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AN INTERCOMPARISON OF THE CHEMICAL AND METEOROLOGICAL DATA GENERATED
FROM AIRCRAFT AND SHIPBOARD SAMPLING DURING GCE/CASE/WATOX

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from Aircraft and Shipboard Sampling During GCE/CASE/WATOX

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ABSTRACT. We compared 16 selected chemical and meteorological parameters from the NOAA ship Mt. Mitchell and the NOAA King Air research aircraft during the GCE/CASE/WATOX experiment. The aircraft made 8 research flights during July, 1988 in coordination with the ship. The first four were off the U. S. East Coast near Newport News, VA and the last four were near the island of Bermuda. We used only the low altitude portions of the aircraft flights in our ship to aircraft intercomparisons. Generally, the aircraft and shipboard measurements were within expected accuracy limits. Selected exceptions for individual species and days are noted and discussed in the text.

1. INTRODUCTION

The combined 1988 Global Change Expedition (GCE), Coordinated Air-Sea Experiment (CASE), and Western Atlantic Ocean Experiment (WATOX) examined the biogeochemical cycles of carbon, nitrogen, sulfur, and trace metals in and above the North Atlantic Ocean. GCE/CASE/WATOX included research cruises aboard the NOAA ship Mt. Mitchell and the private schooner Flertje, a series of flights using the NOAA King Air research aircraft, and surface atmospheric sampling on the island of Bermuda (Pszenny et al., 1991).

CASE was an important component of the GCE/CASE/WATOX. It offered the opportunity to coordinate the sampling of many trace gases and particulates aboard an aircraft and a ship. The NOAA King Air flew 8 research missions. Portions of each mission were at low altitude (150 m above sea level) near the Mt. Mitchell. The GCE/CASE/WATOX scientists gathered parallel meteorological, trace gas, and aerosol samples aboard both platforms. They collected paired samples of a subset of these species. Since different groups collected the paired samples and used varied equipment to do so, it is appropriate to compare the results they obtained. This paper is not intended as an exhaustive quality control document for GCE/CASE/WATOX. The quality control procedures used in individual data sets are described in the contributions which follow. But, we do compare the trace species measurements when paired data were available.

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2. SETTING

2.1 Sampling Platforms

We used the NOAA ship Mt. Mitchell as our marine surface sampling platform. Figure 1 is a schematic drawing of the Mt. Mitchell. A 10-m sampling tower is shown, mounted on the bow of the ship.

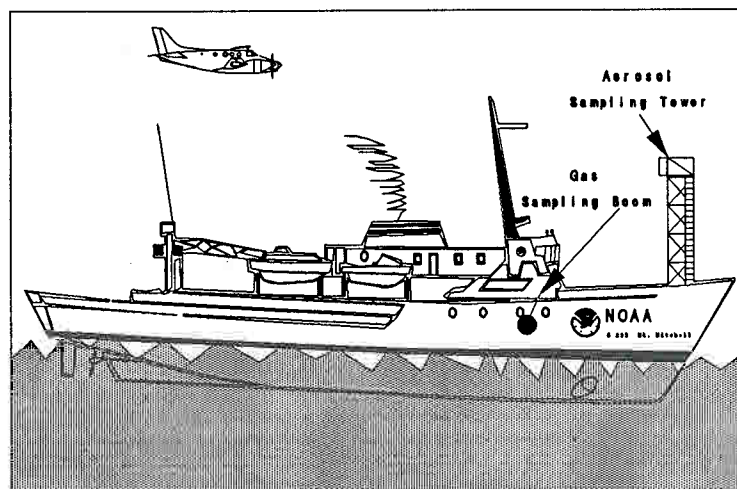


Figure 1.--A schematic of the NOAA ship Mt. Mitchell.

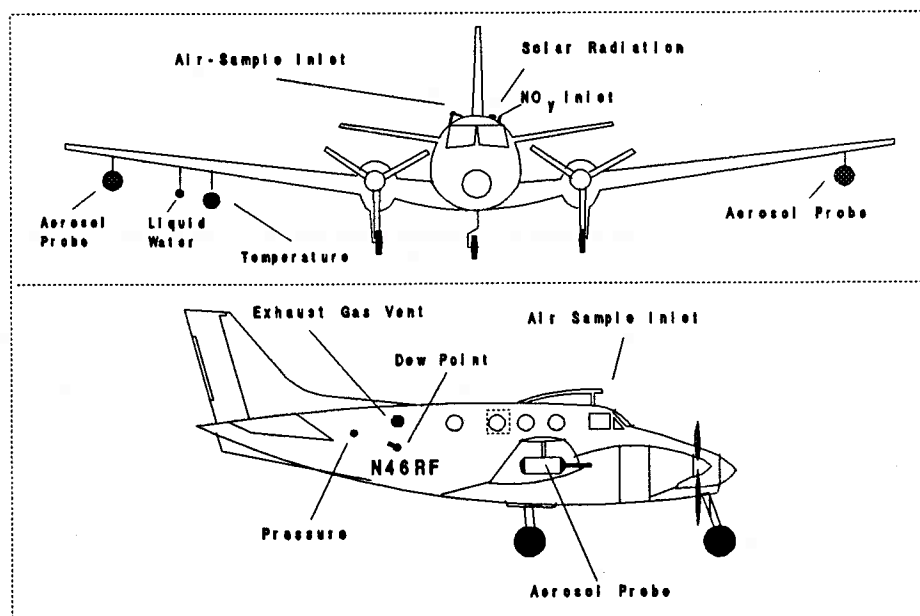


Figure 2.--A schematic of the NOAA King Air research aircraft.

We also used the NOAA King Air (KA) research aircraft as our airborne sampling platform. Figure 2 is a schematic drawing of the KA. Wellman et al. (1989) gave a detailed description of the aircraft.

2.2 Locations and Dates

The Mt. Mitchell left port at Norfolk, VA on 15 July 1988. It steamed to a sampling position about 150 km off the U. S. East Coast (35.7°N , 74.5°W), arriving on 16 July 1988. The ship remained near this location through 21 July 1988. It then steamed to the vicinity of Bermuda (32.0°N , 63.7°W), arriving on 24 July 1988. It stayed near this location through 28 July before docking in Bermuda. We called this leg I of the GCE/CASE/WATOX experiment. Legs II, III, and IV of the experiment were a continuation of the ship cruise and did not involve the aircraft. Figure 3 shows the track of the Mt. Mitchell during Leg I.

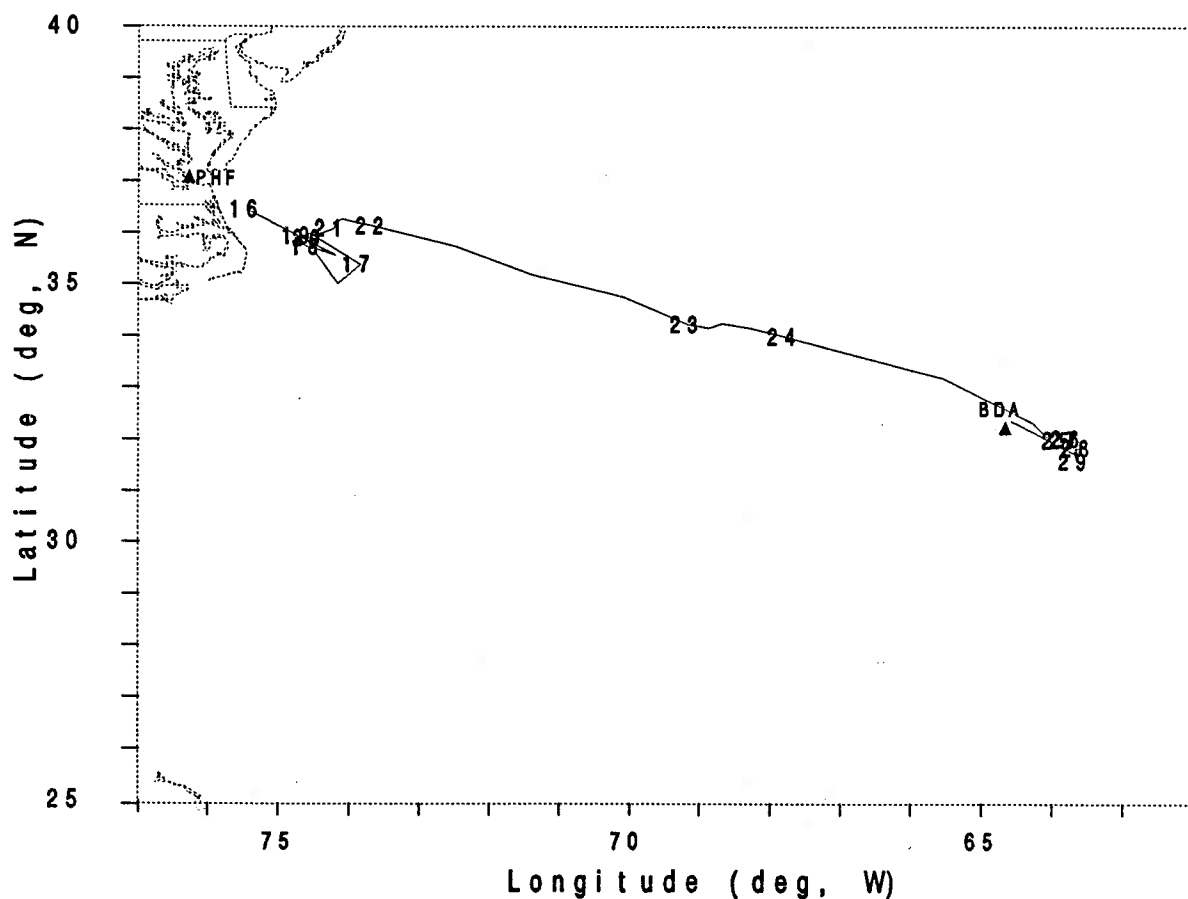


Figure 3.--The track of the Mt. Mitchell during Leg I of GCE/CASE/WATOX.

The KA made eight research flights during GCE/CASE/WATOX. Table 1 lists the dates and times of these eight missions. The first four flights were from Newport News, VA (NPN) to the vicinity of the Mt. Mitchell. The last four flights were from the island of Bermuda (BDA) to the vicinity of the Mt. Mitchell. Figure 4a-b shows a typical aircraft flight pattern from the U.S. East Coast and the Bermuda missions. Gunter and Boatman (1989) gave a complete summary of the aircraft measurements made during GCE/CASE/WATOX.

