Winter 1998

Physical and chemical climate in the Nepal Himalaya

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PHYSICAL AND CHEMICAL CLIMATE IN THE NEPAL HIMALAYA

BY

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DISSERTATION

Submitted to the University of New Hampshire
in Partial Fulfillment of the Requirements
for the Degree of

Doctor of Philosophy
in Earth Sciences

December, 1998

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DEDICATION

To my wife, daughter, and parents
for their support, encouragement
and patience

ACKNOWLEDGMENTS

I would, first and foremost, like to extend my profound thanks to my committee members for their guidance, suggestions, enthusiasm, questions and general assistance offered during the many phases of this investigation: Dr. Cameron Wake (Dissertation Director), Dr. Jack Dibb, Dr. Paul Mayewski, Dr. Robert Talbot and Dr. Barry Keim.

I am grateful to the Climate Change Research Center and the Earth Sciences Department at the University of New Hampshire for all the assistance provided. I am thankful to Sallie Whitlow for analyzing numerous samples. I am grateful to the Department of Hydrology and Meteorology, Nepal for their support to my field study between September, 1996 and November, 1997 and for providing meteorological data. I am indebted to David Averill for his support in the 1994 field season and during my entire stay in New Hampshire. I am very thankful to my field observers Dorzee Sherpa, Phortse and Karna Jirel, Jiri for their untiring work in collecting the data.

This research was conducted while I was funded as a research assistant through several grants awarded to Dr. Paul Mayewski, Dr. Cameron Wake and Dr. Jack Dibb. The latter part of the study was supported by START fellowship. The Department of Earth Sciences, University of New Hampshire provided partial support to cover travel expenses.

TABLE OF CONTENTS

APPENDICES

LIST OF TABLES

LIST OF FIGURES

ABSTRACT

PHYSICAL AND CHEMICAL CLIMATE IN THE NEPAL HIMALAYA

by

Arun Bhakta Shrestha

University of New Hampshire, December, 1998

The physical climate of the Himalaya is governed by monsoon circulation, a primary source of moisture to the region. The moisture input during summer monsoon circulation amounts to more than 70% of total moisture input to the region. Monsoon precipitation forms direct input to the flow of several important river systems of the Himalaya region and the Indo-Gangetic plain, while it also feeds the vast glaciers and snow fields in the Himalayas, thus maintaining the river flow during dry seasons. The agro-based economy of the Himalaya and the Indo-Gangetic plain can be adversely effected by long-term fluctuations in the life sustaining rains of the summer monsoon. Model and observational studies show that the south Asian monsoon can be highly influenced by physical factors within and beyond the region (e.g., extent of snow cover in Eurasia and in the Himalaya, El Niño Southern Oscillation, location of monsoon trough, stratospheric circulation, etc.) as well as by the chemical climate (e.g., greenhouse gases, sulfate aerosol, etc.). To date, little research has focused on the physical and chemical climate in the Himalaya, while not much is known about the chemical climate there. This dissertation is a first attempt to investigate both the physical and chemical aspects of climate in the

Nepal Himalaya with the objective of documenting current conditions and past trends as well as laying the foundation for investigations of their inter-relationship in the future.

Temperature and precipitation records from a network of meteorological station in Nepal were analyzed over the period 1971-1994. Temperature records show significant warming trends after the mid-1970's. The temperature trend in Nepal is in broad agreement with global temperature trends, while it differs from temperature trends in the neighboring Indian plains. Within Nepal, the warming trend is greater in the high elevation regions , suggesting that the mountains are more sensitive to climate change. Precipitation records from Nepal show significant variability at annual and decadal time scales, whereas long-term trends are lacking. The precipitation record from Nepal shows significant correlation with several large scale climatological parameters such as Southern Oscillation Index, sunspot numbers, temperature over Tibetan Plateau, and temperature over the Indian Ocean and southern India, while it differs considerably from the all-India precipitation record. Among others factors, El Niño events have a dominant influence on the precipitation variations in the Himalaya.

Aerosol and precipitation chemistry of the eastern Himalaya are characterized by marked seasonal fluctuations. The concentrations of water soluble major ions are low and comparable to remote tropospheric concentrations during late monsoon and post-monsoon, while concentrations during pre-monsoon and early monsoon are considerably higher, comparable to concentrations at urban sites in Europe and North America. The seasonal changes in concentrations are clearly related to changes in atmospheric circulation. Regional-scale thermally-driven valley wind systems are responsible for transporting pollutants to the Himalaya during the pre-monsoon season, whereas monsoon circulation transports pollutants during the early period of the summer monsoon. Onset of the active monsoon (mid-June) coincides with a dramatic drop in major ion concentrations and the low concentrations are maintained over the post-monsoon.

