Tellus (1983), *35B*, 121–130

Some characteristics of chemical precipitation

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(Manuscript received January 8; in final form June 23, 1982)

ABSTRACT

Two very dissimilar cold frontal occlusions deposited 6 mm and 60 mm of rain respectively at an observatory in the rural northeastern U.S.A. Water precipitation was carefully observed, in conjunction with other meteorological parameters, through both storm systems.

The concentration of several ionic substances were measured in specimens of rain water collected during meteorologically homogeneous portions of the two storms.

The rate of chemical precipitation was determined with respect to water precipitation. This rate was constant during the prefrontal stage of both storms, and increased following cold frontal passage, in the convective side of the systems. Analyses of the time rate of chemical precipitation showed it to be constant during the continuous rain of April 5.

Examinations of the ratio of chloride to sodium through the time history of the storms shows them to agree with a transport model postulated by Rossby and Egnér (1955).

Although the two storms varied considerably with respect to convectivity and prevailing flow, the total amounts of the several ions precipitated were directly proportional to the amount of water precipitated.

1. Introduction

The study of chemical precipitation began almost coincidentally with the formalization of nitrogen chemistry. Reviews of early work in chemical precipitation can be found in Eriksson (1953) and Rankhama and Sahama (1958). Houghton (1938) performed analyses of cloud and precipitation water as part of studies of nucleation. Rossby combined his interest in atmospheric circulation with Egnér's interest in agricultural meteorology (Rossby and Egnér, 1955) to begin the Scandinavian study of chemical climate which has been the basis for many recent programs studying the deposition of chemical compounds on the earth's surface.

The studies of chemical precipitation during the first three-fourths of this century generally relied on monthly or weekly collections of precipitation water. This allowed estimation of some of the source of precipitation material and some accounting for the balance of chemical substances precipitated and those materials in run-off water (Eriksson, 1955).

Junge and Werby (1958) and Lodge et al. (1968) made extensive surveys of precipitation chemistry in North America. More recently, several investigators have turned to so-called "event" sampling of precipitation which allows some comparison of precipitated material with the source region of the precipitating air mass (Wilson and Mohnen, 1981; Wilson et al., 1982). Results of more recent work were presented at the Conference on Long Range Transport of Air Pollution, Albany, NY, April 1981.

A pioneering study was performed in Hawaiian showers by Mordy (1953). He studied relatively homogeneous rains which resulted from lifting of the air as trade winds passed over the Koolau Ridge on the island of Oahu. He found that during showers of a few hours duration, nitrogen compounds (expressed as nitrate N plus ammonia N) were precipitated at a constant time rate, although the time rate of water precipitation changed.

This paper describes meteorological and chemical observations made during two very dissimilar cold frontal occlusions. These observations were made to determine if changes in

identifiable meteorological parameters resulted in changes in the rate or magnitude of chemical precipitation and to examine the variation of chemical precipitation on a time scale approximating that of changes in precipitation processes. The presence of a meteorological observer allowed determination of whether cloud type, height, thickness and near surface meteorological parameters influenced chemical precipitation rates in a single storm system.

2. Instrumentation and observational technique

The observations were performed in a rural area 50 km northwest of the Albany National Weather Service Station, NY, U.S.A. The observation site is in a basin at altitude 250 m, shielded from strong winds by a 330 m altitude ridge from N through W to SSE. The precipitation was collected in a field of approximately 200 m × 300 m surrounded by woodlots. Because of the nature of the topography of the area, surface winds are quite light, and rain falls vertically in most situations. When squall lines and cold fronts pass, stronger winds can be observed moving the trees of the ridge shielding the site, and the ridge lines provide scale for estimating cloud motion and height.

A 27.5 cm diameter collecting funnel was placed about 1 m above the ground before onset of precipitation. Cloud type and appearance, temperature and dewpoint, winds, distant lightning, and Aitken nucleic concentration were observed prior to the onset of precipitation. The collecting funnel was frequently rinsed with distilled water prior to the beginning of precipitation, and the rinse water saved for analysis, and detection of possible contamination.

