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Sensitivity of aerosol and cloud properties to coupling strength of marine boundary layer clouds over the northwest Atlantic

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Table S1. Sensitivity test results for aerosol and other atmospheric properties investigated across the MinAlt-BCB pairs. Δq_t and $\Delta\theta_\ell$ (left/right) were varied (the original criteria are reported in the first column) and the column headers are interpreted as such: for the 0.6/1.0 column, strong coupling is for $\Delta\theta_\ell \leq 1.0$ K and $\Delta q_t \leq 0.6$ g kg^{-1}; moderate coupling with high $\Delta\theta_\ell$ is for $\Delta\theta_\ell > 1.0$ K and $\Delta q_t \leq 0.6$ g kg^{-1}; moderate coupling with high Δq_t is for $\Delta\theta_\ell \leq 1.0$ K and $\Delta q_t > 0.6$ g kg^{-1}; and weak coupling is for $\Delta\theta_\ell > 1.0$ K and $\Delta q_t > 0.6$ g kg^{-1}. Flight data for MinAlt-BCB pairs were grouped based on the new criteria, and the values reported are the means of the grouped data for each coupling regime. The Δ calculation refers to the MinAlt value minus the BCB value.

	0.8/1.0	**0.6/1.0**	**0.7/1.0**	**0.9/1.0**	**1.0/1.0**	**1.0/1.2**	**0.8/0.8**	**0.8/0.9**	**0.8/1.1**	**0.8/1.2**	**0.6/0.8**	**0.5/0.5**
# points												
Strong coupling	293	274	286	302	310	320	287	289	297	303	268	210
Moderate coupling, high $\Delta\theta_\ell$	56	53	56	57	57	47	62	60	52	46	59	92
Moderate coupling, high Δq_t	42	61	49	33	25	27	35	38	42	44	54	63
Weak coupling	20	23	20	19	19	17	27	24	20	18	30	46
Δscat												
Strong coupling	2.2	1.9	2.2	2.2	2.2	2.2	2.2	2.2	2.2	2.2	2.2	2.2
Moderate coupling, high $\Delta\theta_\ell$	3.5	3.5	3.4	3.4	3.4	3.9	3.3	3.3	3.7	3.9	3.4	3.0
Moderate coupling, high Δq_t	2.4	2.5	2.5	2.4	2.7	2.8	2.6	2.5	2.4	2.5	2.6	2.1
Weak coupling	3.5	3.3	3.5	3.5	3.5	3.4	3.0	3.2	3.5	3.4	2.9	3.1
ΔIntV												
Strong coupling	2.5	1.5	2.5	2.5	2.5	2.4	2.5	2.5	2.5	2.5	2.4	2.5
Moderate coupling, high $\Delta\theta_\ell$	2.1	2.1	2.1	2.2	2.2	2.3	2.2	2.2	2.2	2.2	2.2	2.1
Moderate coupling, high Δq_t	1.9	2.5	2.1	1.9	2.3	2.3	1.8	1.8	1.9	1.9	2.6	2.5
Weak coupling	2.8	2.8	2.8	2.6	2.6	2.6	2.6	2.7	2.8	2.9	2.6	2.8
$\Delta N_{a>3\mu m}$												
Strong coupling	0.32	0.20	0.32	0.31	0.31	0.30	0.32	0.32	0.32	0.31	0.33	0.35
Moderate coupling, high $\Delta\theta_\ell$	0.33	0.35	0.33	0.32	0.32	0.37	0.32	0.32	0.34	0.38	0.34	0.29

