Table S1. 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 S2. Same as Table S1, 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 S3. 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 S4. 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 S5. 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 S6. Results of Welch's t-tests for nine different chemical species, pH, and Cl^-:Na^+ comparing species concentrations within different degrees of coupling. Cells colored green indicate statistical significance ($\alpha < 0.05$). Refer to Tables S3-S5 (also Table 5) 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		

pH

	Strong	Moderate, high $\Delta\theta_\ell$	Moderate, high Δq_t
Moderate, high $\Delta\theta_\ell$	0.08		

Moderate, high Δq_t	0.07	0.84	
Weak	0.47	0.63	0.56

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

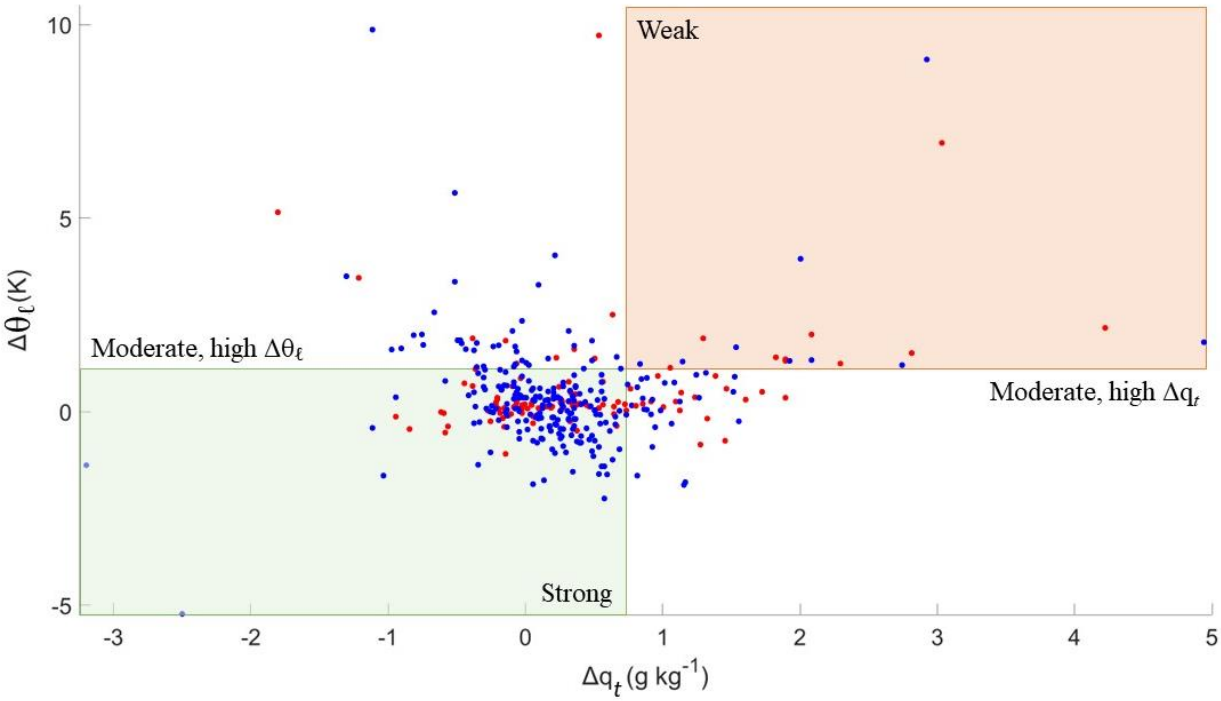

Figure S1. Scatterplot of $\Delta\theta_\ell$ vs. Δq_t values for the BCB and MinAlt pairs divided into the four coupling regimes, where winter pairs are indicated by blue points and summer pairs are indicated by red points. Refer to Table 3 for the number of points in each coupling regime categorized by season.