The onset of rain was usually foretold by the sound of rain on the trees in the adjacent woodlots. The time of beginning of precipitation was noted, as were drop size, cloud base height, and cloud appearance. As rain continued, repeated meteorological observations were made and noted. The observer attempted to "forecast" or anticipate changes in the nature of precipitation while observing approaching clouds or rain shafts. When it appeared that a change in rain rate, shower characteristics, cloud base height or wind speed was imminent, the observer would retrieve the collected specimen, rinse the collecting funnel with distilled water, and reset the collector. This was followed by a complete set of meteorological observations of temperature, dewpoint, pressure, visibility, cloud characteristics, and Aitken nuclei concentration.

An immediate analysis of precipitation pH and ammonia nitrogen concentration was performed on site if 30 ml or more of precipitation water was collected. Three (3) ml of sample were used for colorimetric pH determination. An additional 10 ml were transferred to a polyethylene container and the pH determined with a glass electrode. This aliquot was diluted with 10, 20 and 10 ml of distilled water, and the pH measured at each step. This always yielded a linear decrease in hydrogen ion concentration. Sodium hydroxide was added to the resulting 50 ml of diluted precipitation until a neutral solution was achieved. Nessler's reagent was added and the ammonia nitrogen concentration determined colorimetrically with a Hellige Aquatester (Hellige, Inc. LIC, NY). The author considered immediate analysis of ammonia N to be of more importance than absolute accuracy as conversion among nitrogen compounds is suspected in rain water. Good agreement was achieved with later ion analysis. The residue of the specimen was then stored in a cool (+2 °C to +8 °C) place for later analysis by ion chromatography. When less than 30 ml was collected, the entire specimen was saved for ion chromatography analyses and pH was determined on the residual sample in the laboratory.

Observations continued beyond the time of cessation of precipitation, to determine characteristics of the incoming air mass.

3. Results of experiments; weak cold frontal occlusion

A weak cold frontal occlusion produced precipitation in the area on April 5, 1981. This system was typical of those often encountered in the Northeastern United States, with a prolonged, bleak, cloudy period producing a total of 6 mm of precipitation. The winds at 850 and 700 mb were missing from the 12 GMT April 5 (0700 April 5 EST) Albany sounding but 500 mb winds were SW, and surface wind SSE. The 00 GMT April 6

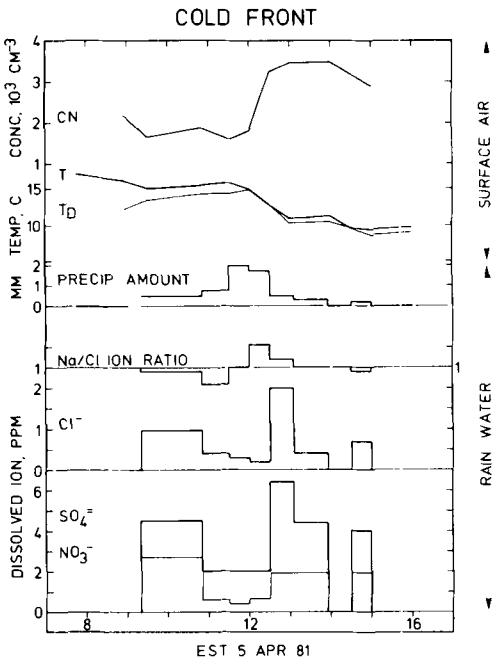

Fig. 1. A chronology of events surrounding the passage of a cold front on April 5, 1981. The sodium to chloride ratio is the molar ratio, which would be unity for pure sodium chloride. The thick line denotes the amount of dissolved sulfate and the thinner line, dissolved nitrate. A total of 6 mm of water precipitation occurred, the major part falling concurrent with the passage of the front.

sounding (1900 EST April 5) showed winds of 250° at 850 and 700 mb, and a WNW surface wind following passage of the cold front. The results of surface observations are shown in Fig. 1.

