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Interrelationships of Chemical and Physical Information at Quillayute, Washington for 1974

December, 1975

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BNWL-1966
Environmental Control Technology
and Earth Sciences

INTERRELATIONSHIPS OF CHEMICAL AND PHYSICAL INFORMATION AT QUILLAYUTE, WASHINGTON FOR 1974

by

J. D. Ludwick
T. D. Fox
C. W. Thomas
L. L. Wendell

December, 1975

Battelle
Pacific Northwest Laboratories
Richland, Washington

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INTERRELATIONSHIPS OF CHEMICAL AND PHYSICAL INFORMATION AT QUILLAYUTE, WASHINGTON FOR 1974

ABSTRACT

A large number of radioactive and non-radioactive airborne constituents are being measured continually at our coastal air-reference station located in northwestern Washington State. Important correlations have been observed between many of these materials and also with atmospheric parameters. The apparent relationships between changing concentrations and meteorological parameters are being tested using more thorough techniques of investigation, including back trajectory computer analyses.

The permanent air sampling and monitoring station located at the Quillayute Air Base in northwest Washington State is in its twenty-first month of operation and has completed its first full year of data collection. Considerable expansion and improvement of this facility has taken place during the past year. The continuous air monitoring program which was initiated with the condensation nuclei detector and ozone monitor has been expanded to include real-time measurement of NO and NO_x; CO as well as CH_4 and total hydrocarbons. Sampled air is drawn down a 32 meter-high stack utilizing a high-volume pump (2.25 m^3/min). This establishes a linear velocity of 760 m/min across the face of an IPC filter, for high-efficiency particulate collection. Daily particulate samples contain sufficient ^7Be and other radioisotopes to allow their accurate measurement for some of our daily correlation analysis.

A separate high-volume 17 m^3/min air sampling unit is also used for sample collection and measurement of some 30 radionuclides. This system is also used for other special studies.

In order to better coordinate certain pollutant sampling with wind information, a small wind station was mounted at a 22-meter height at the air reference station. This provides supplemental as well as comparative information with the hourly surface values reported by the U. S. Weather Bureau at the Quillayute Airport site.

There has been essentially no land use change in the western fetch to the ocean, some 3 miles away since the establishment of the air-reference station at its present location. In general, no land use change is noticeable within 10 miles of the site in any direction.

A considerable effort was expended this year in improving the method of handling the huge amount of data coming from measurements of the many natural and anthropogenic pollutant materials on the daily and weekly analytical programs. Most of the data is presently punched onto cards which are then available for comparison or individual readout in simplified computer programs. Occasional instrumental failures have resulted in incomplete data.

Figures 1 - 11 represent the composite computer display of data from one of the major portions of the program in calendar year 1974. Interesting correlations and anticorrelations are present in these displays; however, their prominance is not always self evident. Figures 12 and 13 represent a portion of the data replotted for better intercomparison purposes. The correlations between the ozone and radionuclide data are quite evident and indicate upper altitude air sources.

Seasonal fluctuations in the materials whose source region can be traced to the stratosphere are illustrated in Figure 14. Spring intrusions of stratospheric material into the troposphere appear evident on this display.

The concentrations of a wide spectrum of fallout and natural radionuclides were measured in surface air at the Quillayute, Washington reference station during 1974. Aerosol samples were

obtained by passing large volumes of air (17 m^3 per minute) from a 12-m-high stack through IPC filter paper for 4-7 day periods. IPC filters had been tested and found to be essentially absolute for removing the airborne radionuclides at filtering face velocity of greater than 670 linear meters per minute. The radionuclide concentrations measured at Quillayute during 1974 were about 1/3 the concentrations measured at Richland, Washington. This relatively large difference is believed due to the orographic lifting of the Olympic and Cascade mountains with subsequent vertical mixing of higher altitude and surface air prior to sampling at Richland. Since the fallout radionuclide concentration increases with altitude, the vertical mixing caused by movement over the mountains could result in higher radionuclide concentrations at Richland. The concentrations of radionuclides associated with weapons debris observed in surface air in 1973 at Quillayute, Washington were the lowest recorded in the northwestern United States since measurements began in 1961. Previous data has shown that material inserted into the stratosphere during one year will be measured in surface air the following year at the mid-northern latitudes. During 1974 the concentration of weapons associated radionuclides in air at Quillayute, Washington increased 5-20 times over that measured during 1973 as a result of the June 1973 nuclear tests. (see Figure 15). In June 1974 a similar Chinese nuclear test was conducted. Most of the debris was injected into the stratosphere, and the fresh debris in the troposphere from this test was about one-fifth of that measured after the June 1973 nuclear test.

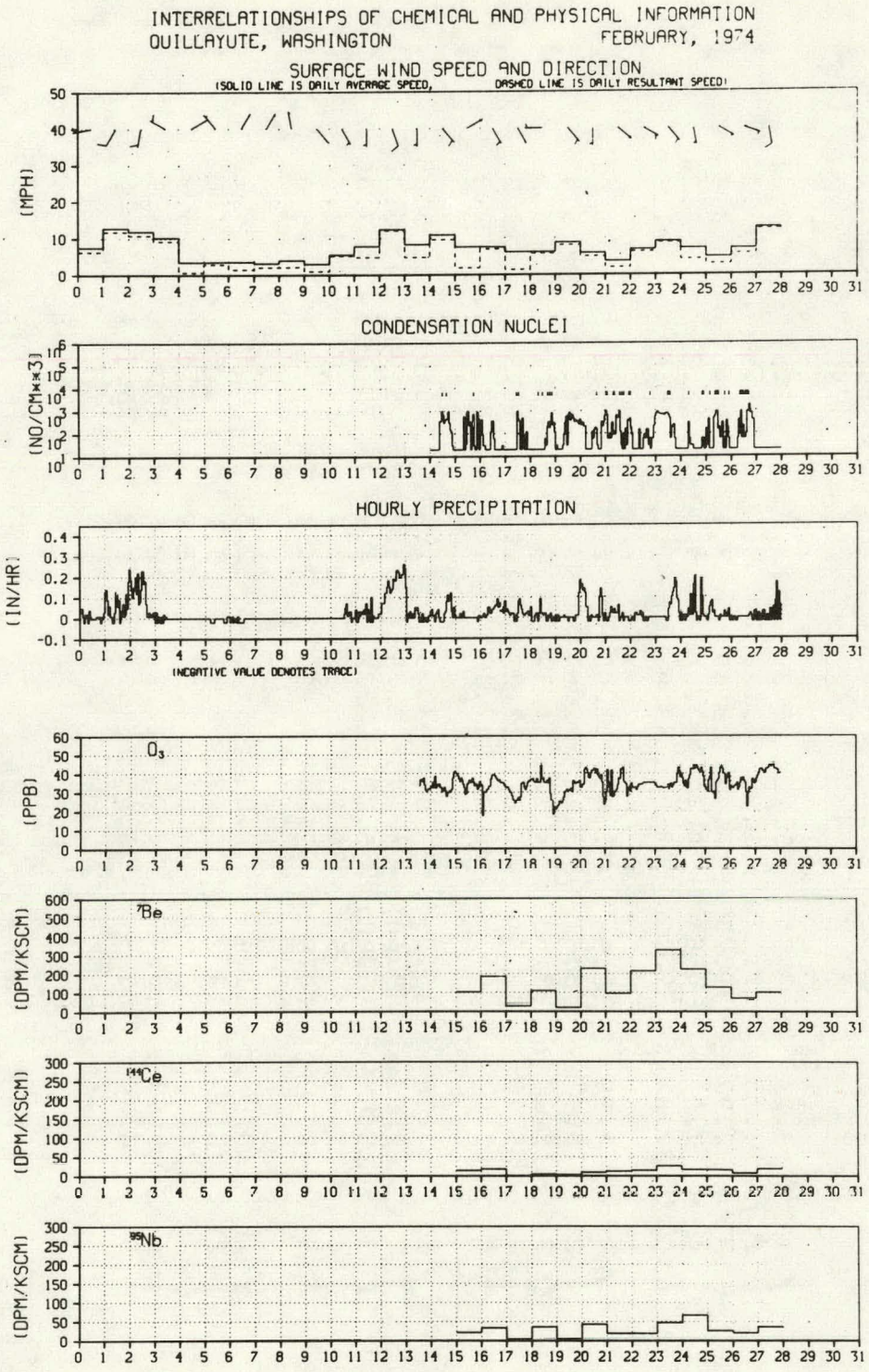
Two high-volume Andersen Cascade impactors with a total air flow of 1.13 m^3/sec are operated at Quillayute and allow our measurements of the radionuclide distribution versus particle size. The impactors separate the atmospheric aerosol into five fractions: greater than 7 microns, 3.3 to 7.0 microns, 2.0 to 3.3 microns, 1.1 to 2.0 microns, and a backup filter of less than 1.1 microns. For the radionuclide particle size studies eight weeks of samples were combined. Results from

measurement of these fractions are shown in Table 1. Measurements made during 1973 show 83 percent of the ^{7}Be activity, 58 percent of the ^{137}Cs activity, and 44 percent of the fresh medium-lived nuclear debris (from the June 1973 Chinese nuclear test) to be associated with particles less than 1.1 micron. During 1974 our measurements showed almost identical results for ^{7}Be but a much smaller size range for ^{137}Cs. About 82 percent of the ^{7}Be activity and 71 percent of the ^{137}Cs activity was associated with particles less than 1.1 micron. This reflects the fact that the ^{137}Cs was almost entirely from stratospheric origin. These data together with concentration data indicate that most of the ^{137}Cs in surface air was from stratospheric nuclear debris inserted in 1973 by the Chinese test. Thus, the stratospheric input completely overwhelmed the small contribution of fresh nuclear debris from the 1974 nuclear test which would be associated with relatively large tropospheric aerosol particles.