Moderate coupling, high Δq_t	0.15	0.15	0.15	0.16	0.15	0.15	0.15	0.15	0.15	0.15	0.16	0.22
Weak coupling	0.53	0.45	0.53	0.57	0.57	0.60	0.41	0.46	0.53	0.56	0.36	0.31
N_d												
Strong coupling	344	366	346	344	348	348	345	345	343	343	351	356
Moderate coupling, high $\Delta\theta_\ell$	419	422	421	419	419	439	411	412	432	441	412	376
Moderate coupling, high Δq_t	329	318	334	343	294	285	373	362	345	336	767	371
Weak coupling	275	279	275	270	270	280	263	267	275	286	267	254
MinAlt σ_w												
Strong coupling	0.86	1.17	0.87	0.86	0.86	0.86	0.85	0.85	0.86	0.86	0.86	0.85
Moderate coupling, high $\Delta\theta_\ell$	1.00	1.00	0.99	0.99	0.99	0.97	1.01	1.02	0.96	0.97	1.02	0.97
Moderate coupling, high Δq_t	0.81	0.78	0.77	0.73	0.79	0.79	0.84	0.84	0.81	0.80	0.80	0.86
Weak coupling	0.49	0.58	0.55	0.52	0.52	0.50	0.57	0.53	0.55	0.53	0.60	0.63
BCB σ_w												
Strong coupling	0.70	0.86	0.71	0.70	0.69	0.70	0.71	0.71	0.70	0.71	0.72	0.71
Moderate coupling, high $\Delta\theta_\ell$	0.64	0.68	0.64	0.65	0.65	0.62	0.64	0.64	0.64	0.61	0.67	0.67
Moderate coupling, high Δq_t	0.81	0.72	0.75	0.83	0.99	0.97	0.79	0.82	0.81	0.81	0.70	0.67
Weak coupling	0.49	0.44	0.49	0.48	0.48	0.45	0.60	0.54	0.49	0.47	0.55	0.72
BCB - MinAlt σ_w												
Strong coupling	-0.15	-0.31	-0.15	-0.16	-0.16	-0.17	-0.15	-0.14	-0.16	-0.16	-0.14	-0.15
Moderate coupling, high $\Delta\theta_\ell$	-0.34	-0.32	-0.35	-0.34	-0.34	-0.36	-0.37	-0.38	-0.33	-0.36	-0.35	-0.30
Moderate coupling, high Δq_t	0.01	-0.06	-0.02	0.10	0.20	0.19	-0.04	-0.03	0.01	0.01	-0.10	-0.19
Weak coupling	-0.05	-0.14	-0.05	-0.05	-0.05	-0.05	0.03	0.01	-0.05	-0.06	-0.05	0.09

Table S2. Statistics for aerosol and other atmospheric properties investigated across the MinAlt-BCB pairs (Δ calculation refers to the MinAlt value minus the BCB value) for the summer flights, except for MinAlt σ_w and BCB σ_w, which are the average σ_w for each respective leg. Each property is broken down into the different degrees of coupling (n = number of points used in each coupling category).