At the beginning of the experiment, 0745 EST April 5, 1981, it was overcast and calm with a surface temperature of 17 °C and visibility of 25 km. A very brief shower, occurred at 0855, consisting of large drops several centimeters apart, which did not wet the ground. The funnel/collector was rinsed, and a shower began coincident with strong southwesterly gusts at 0920. The initial drops indicated pH 4.5. These gusts and intermittent showers continued until 1050; the first specimen was then collected and the funnel collector rinsed and reset in anticipation of light rain which began just before 1100. This rain continued with lulls, but never totally ceased, from 1105 through 1130 when the collector was changed, rinsed and reset. These initial showers had a pH slightly above 4.0. The showers continued as surface air cooled and approached saturation. A visibility of 8 km was observed in saturated air at 15 °C concurrent with a minimum barometric pressure of 977.4 mb at noon, when the collector was changed, rinsed and reset. The temperature decreased to 13 °C as the barometric pressure increased slightly at 1230, after which the collector was again changed, rinsed and reset. The acidity of this precipitation water was again slightly above pH 4.0. The rain continued, with lulls but never ceasing through 1305 when the collector was changed, rinsed and reset immediately after cessation of rain. This precipitation water was also slightly above pH 4.0. At 1315 the sky brightened, but showers began, which continued through 1355 in gusty winds with rising cloud bases. By 1430 the temperature had decreased to 9.5 °C and another intermittent shower began which ceased completely just before 1500 EST. No additional precipitation occurred after 1500, while temperature continued to decrease, and winds became northwesterly at the surface. All collected rain water samples were stored at about 2 °C, and analyzed by ion chromatography. The preceding observations uniquely describe cold front passage. The front passed between 1200 and 1230 EST, concurrent with greatest rain fall. The chemical precipitation data in Fig. 1 shows that the chloride, sulfate, and nitrate ion concentration decreased but the sodium to chloride ion ratio remained quite near unity before frontal passage. Chloride ion concentration increased in the post frontal shower. This increase in chloride ion concentration was accompanied by increases in sulfate and nitrate ion concentration. Measurements showed hydrogen ion concentrations corresponding to pH slightly above 4 in prefrontal rain, and slightly below pH 4 in post frontal rain.

The precipitation of nitrate, sulfate, chloride and sodium ions, relative to precipitation of water are shown in Fig. 2. There appear to be two steady but different rates of precipitation of ionic mass relative to water mass. When compared to the meteorological events described, these are seen to be a prefrontal (advective) rate, and a five times greater postfrontal or convective rate. The prefrontal, occluded part of the system will be referred to as the "advective side" and the latter portion of the storm, influenced by the cold front, as the "convective side", in interest of conciseness. The