In general, the wealth of data accumulated by the continuous operation of the Quillayute air reference station will provide unique opportunities for study of the correlations of large scale meteorological processes with natural and anthropogenic tracer pollutants.

Table 1. Distribution of Atmospheric Aerosols at Quillayute, Washington.

		SEPT OCT 73	NOV DEC 73	JAN FEB 74	MAR APR 74	MAY JUN 74	JULY AUG 74	$\bar{X}$
^{7}Be	1	3.84	8.31	1.21	3.39	3.03	1.87	3.61
	2	3.13	2.17	2.08	2.28	3.25	5.94	3.14
	3	2.97	2.20	3.44	3.46	3.19	5.58	3.47
	4	7.51	4.54	9.40	7.66	9.33	11.3	8.29
	5	82.6	82.8	83.9	83.0	81.3	75.6	81.5
^{95}Nb	1	7.99	11.9	1.45	3.65	2.31	2.83	5.02
	2	10.7	5.61	3.14	2.64	3.78	6.50	5.40
	3	12.1	3.63	4.22	4.34	3.80	5.19	5.55
	4	8.28	12.5	12.8	12.2	12.7	11.9	11.7
	5	61.0	66.3	78.4	77.1	77.5	73.4	72.3
^{137}Cs	1	7.10	15.2	2.94	3.03	2.97	3.88	5.85
	2	11.8	9.60	4.14	2.79	3.90	5.70	6.32
	3	15.6	6.78	5.71	4.21	4.80	5.25	7.06
	4	17.2	7.91	20.2	15.0	13.4	14.0	14.6
	5	48.3	60.5	67.0	74.9	74.4	71.1	66.0
^{144}Ce	1	10.0	11.4	2.01	3.05	2.30	2.37	5.18
	2	10.8	13.2	5.66	2.31	3.14	4.58	6.62
	3	14.3	5.97	4.02	4.54	3.94	5.42	6.37
	4	9.93	6.96	12.7	12.2	12.6	12.5	11.1
	5	54.9	62.5	75.6	77.9	78.2	75.3	70.7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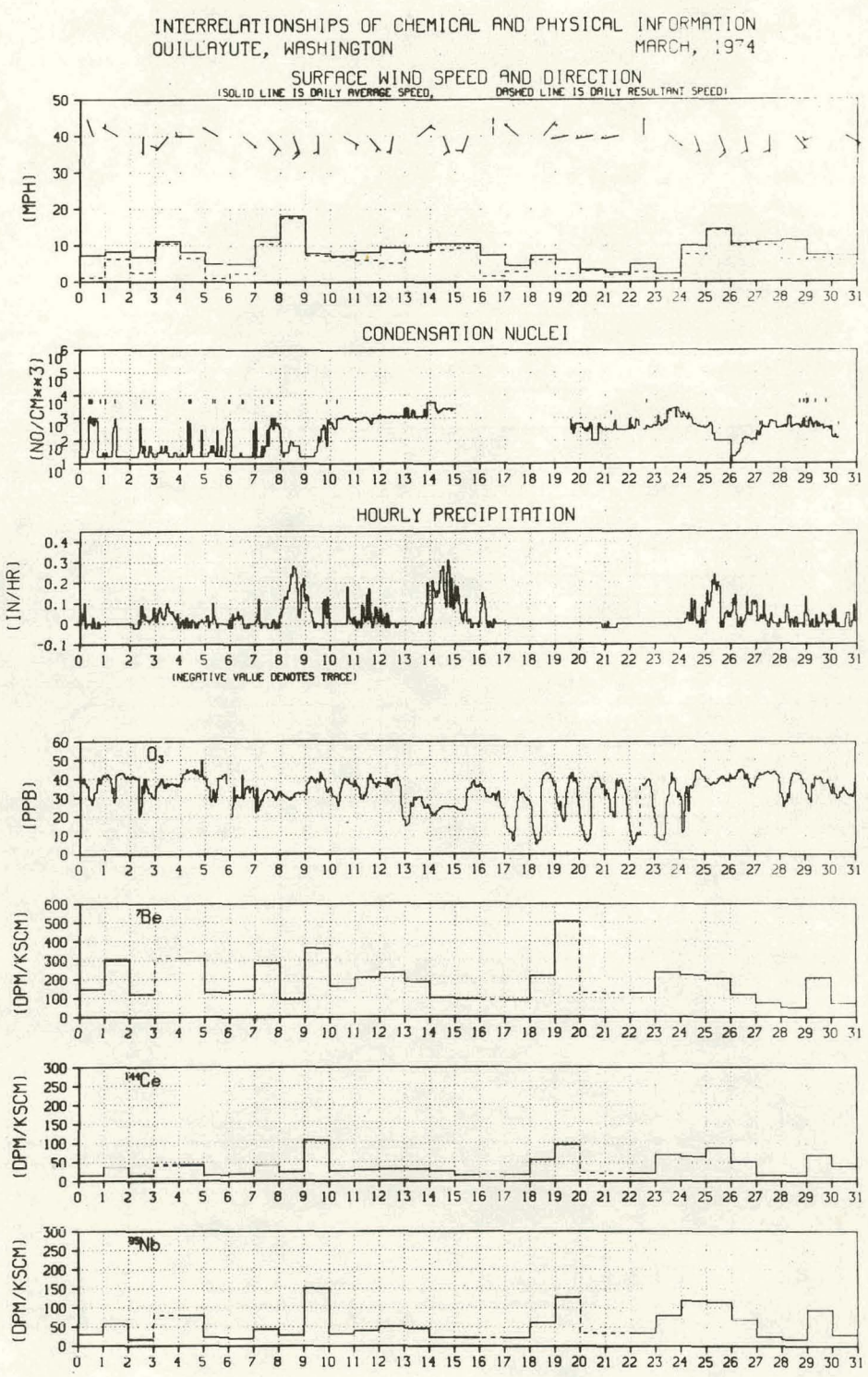
QUILLAYUTE, WASHINGTON
FEBRUARY, 1974
SURFACE WIND SPEED AND DIRECTION
(SOLID LINE IS DAILY AVERAGE SPEED, DASHED LINE IS DAILY RESULTANT SPEED)
(MPH)
CONDENSATION NUCLEI
(NO/CM**3)
HOURLY PRECIPITATION
(IN/HR)
(NEGATIVE VALUE DENOTES TRACE)
(PPB)
O_3
(DPM/KSCM)
^{7}Be
^{144}Ce
^{95}Nb

