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Supporting Information

Sources of Nitrate in Stratocumulus Cloud Water: Airborne Measurements during the 2011 E-PEACE and 2013 NiCE Studies

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Supporting Information Summary: 6 Pages including Cover Page, 3 Tables, 3 Figures

Table S1. Precision of the ion chromatography (IC) method in measuring the ionic components in cloud water. The IC analysis was repeated for solutions with known concentrations to get the relative standard deviation (RSD). Concentration units used for the calculations are ppm.

Ion	# of Samples	RSD (%)
Cl^-	8	12.8
NO_3^-	8	5.6
SO_4^{2-}	9	3.4
Oxalate	8	4.6
Na^+	8	20.5
NH_4^+	8	8.9
K^+	8	13.2

Table S2. Limits of detection (LOD) for IC analyses of cloud water samples and the average concentrations measured in the blanks (standard deviations in parentheses). Standard solutions with different known concentrations of the solute were analyzed. The blanks were collected after rinsing the cloud water collector several times with de-ionized water. The "*" symbol denotes concentration in blanks that are below detection and therefore, the LOD was used as the blank.

Ion	LOD (ppm)	Blank (ppm)
Cl^-	0.050	0.057 (0.020)
NO_3^-	0.005	0.005*
SO_4^{2-}	0.009	0.013 (0.004)
Oxalate	0.005	0.005*
Na^+	0.005	0.022 (0.017)
NH_4^+	0.010	0.032 (0.039)
K^+	0.010	0.011 (0.005)

Table S3. Initial gas phase mixing ratios [ppb] for the three model cases compared in Figure 6 and Figures S2-S3. The mixing ratios are based on [a]Eyring et al. (2010), [b]Ervens et al. (2004), [c]Kato et al. (2007), and [d]Hays et al. (2002). Since the latter two studies were performed immediately near the emissions sources, mixing ratios were scaled down by up to an order of magnitude in order to account for plume age (e.g., cf. Fig. 1 in Eyring et al., 2010). Note that the exact mixing ratios of organic compounds (e.g., toluene, isoprene) are only listed in order to differentiate the different cases. Since OH concentrations are kept constant throughout the model simulations, their exact values have little impact on the model results. The initial sub-cloud aerosol mass concentrations for sulfate, nitrate and ammonia are based on C-ToF-AMS measurements during NiCE. 'Total mass' accounts for the total sub-cloud aerosol mass concentration, based on C-ToF-AMS measurements. In the model, the difference between the sum of the three inorganic ions and total mass was described as slightly soluble organic mass.

	Low Cl^- Marine[a]	Ship Plume[c]	Fire[a,b,d]
SO_2	0.28	14.5	1
O_3	36	100	90
H_2O_2	0.5	1	1
NH_3	2	50	14
HNO_3	1.13	5	3
NO_x	28	50	1
Toluene	0.87	5	1.27
Ethylene	0.71	8	0.12
Isoprene	0.025	2	0.053
CO	4.8	28	200
Initial aerosol composition [$\mu g\ m^{-3}$]			
SO_4^{2-}	1.9	2.1	1.04
NO_3^-	0.074	0.07	0.05
NH_4^+	0.4	0.5	0.38
Total mass	3.3	3.17	2.1