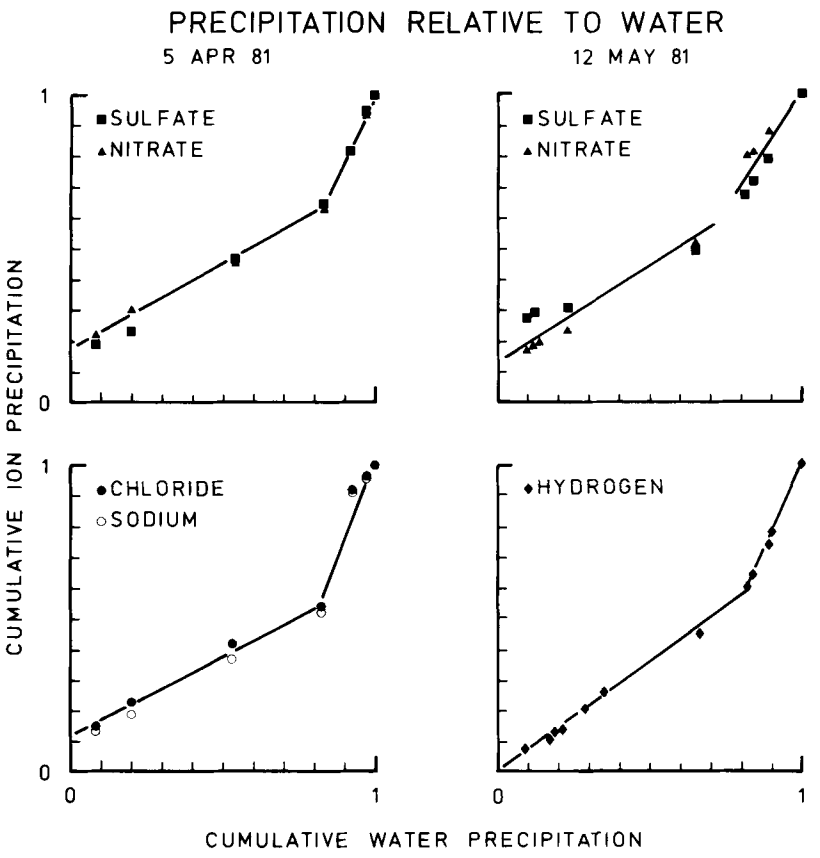

Fig. 2. Cumulative precipitation of dissolved ion mass, with respect to precipitated water mass. The amount of precipitated mass is normalized to a total of one for each storm. The results obtained April 5 are plotted in the left figures; results from May 10–12, 1981, are shown in the two right figures.

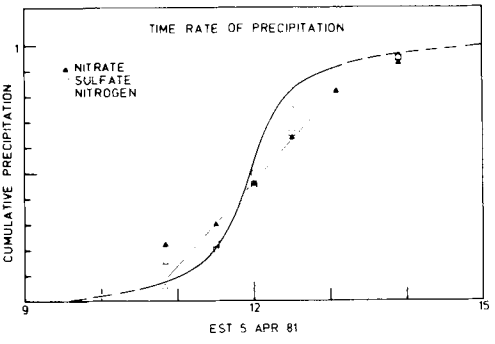

Fig. 3. The cumulative precipitation of water, nitrate ion, sulfate ion, and total (ammonia plus nitrate) nitrogen during the rain of April 5, 1981. The cumulative water precipitation is plotted as a heavy line. The broken portions of the line indicate intermittent rain, while the solid portion indicates steady rain.

straight line through the advective data points has an intercept somewhat greater than zero for all four ionic compounds. This may reflect a steady removal rate of ionic substance in the precipitating cloud, following collection of sub-cloud material or very large nuclei by the initial rainfall.

The relative water precipitation, and the relative precipitation of sulfate, nitrate, and the sum of nitrate and ammonia nitrogen are plotted as a function of time, similar to the analysis of Mordy (1953) in Fig. 3. Steady rain is denoted by the solid line and intermittent rain by the broken line. If the intercept value obtained for sulfate and nitrate in Fig. 2 is subtracted, the resulting linear plot shows them to be precipitated at a constant time rate, during steady rain, in both the pre- and postfrontal situations. The sum of nitrate and ammonia nitrogen

is more closely related to the amount of water precipitated, in disagreement with Mordy's work.

One would not expect any similarity in precipitation of ionic material by such different meteorological events as Hawaiian orographic rain and a midlatitude, cold frontal occlusion, but in both cases, a similarity in time rate of chemical precipitation exists.

In reality the rain-generating processes are quite similar. Moist tropical air is forced to rise over the Hawaiian Koolaus, providing the constant source of moisture and nitrogen described by Mordy. The rain received from cold frontal occlusions is generated by the forced rise of moist air, flowing from the south to west quarter, over a wedge of drier surface air flowing from the south to east quarter. This strong surface flow, generally from the southeast direction, and quite dry, is the result of circulation about a high-pressure system east of the station, according to Danielson (1975). The surface air is not water-saturated at the onset of precipitation, with the initial rain falling through the wedge of dry air. As the preceding high-pressure system moves eastward and is displaced by the low, the surface air becomes increasingly moist, although the surface wind does not change direction. This is accompanied by a lowering of cloud base, heavier rain, and humidity approaching saturation at the surface, as the front passes. Following the passage of the cold front, the above surface air becomes cooler, the residual heat and moisture near the surface generate instability, and the precipitation results from convective processes. The Hawaiian showers have similar small convective cells imbedded, due to release of latent heat, which probably account for the changes in the rate of water precipitation observed by Mordy.