Figure 5 depicts the altitudes and events common to each aircraft flight. The KA began each mission by climbing to 2,600 m above mean sea level (MSL). We exposed filter packs at this altitude, while flying to the Mt. Mitchell. The KA descended to 150 m, MSL over the Mt. Mitchell, with 15-min stops at 2,300 m, MSL and 760 m, MSL. We exposed a second set of filter packs at 150 m, MSL above the Mt. Mitchell. The KA flew past the Mt. Mitchell one time at 30 m, MSL. It returned to base at 150 m, MSL, while exposing a third filter set. The aircraft filter data in this comparison are from the two 150 m exposures. The aircraft continuous analyzer data are from the single 30 m fly-by interval. Ship filter data include 1-hour exposures overlapping those of the aircraft and half-day exposures bracketing the flight times.

3. MEASUREMENT TECHNIQUES

Tables 2 and 3 list the measurement techniques used to generate the data compared in this study. These tables specify the estimated precisions, detection limits, and response times for the various sampling systems.

4. INTERCOMPARISONS

4.1 Position

Both the KA and the Mt. Mitchell carried Long Range Aid to Navigation equipment (LORAN) for determining their position. Tables 2 and 3 suggest that they are accurate within ± 300 m. The two platforms agreed within 0.0067° (750 m). Considering that the aircraft flew several hundred meters upwind of the ship during the intercomparison period, this agreement is excellent.

4.2 Pressure

Table 2 shows that the Mt. Mitchell carried a barometer accurate to ± 2 mb; Table 3 suggests that the KA had a pressure transducer accurate to ± 2 mb. We show the results of our pressure comparison between the KA and the Mt. Mitchell in Figure 6. The KA averaged 3.9 mb (± 1.1 mb) lower than the ship. As shown in Fig. 5, the KA flew 30 m (3 mb) above the Mt. Mitchell during each intercomparison period. Therefore, the pressure sensors agreed within their ± 2 mb accuracies.

Table 1.--The dates, intercomparison times, fly-by times, and filter exposure times during GCE/CASE/WATOX

Date, 1988	Intercomparison Time (CUT)	Fly-by Time (CUT)	Filter Exp. Ship	Time (CUT) Aircraft	Group
<u>East Coast</u>					
17 July	15:30-16:30	16:24	10:30-22:00	---	AES
			14:33,15:45 ¹	---	AOML
18 July	15:18-16:18	16:06	14:32,15:45 ¹	15:25-16:22 ²	UVA-a
			---	16:43-17:20 ²	UVA-a
			22:00-22:00 ³	---	UVA-b
			10:22-22:20	---	AES
			14:38,15:49 ¹	---	AOML
			14:37,15:49 ¹	15:09-15:59 ⁴	UVA-a
19 July	16:18-17:18	17:06	---	16:10-16:51 ²	UVA-a
			22:00-22:00 ³	---	UVA-b
			10:17-22:10	---	AES
			14:31,15:52 ¹	---	AOML
			14:35,15:49 ¹	16:13-17:03 ²	UVA-a
			---	17:12-18:02 ²	UVA-a
21 July	14:48-15:48	15:36	22:00-22:00 ³	---	UVA-b
			10:19-21:58	---	AES
			14:37,15:50 ¹	---	AOML
			14:37,15:49 ¹	14:45-15:35 ²	UVA-a
			---	15:45-16:35 ²	UVA-a
			22:00-22:00 ³	---	UVA-b
<u>Bermuda</u>					
26 July	14:12-15:12	15:00	10:20-22:00	---	AES
			12:50,14:30 ¹	---	AOML
			12:50,14:30 ¹	13:57-14:47 ²	UVA-a
			---	15:03-15:53 ²	UVA-a
			21:00-21:00 ³	---	UVA-b
27 July (1)	13:00-14:00	13:54	09:28-22:10	---	AES
			12:16,14:07 ¹	---	AOML
			12:15,14:01 ¹	12:55-13:45 ²	UVA-a
			---	13:56-14:46 ²	UVA-a
			23:00-09:00 ⁵	---	UVA-b
27 July (2)	19:12-20:12	20:06	---	---	AES
			19:35,21:08 ¹	---	AOML
			19:35,21:07 ¹	19:08-19:58 ²	UVA-a
			---	20:08-20:58 ²	UVA-a
			---	---	UVA-b
28 July	13:30-14:30	14:18	08:55-24:04	---	AES
			---	---	AOML
			---	13:21-14:11 ²	UVA-a
			---	14:21-15:11 ²	UVA-a
			---	---	UVA-b

¹These are beginning times for each 1-hour exposure.

²Total and Separated filter pack exposure.

³This was a 24-hour exposure.

⁴Total filter pack exposure.

⁵This was a 34-hour exposure.

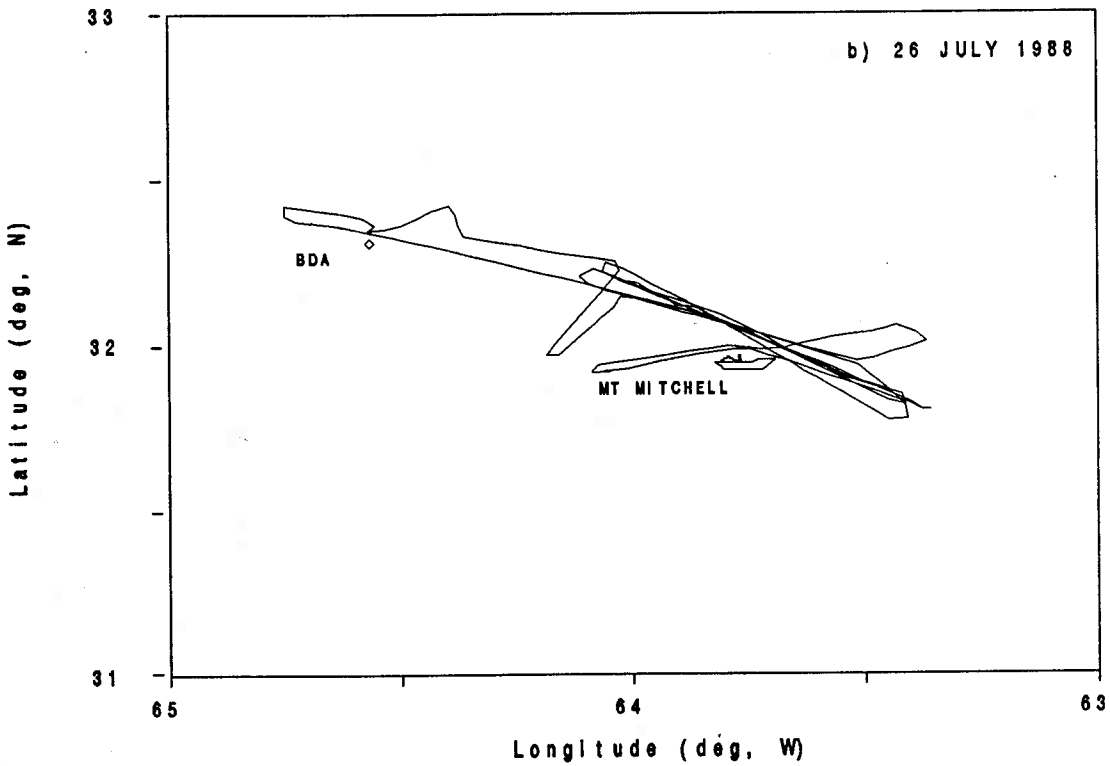
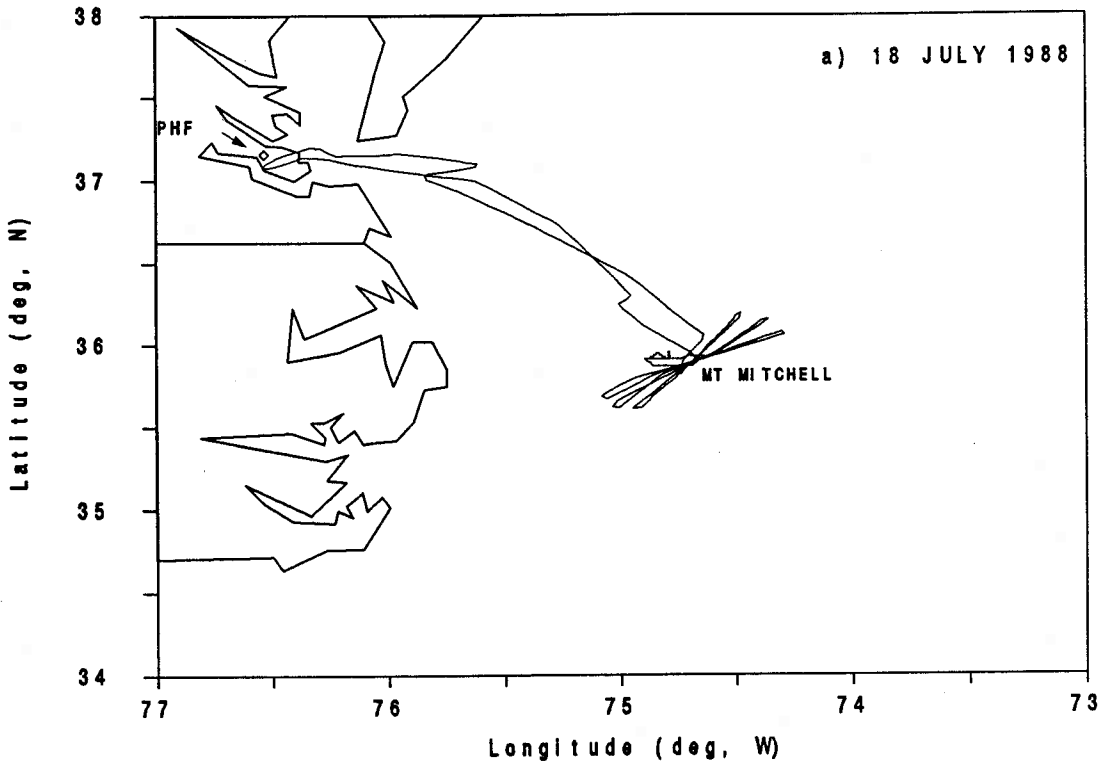


Figure 4.--Typical King Air flight tracks from a) the U.S. East Coast flights, and b) the Bermuda missions.