Figure 1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QUILLAYUTE, WASHINGTON
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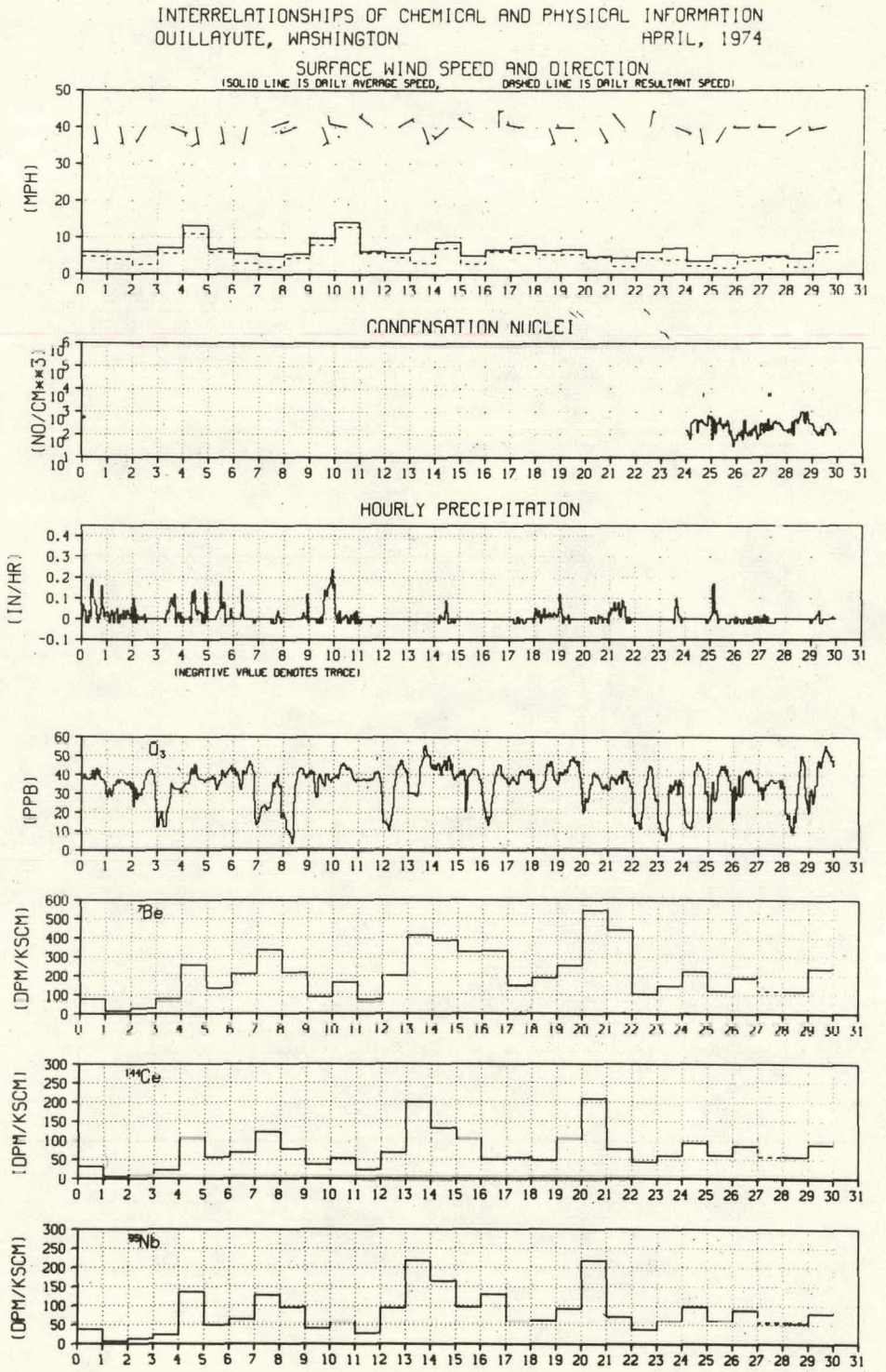
HOURLY PRECIPITATION
(IN/HR)
(NEGATIVE VALUE DENOTES TRACE)
(PPB)
O3
(DPM/KSCM)
7Be
144Ce
95Nb

Figure 2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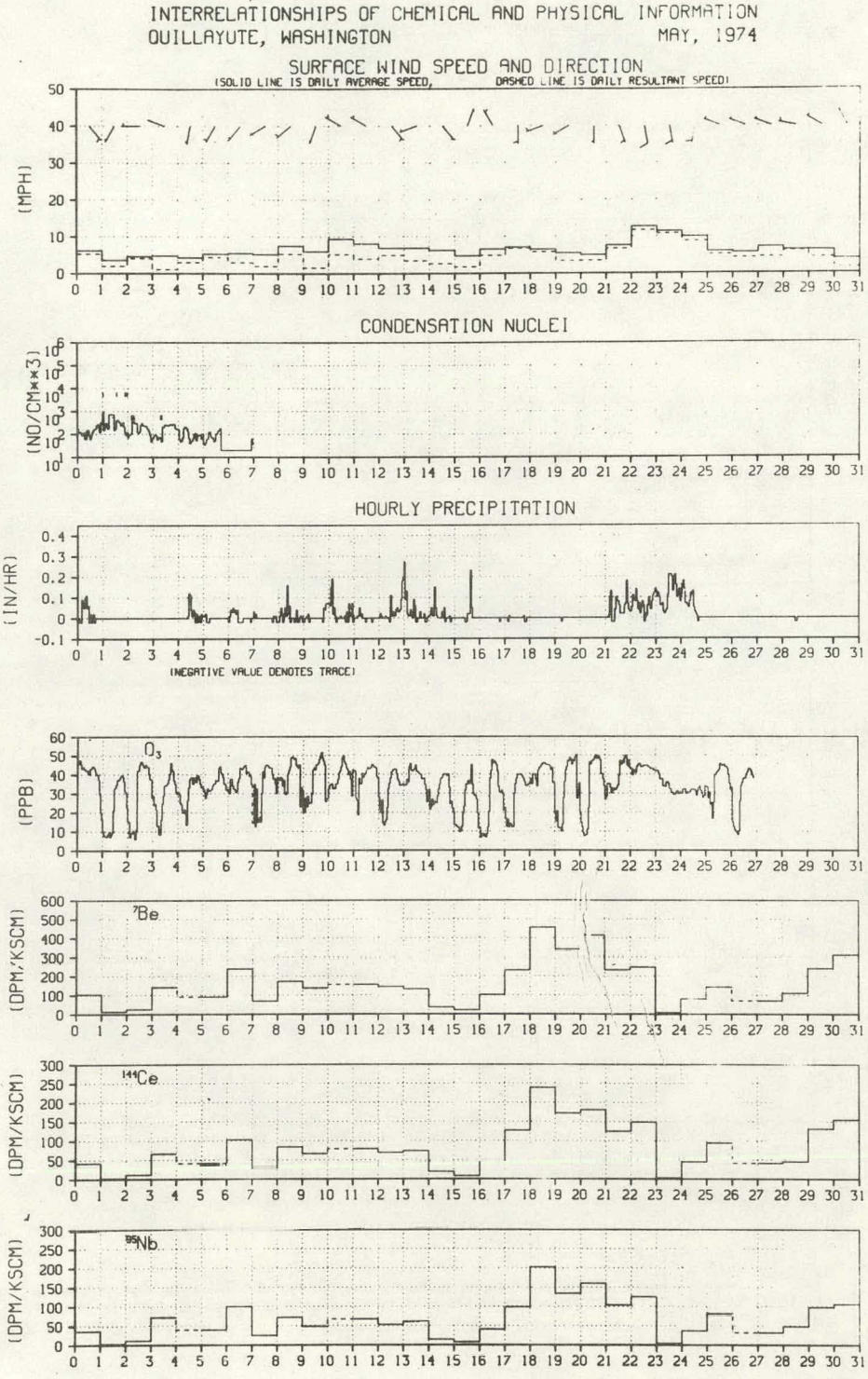
HOURLY PRECIPITATION
(IN/HR)
(NEGATIVE VALUE DENOTES TRACE)
(PPB)
O3
(DPM/KSCM)
7Be
144Ce
95Nb