The chronology of the April 5 storm (Fig. 1) shows relatively high concentrations of chloride nitrate and sulfate in the initial showers, while the rain is falling through the large wedge of dry air. The concentrations diminish as cloud base lowers, and then increase sharply following frontal passage. The cumulative rates of ionic precipitation with respect to water increase sharply with passage of the cold front (Fig. 2) but the rate of sulfate and nitrate precipitation with respect to time (Fig. 3) remains constant. This may indicate that there is an ample supply of sulfate and nitrate available and that the amount precipitated is regulated by its rate of entrainment into the cloud and precipitation water.

We know little about sulfur and nitrogen compound vapor concentrations at typical cloud levels. Georgii (1970) measured 5 μg^{-3} of SO_2 at 3–5 km over Germany, and 1 μg^{-3} over Colorado at 2–3 km above the surface. The lowest level of SO_2 concentration reported in the literature is 0.3 μg^{-3} measured at 3.4 km by Bodhaine et al. (1980) at Mauna Loa. Attempts to measure SO_2 concentrations at 1.5 km, elevation about 150 km north of this experiment site failed, as concentrations were always apparently lower than the 5 ppb threshold of the instrument used (Mohnen, private communication).

One might assume, quite conservatively that 1 to 3 ppb of sulfur dioxide would generally be present at cloud-forming levels. This would have the potential of generating 3.4 to 10.2 $\times$ 10 ^{6} g^{-3} of sulfate if oxidation were complete. A typical cloud formed in cold frontal occlusion would contain from 0.1 to 1.0 g^{-3} of condensed water, of which 10%, or from 0.01 to 0.1 g^{-3} of water might be precipitated. There is then a potential to precipitate sulfate ion in rain water at concentrations of 30 to 1000 ppm from such storms, and 3 to 100 ppm of sulfate can be precipitated if only 10% of the available sulfur is collected. There is indeed an ample supply of sulfur available, similar to the constant supply of nitrogen hypothesized by Mordy. It would appear that this linear time rate of sulfate and nitrate precipitation is attributable to a steady flux of nuclei into the cloud, and a constant (at constant temperature) time rate of diffusion of vapors into the cloud droplets. The dissolved vapors can then oxidize into additional sulfate and nitrate while residing in the cloud and precipitating to the surface in rain drops.

4. Results of experiments: prolonged moist advection preceding a cold front

A period of advection of warm, moist air over the station followed by a trailing cold front occurred at the collection site during May 10–12, 1981. A chronology of meteorological events, and precipitated chemicals are shown in Fig. 4.

Very warm, very moist air began moving over the station from the south on the evening of May 10, following a period of clear dry weather. Surface winds were from the southeast and several showers occurred, as the overcast lowered on May 11. At 1900 EST May 11, a saturated layer extended from