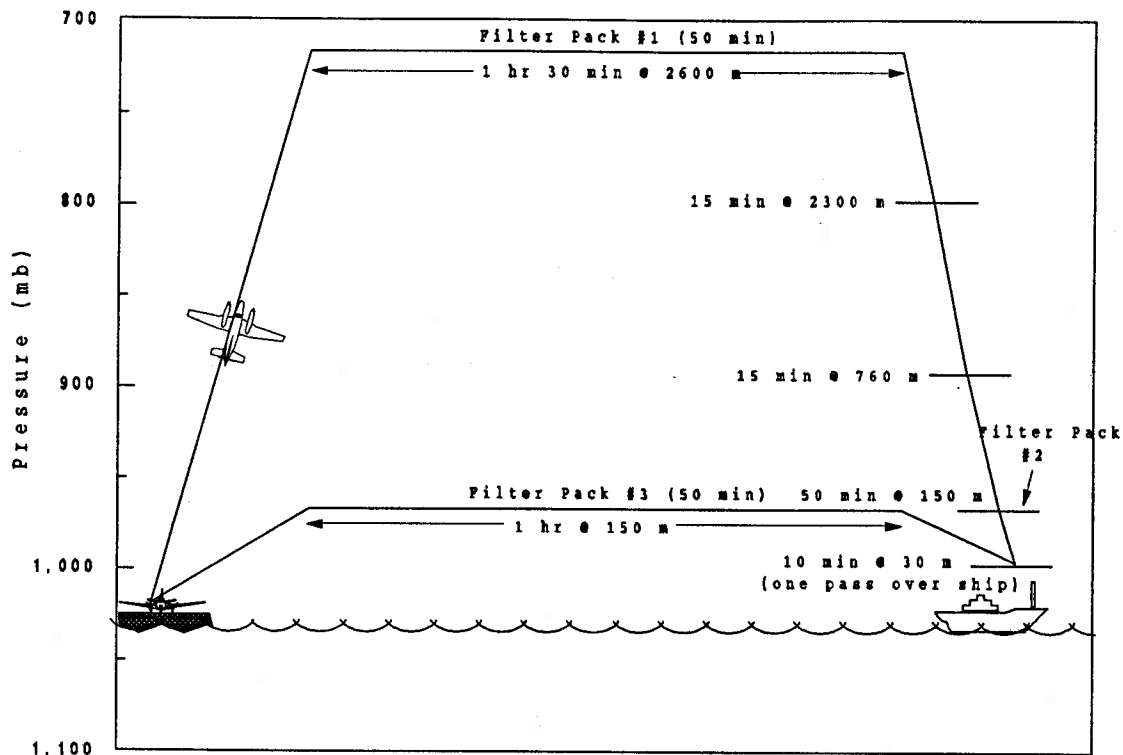


Figure 5.--The altitude and events common to each aircraft flight made during GCE/CASE/WATOX.

4.3 Temperature

The KA carried a platinum resistance temperature sensor accurate to 0.5°C (Table 3); the Mt. Mitchell had a sling psychrometer accurate to 1°C (Table 2). Figure 7 presents the results of the temperature intercomparison between the KA and the Mt. Mitchell. The KA averaged 0.4°C ($\pm 0.7^{\circ}\text{C}$) higher than the Mt. Mitchell. This is within the stated accuracies for both the ship and aircraft.

4.4 Dew Point

The Mt. Mitchell carried a sling psychrometer that measured the wet bulb temperature (Table 2); the KA had a cooled mirror dew point sensor (Table 3). Figure 8 shows the results of the dew point intercomparison between the KA and the Mt. Mitchell. The KA averaged 3.1°C ($\pm 0.7^{\circ}\text{C}$) lower than the Mt. Mitchell. Since dew point typically decreases with altitude, we expected that the aircraft's dew point could be as much as 2°C lower than the one aboard ship. The fact that it averaged 3.1°C lower, suggests a bias in either the aircraft or ship system. The ship system measures atmospheric moisture as the wet bulb temperature. It reaches this temperature via evaporative cooling at constant pressure. If the observer fails to allow sufficient time for completion of the cooling process, the psychrometer wet bulb temperature will be too high. We suspect that this may partially account for the difference in dew point between the two platforms.

Table 2.--The shipboard scientific instruments compared in this study

Response Species	Group	Method	Average			
			Precision	Detection		
				Limit	Time	
<u>Continuous Samplers</u>						
Temperature	NOAA	psychrometer	1° C	0.1° C	5 min	
Dew Point	NOAA	psychrometer	1° C	0.1° C	5 min	
Pressure	NOAA	barometer	2 hPa	0.1 hPa	<1 sec	
Ozone	AOML	uv photometric	2 ppbv	0.1 ppbv	1 min	
SO ₂	NOAA	pulsed fluorescence	1.5 ppbv	1.0 ppbv	1 min	
NO _y	York U.	chemiluminescence	0.2 ppbv	0.05 ppbv	1 min	
Wind Speed	NOAA	anemometer	2 m s ⁻¹	0.1 m s ⁻¹	1 min	
Wind direction	NOAA	anemometer	10 degree	1 degree	1 min	
Position	NOAA	Loran-C	300 m	30 m	1 sec	
<u>Discrete Samplers</u>						
SO ₂	UVA-a	rayon/IC	50 pptv	50 pptv	1 hr	
	AES	nylon, impreg/IC	60 pptv	30 pptv	12 hr	
	AOML-a	rayon/IC	25 pptv	20 pptv	12 hr	
SO ₄ ²⁻	UVA-a	quartz/IC	0.5 nml/scm	0.37 nml/scm	1 hr	
	AES	teflon/IC	0.7 nml/scm	0.6 nml/scm	12 hr	
	AOML-a	quartz/IC	0.9 nml/scm	2.5 nml/scm	12 hr	
MSA	UVA-a	quartz/IC	0.07 nml/scm	0.13 nml/scm	1 hr	
	AOML-a	quartz/IC	0.3 nml/scm	0.1 nml/scm	12 hr	
NO ₃ ⁻	UVA-a	quartz/IC	0.11 nml/scm	0.21 nml/scm	1 hr	
	AES	teflon/IC	0.6 nml/scm	0.6 nml/scm	12 hr	
	AOML-b	teflon/IC	2.7 nml/scm	2.2 nml/scm	1 hr	
HNO ₃	UVA-a	rayon/IC	4.0 pptv	8.0 pptv	1 hr	
	AOML-a	rayon/IC	20 pptv	25 pptv	12 hr	
NH ₄ ⁺	UVA-a	quartz/colorim	4.1 nml/scm	8.1 nml/scm	1 hr	
	UVA-b	zeflour/colorim	4 nml/scm	2 nml/scm	24 hr	
	AOML-b	quartz/IC	1.5 nml/scm	1.9 nml/scm	1 hr	

4.5 Horizontal Wind

The Mt. Mitchell measured the horizontal wind via anemometry; the KA used its position, true airspeed, and heading to compute the horizontal wind.

Table 3.--The airborne scientific instruments compared in this study

Detection Species	Response Group	Method	Average			
			Precision	Limit		Time
<u>Continuous Samplers</u>						
Temperature	NOAA	platinum res.	0.5° C	0.1° C		<1 sec
Dew Point	NOAA	hygrometer	1° C	0.1° C		2 sec
Pressure	NOAA	transducer	2 hPa	0.1 hPa		<1 sec
Ozone	NOAA	uv photometric	2 ppbv	1.0 ppbv		1 hr
SO ₂	NOAA	pulsed fluorescence	0.1 ppbv	0.05 ppbv		1 min
NO _y	NOAA	chemiluminescence	0.1 ppbv	0.1 ppbvq		1 min
Wind Speed	NOAA	computed	2 m s ⁻¹	0.1 m s ⁻¹		1 min
Wind direction	NOAA	computed	10 degree	1 degree		1 min
Position	NOAA	Loran-C	300 m	30 m		1 sec
<u>Discrete Samplers</u>						
SO ₂	UVA-a	rayon/IC	50 pptv	50 pptv		50 min
SO ₄ ²⁻	UVA-a	quartz/IC	0.5 nml/scm	0.37 nml/scm		50 min
MSA	UVA-a	quartz/IC	0.3 nml/scm	0.13 nml/scm		50 min
NO ₃ ⁻	UVA-a	quartz/IC	0.5 nml/scm	0.21 nml/scm		50 min
HNO ₃	UVA-a	rayon/IC	0.9 pptv	0.45 pptv		50 min
NH ₄ ⁺	UVA-a	quartz/colorim	10 nml/scm	8.1 nml/scm		50 min

Figure 9 is a wind rose depicting the results from each intercomparison. We plotted the wind directions and speeds measured by the KA (x) and the Mt. Mitchell (+) as the aircraft passed 30 m above the ship. The average wind direction difference between the KA and Mt. Mitchell was -2° ($\pm 11^\circ$). This is within the $\pm 10^\circ$ accuracy given in Tables 2 and 3. The average difference in wind speed between the KA and Mt. Mitchell was 3.4 m s⁻¹ (± 2.5 m s⁻¹). Figure 9 shows that the KA wind speeds were consistently higher than those aboard the Mt. Mitchell. This is plausible, since wind speed typically increases to its geostrophic value at the top of the marine boundary layer, according to the Ekman equations (Hess, 1959). Considering this fact, the wind speed accuracies given in Tables 2 and 3 for the KA and Mt. Mitchell are reasonable.

4.6 Ozone

We measured ozone aboard the KA and Mt. Mitchell using uv-photometric analyzers. The KA carried a Thermo-Environmental Inc. (TECO) model 49 ozone analyzer. The Mt. Mitchell had a Dasibi model 1008-AH. Both devices were compared to a standard ozone instrument, traceable to the National Institute for Standards and Testing (NIST). Neither analyzer had a filter for removing aerosol particles.