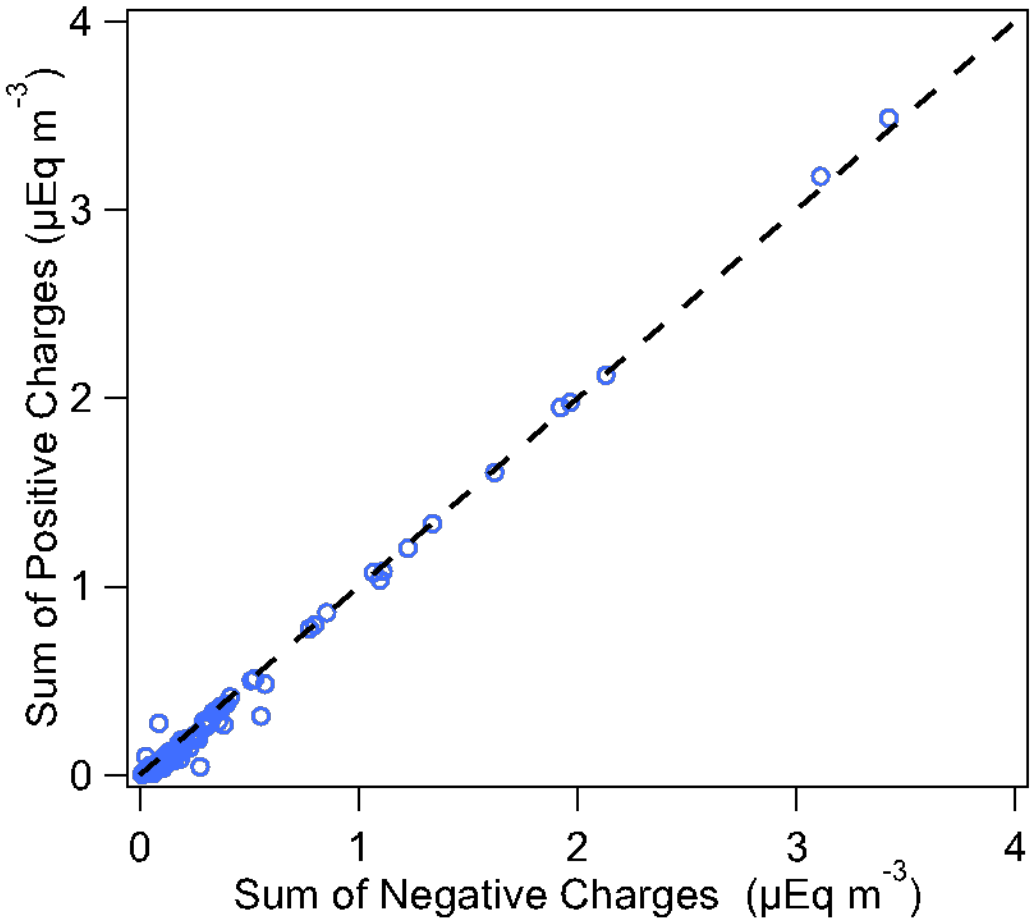

Figure S1. Charge balance for the ionic species measured by IC in the cloud water samples. The slope of the fit between the positive and negative ions was found to be 1.02 ± 0.01 (y-intercept = -0.04, r^2 = 0.99).The dashed black line shows the 1:1 line.