The variations in the aerosol concentrations at time scales greater than a week are well reflected in precipitation chemistry, whereas small scale variations are not. Since glaciochemical records, are generally interpreted on a seasonal or longer temporal resolution, the relationship between aerosol and precipitation chemistry supports the potentials of using glaciochemical data to document change in chemical composition of the Himalayan atmosphere in the past. Multivariate (EOF) analysis on the aerosol data reveals that the two transport mechanisms, thermal valley wind system and monsoon circulation, although physically different, result in similar transport pathways and source regions impacting the interior of the Himalaya.

CHAPTER ONE

INTRODUCTION

I. INTRODUCTION

Earth's climate is dependent upon the radiative balance of the atmosphere, which in turn is controlled by the input of solar radiation and the atmospheric abundance of radiatively active trace gases (i.e., greenhouse gases), clouds, and aerosols. Changes in any part of the system effects all the other components and results in climate change. Initiatives within international scientific communities over recent decades has resulted in an improved understanding of eaarths complex climate system with a focus on the effect of anthropogenic perturbations to the natural state (e.g., IPCC, 1990). Several projections and prognoses regarding future climate change have evolved over the decades. Despite great strides that have been made towards understanding Earth's climate, there are several uncertainties which limit the confidence of prediction on regional scales. These uncertainties are largely due to the lack of instrumental meteorological records and other environmental observations in those regions. Combined with a relatively poor understanding of natural and anthropogenic controls concerning the climate system on regional scales, the Himalaya and its surroundings represent such a region.

The Himalayas and the Tibetan Plateau play a central role in the regional scale atmospheric circulation pattern in Asia. Intense solar radiation in the highlands creates high temperatures and reinforces the low pressure center over northern India (Flohn, 1968), which drives summer monsoon circulation over the Indian sub-continent. The extensive Himalayan range elongates in a east-west direction, forming a barrier between tropical and polar air masses (Nieuwolt, 1977). During winter, the Himalaya and the Tibetan Plateau deflect strong westerly winds along their northern and southern margins, while in summer the Himalaya restricts the penetration of south-easterly monsoon air masses. The elevated topographies of the Himalayas together with the input of moisture by monsoon circulation give rise to a vast area of glaciers and snow fields, which form the headwaters of several socio-economically important river systems (e.g., Ganges and Brahamaputra). Large fluctuations in climate (e.g., changes in temperature and moisture input) could therefore have an adverse effect on a regional scale, causing droughts and floods.

Links between monsoon and other global scale climate related phenomena (e.g., Eurasian snow cover, El Niño Southern Oscillation, etc.) extend the implications of climatic variations in the Himalaya and in the Tibetan Plateau beyond the regional scale (Dey and Bhanu Kumar, 1982; Dey and Bhanu Kumar, 1983; Torgersen, 1984; Barnett et al., 1988). Climatic changes in the Himalayan region could be a reflection of large scale climate changes or could even be driving them. Unfortunately, the instrumental meteorological records from high elevations (i.e., greater than 4000 m a.s.l.) of the Himalaya are relatively short (i.e., 5-7 years; Grabs, 1992) and therefore provide only limited information on changes in high elevation climate. Meteorological data from the Tibetan Plateau are also limited. Studies on climatic trends, therefore, have to rely on records from stations south of the Himalaya or from outside the Tibetan Plateau.

The atmospheric composition over the Himalayas should reflect the interaction of two major circulation systems: the summer south-easterly monsoon circulation and the winter westerly circulation. The circulation system in the Himalayas is also characterized by regional thermally driven valley wind systems. Due to high radiation and intense heating of the high elevation regions a regional scale, valley wind systems dominate during the day time (e.g., Hindman, 1995; Hindman et al., 1996). Due to proximity to the ground surface, this circulation is likely to pick-up and transport a considerable amount of pollutants (Nakajima, 1976). Further, the intensity of this circulation system changes with season, which should have an important impact on the atmospheric composition.