Parameter	Degree of Coupling	Mean	Standard Deviation	Min	25%	50%	75%	Max	n
Δscat	Strong	2.6	2.6	0.02	0.75	1.8	3.3	13.9	106
	Moderate, high $\Delta\theta_\ell$	4.0	3.8	0.08	1.4	3.3	5.9	13.3	12
	Moderate, high Δq_t	2.3	2.2	0.42	0.80	1.5	3.4	9.2	16
	Weak	3.8	3.7	0.01	0.18	3.0	7.7	10.8	11
ΔIntV	Strong	3.0	2.7	0.02	1.2	2.0	4.0	13.6	115
	Moderate, high $\Delta\theta_\ell$	3.2	3.0	0.38	0.75	2.2	5.8	9.3	12
	Moderate, high Δq_t	1.6	1.6	0.01	0.17	1.0	3.4	4.3	17
	Weak	2.7	2.0	0.18	1.1	2.5	4.2	5.8	11
$\Delta N_{>3\mu m}$	Strong	0.47	0.55	0.00	0.10	0.32	0.61	3.3	115
	Moderate, high $\Delta\theta_\ell$	0.42	0.33	0.00	0.12	0.41	0.68	1.1	12
	Moderate, high Δq_t	0.20	0.18	0.02	0.05	0.12	0.30	0.61	17
	Weak	0.14	0.14	0.02	0.04	0.09	0.27	0.41	11
N_d	Strong	249	133	45	169	216	335	774	90
	Moderate, high $\Delta\theta_\ell$	256	140	45	114	330	349	440	9
	Moderate, high Δq_t	290	118	25	211	287	384	450	12
	Weak	209	112	50	77	220	293	392	10
MinAlt σ_w	Strong	0.51	0.24	0.00	0.33	0.51	0.66	1.2	118
	Moderate, high $\Delta\theta_\ell$	0.43	0.36	0.00	0.15	0.36	0.67	1.2	12
	Moderate, high Δq_t	0.61	0.25	0.30	0.40	0.59	0.74	1.2	18
	Weak	0.38	0.20	0.11	0.16	0.35	0.52	0.73	11
BCB σ_w	Strong	0.58	0.68	0.00	0.22	0.41	0.69	4.0	118
	Moderate, high $\Delta\theta_\ell$	0.47	0.36	0.00	0.22	0.37	0.76	1.1	12
	Moderate, high Δq_t	0.76	0.81	0.00	0.26	0.45	0.97	3.3	18
	Weak	0.32	0.30	0.00	0.00	0.22	0.50	0.97	11
BCB - MinAlt σ_w	Strong	0.08	0.73	-1.2	-0.26	-0.10	0.19	3.6	116
	Moderate, high $\Delta\theta_\ell$	0.02	0.43	-0.83	-0.25	0.08	0.32	0.59	10
	Moderate, high Δq_t	0.15	0.81	-0.64	-0.26	-0.09	0.20	0.00	18
	Weak	-0.07	0.33	-0.52	-0.36	0.02	0.15	0.50	11

Table S3. Same as Table S2, except for winter flights only.

Parameter	Degree of Coupling	Mean	Standard Deviation	Min	25%	50%	75%	Max	n
Δscat	Strong	2.0	1.7	0.00	0.79	1.6	2.7	10.3	168
	Moderate, high $\Delta\theta_\ell$	3.3	3.5	0.07	0.90	2.2	4.3	14.6	40
	Moderate, high Δq_t	2.5	2.1	0.01	0.67	2.3	4.3	6.7	23
	Weak	3.1	2.8	0.54	0.89	1.6	6.5	7.1	9
ΔIntV	Strong	2.3	2.5	0.02	0.45	1.5	3.2	13.0	173
	Moderate, high $\Delta\theta_\ell$	1.8	1.8	0.00	0.44	1.3	2.6	7.5	42
	Moderate, high Δq_t	2.0	2.1	0.12	0.74	1.3	2.4	8.3	24
	Weak	2.9	2.7	0.30	0.84	2.3	5.0	7.6	9
$\Delta N_{>3\mu m}$	Strong	0.22	0.52	0.00	0.04	0.09	0.19	4.9	173
	Moderate, high $\Delta\theta_\ell$	0.30	0.69	0.00	0.03	0.10	0.23	3.6	42
	Moderate, high Δq_t	0.11	0.08	0.00	0.10	0.10	0.13	0.35	24
	Weak	0.93	2.1	0.01	0.06	0.06	0.92	6.0	9
N_d	Strong	402	238	19	226	367	550	954	148
	Moderate, high $\Delta\theta_\ell$	457	246	73	243	409	670	962	39
	Moderate, high Δq_t	354	171	40	235	363	512	671	19
	Weak	358	222	81	116	419	560	606	8
MinAlt σ_w	Strong	1.1	0.46	0.00	0.82	1.1	1.4	2.4	175
	Moderate, high $\Delta\theta_\ell$	1.1	0.52	0.00	0.97	1.2	1.5	2.2	44
	Moderate, high Δq_t	0.95	0.49	0.00	0.63	0.85	1.4	1.9	24
	Weak	0.75	0.46	0.00	0.32	0.95	1.0	1.3	9
BCB σ_w	Strong	0.79	0.57	0.00	0.33	0.80	1.2	2.5	175
	Moderate, high $\Delta\theta_\ell$	0.69	0.67	0.00	0.00	0.68	1.1	2.2	44
	Moderate, high Δq_t	0.86	0.74	0.00	0.10	0.81	1.2	2.5	24
	Weak	0.71	0.63	0.00	0.10	0.86	1.4	1.6	9
BCB - MinAlt σ_w	Strong	-0.30	0.57	-2.0	-0.58	-0.23	0.10	1.1	169
	Moderate, high $\Delta\theta_\ell$	-0.42	0.57	-2.2	-0.88	-0.29	0.00	0.53	43
	Moderate, high Δq_t	-0.09	0.68	-1.6	-0.57	-0.05	0.24	1.6	24
	Weak	-0.04	0.62	-1.1	-0.46	0.00	0.38	0.90	9