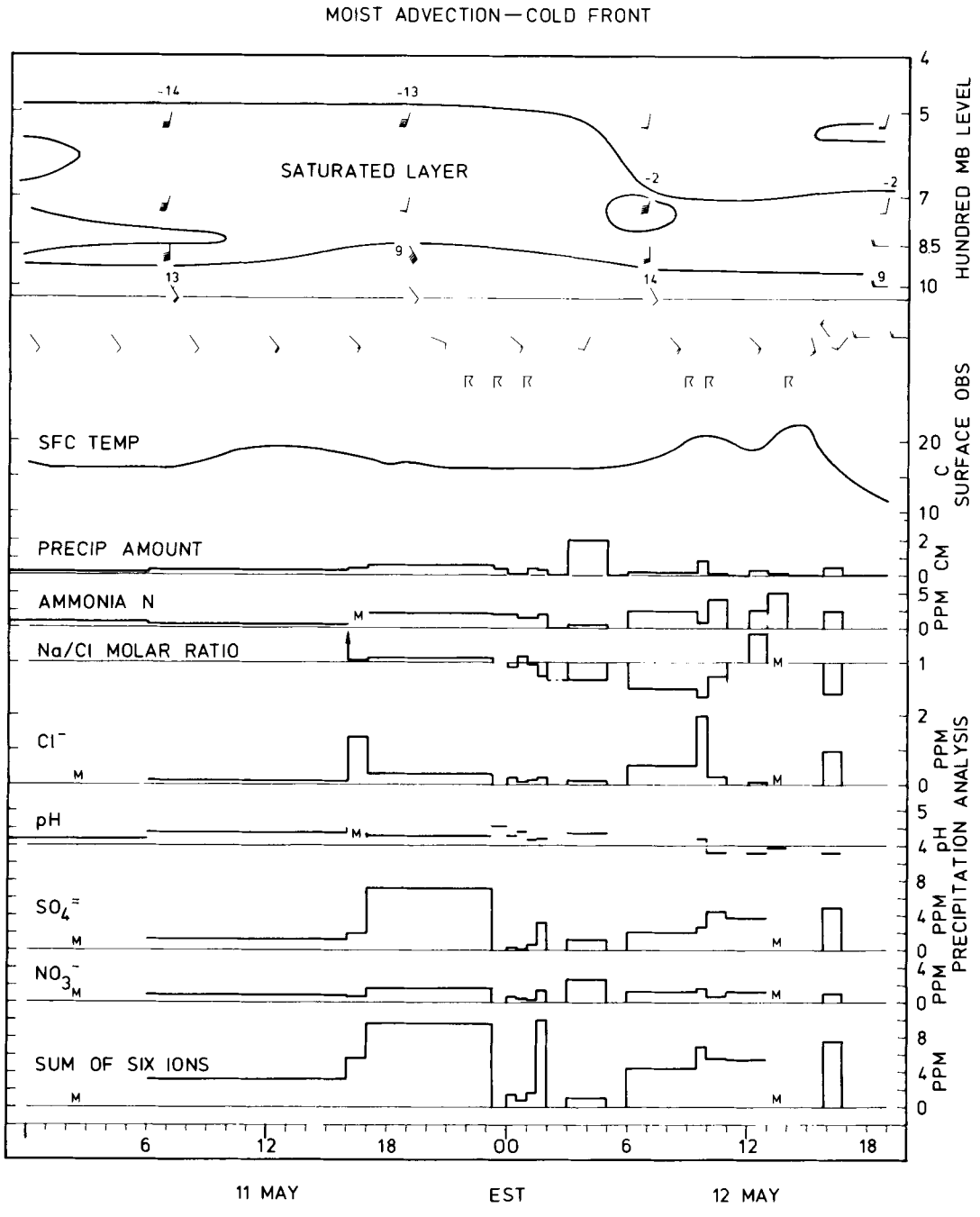

Fig. 4. A chronology of meteorological events and chemical precipitation accompanying a cold frontal occlusion which precipitated 59.9 mm of rain. The sum of six ions is the arithmetic sum of sulfate, chloride, nitrate, ammonium, sodium and potassium, used as an index of the total dissolved material precipitated. Specimens which were not chemically analyzed are designated by M.

below 850 mb to above 500 mb, with 20–25 m s^{-1} winds from the south at all levels in the saturated air as shown at the top of Fig. 4. A thundershower began at 2300 on May 11; showers and thundershowers continued through the night with heavy rains ceasing at 0500 May 12. The sample collector was retrieved, rinsed and reset six times during these thundershowers as rain and cloud conditions appeared to vary. There was complete cessation of rain from 0500 to 0600, and the Albany radiosonde of 0700 EST shows the saturated layer to have thinned, extending from 900 to 700 mb. Several very brief showers occurred, intermittent with weak sunshine from 0700 through 0930, when a heavy shower began, preceded by lightning. Additional showers and lulls occurred throughout the afternoon, ending with a final heavy shower accompanied by lightning and gusty, westerly winds at the surface between 1550 and 1645. The 1900 EST sounding showed westerly winds at the 1000 and 850 mb layers, with southerlies persisting at 700 and 500 mb. The saturated layer still extended from 900 to 700 mb, and an additional saturated layer was just below 500 mb. The surface observation indicates that the warm, moist air was present at the surface after 0700 EST and that the cold front passed at the surface at about 1600 EST. The storm precipitated a total of 59.9 mm of water.

The collected specimens were analyzed for ammonia, nitrogen and hydrogen ion concentration immediately after collection, and by ion chromatography for sulfate, nitrate, chloride, ammonium, sodium and potassium after 3 to 5 days' storage at 2 °C. This analysis indicates that the advective side of the system generally had a hydrogen ion concentration corresponding to a pH higher than 4, and that the convective showers accompanying the cold front were more acidic. The Na/Cl ionic ratio was near unity in the moist advection portion of the storm but varied greatly in the more convective showers.