The composition of the atmosphere over the region, however, is poorly known due to limited amount of monitoring programs outside of major cities. Ideal sites for monitoring atmospheric composition and its evolution should be free from local pollution and representative of the remote troposphere (Adams et al., 1977; Huebert and Lazrus, 1980). Short term aerosol sampling during monsoon and post-monsoon seasons by Ikegami et al. (1978), Wake et al. (1994), and Shrestha et al. (1997) has indeed shown low pollutant concentrations in the Himalayan atmosphere suggesting these sites provide a representation of the remote troposphere. However these studies, limited on temporal scales (i.e., weeks), do not provide useful information on the seasonal variations in the atmospheric composition that are expected due to dramatic changes in atmospheric circulation on seasonal time scales. Furthermore, the continued population growth and increasing industrialization of Asia is expected to result in large increases in anthropogenic emissions and air pollution (Foell et al., 1995; Zhao and Sun, 1986; Galloway et al., 1987; Carmichael et al., 1995). A systematic monitoring of atmospheric composition in the remote Himalayas will serve as a baseline to monitor the influence of anthropogenic activities on atmospheric composition.

The focus of the research presented in this dissertation was the investigation of climatological variation in the Himalayas and its vicinity as inferred by instrumental meteorological records and to investigate seasonal and spatial variations in the atmospheric composition in the Himalayas. The second objective of the research was to investigate the relationship between variations in atmospheric composition and their relationship with meteorological variables, as well as precipitation chemistry. This objective was aimed at providing important information for the climatic interpretation of glaciochemical data currently being collected from vast glaciated regions of the Himalayas. Temperature and precipitation trends were analyzed using records from a network of meteorological stations in Nepal. A short-term (two weeks) aerosol and snow chemistry sampling program was conducted at Hidden Valley in the Himalayas of western Nepal in the summer of 1994. A year-long sampling of aerosol chemistry and SO_2 was conducted simultaneously at a remote Himalayan site (Phortse) and a rural Middle-Mountain site (Jiri). Within the year long sampling in Phortse, three short-term intensive sampling programs, which also included precipitation sampling for chemical studies, were also conducted.

The body of this dissertation consists of five sections (II through VI), each of which represents a paper that has already been published (Shrestha et al., 1997; Chapter IV), has been accepted for publication (Shrestha et al., press; Chapter II), or is being prepared for submission to international scientific journals for publication. As such, each section contains its own abstract, introduction, review of methods, presentation and discussion of results, conclusions, and acknowledgments. A complete list of references is included at the end of this dissertation. The first two papers focus on physical climate variations in the Himalayas and the final three papers deal with aerosol and precipitation chemistry in the Himalayas. Combined, these papers are the first in depth studies on climatic trends and chemistry of the atmosphere in the Nepal Himalaya and thus provide a significant contribution to the improved understanding of regional climate dynamics and their relationship to larger scale climatic phenomena.

Chapter II describes temperature trends in the Himalayas based on meteorological station records from Nepal. The data reveal warming trends after 1977, ranging from 0.06° to 0.12 °C per year in most of the Middle Mountain and Himalayan regions, while the Siwalik and Terai (southern plains) regions show warming trends at less than 0.03 °C per year. The Kathmandu record, the longest in Nepal (1921-1994), while partially influenced by recent urbanization also shows features similar to temperature trends in the northern hemisphere, suggesting links between regional trends and global scale phenomena.

Chapter III provides an analysis based on precipitation records from stations in Nepal for the period 1959-1994. The data show variability at inter-annual and decadal time scales both in the country-wide and sub-regional spatial scales in annual, monsoon, pre-monsoon, and winter precipitation. Associations between all-Nepal precipitation and large scale phenomenon such as El Niño and to some extent with sunspot numbers are found. The year 1992 is the driest one in the record and is likely related to the Mt. Pinatubo eruption.

Chapter IV describes aerosol and surface snow samples collected from Hidden Valley in the Dhaulagiri region of western Nepal during the summer monsoon of 1994. While the samples are temporally limited, they represent the first detailed investigation of aerosol and snow chemistry and their relationship with meteorological variations and variations in synoptic scale circulation.

In Chapter V, a year long record (Sep. 1996 to Nov. 1997) of the water soluble components of atmospheric aerosol at a remote Himalayan site and a rural Middle-Mountain site in Nepal is presented. This study yields insights into the seasonal variations in the Himalayan atmospheric composition. Low concentrations during the second half of monsoon and post-monsoon seasons, gradually increasing concentrations during winter and significantly higher concentrations (up to 20 times higher than low concentrations) during pre-monsoon and early-monsoon seasons are characteristic of the seasonal variations. The seasonal variations in aerosol concentrations are related to the variations in the atmospheric circulation and precipitation.

Chapter VI describes three short-term intensive sampling campaigns conducted at Phortse in the post-monsoon (September) of 1996, pre-monsoon (March) of 1997 and Monsoon (August) of 1997. This study is the most detailed investigation on the relation between aerosol precipitation chemistry so far in the Nepal Himalaya. Results show that precipitation chemistry in general is similar to aerosol chemistry, indicating that glaciochemical data from the Himalayas can be used to interpret atmospheric composition of the past.