Table S4. Mean cloud water sample concentrations (μg m^{-3}), pH, and Cl^-:Na^+ mass ratio. Each chemical species is broken down into the different degrees of coupling (n = number of points used in each coupling category; n_{pH} = number of points used in pH analysis).

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t	Weak
Cl^-	45.6	32.0	52.2	4.1
Na^+	27.9	18.6	29.9	2.5
Mg^{2+}	3.3	2.2	3.6	0.35
K^+	0.56	0.37	0.59	0.05
nss-Ca^{2+}	0.53	0.18	0.17	0.03
nss-SO_4^{2-}	2.6	1.5	1.7	1.2
NO_3^-	6.0	2.7	3.3	1.5
Oxalate	0.10	0.01	0.03	0.01
NH_4^+	0.89	0.37	0.33	0.56
pH	4.9	4.6	5.3	4.4
Cl^-:Na^+	1.7	1.7	1.7	1.5
n	40	17	4	6
n_{pH}	16	11	3	2

Table S5. Mean cloud water sample concentrations (μg m^{-3}) , pH, and Cl^-:Na^+ mass ratio during the summer flights. Each chemical species is broken down into two different degrees of coupling (strong and 'moderate, high $\Delta\theta_\ell$'), since there were not any samples that fit the criteria for the other two degrees of coupling ('moderate, high Δq_t' and weak). (n = number of points used in each coupling category; n_{pH} = number of points used in pH analysis)

	Strong	Moderate, high $\Delta\theta_\ell$
Cl^-	39.3	4.6
Na^+	24.0	2.7
Mg^{2+}	2.5	0.35
K^+	0.50	0.06
nss-Ca^{2+}	0.65	0.10
nss-SO_4^{2-}	4.0	0.22
NO_3^-	9.1	0.58
Oxalate	0.27	0.00
NH_4^+	2.0	0.03
pH	5.2	5.4
Cl^-:Na^+	1.6	1.7
n	10	1
n_{pH}	4	1

Table S6. Mean cloud water sample concentrations (μg m^{-3}), pH, and Cl^-:Na^+ mass ratio during the winter flights. Each chemical species is broken down into the different degrees of coupling (n = number of points used in each coupling category; n_{pH} = number of points used in pH analysis).

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t	Weak
Cl^-	47.7	33.7	52.2	4.1
Na^+	29.2	19.6	29.9	2.5
Mg^{2+}	3.6	2.3	3.6	0.35
K^+	0.58	0.39	0.59	0.05
nss-Ca^{2+}	0.49	0.19	0.17	0.03
nss-SO_4^{2-}	2.1	1.6	1.7	1.2
NO_3^-	5.0	2.8	3.3	1.5
Oxalate	0.05	0.01	0.03	0.01
NH_4^+	0.57	0.39	0.33	0.56
pH	4.8	4.5	5.3	4.4
Cl^-:Na^+	1.7	1.7	1.7	1.5
n	30	16	4	6
n_{pH}	12	10	3	2

Table S7. Results of Welch's t-tests for nine different chemical species, pH, and Cl^-:Na^+ comparing species concentrations within different degrees of coupling. Bolded values indicate statistical significance ($\alpha < 0.05$). Refer to Tables S4-S6 (also Table 4) for the number of points within each coupling category.