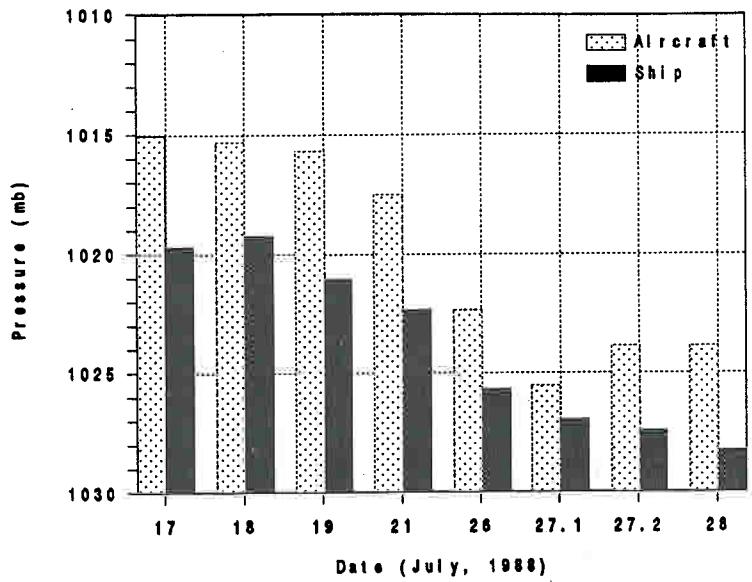


Figure 6.--The ambient pressures that the KA and Mt. Mitchell measured during each of their 8 intercomparisons.

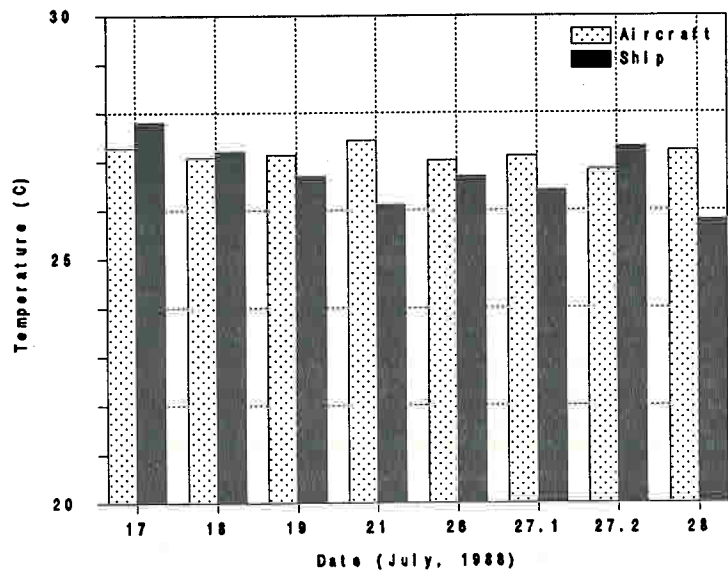


Figure 7.--The temperatures that the KA and Mt. Mitchell measured during each of their 8 intercomparisons.

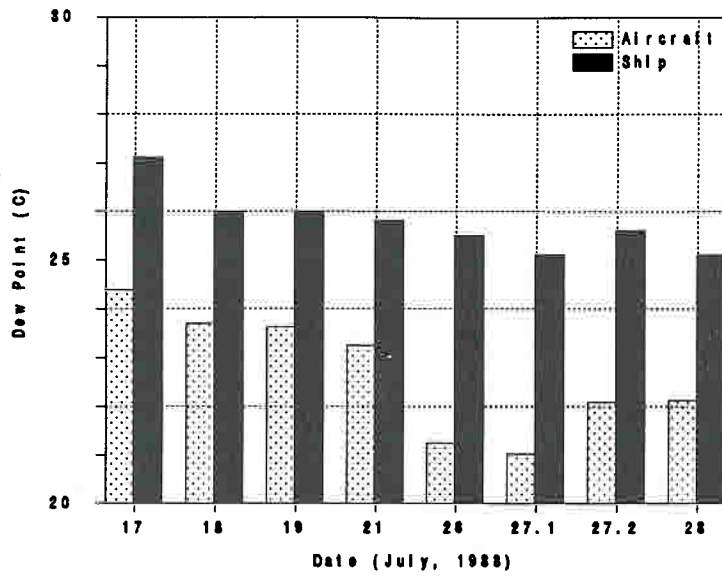


Figure 8.--The dew points that the KA and Mt. Mitchell measured during each of their 8 intercomparisons.

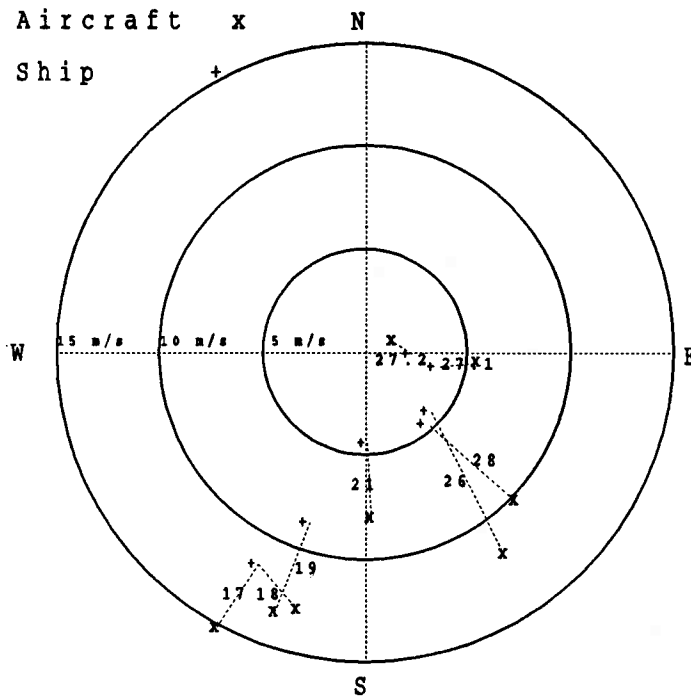


Figure 9.--The horizontal wind speeds and directions that the KA and Mt. Mitchell measured during each of their 8 intercomparisons. Numbers represent the dates of each intercomparison (see Table 1).

Figure 10 shows the results of the 8 intercomparisons between the two systems. Six of the eight comparisons were favorable and agreed within 0.5 ppbv (± 1.3 ppbv). The ship data were unavailable during the second flight on 27 July 1988. In the 18 July 1988 comparison, the Mt. Mitchell sensor reported a concentration of 110 ppbv; the KA device read 18 ppbv. Figure 10 shows that the KA analyzer measured a concentration typical of those encountered in marine air; the Mt. Mitchell sensor measured a concentration that was higher than any other observed.

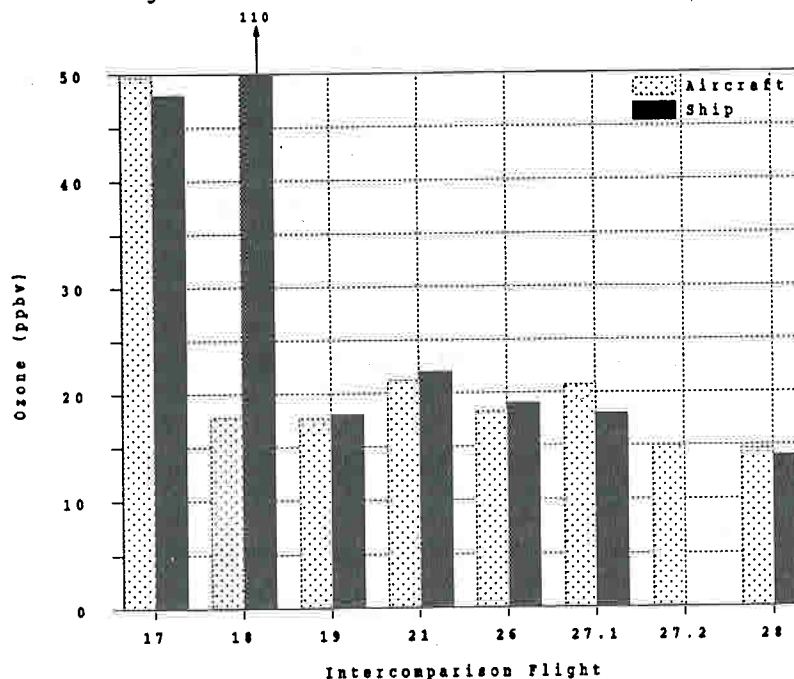


Figure 10.--The ozone concentration that the KA and Mt. Mitchell measured during each of their 8 intercomparisons. The AOML value for 18 July is believed to be spurious, due to condensation in the instrument inlet tube.

A scan of the complete Mt. Mitchell ozone record revealed occasional short intervals prior to 21 July 1988 when the ozone concentration rose above the 100 ppbv level. The reason for these excursions was probably water condensing in the intake tubes to the Mt. Mitchell system. The excursions ceased on 20 July, after the sensor was moved from an air conditioned environment to the hot pump room aboard the ship.

4.7 Sulfur Oxides

Sulfur Dioxide (SO_2). The instrumentation aboard the KA included a continuous analyzer (TECO, model 43s) for the sampling of SO_2 (Luria et al., 1988; Luria et al., 1989) and two three-stage-tandem 90-mm filter packs. We sampled atmospheric aerosols on an upstream quartz filter and alkaline reactive gases of sulfur (SO_2), nitrogen (HNO_3), and chlorine (HCl) on two downstream Rayon filters impregnated with K_2CO_3 (Bardwell et al., 1991). One filter system sampled whole air (total system), while the other was downstream from a cyclone separator (separated system). The separated system removed large particles with a 50% cut-off at approximately $0.8 \mu\text{m}$ diameter (Boatman

et al., 1989). Although we expected the aerosol composition in the parallel systems to be different, we presumed that the paired SO₂ filter analyses would be similar.

The continuous SO₂ instrument employs the pulsed fluorescence method; it is an improved version of the original pulsed fluorescence device, designed to measure low SO₂ levels. According to the manufacturer's specifications, instrument precision is ±100 pptv and the minimum detection limit is 50 pptv (95% response time). Its rejection rate for NO is 1:50, and it is unaffected by ambient levels of NO₂, O₃, CO, or CO₂. Still, the device is sensitive to water vapor density and temperature changes.

Although the zero-level response of a prior continuous SO₂ monitor used aboard the KA (TECO model 43a, ppbv range) was insensitive to altitude (pressure) changes (Boatman et al., 1988), we found that this was not so for the TECO model 43s (sub-ppbv range). Apparently, changes in temperature and humidity at the various sampling altitudes shift the baseline by a value equivalent to several tenths of a ppbv. Even zero checks at each altitude could not overcome this offset; the humidity and temperature of the pure air used to calibrate the instrument could not be adjusted during flight. To compensate for the variable zero level, we made a zero offset correction for each sampling leg. The correction assumes that the lowest reading of that segment is zero.