This storm had considerable variation in its rate and duration of precipitation. Although the observer was continuously present at the site from 2300, May 11, through cessation of precipitation, it was neither always possible to identify individual shower cells, due to convectivity buried in the upper layers, nor to uniquely identify brief rainless periods. This makes a time-rate analysis of such a storm impossible. The large changes in concentration of several ions observed at around 0200 and 1030 indicate the importance of identifying precipitation conditions and resetting the collector, as a few minutes lag, or lead in these collections would have greatly influenced the concentrations measured at these times.

It is possible to perform a cumulative analysis of ionic precipitation with respect to water precipitation with the observations of May 11–12, as shown on the right side of Fig. 2. There is again an unmistakable change in the rate of ionic precipitation, with respect to water precipitation. The precipitation falling during the moist advection period, prior to cessation of rain at 0630, precipitated ions at about one-half the rate with respect to water as that which fell in the more convective showers associated with the cold front. The Na/Cl ionic ratio also differs more frequently from unity during this period.

5. Discussion

Two very different storms were studied. The first was a weak cold frontal occlusion with predominantly westerly to southwesterly flow, weak convection, and no lightning, or other electrical activity. The second storm was a case of strong advection of warm, moist air from the south, with strong convection and frequent lightning and thundershowers imbedded throughout the storm system. The April cold front deposited a total of 6.1 mm of rain in 6 hours and the May storm deposited 59.9 mm of water over a period of nearly 48 hours. The two storms had a very similar trend in chemical precipitation with the final convective stage of the storms precipitating chemicals at a rate of 2 to 5 times greater than the initial stages of the storms. The cumulative precipitation of several substances, derived from the same data base as Figs. 2 and 3, is shown in Table 1.

The total amount of chloride, nitrate, sulfate, and potassium precipitated is in proportion to the total water precipitated for both cases, although the short term precipitation rates varied considerably in both storms. The data of Table 1 indicate that, although the precipitation rate, with respect to water, of the several substances is greater in the convective, or postfrontal portion of both systems, the total precipitation of most substances is greater in the early advective portion of both systems. Only chloride seems counter to this trend.

Table 1. *Cumulative precipitation*

Storm character	H_2O (mm)	N $\mu g\ cm^{-2}$	Cl^-	NO_3^-	$SO_4^=$	Na^+	NH_4^+	K^+	H^+
Moist advection	41.8	0.55	0.88	2.70	6.71	0.55	0.51	0.71	0.193
Convective	18.1	0.32	2.38	2.30	4·12	0·20	0·23	0·21	0·198
Total May 10–12, 81	59.9	0.87	3.26	5.00	10.83	0.75	0.74	0.92	0.391
Pre-frontal	5.04	—	0.17	0.39	1.14	0.12	0.18	0.06	—
Post-frontal	1.10	—	0.15	0.22	0.60	0.11	0.04	0.03	—
Total April 5, 81	6.1	—	0.32	0.61	1.74	0.23	0.22	0.09	—

Rossby and Egnér (1955) show a figure in which they generalize the behavior of sodium and chloride as a storm system passes from sea to land. The initial ratio of sodium to chloride in maritime rain approximates that in sea water, but the more volatile chloride is rapidly diminished as the system moves over land and is removed from the chloride source. Further inland, continental HCl, $CaCl_2$ and $MgCl_2$ are introduced into the system, again raising the Cl/Na ratio to or above the sea water ratio.

The ratio of chloride to sodium precipitated in the advective and convective portions of the two storm systems is shown in Table 2. The weakly convective cold frontal precipitation of April 5 nearly retains the maritime ratio throughout. The initial advective stage of the May 10–12 precipitation has a chlorine to sodium ratio very near to that of sea water, but the convective stages of this storm vary significantly, having large amounts of excess chloride, as Rossby and Egnér hypothesized.