Cl^-

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t
Moderate, high $\Delta\theta_\ell$	0.36		
Moderate, high Δq_t	0.57	0.35	
Weak	**$3.54E^{-5}$**	**0.01**	0.15

Na^+

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t
Moderate, high $\Delta\theta_\ell$	0.30		
Moderate, high Δq_t	0.61	0.34	
Weak	**$1.07E^{-4}$**	**0.01**	0.14

Mg^{2+}

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t
Moderate, high $\Delta\theta_\ell$	0.24		
Moderate, high Δq_t	0.88	0.45	
Weak	**$6.41E^{-5}$**	**0.01**	0.12

K^+

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t
Moderate, high $\Delta\theta_\ell$	0.27		
Moderate, high Δq_t	0.61	0.34	
Weak	**$5.52E^{-5}$**	**0.01**	0.15

nss-Ca^{2+}

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t
Moderate, high $\Delta\theta_\ell$	0.06		
Moderate, high Δq_t	0.06	0.94	
Weak	**$1.30E^{-3}$**	0.21	0.31

nss-SO_4^{2-}

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t
Moderate, high $\Delta\theta_\ell$	0.21		
Moderate, high Δq_t	0.74	0.64	
Weak	0.25	0.78	0.54

NO_3^-

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t
Moderate, high $\Delta\theta_\ell$	0.07		
Moderate, high Δq_t	0.46	0.65	
Weak	**$9.57E^{-3}$**	0.37	0.36

Oxalate

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t
Moderate, high $\Delta\theta_\ell$	0.14		
Moderate, high Δq_t	0.35	0.50	
Weak	0.18	0.71	0.61

NH_4^+

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t
Moderate, high $\Delta\theta_\ell$	0.09		
Moderate, high Δq_t	0.07	0.84	
Weak	0.47	0.63	0.56

pH

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t
Moderate, high $\Delta\theta_\ell$	0.08		
Moderate, high Δq_t	0.96	0.62	
Weak	0.16	0.75	0.58

Cl^-:Na^+

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t
Moderate, high $\Delta\theta_\ell$	0.79		
Moderate, high Δq_t	0.49	0.43	
Weak	0.30	0.34	0.22