Additionally, we observed a baseline drift in several cases, while sampling at constant altitude. To account for the drift, we computed a baseline correction, using the two lowest readings near the beginning and near the end of each constant-altitude flight leg. The consequence of this procedure is that the reported SO₂ concentrations are upper limits. Temperature and dew point fluctuations between ground level and the higher sampling level were 20-40 times greater than fluctuations within a constant-altitude sampling leg; we estimated that temperature and dew point influences on the zero level of the SO₂ analyzer within a constant-level flight leg were less than 10 pptv.

The Mt. Mitchell carried a Thermo-Electron 43A continuous analyzer with an accuracy of ±1.5 ppbv and a detection limit of 1 ppbv. This device suggested that SO₂ concentrations were below its detection limit during each intercomparison period. The Mt. Mitchell also had multi-stage filter pack systems aboard for the measurement of atmospheric aerosols, HNO₃, and SO₂.

The agreement between the two aircraft filter systems (see Table 4) is excellent [$SO_2(\text{separated}) = 1.08 \times SO_2(\text{total}) + 6.76$, $r^2 > 0.99$]. The slope of the linear fit suggests that SO₂ measured with the separated filter pack was 8% higher, on average. This observation suggests that some SO₂ may have been scavenged from the air stream by the accumulation of alkaline sea-salt aerosol on the upstream filter of the total aerosol sampler. This resulted in a small negative bias in SO₂ from that system (see Bardwell et al., 1991). Since the data set from the total filter pack was more complete, the comparison with the continuous monitor is based on it.

Table 4.--The average SO₂ concentrations (pptv) measured aboard the KA.

Flight leg	Continuous	Total Filter Pack	Separated Filter Pack
17.1	119±110	160±10	--
17.2	350±260	670±30	630±50
17.3	1460±119	1890±40	2070±60
18.1	90±40	0±10	--
18.2	140±90	130±30	--
18.3	1590±1340	1470±40	--
19.1	80±40	50±10	--
19.2	140±100	130±30	140±50
19.3	220±240	190±30	220±50
21.1	60±30	0±10	--
21.2	130±20	70±30	80±50
21.3	70±50	40±40	30±40
26.1	90±40	20±10	--
26.2	60±30	40±10	60±50
26.3	70±30	30±10	40±50
27.1.1	70±30	20±10	--
27.1.2	100±80	90±10	170±50
27.1.3	60±70	40±10	40±50
27.2.1	60±30	10±10	--
27.2.2	40±20	20±10	20±50
27.2.3	50±20	20±10	10±50
28.1	40±20	20±40	--
28.2	50±40	30±10	30±50
28.3	50±30	10±10	70±50

Figure 11 presents a comparison between the airborne filter analyses and the corresponding continuous analyzer averages calculated for the same period. This figure shows that the data from the two independent methods have a small bias. The correlation between the integrated continuous analyzer values and the filter data is very good, when considering the entire 24 data points ($r^2 > 0.94$). Also, if we exclude the three highest measurements, and compute the correlation coefficient for SO₂ < 250 pptv, the agreement remains very good ($r^2 > 0.9$); the slope of the linear fit is nearly 1. But, we see that the continuous method averaged 35 pptv higher than the filter pack technique. A portion of this bias probably relates to the procedure used to zero the continuous analyzer that, as previously noted, results in higher estimates for SO₂ concentrations. Other unidentified factors also may have contributed to these small differences between the two techniques.

Figure 12 is a scatterplot comparing the airborne filter-derived SO₂ concentrations with their shipboard counterparts from the Canadian Atmospheric Environment Service (AES), the University of Virginia (UVA), and the NOAA Atlantic Oceanographic and Meteorological Laboratory (AOML). We derived sulfur dioxide values from the AES filter system using the total sulfur loading on the nylon and Whatman-41 filters (Anlauf et al., 1985). But, for these samples, loadings on the latter were quite low. This comparison used the 7 airborne filters exposed nearest to the ship (see fig. 5). Figure 12

shows a reasonable correspondence between the KA and UVA data sets [$\text{SO}_2(\text{UVA-a}) = 0.89 \times \text{SO}_2(\text{KA}) + 0.50$, $r^2 > 0.99$]. The agreement between the KA and AES data is less comforting [$\text{SO}_2(\text{AES}) = -0.06 \times \text{SO}_2(\text{KA}) + 2.37$, $r^2 = 0.21$]. There is a suggestion that the AES system gave systematically lower SO_2 concentrations than those of the KA and UVA. We suspect an exposure problem with the AES Whatman-41 filters. Finally, figure 12 suggests that the KA total filter pack measured more SO_2 than did the AOML-a system [$\text{SO}_2(\text{AOML-a}) = 0.45 \times \text{SO}_2(\text{KA}) + 3.29$, $r^2 = 0.83$].

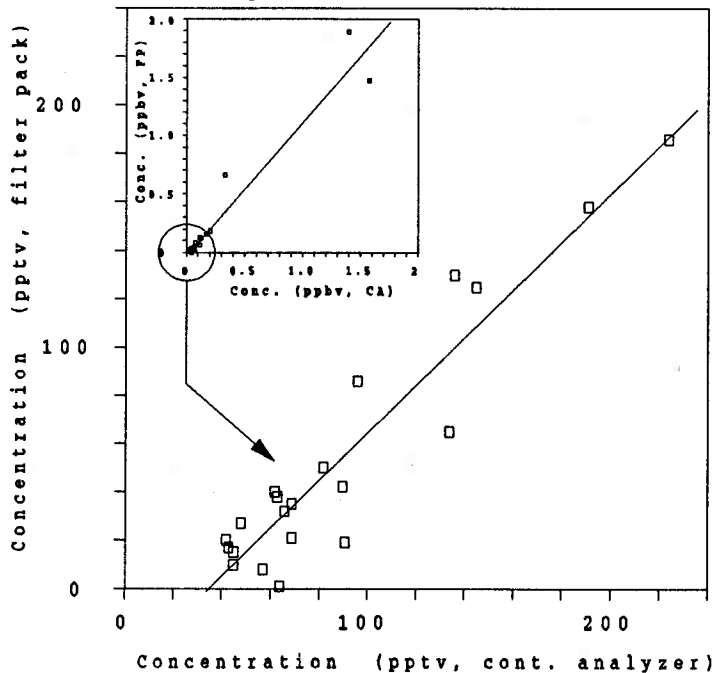


Figure 11.--A comparison of the SO_2 concentrations measured aboard the KA using the continuous analyzer and the filter pack. The inset is for SO_2 concentrations between 0 and 2 ppbv.

Sulfate (SO_4^{2-}). We sampled SO_4^{2-} aerosol particles aboard the KA using quartz filters installed in both the total and separated filter systems; UVA quantified the amount of non-sea-salt SO_4^{2-} on each filter using the technique of Bardwell et al. (1991). We also sampled SO_4^{2-} aerosol particles aboard the Mt. Mitchell with quartz (UVA, AOML-a) and Teflon (AES) filters. UVA analyzed one set of filters (Bardwell et al., 1991), AES analyzed a second set (Anlauf et al., 1985), and AOML analyzed a third set.

Figure 13 shows histograms of our non-sea-salt SO_4^{2-} concentration measurements for the 8 available flights. Close inspection reveals that the KA total and separated systems were in good agreement, except on 28 July. The total system collected slightly more SO_4^{2-} in 12 of the 14 comparisons. We expected the total system to yield more SO_4^{2-} , since the separator preferentially removed aerosol particles larger than about $0.8 \mu\text{m}$ in diameter. The differences noted on 28 July suggest that, on this day, a large fraction of the non-sea-salt sulfate was associated with coarse-fraction aerosol particles. The aircraft also saw the highest sea-salt concentrations and the biggest Cl^- losses on this day (Keene et al., 1991).

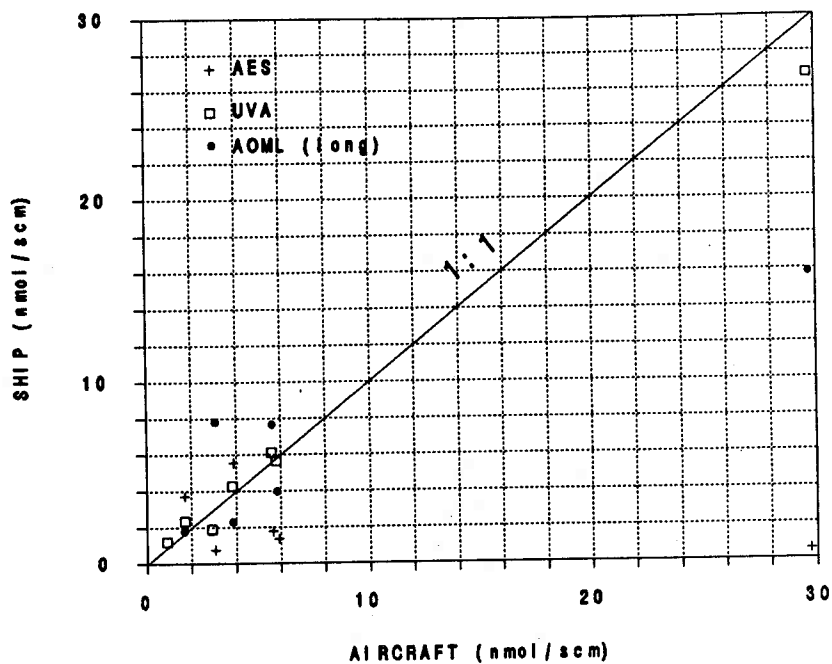


Figure 12.--A comparison between the SO_2 concentrations measured with filter packs aboard the KA and on the Mt. Mitchell.