Figure 3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MAY, 1974
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CONDENSATION NUCLEI
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HOURLY PRECIPITATION
(IN/HR)
(NEGATIVE VALUE DENOTES TRACE)
(PPB)
O_3
(DPM/KSCM)
^{7}Be
^{144}Ce
^{95}Nb
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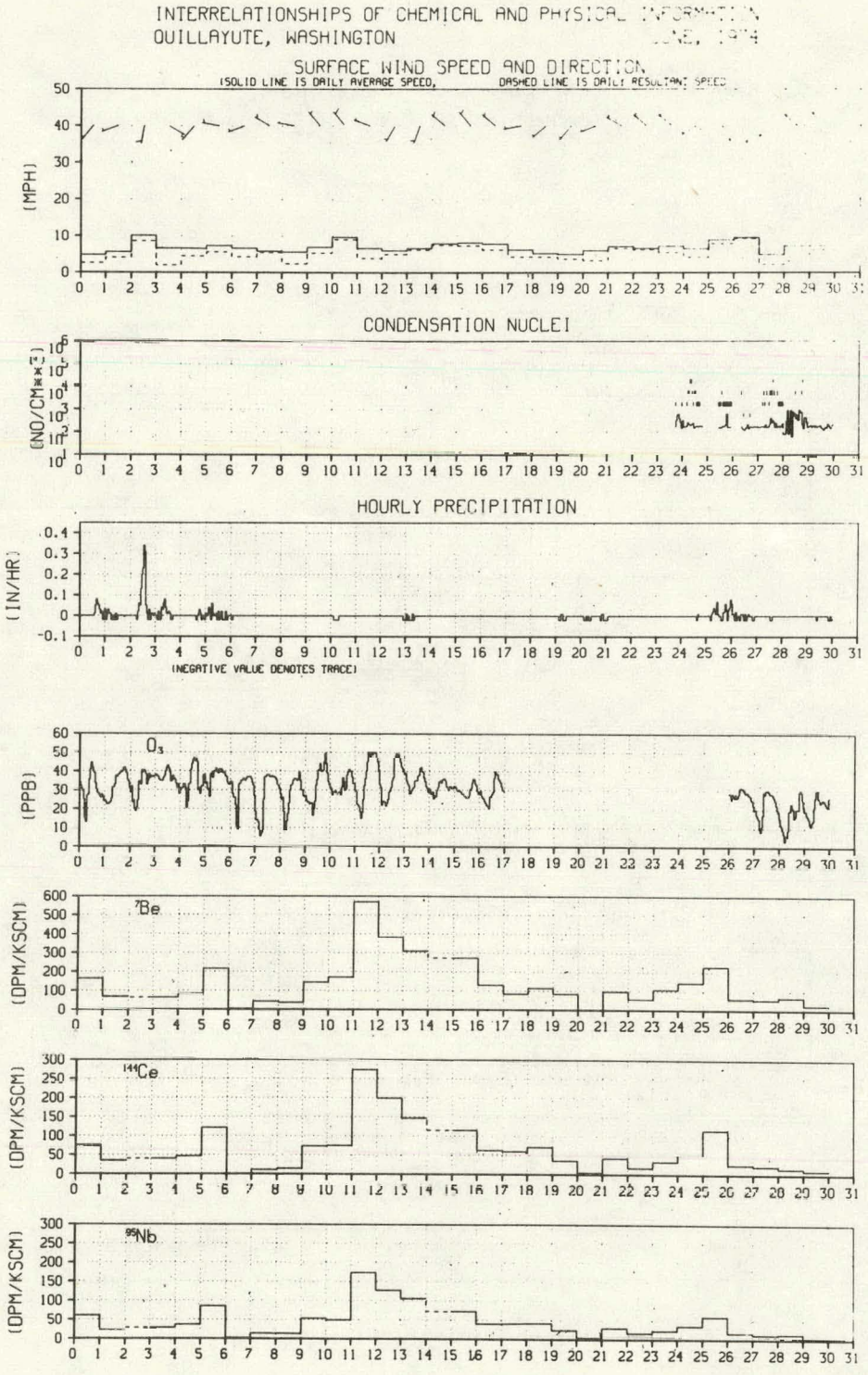
Figure 4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CONDENSATION NUCLEI
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(IN/HR)
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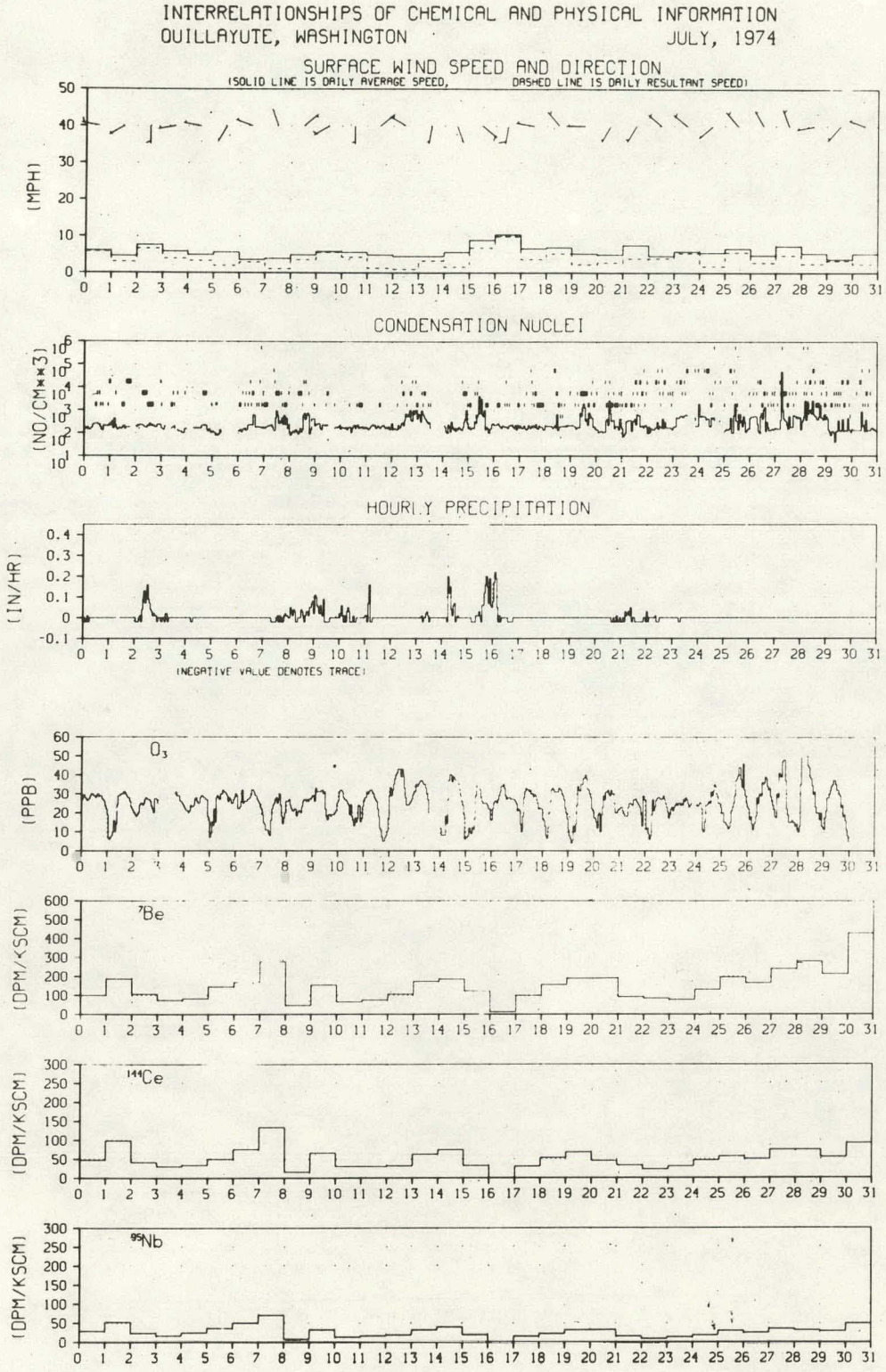
(PPB)
O3
(DPM/KSCM)
7Be
144Ce
95Nb

Figure 5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CONDENSATION NUCLEI
(NO/CM**3)
HOURLY PRECIPITATION
(IN/HR)
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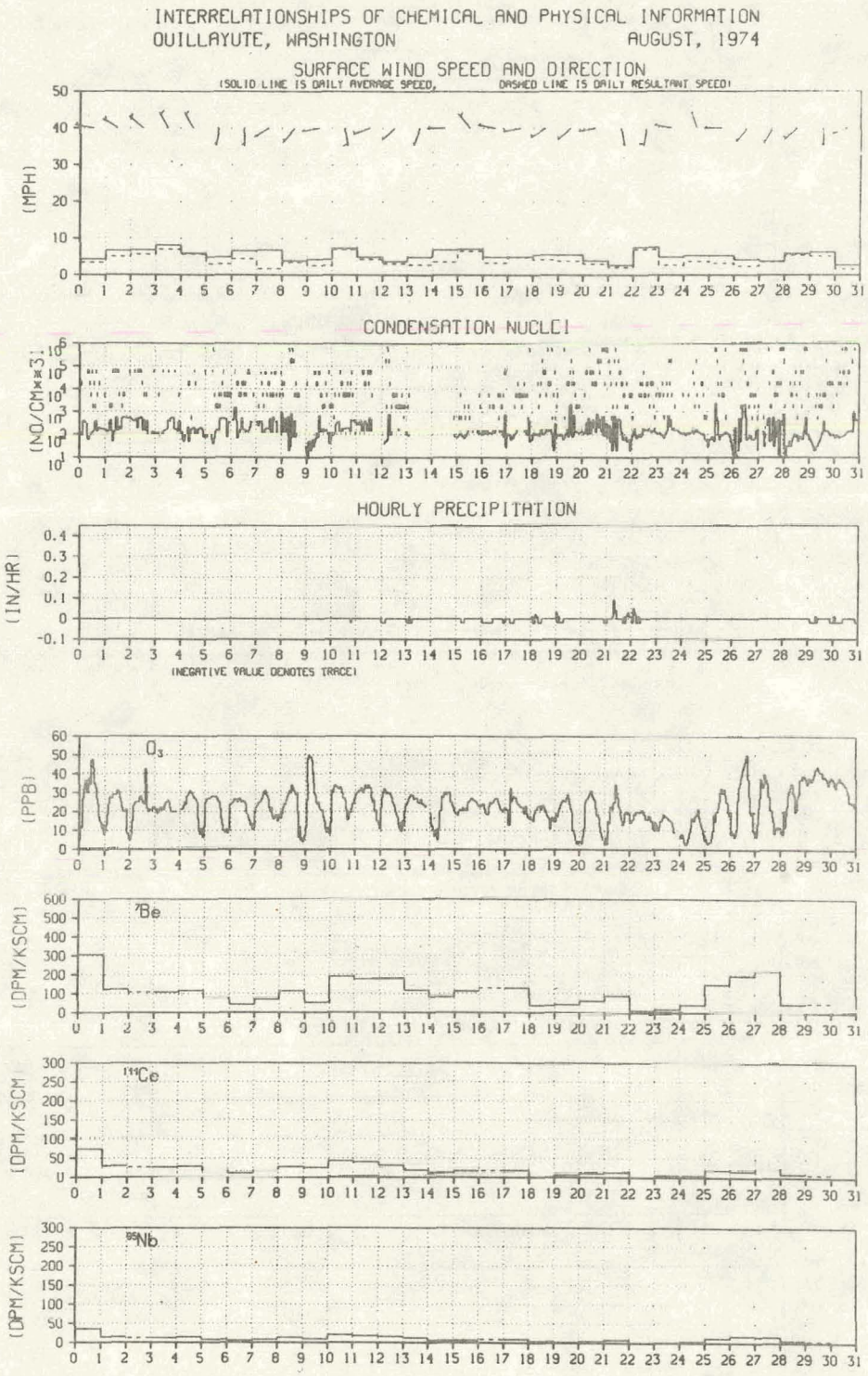
O3
(PPB)
7Be
(DPM/KSCM)
144Ce
(DPM/KSCM)
95Nb
(DPM/KSCM)