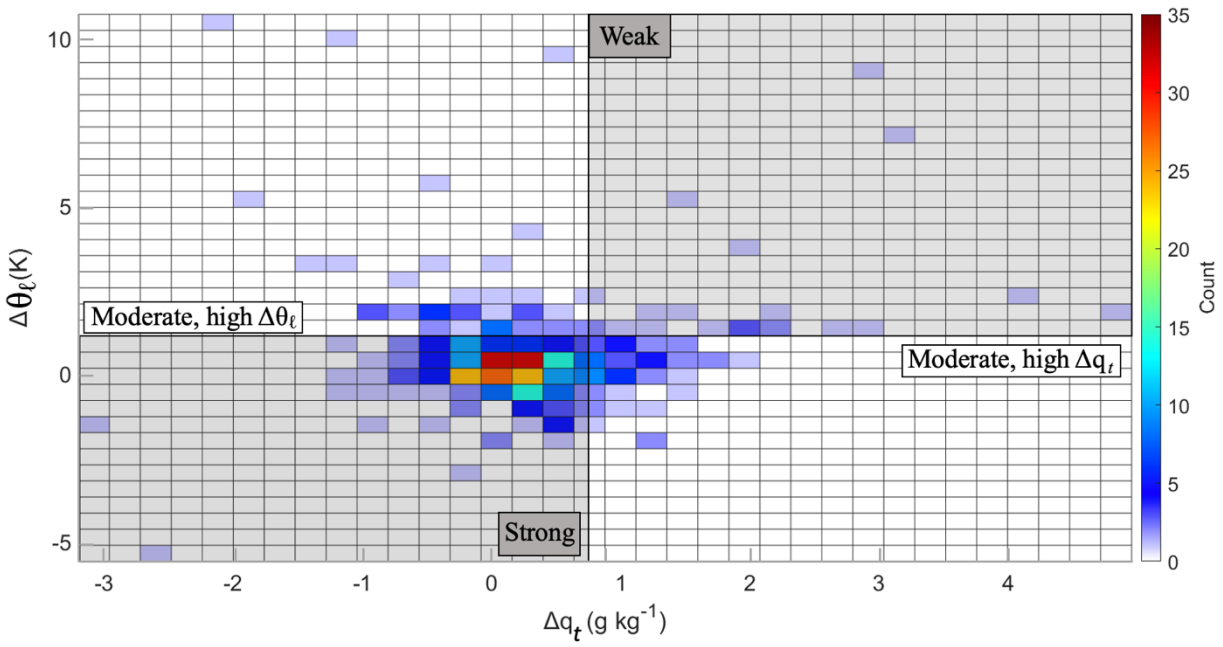

Figure S1. Joint frequency histogram of $\Delta\theta_\ell$ versus Δq_t for MinAlt-BCB pairs, categorized into four coupling regimes. There is a total of 411 pairs plotted (strong coupling = 293, moderate coupling with high $\Delta\theta_\ell$ = 56, moderate coupling with high Δq_t = 42, weak coupling = 20). Figure 4 shows this information visually also but breaks down the MinAlt-BCB pairs into seasons.

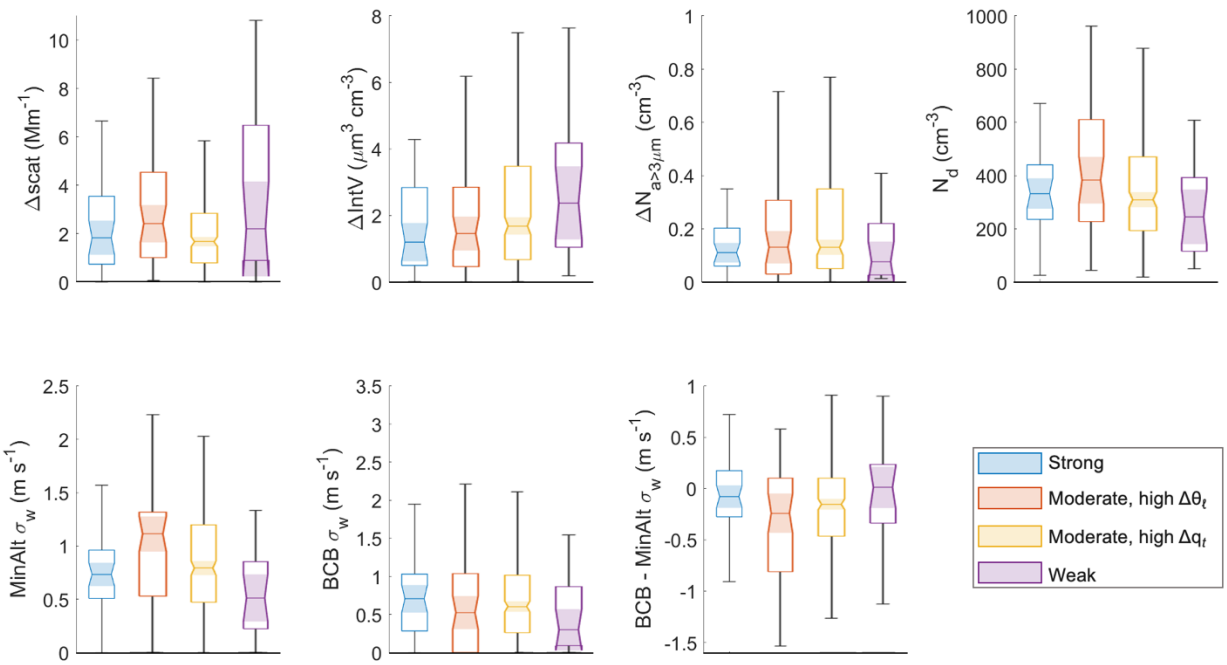

Figure S2. Notched box plots of various atmospheric properties investigated across MinAlt-BCB pairs. Refer to Table 3 for additional statistics and total number of points. The notches of the boxes assist in the determination of statistical significance between multiple medians (the shading indicates where the notches begin and end). If notches/shading do not overlap, the medians are statistically different from one another (also referred to as statistically significant).