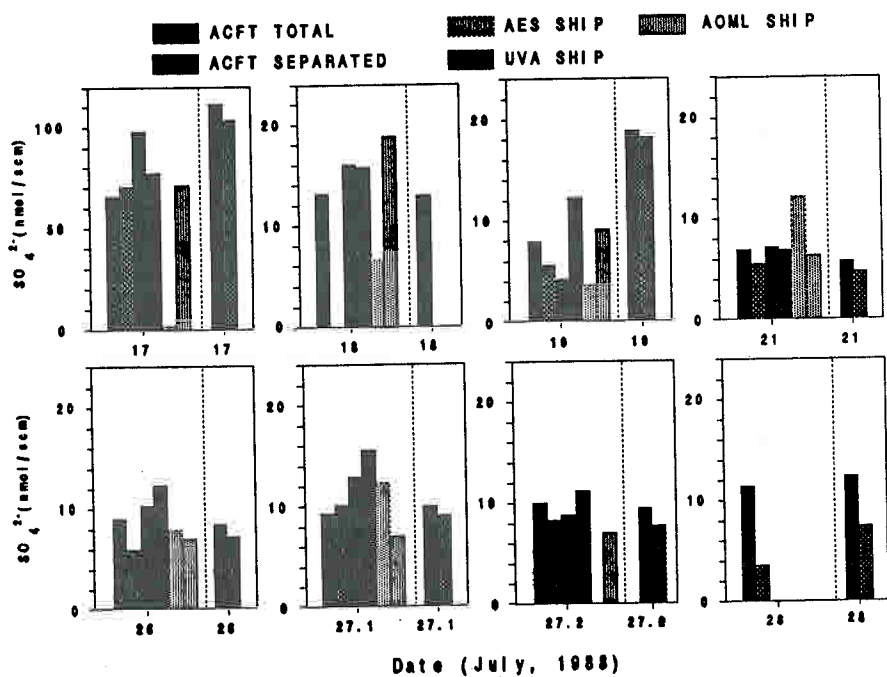


Figure 13.--A comparison between the non-sea-salt SO_4^{2-} concentrations measured with filter packs aboard the KA and on the Mt. Mitchell. Those to the right of each dotted line are for filters exposed while the aircraft returned to base.

The comparison between the UVA, AES, and AOML shipboard filters in fig. 13 yields mixed results. The AES system measured lower SO_4^{2-} concentrations than the other two in 5 of the 6 comparisons. The small size of our comparison set precludes a more detailed statistical summary.

The comparison between the shipboard and aircraft filter systems in fig. 13 also gives mixed results. We exposed two UVA filter packs aboard the ship during each aircraft flight, one during the fly-by and a second afterward (see fig. 5). The comparison reveals that UVA's shipboard SO_4^{2-} concentrations were generally 10-20% higher than those aboard the KA. This seems reasonable.

We exposed the KA filters at 150 m above the sea, where coarse-mode aerosol particles are significantly smaller in number than at the sea surface. The comparison also reveals that the AES's shipboard SO_4^{2-} concentrations were more variable than the other three systems.

The small number of samples compared here limits our ability to do a rigorous statistical assessment of differences between the paired data. Some differences observed in the data were real and resulted from temporal and spacial variability in the sampled air masses. We saw artifacts but the results were consistent. This was a useful effort because it proves the difficult nature of making comparable and representative measurements of non-sea-salt SO_4^{2-} from a ship and aircraft.

Methanesulfonic Acid ($\text{CH}_3\text{SO}_3\text{H}$, MSA). We measured MSA aboard the KA using the total and separated filter systems and the analysis technique described by Bardwell et al. (1991). We measured MSA aboard the Mt. Mitchell using the Bardwell et al. procedure and a similar one developed at AOML. Figure 14 depicts the results of the daily comparisons we made.

Examining first the comparison between the KA filter systems, we see that the two agreed within 10% in 5 of 14 cases. In the remaining 9 cases, the separated system measured smaller MSA concentrations than those seen with the total system. Although MSA is present as sub-micron aerosol particles (Saltzman et al., 1983), we observed significant percentages (17 to 32%) in the coarse-fraction aerosol ($>1.0 \mu\text{m}$ diameter) for several cascade impactor samples collected during the eastern North Atlantic portion of the cruise. These observations are consistent with those from the aircraft, and suggest that, at times, significant amounts of MSA may be associated with the coarse marine aerosol.

Examining second the comparison between the shipboard filter systems, we see that the two agreed within about 20% in all 7 cases. In 5 of the 7 cases, the UVA-a system measured more MSA than the AOML-a one. We speculate that differences in the sampling technique between the two systems (UVA-1hour; AOML-12 hours) may have caused this effect.

Examining third the comparison between the KA and Mt. Mitchell systems, we see that the KA total system and the AOML-a system agreed within 10% in 2 of 7 cases. The KA system was as much as 20% lower than the AOML-a one in the remaining 5 cases. The KA system agreed within 10% of the UVA system in only 1 of 7 cases; it was as much as 40% lower than the UVA-a system in the remaining 6 comparisons. The KA separated system was always lower than the shipboard UVA-a device; it was also lower than the AOML-a system, except on 17 July 1988.

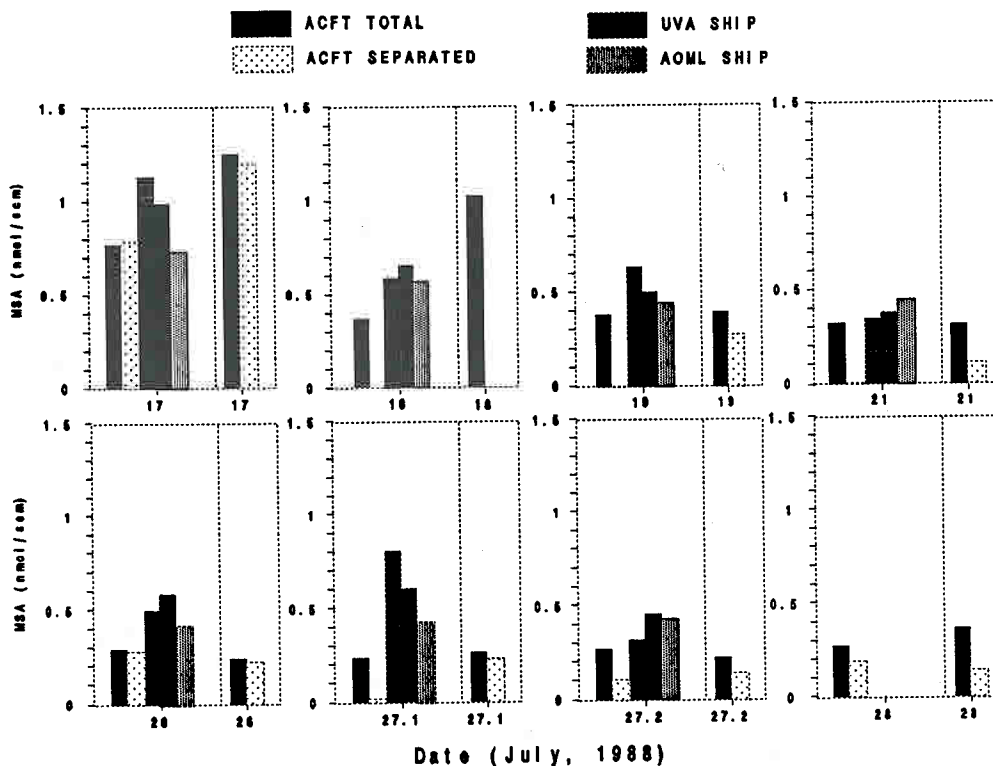


Figure 14.--A comparison between the MSA concentrations measured with filter packs aboard the KA and on the Mt. Mitchell. Those to the right of each dotted line are for filters exposed while the aircraft returned to base.

We know that the population of aerosol particles decreases with altitude in the marine boundary layer (Kim et al.; 1991) due to gravitational settling. Therefore, it is reasonable to suggest that MSA concentrations at the sea surface should be higher than those aloft, due to their association with the marine aerosol. This was the case in all the KA total vs UVA comparisons and in 5 of 7 KA total vs AOML-a comparisons. A quantitative statistical comparison of these diverse sampling systems is not possible, due to the sparseness of data.

4.8 Nitrogen Oxides

Reactive Nitrogen Species (NO_y). We define NO_y as the sum of NO , NO_2 , HNO_3 , PAN, RONO_x , HO_2NO_2 , $2x\text{N}_2\text{O}_5$, and NO_3^- (see Dickerson et al.; 1987, Pickering et al.; 1988). We measured NO_y aboard the KA with a single detector $\text{NO}-\text{O}_3$ chemiluminescence device, utilizing heated molybdenum for NO_y to NO conversion (Ray et al., 1991). We measured NO_y aboard the Mt. Mitchell with a luminol- NO_2 chemiluminescence instrument that used gold and chromium trioxide for NO_y to NO and NO to NO_2 conversion, respectively (Hastie et al., 1991).

Figure 15 shows a comparison of the two instruments during each of 8 flights. In one case (27 July 1988, flight 2) the shipboard system was not working. In 5 of the remaining 7 cases, the aircraft and shipboard systems agreed within 20%. The best-fit linear regression line for these 5 cases is

NO_y (KA, ppbv) = 0.145 + 0.874 x NO_y (Mt. Mitchell, ppbv) with $R^2 = 0.84$.
 This reveals two biases. The first, an offset of 0.145 ppbv, could be due to a calibration error or to incorrect baseline zero subtraction in one or both instruments. The second, a proportional offset dependant on NO_y magnitude, may be due to calibration differences, sample line losses, or differences caused by the inlet filters.