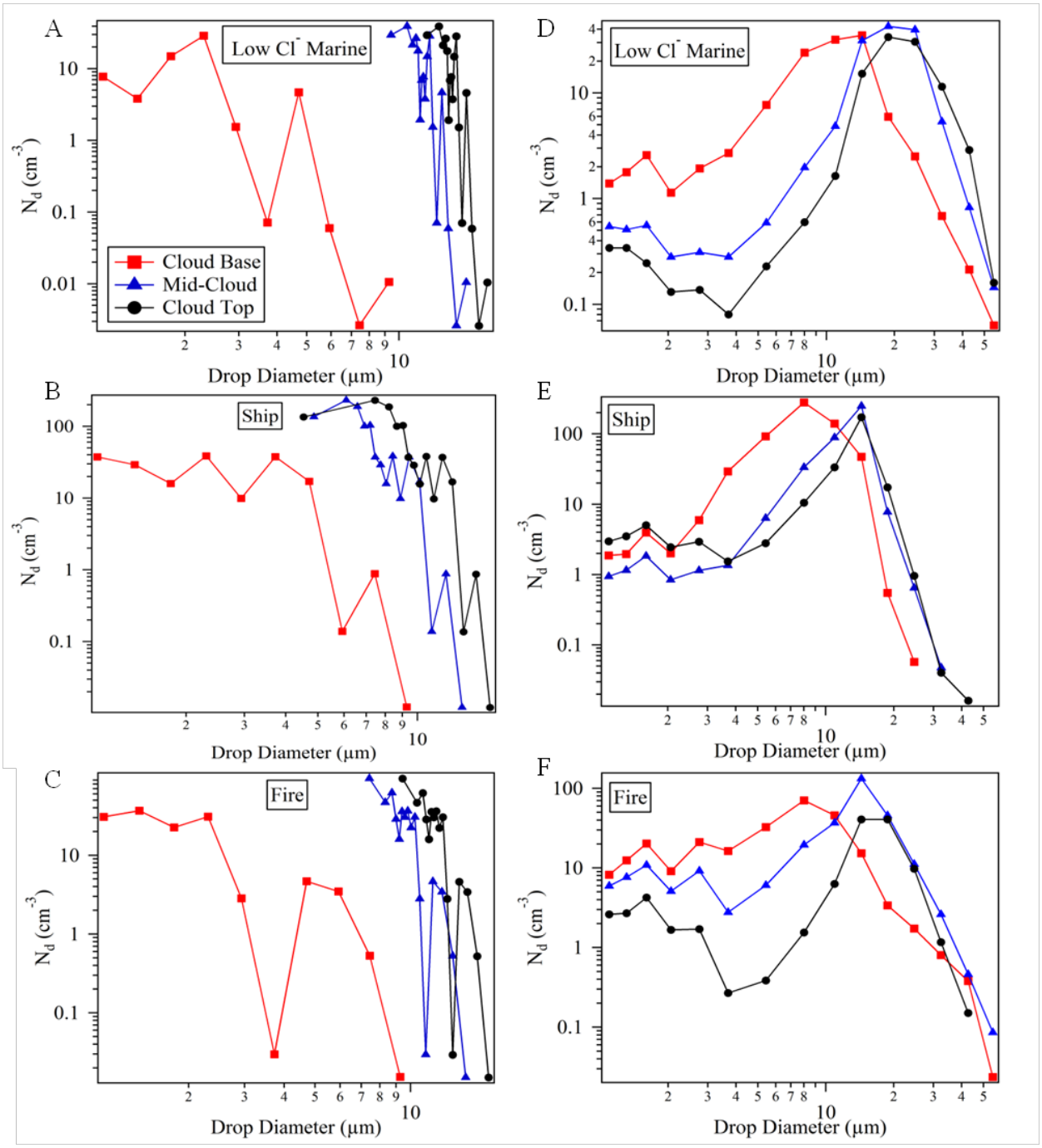

Figure S2. (A-C) Modeled drop size distributions at three different heights (near cloud base, center of cloud and cloud top) for the three model cases compared in Figure 6. (D-F) Corresponding drop size distributions for the three model cases as measured by the CAS during NiCE. N_d represents drop number concentration.

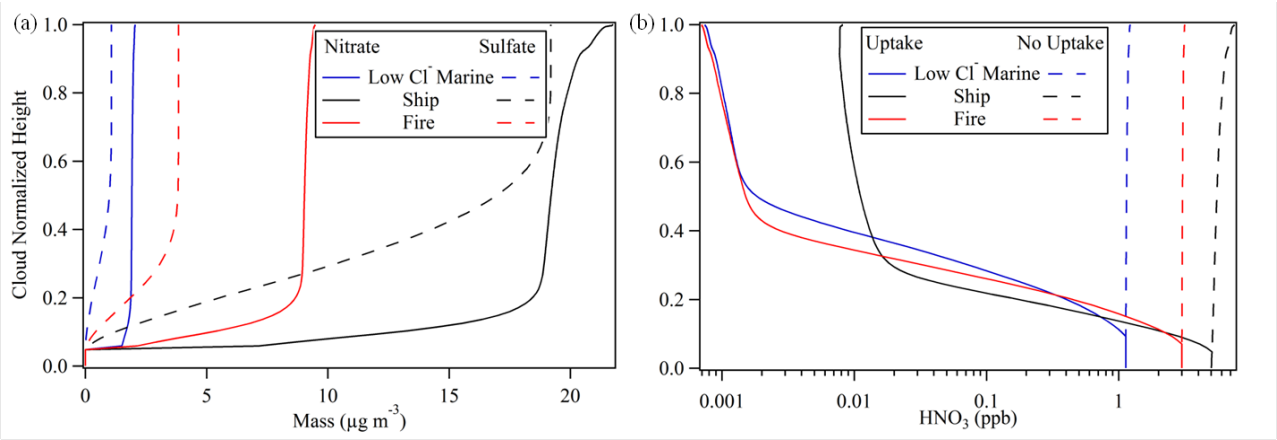

Figure S3. Vertical variation of (a) nitrate and sulfate concentrations and (b) HNO_3 (with and without uptake by cloud droplets), in the three model simulations compared in Figure 6. The figures clearly show that the majority of HNO_3 is scavenged by the cloud droplets. All nitrate that is present in cloud water at cloud top is assumed to remain in the particle phase.

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