Figure 6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(SOLID LINE IS DAILY AVERAGE SPEED, DASHED LINE IS DAILY RESULTANT SPEED)
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HOURLY PRECIPITATION
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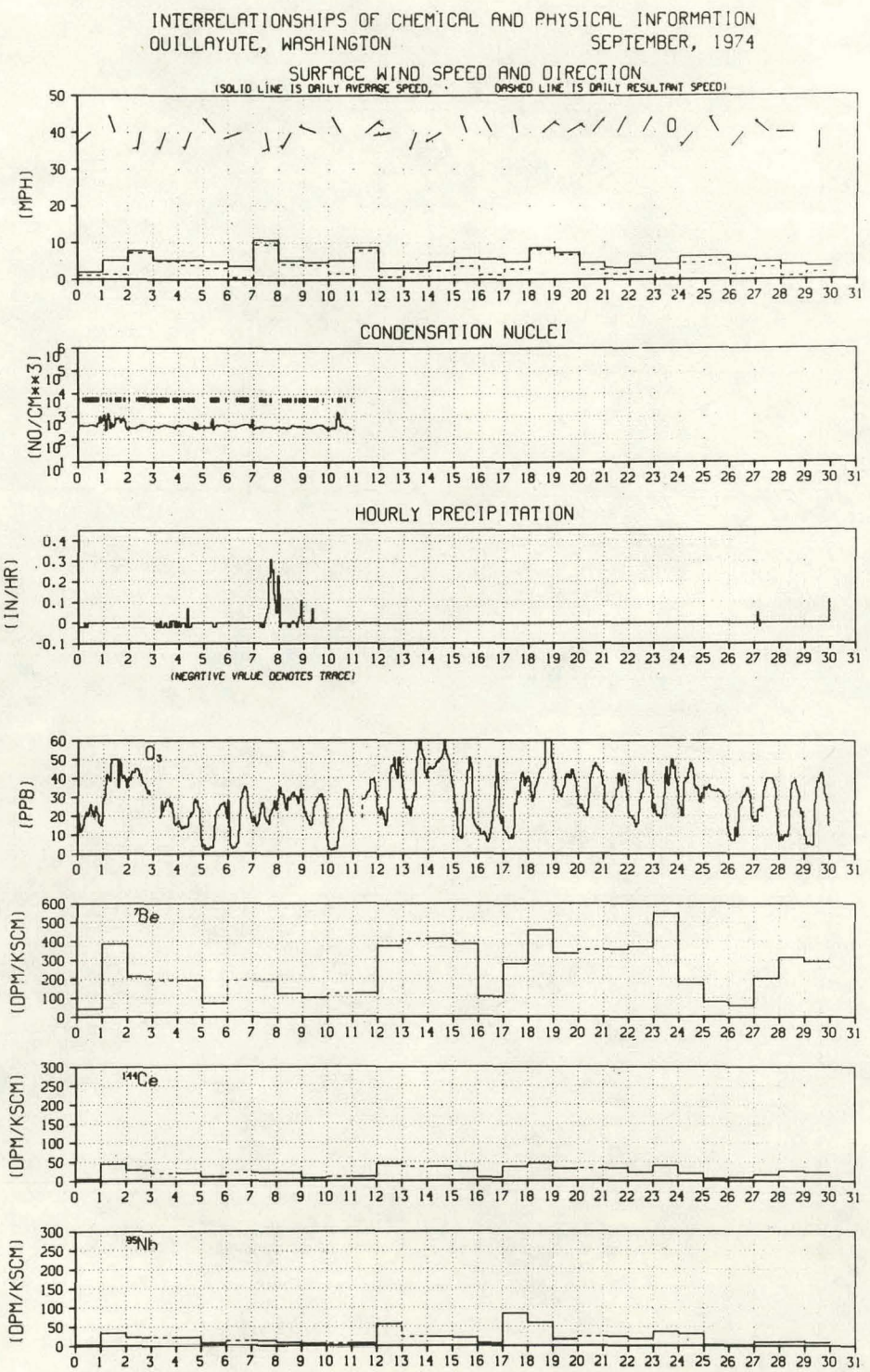
(PPB)
O_3
(DPM/KSCM)
^{7}Be
^{144}Ce
^{95}Nb

Figure 7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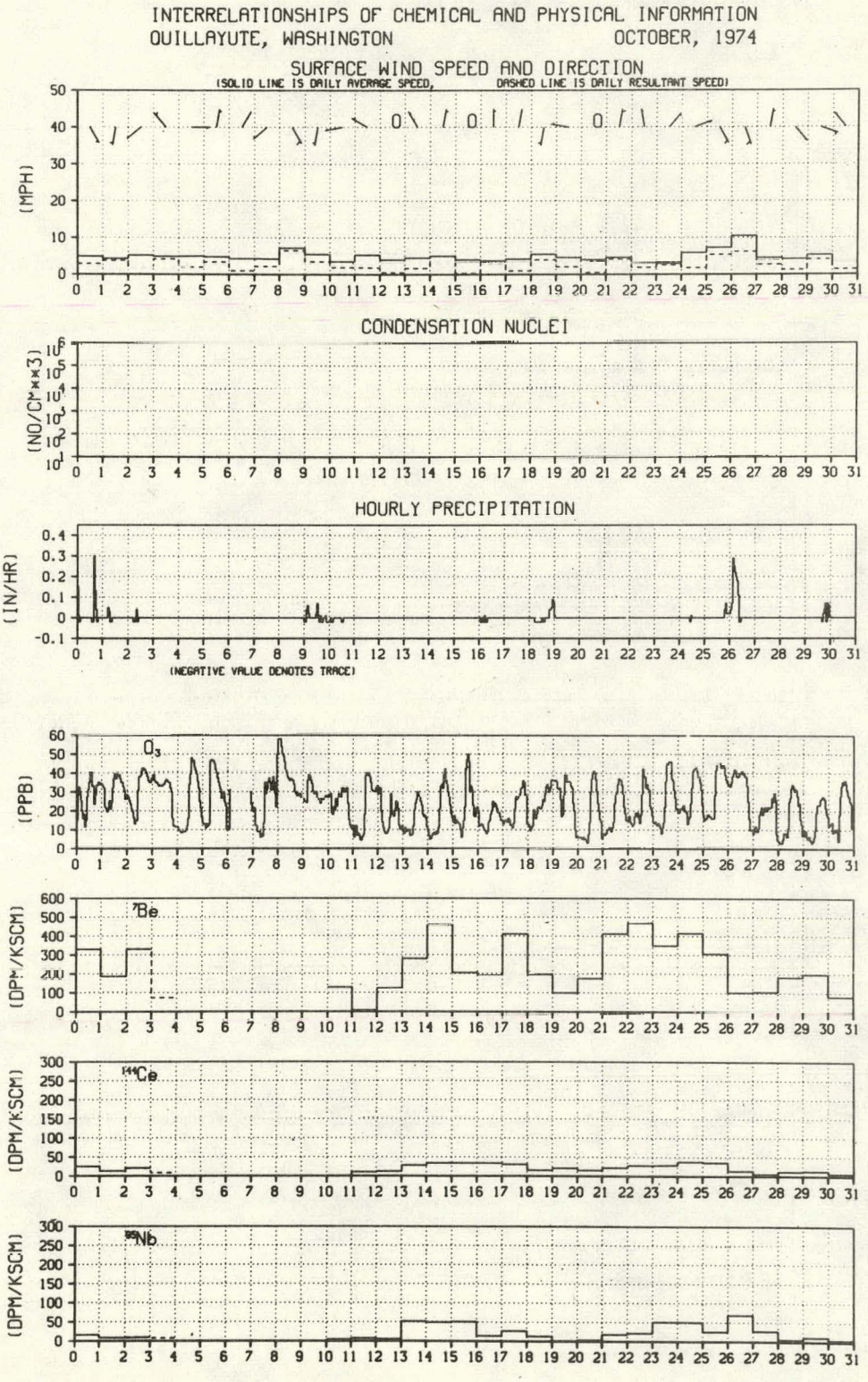
(MPH)
CONDENSATION NUCLEI
(NO/CM**3)
HOURLY PRECIPITATION
(IN/HR)
(NEGATIVE VALUE DENOTES TRACE)
(PPB)
O_3
(DPM/KSCM)
^{7}Be
^{144}Ce
^{95}Nb