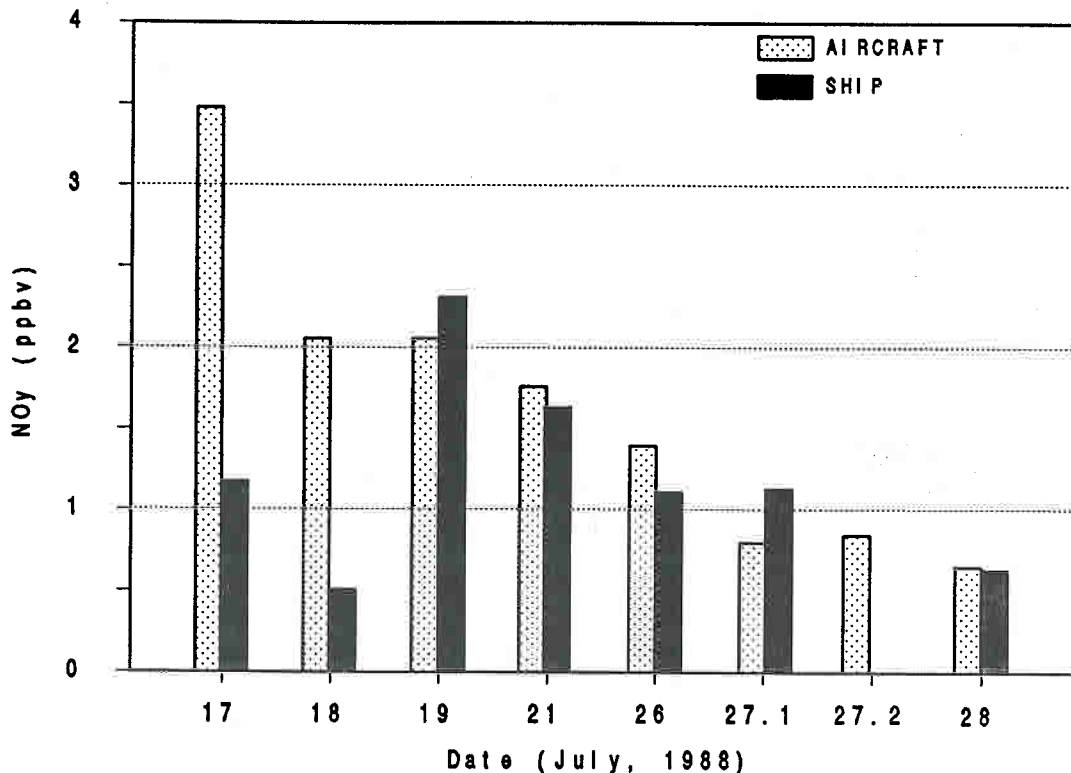


Figure 15.--A comparison between the NO_y concentrations measured continuously aboard the KA and on the Mt. Mitchell.

Figure 15 also reveals a substantial difference between the KA and Mt. Mitchell NO_y concentrations on 17-18 July 1988. Several factors suggest that the shipboard measurement was too low on these days. First, the KA values were more consistent with the nearshore concentrations measured in this and previous studies (Misanchuk et al., 1987). The Mt. Mitchell concentrations were more typical of the cleaner values found near Bermuda. Second, the shipboard values shift into agreement with the aircraft ones on 19 July 1988 (Hastie et al., 1991). Third, other trace species reflected a continental influence on 17 and 18 July 1988 (see fig. 10).

Nitric Acid Vapor (HNO_3). We measured HNO_3 with impregnated Rayon filters aboard the KA and Mt. Mitchell. Bardwell et al. (1991) described our sampling and analysis technique. We exposed one HNO_3 filter in the total system aboard the KA during each of its 150 m sampling runs. We exposed two shipboard HNO_3 filters at times that encompassed the 150 m sampling run of the aircraft.

Figure 16 shows the results of our HNO_3 intercomparison. The two shipboard filters from each case agreed within several percent, except on 17

July 1988. Since HNO_3 concentrations were at their highest observed values on this day, we suspect that HNO_3 variability was also high. Aircraft HNO_3 concentrations were significantly higher than those aboard ship during 3 of the 4 U. S. East Coast flights and one of the Bermuda flights. They were about equal to one another during 2 of the 4 Bermuda flights. The aircraft HNO_3 concentrations were significantly lower than the shipboard measurements on 17 July 1988. Generally, airborne samples were equal to or greater than those aboard ship in 6 of the 7 available cases. Keene et al. (1991) suggested that this is the result of heterogeneous chemical reactions on sea salt aerosols, which are most prevalent near the surface of the sea (Kim et al.; 1991).

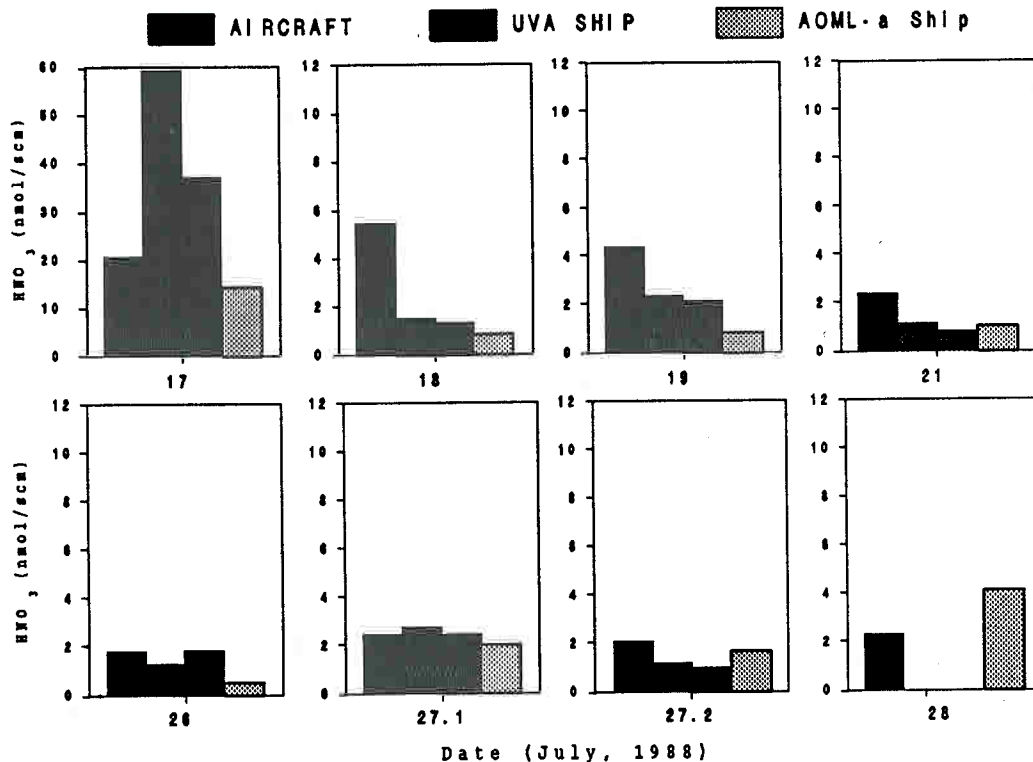


Figure 16.--A comparison between the HNO_3 concentrations measured with filter packs aboard the KA and on the Mt. Mitchell. Note the different vertical scale for 17 July.

Particulate Nitrate (NO_3^-). We measured particulate nitrate on the KA (quartz filters) and aboard the Mt. Mitchell (UVA-a, AOML-b - quartz filters and AES - Teflon filters). UVA analyzed the aircraft filter set and one of the shipboard filter sets using the ion chromatographic technique described by Bardwell et al. (1991). AOML and AES analyzed their filters by ion chromatography.

Figure 17 shows a comparison of our results; shipboard measurements were not available on 28 July 1988. The KA separated filters measured less NO_3^- than their total counterparts in all 14 available cases. We interpret this to mean that a portion of the aerosol NO_3^- was on particles $>0.8 \mu\text{m}$ in diameter. The UVA-a, AOML-b, and AES shipboard filter samples compared favorably, except on 17 July 1988. On this day, the UVA-a NO_3^- concentrations were much higher than the AOML-b ones and the AOML-b ones were much higher than those of the

AES. If we exclude the 17 July 1988 samples as outliers, the UVA-a NO_3^- concentrations relate to those of AOML-b through the equation $\text{NO}_3^-(\text{UVA-a}) = 1.97 + 0.83 \times \text{NO}_3^-(\text{AOML-b})$ with $R^2 = 0.58$. Similarly, the UVA-a NO_3^- concentrations relate to those of AES through the equation $\text{NO}_3^-(\text{UVA-a}) = -1.34 \times \text{NO}_3^-(\text{AES}) + 22.41$ with $R^2 = 0.34$. Finally, the AOML-b NO_3^- concentrations relate to those of AES by the equation $\text{NO}_3^-(\text{AOML-b}) = -0.82 \times \text{NO}_3^-(\text{AES}) + 16.41$ with $R^2 = 0.15$. We conclude that the NO_3^- data collected aboard ship by the UVA-a and AOML-b systems are similar. Those collected by AES are different. The AES collected 12-hour samples, while UVA-a and AOML-b collected 50-min to 1-hour samples. The comparison of 17 July 1988 is an outlier.

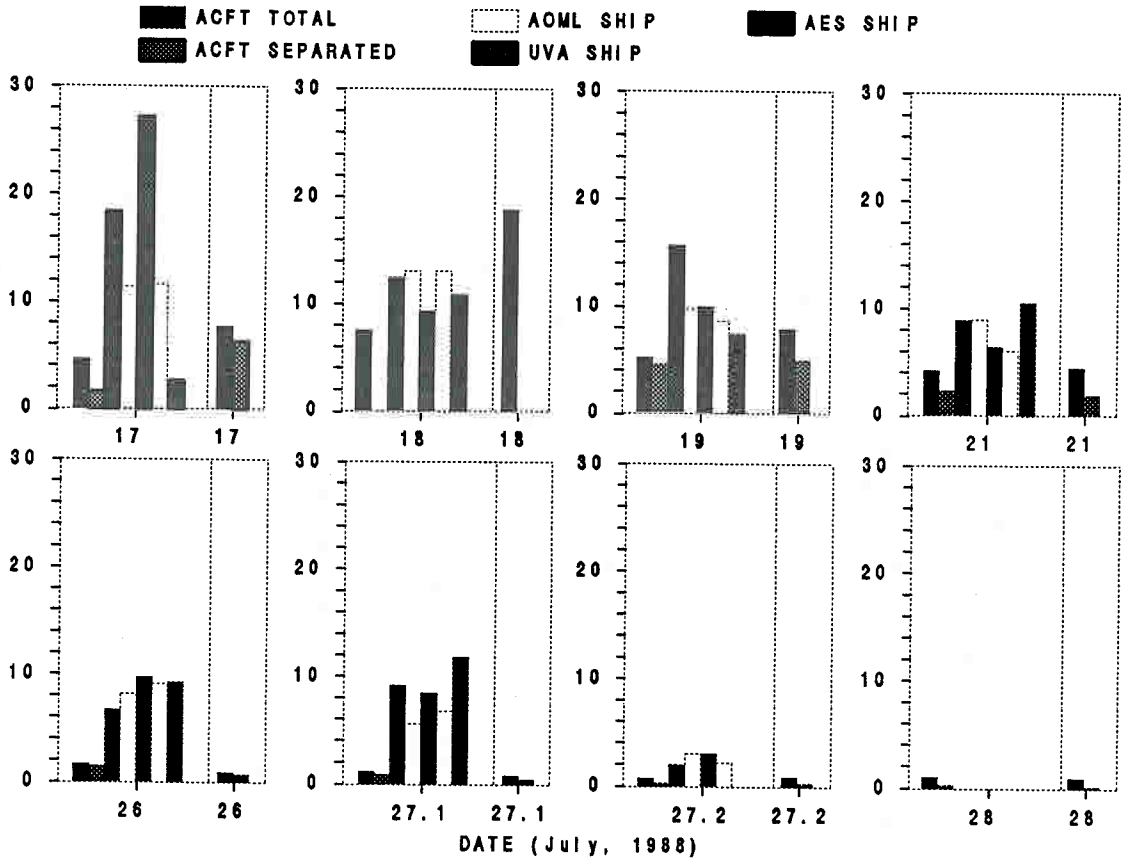


Figure 17.--A comparison between the NO_3^- concentrations measured with filter packs aboard the KA and on the Mt. Mitchell. Those to the right of each dotted line are for filters exposed while the aircraft returned to base.