The meteorological data indicates that during the early portion of both storms, surface and near surface winds were from the southeast, and the air was not saturated near the surface. Moist air from the southwest to west, on April 5, and from the south on May 10–12, was forced to rise over this air, becoming saturated and producing rain. This southeasterly flowing air isolated the moist air from the surface, allowing maritime conditions to persist in the precipitation as it travelled the several hundred kilometers from the Atlantic to the observation station. The April 5 record in Fig. 1 shows that only the final half millimeter of precipitation received significant input of continental air to produce an excess amount of chloride.

The warm moist air of May 11–12 was obviously maritime in origin. Lightning was observed in the system prior to the convective phase which began around 1000 on May 12, but this was due to release of latent heat within the system originating the convective cycle aloft, similar to maritime tropical thundershowers, rather than due to differential heating as in continental thundershowers. Strong mixing of surface air into the system by convective activity, during the intermittent sunny periods following 0900 May 12, caused the final convective portion of the storm to be greatly enriched in chloride, in agreement with Rossby–Egnér hypotheses.

Table 2. *Chloride to sodium ratio (using Rossby notation)*

	Storm character	Cl^- ppm	Na^+ ppm	Ratio Cl^-/Na^+
April 5, 81	Advective	0.17	0.12	1.41
	Convective	0.15	0.11	1.36
	Total	0.32	0.23	1.39
May 10–12, 81	Advective	0.88	0.55	1.60
	Convective	2.38	0.20	11.90
	Total	3.26	0.75	4.35

6. Conclusions

Two precipitation systems, representing different synoptic situations, showed very similar characteristics when cumulative chemical precipitation was compared with cumulative water precipitation. The total precipitation of chloride, nitrate, potassium and sulfate by the two storms was nearly uniform with respect to water precipitation.

The results obtained show considerable variation of concentration and precipitation rate of several substances within storms, even though the total deposition of most ions is proportional to the amount of water precipitation as shown by Wilson and Mohnen (1981). The similarity in chemical precipitation rate among such divergent precipitation regimes may of course be due to chance, as only the data of these two systems is available.

An observer at the surface can only hypothesize relative to the processes occurring within clouds above his station. The observations of the two storms described are used to formulate these concluding hypotheses.

The rate of chemical precipitation with respect to water precipitation is a characteristic of the portion of the storm system from which the precipitation is falling. The chemical precipitation rate is greater in the convective rain following frontal passage than in prefrontal rail, but firm numerical values of precipitation rate require several more observations.

Although the rate of precipitation of sodium, chloride, sulfate and nitrate is linear during the prefrontal stages of precipitation, extrapolating this line from the first collection to zero collection shows a positive intercept. This may be a result of preferential nucleation of larger particles during early stages of precipitation, or wet scavenging of additional material below cloud base. Hydrogen ion precipitation also proceeds at a linear rate, but this line passes through the origin. This would indicate that little or no hydrogen ion is collected by scavenging and that the initially favored large nuclei are soluble particles of maritime origin.

The linear rate of precipitation of nitrate and sulfate with respect to time may indicate that these ions result from diffusion of nitrogen and sulfur vapors into cloud drops, and the rate of precipitation depends on the rate of oxidation.

The Rossby–Egnér (1955) model is an important tool for evaluation of continental input to chemical precipitation. Examination of the chloride to sodium ratio in precipitation, coupled with analysis of meteorological parameters can be used to identify events of continental chemical input to the precipitating system. Such analysis cannot be reliably performed on precipitation specimens which represent inhomogeneous precipitation mechanisms.

This work, and these concluding hypotheses, are based on intensive study of but two storms, and may only be the result of chance occurrences. However, it would seem productive to test these hypotheses in similar and dissimilar precipitation occurrences in the course of other precipitation studies.

7. Acknowledgements

This work was supported in part by NOAA-ERL-ARL-GMCC under contract number RAD 00023. The ion chromatographic analysis of collected rain water specimens was done at the ASRC laboratory by D. Bird and A. M. Pitaniello. The author's analysis of the meteorological situations accompanying these precipitation events were verified through chart analysis by K. Kebschull. The author thanks V. Mohnen, R. Castillo, J. Kadlecek, and S. Barnard for many helpful suggestions in preparing the manuscript.

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