Figure 8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OCTOBER, 1974
SURFACE WIND SPEED AND DIRECTION
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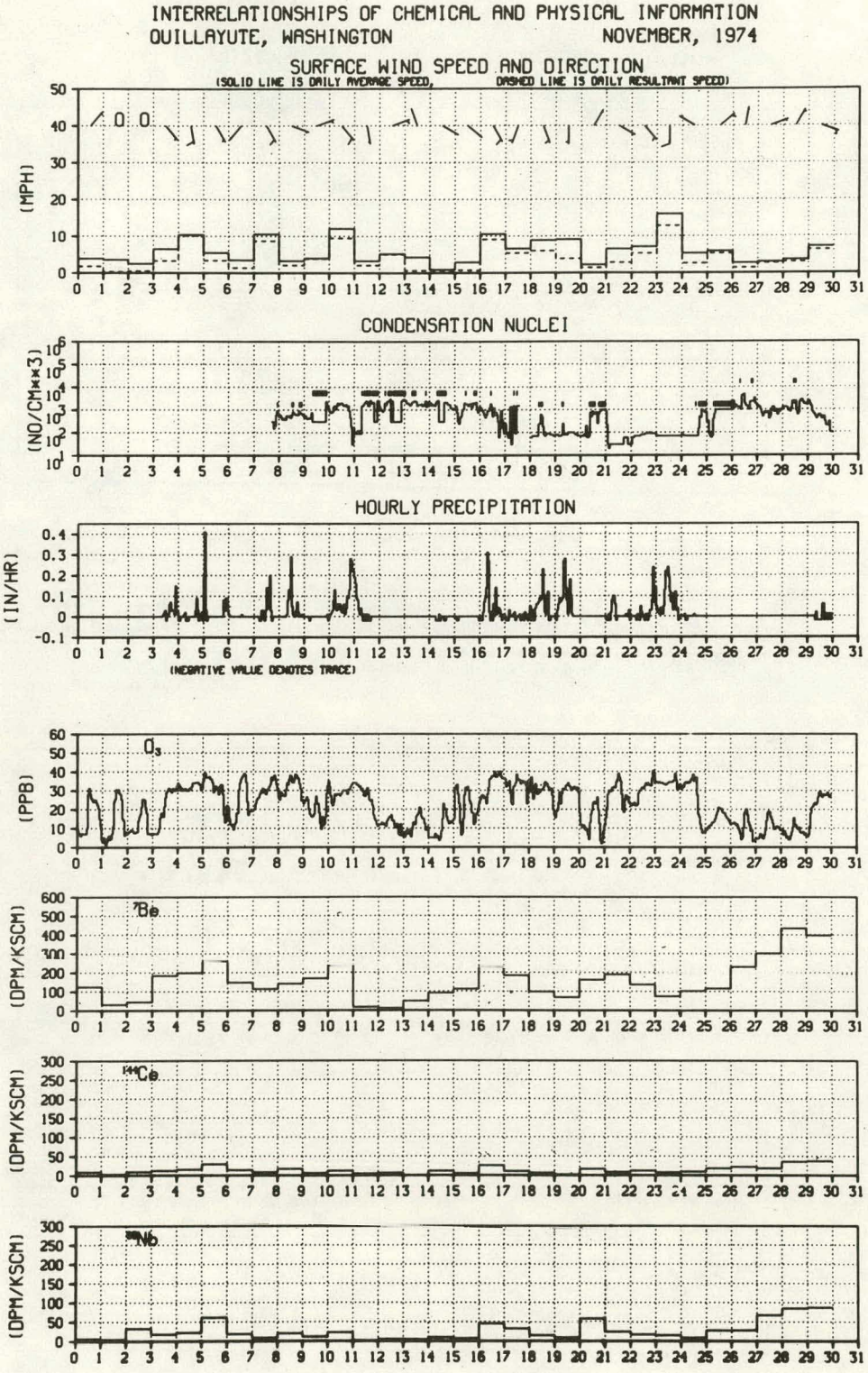
(MPH)
CONDENSATION NUCLEI
(NO/CM**3)
HOURLY PRECIPITATION
(IN/HR)
(NEGATIVE VALUE DENOTES TRACE)
(PPB)
O3
(DPM/KSCM)
7Be
144Ce
95Nb

Figure 9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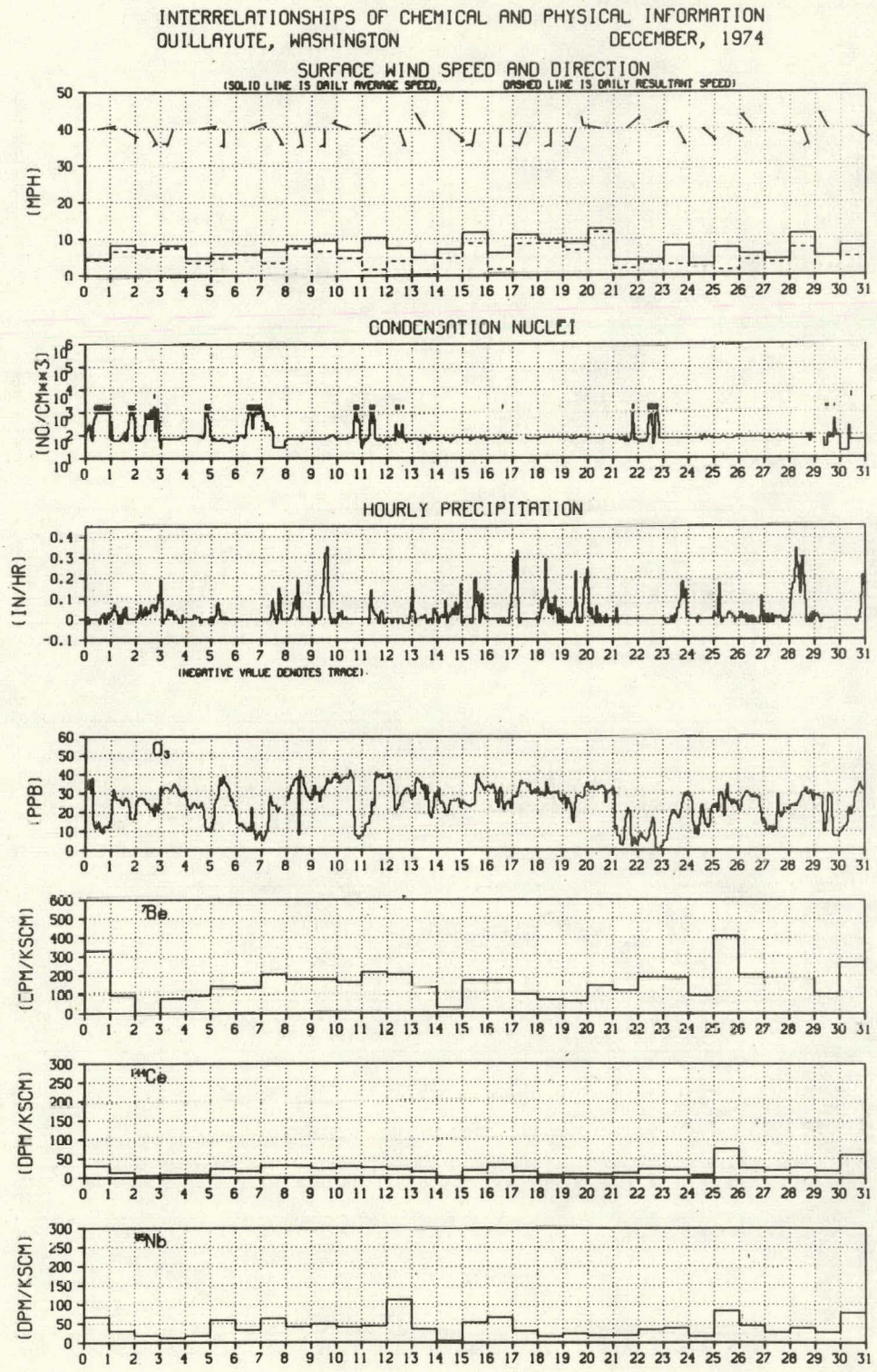
QUILLAYUTE, WASHINGTON NOVEMBER, 1974
SURFACE WIND SPEED AND DIRECTION
(SOLID LINE IS DAILY AVERAGE SPEED, DASHED LINE IS DAILY RESULTANT SPEED)
(MPH)
CONDENSATION NUCLEI
(NO/CM**3)
HOURLY PRECIPITATION
(IN/HR)
(NEGATIVE VALUE DENOTES TRACE)
(PPB)
(DPM/KSCM)
(DPM/KSCM)
(DPM/KSCM)