The NO_3^- concentrations from the KA were significantly lower than those observed by the three systems aboard ship. We agree with Keene et al. (1991) that this is due to the scavenging of HNO_3 by sea-salt aerosol. Because of this gradient of NO_3^- with altitude, we could not further assess the quality of the airborne vs shipboard NO_3^- measurements.

4.9 Ammonium Ion (NH_4^+)

We measured NH_4^+ aboard the KA using both the total and separated filter

systems. We analyzed the quartz filters from each pack for NH_4^+ by colorimetry, using the indophenol blue technique and the procedures described by Bardwell et al. (1991). We also measured NH_4^+ aboard the Mt. Mitchell with three systems. UVA supplied the first system (-a) and we employed the Bardwell et al. analysis technique. AOML supplied the second system (-b) and we utilized an automated colorimetry technique similar to that of Bardwell et al. UVA also supplied a third system (-b) for the collection of selected organic nitrogen species. This system exposed 47 mm diameter 1.0 μm pore size Zeflour filters mounted in polycarbonate cassettes, which we analyzed for NH_4^+ on a Technicon Auto-Analyzer II, using a GPCP technique. Gorzelska and Galloway (1991) and Galloway et al. (1982) gave methodological details.

Figure 18 shows the results of our comparison. The airborne total and separated filter packs showed reasonable agreement. We noted no propensity toward higher NH_4^+ concentrations from the total system. The best-fit linear regression equation for the 12 available data pairs was $\text{NH}_4^+(\text{total}) = -4.14 + 1.18 \times \text{NH}_4^+(\text{separated})$ with $R^2 = 0.97$. This suggests that coarse-mode particles contained about 18% of the total NH_4^+ .

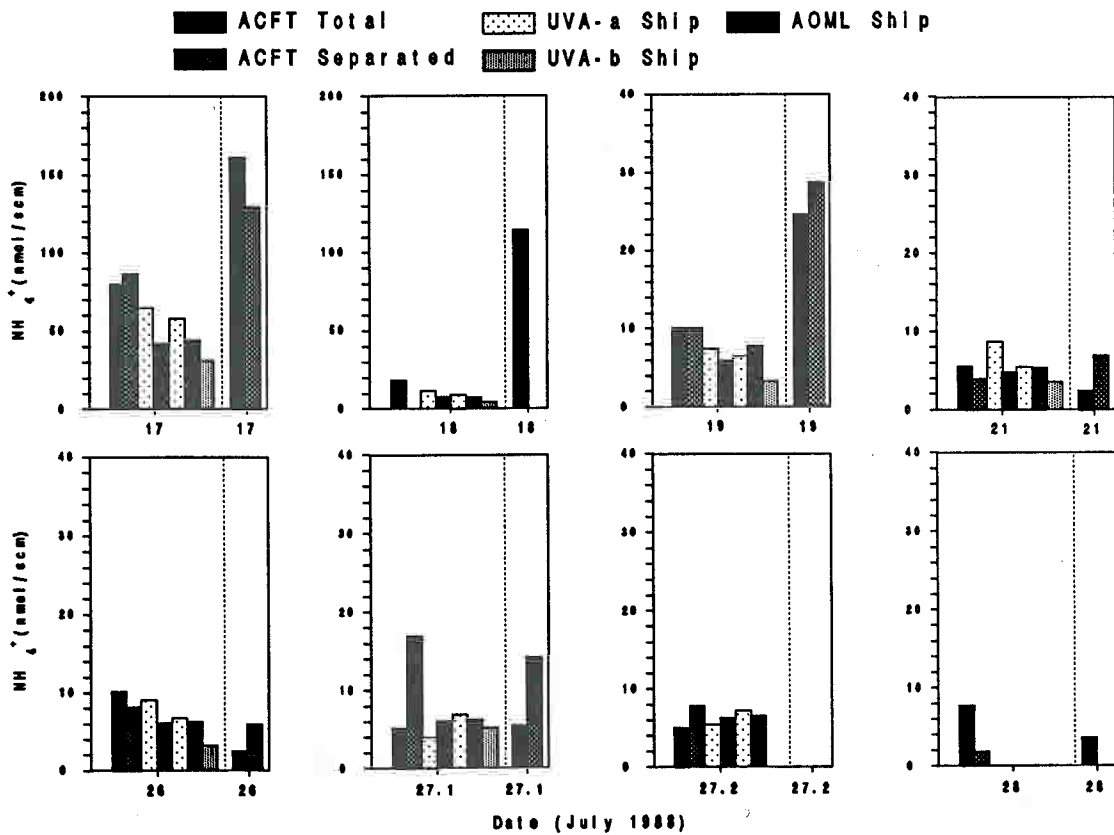


Figure 18.--A comparison between the NH_4^+ concentrations measured with filter packs aboard the KA and on the Mt. Mitchell. Those to the right of each dotted line are for filters exposed while the aircraft returned to base.

Our comparison of the shipboard data suggests that the UVA-a system collected 1.5 - 2 times more NH_4^+ than the AOML-b and UVA-b ones, respectively. The best-fit linear regression equations for the two comparisons are $\text{NH}_4^+(\text{UVA-a}) = -2.25 + 1.48 \times \text{NH}_4^+(\text{AOML-b})$ with $R^2 = 0.98$ and $\text{NH}_4^+(\text{UVA-a}) = 0.02 + 2.07 \times \text{NH}_4^+(\text{UVA-b})$ with $R^2 = 0.98$. We expected lower NH_4^+ concentrations from the UVA-b system, since it collected 24-hour, rather than 1-hour, samples.

We also compared the two aircraft systems with the three aboard ship. The best-fit regression equation that resulted was $\text{NH}_4^+(\text{aircraft}) = 1.02 + 2.32 \times \text{NH}_4^+(\text{ship})$.

5. CONCLUSIONS

We compared selected trace chemical measurements made during the GCE/CASE/WATOX experiment by the NOAA ship Mt. Mitchell and the NOAA King Air research aircraft. The aircraft made 8 research flights in coordination with the ship. Our analyses included only the low altitude portions of each aircraft flight. Tables 2 and 3 summarize the average expected precisions and detection limits, and the average response or exposure times for the sampling systems.

The baseline variables of position, pressure, temperature, dew point, and horizontal wind agreed, within their stated accuracies. We suspect that the dew point measurements aboard ship may, at times, be slightly ($<3^\circ\text{C}$) too high.

Ozone concentrations aboard the two platforms agreed well on and after 21 July 1988. Prior to this day, the ozone concentrations aboard ship were occasionally greater than 100 ppbv, due to condensation in the instrument inlet lines.

The sulfur dioxide concentrations measured aboard the aircraft agreed with one another. The aircraft's continuous analyzer averaged 35 pptv higher in concentration than concurrent filter data. Sulfur dioxide concentrations aboard the aircraft compared favorably to those on the ship, except for the AES filter packs. We suspect an exposure problem with the AES Whatman-41 filters.

The sulfate concentrations measured aboard the aircraft were in good agreement, except on 28 July 1988. On this day more coarse-fraction aerosol particles may have been present. The AES filter system aboard the ship measured less sulfate than the UVA-a and AOML-b ones. The shipboard sulfate concentrations were 10-20% higher than those in the aircraft. We exposed the aircraft filters 150 m above the sea, where coarse-mode aerosol particles are significantly smaller in number than at the sea surface.

The MSA measurements aboard the aircraft showed smaller concentrations from the separated system. This suggests that, at times, significant amounts of MSA may be associated with the coarse marine aerosol. The shipboard MSA measurements agreed within 20%. But, the UVA-a sampler measured consistently more MSA than the AOML-b one. The aircraft MSA concentrations were usually less than those aboard ship. We know that the population of aerosol particles

decreases with altitude in the marine boundary layer (Kim et al., 1991) due to gravitational settling. Therefore, it is reasonable to suggest that MSA concentrations at the sea surface should be higher than those aloft.

Differences in NO_y concentrations between the ship and aircraft were probably due to sampling or calibration variations, except on 17 and 18 July 1988. The shipboard measurements may have been too low on these days.

Shipboard nitric acid vapor measurements agreed within several percent. Aircraft HNO_3 concentrations were significantly higher than those aboard ship. Keene et al. (1991) suggested that this is the result of heterogeneous chemical reactions on sea salt aerosol particles.

The filters exposed behind the aerosol separator aboard the aircraft measured less nitrate than their total counterparts. The shipboard nitrate measurements of UVA-a and AOML-b are similar. Those of AES are different. The AES collected 12-hour samples, while UVA-a and AOML-b collected 50-min to 1-hour samples. The aircraft nitrate concentrations were significantly lower than those aboard ship. This is probably due to the scavenging of HNO_3 by sea-salt aerosol particles.

The airborne NH_4^+ measurements agreed within about 20%. The UVA-a system aboard ship collected 1.5 - 2 times more NH_4^+ than the AOML-b and UVA-b ones, respectively. The aircraft measurements were more than 2 times larger in concentration than those aboard ship.

Because of their difficult logistics, combined ship and aircraft research experiments are rare. The results of Andreae et al. (1988) provide a single example. With this in mind, and considering the obstacles to successful measurement aboard both platforms, we consider our comparison effort a success.

6. ACKNOWLEDGEMENTS

The GCE/CASE/WATOX experiment was the combined effort of many scientists, engineers, and technical personnel. We offer a collective 'job well done' to each of these people. Special thanks go to the flight crew of the NOAA King Air and the staff of the NOAA ship Mt. Mitchell. This field effort would not have occurred without them. We acknowledge Dr's J. Galloway and J. Miller for their expert direction and support of this program.

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