.38	0.60	1.00												
SO <sub>4</sub>	0.46	0.73	0.53	0.52	0.70	0.88	0.57	1.00											
pH	0.53	0.49	0.46	0.40	--	--	0.31	--	1.00										

Table S3. Correlation matrix (r values) for snow water constituent concentrations measured at the sites shown in Table 1. Values are only shown when statistically significant (95%) with a two-tailed Student's T-Test.

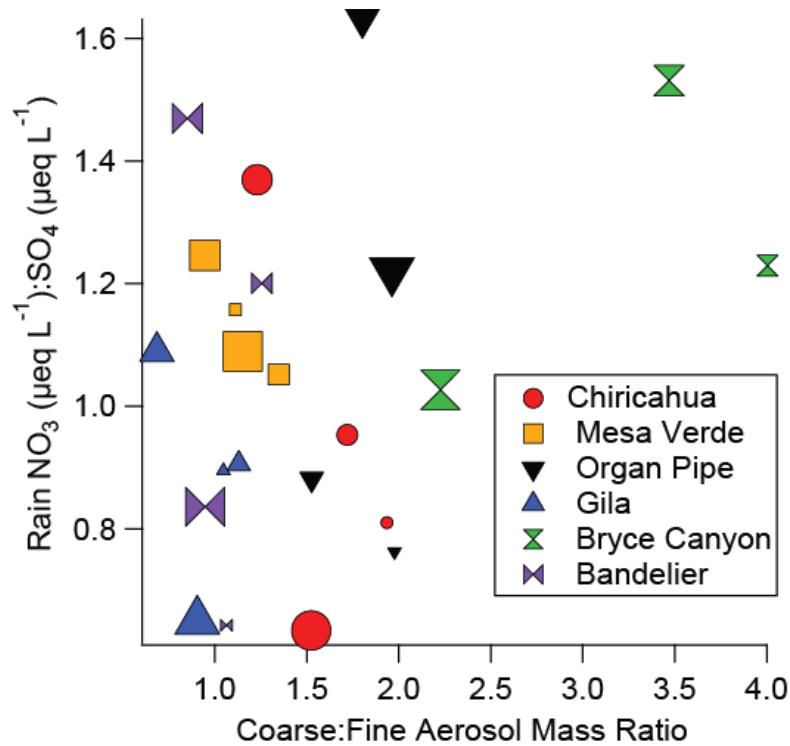
Bryce Canyon Snow (n = 191)										Chiricahua Snow (n = 11)									
Ca	Mg	K	Na	NH <sub>4</sub>	NO <sub>3</sub>	Cl	SO <sub>4</sub>	pH	Ca	Mg	K	Na	NH <sub>4</sub>	NO <sub>3</sub>	Cl	SO <sub>4</sub>	pH		
Ca	1.00									1.00									
Mg	0.93	1.00								0.95	1.00								
K	0.16	0.25	1.00							--	--	1.00							
Na	0.22	0.31	0.94	1.00						0.72	0.87	--	1.00						
NH <sub>4</sub>	0.28	0.36	0.70	0.75	1.00					--	--	--	--	1.00					
NO <sub>3</sub>	0.43	0.46	--	--	0.43	1.00				--	0.62	--	0.62	0.89	1.00				
Cl	0.15	0.26	0.96	0.97	0.74	--	1.00			0.62	0.77	--	0.96	--	--	1.00			
SO <sub>4</sub>	0.45	0.45	--	--	0.37	0.63	--	1.00		--	0.62	--	--	0.86	0.88	--	1.00		
pH	0.58	0.59	0.39	0.43	0.30	--	0.38	--	1.00	0.86	0.83	--	--	--	--	--	--	1.00	
Gila Snow (n = 42)										Mesa Verde Snow (n = 121)									
Ca	Mg	K	Na	NH <sub>4</sub>	NO <sub>3</sub>	Cl	SO <sub>4</sub>	pH	Ca	Mg	K	Na	NH <sub>4</sub>	NO <sub>3</sub>	Cl	SO <sub>4</sub>	pH		
Ca	1.00									1.00									
Mg	0.98	1.00								0.92	1.00								
K	--	--	1.00							0.71	0.77	1.00							
Na	0.93	0.96	--	1.00						0.34	0.52	0.39	1.00						
NH <sub>4</sub>	0.56	0.58	--	0.61	1.00					--	--	0.20	0.32	1.00					
NO <sub>3</sub>	0.87	0.87	--	0.92	0.67	1.00				--	--	0.20	0.26	0.48	1.00				
Cl	0.60	0.63	0.76	0.66	0.44	0.67	1.00			0.19	0.38	0.34	0.91	0.21	0.24	1.00			
SO <sub>4</sub>	0.90	0.93	--	0.95	0.71	0.87	0.63	1.00		0.53	0.57	0.53	0.38	0.51	0.30	0.21	1.00		
pH	--	--	--	--	--	--	--	--	1.00	0.63	0.76	0.56	0.42	--	--	0.29	0.23	1.00	

Table S4. Correlative relationships ( $r$ ) between rain/snow water pH and aerosol constituent mass concentrations at the six study sites. “Coarse:Fine” corresponds to the mass concentration ratio of coarse aerosol ( $\text{PM}_{10} - \text{PM}_{2.5}$ ) to  $\text{PM}_{2.5}$ . Values are only shown when statistically significant (95%) with a two-tailed Student’s T-Test.

Aerosol Species	Bandelier		Bryce Canyon		Chiricahua		Gila		Mesa Verde		Organ Pipe	
	Rain pH	Snow pH	Rain pH	Snow pH	Rain pH	Snow pH	Rain pH	Snow pH	Rain pH	Snow pH	Rain pH	Snow pH
Ca	0.41	--	0.25	0.20	0.23	--	0.23	0.43	0.30	0.19	--	--
Mg	0.23	--	--	--	--	--	--	--	--	--	--	--
K	0.19	--	--	0.21	0.28	--	0.14	0.46	0.26	0.28	--	--
Na	--	--	--	0.32	0.19	--	0.15	0.48	0.35	--	--	--
Fine Soil	0.29	--	0.24	0.15	0.26	--	0.23	0.42	0.32	0.27	--	--
$\text{NO}_3$	--	--	--	--	0.14	--	0.16	--	0.35	--	--	--
Cl	--	--	--	--	--	--	--	--	--	--	--	--
$\text{SO}_4$	--	--	-0.23	--	--	--	-0.35	--	-0.17	-0.40	--	--
Coarse:Fine	--	--	--	0.26	--	--	--	0.36	0.18	0.30	--	--



**Fig. S1.** Annual pH and concentration profiles for snow water in the Southwest. Shaded regions represent when maxima are observed for individual or groups of sites. These results are based on data ranges shown in Table 1 for each site.



**Fig. S2.** Comparison of rain water nitrate:sulfate mass concentration ratios as a function of co-located coarse:fine aerosol mass concentrations. Marker sizes are proportional to the season (DJF < MAMJ < JAS < ON).

## **Statistical Methods**

Basic statistical calculations are conducted in this work including averages, standard deviations, and Pearson correlation coefficients ( $r$ ), which are described by Wilks (2006).

### References

Wilks, D. S.: Statistical methods in the atmospheric sciences, Elsevier, 2nd Ed., 2006.