Figure 10


INTERRELATIONSHIPS OF CHEMICAL AND PHYSICAL INFORMATION
QUILLAYUTE, WASHINGTON
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(MPH)
CONDENSATION NUCLEI
(NO/CM**3)
HOURLY PRECIPITATION
(IN/HR)
(NEGATIVE VALUE DENOTES TRACE)
(PPB)
O3
(CPM/KSCM)
7Be
(DPM/KSCM)
144Ce
(DPM/KSCM)
95Nb

Figure 11

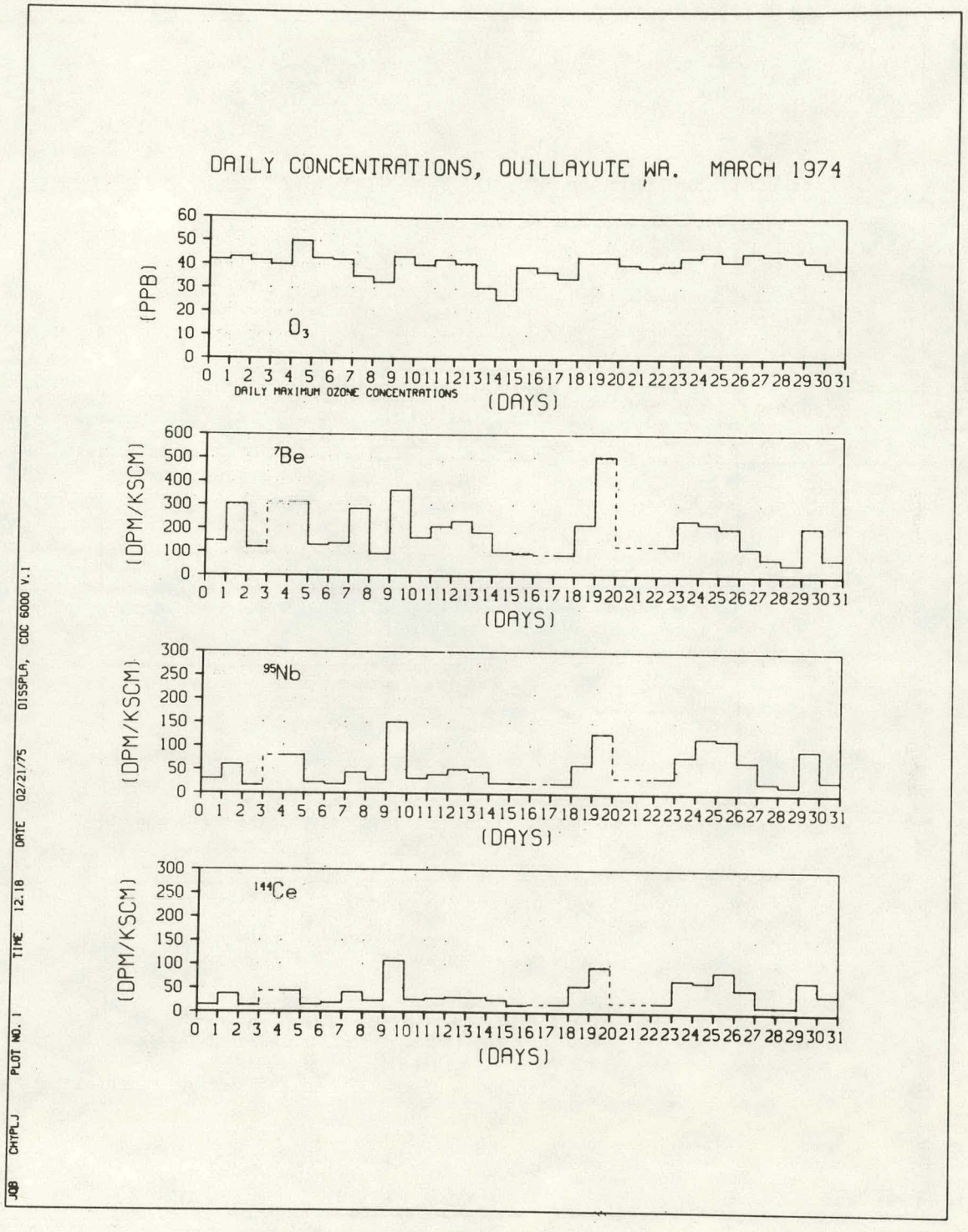
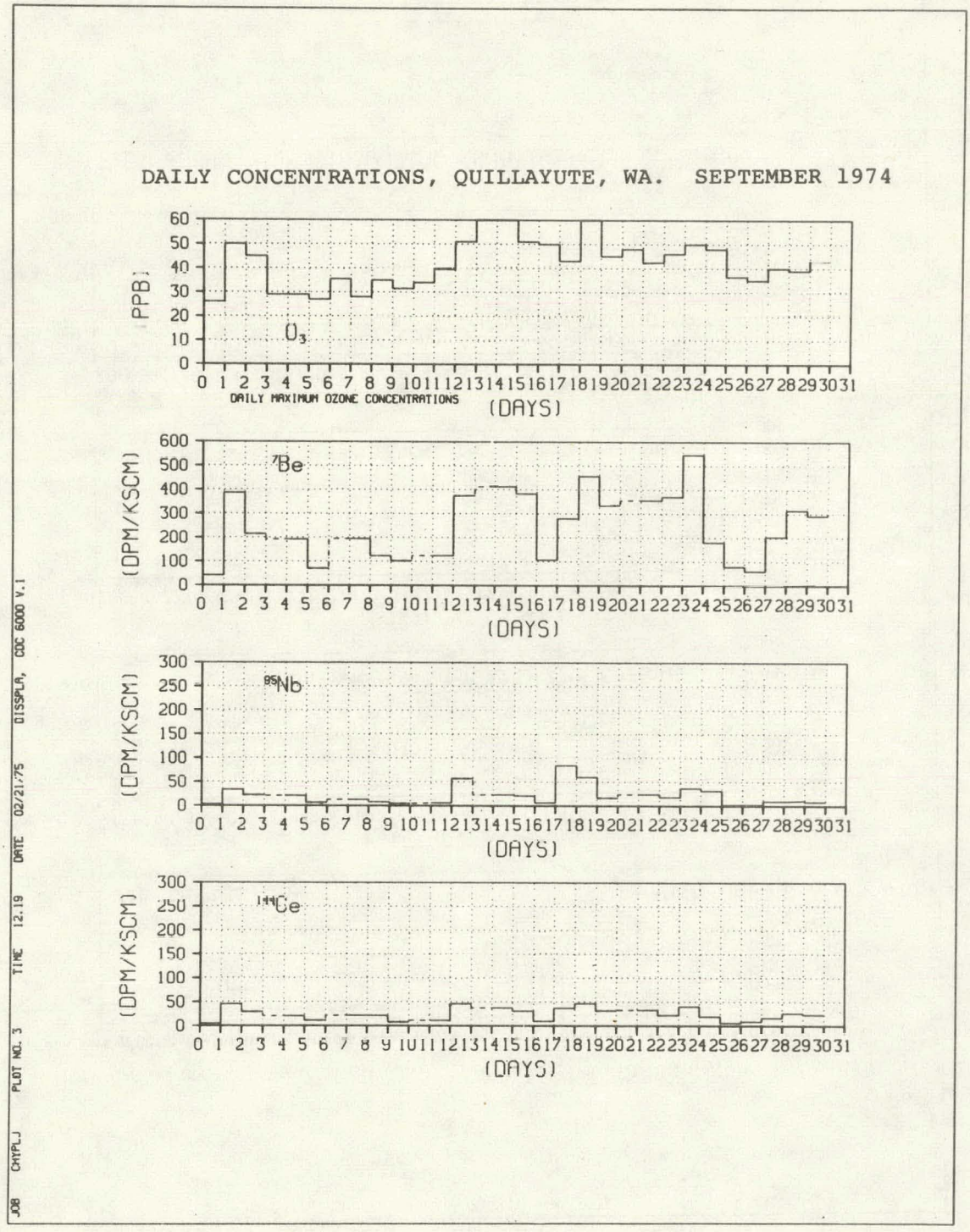

DAILY CONCENTRATIONS, QUILLAYUTE WA. MARCH 1974
(PPB)
O_3
DAILY MAXIMUM OZONE CONCENTRATIONS
(DAYS)
(DPM/KSCM)
^{7}Be
(DAYS)
(DPM/KSCM)
^{95}Nb
(DAYS)
(DPM/KSCM)
^{144}Ce
(DAYS)
JOB CHTPLJ PLOT NO. 1 TIME 12.18 DATE 02/21/75 DISSPLA, CDC 6000 V.1

Figure 12


DAILY CONCENTRATIONS, QUILLAYUTE, WA. SEPTEMBER 1974
(PPB)
O3
DAILY MAXIMUM OZONE CONCENTRATIONS
(DAYS)
(DPM/KSCM)
7Be
(DAYS)
(CPM/KSCM)
95Nb
(DAYS)
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(DAYS)
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Figure 13

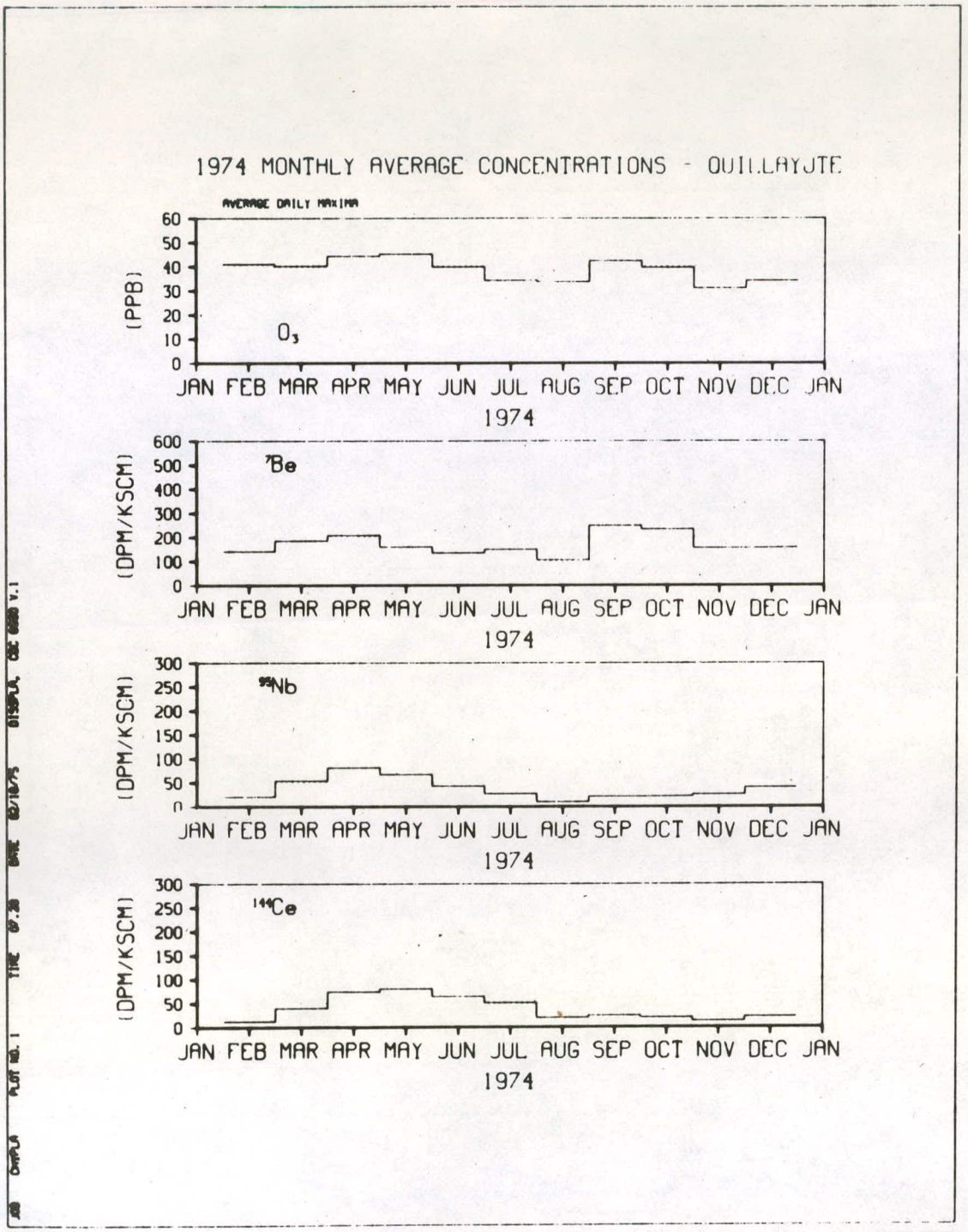
1974 MONTHLY AVERAGE CONCENTRATIONS - QUILLAYUTE
AVERAGE DAILY MAXIMA
(PPB)
O_3
(DPM/KSCM)
^{7}Be
^{95}Nb
^{144}Ce
JAN FEB MAR APR MAY JUN JUL AUG SEP OCT NOV DEC JAN
1974

Figure 14

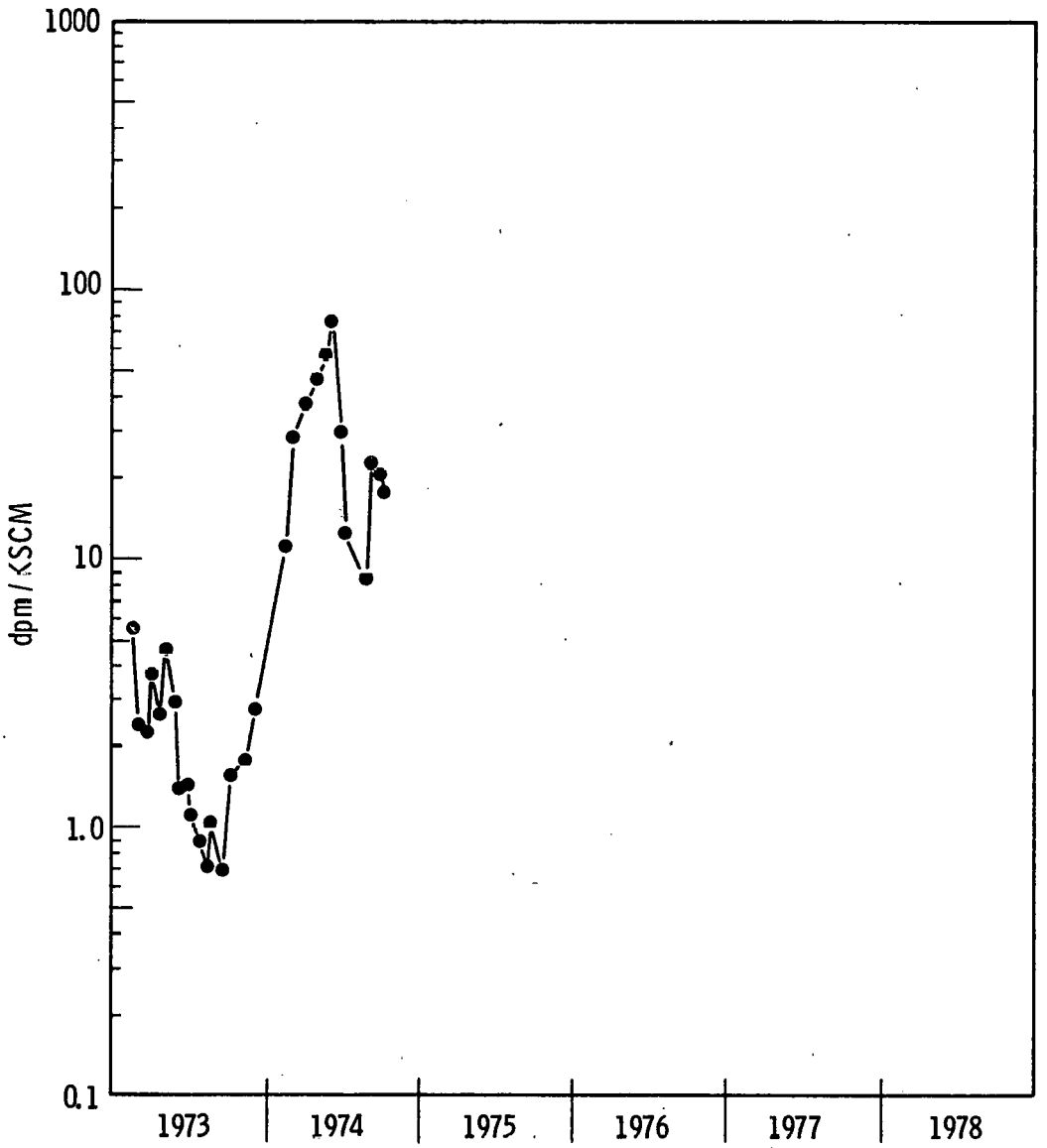

Figure 15. The Concentration of ^{144}Ce in Surface Air at Quillayute, Washington.

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