The final chapter (VII; Concluding Remarks) provide an overall interpretation of the dissertation results, as well as outline the significance and contribution of the research presented here to other programs. This chapter also briefly describes a project to be conducted at two sites in the Nepal Himalaya in the near future, which will include size measurement of major ions and carbonaceous particles in the air and the radiative property of aerosol.

CHAPTER TWO

MAXIMUM TEMPERATURE TRENDS IN THE HIMALAYA AND ITS VICINITY: AN ANALYSIS BASED ON TEMPERATURE RECORDS FROM NEPAL FOR THE PERIOD 1971-94

This chapter has been accepted for publication inthe Journal of Climate
(November 6, 1998)

II. MAXIMUM TEMPERATURE TRENDS IN THE HIMALAYA AND ITS VICINITY: AN ANALYSIS BASED ON TEMPERATURE RECORDS FROM NEPAL FOR THE PERIOD 1971-94

ABSTRACT

Analyses of maximum temperature data from 49 stations in Nepal for the period of 1971-1994 reveal warming trends after 1977 ranging from 0.06° to 0.12 °C per year in most of the Middle Mountain and Himalayan regions, while the Siwalik and Terai (southern plains) regions show warming trends less than 0.03 °C per year. The subset of records (14 stations) extending back to the early 1960's suggests that the recent warming trends were preceded by similar widespread cooling trends. Distributions of seasonal and annual temperature trends show high rates of warming in the high elevation regions of the country (Middle Mountains and Himalaya), while low warming or even cooling trends were found in the southern regions. This is attributed to the sensitivity of mountainous regions to climate changes. The seasonal temperature trends and spatial distribution of temperature trends also highlights the influence of monsoon circulation.

The Kathmandu record, the longest in Nepal (1921-1994), shows features similar to temperature trends in the northern hemisphere, suggesting links between regional trends and global scale phenomena. However, the magnitudes of trends are much enhanced in the Kathmandu as well as in the all-Nepal records. Our analyses suggest that contributions of urbanization and local land use/cover changes to the all-Nepal record are minimal and that the all-Nepal record provides an accurate record of temperature variations across the entire region.

INTRODUCTION

Studies on long-term variations in surface air temperature for the entire globe (Jones et al., 1986c; Hansen and Lebedeff, 1987; Hansen and Lebedeff, 1988) as well as for the hemispheres (Angell and Korshover, 1978; Jones et al., 1986a; Jones et al., 1986b) have shown a rising trend during the last few decades. Similar results have been found for low latitude regions in the Northern Hemisphere.

Mountainous environments are considered sensitive indicators of climate-change (Barry, 1990; Stone, 1992; Beniston, 1994). Several studies in the Himalayas have found that glaciers in the region have retreated considerably in the last two decades (Higuchi et al., 1980; Miller, 1989; Miller and Marston, 1989; Yamada et al., 1992; Kadota et al., 1993). Recent studies have identified the formation and growth of several glacial lakes possibly due to fast retreat of glaciers, which could lead to catastrophic outburst floods (Vuichard and Zimmermann, 1987). It is possible that global warming is responsible for the recent glacial retreat in the Himalayas, although precipitation changes may also be important. However, Seko and Takahashi (1991) have suggested that over the last decade, glacier fluctuation in the Khumbu Himal followed the fluctuations in air temperature more than those in precipitation. Mayewski and Jeschke (1979) have suggested that Himalayan glaciers have been retreating since 1850 AD. It is possible that the cooling in 1940 observed in global record caused re-advancing of these glaciers and the warming after the mid-1970's resulted in accelerated shrinking in the past two decades.

The Himalaya and Tibetan Plateau play an important role in regional climate, most particularly with respect to monsoon circulation. Links between the monsoon and other global scale phenomena extend the implications of climatic variations in the Himalaya and in the Tibetan Plateau beyond the regional scale (Dey and Bhanu Kumar, 1982; Dey and Bhanu Kumar, 1983; Barnett et al., 1988). Climatic changes in the Himalayan region could be a reflection of large scale climate changes or could even be driving them.

It is therefore important to understand the climatic trends in the Himalaya and their relationship with global trends. Unfortunately, the instrumental meteorological records from high elevations (i.e. greater than 4000 m a.s.l.) of the Himalaya are relatively short (i.e. 5-7 years; Grabs, 1992) and therefore provide only limited information on changes in high elevation climate. Meteorological data from the Tibetan Plateau are also rare. Studies on climatic trends, therefore, have to rely on records from stations south of the Himalaya or from outside the Tibetan Plateau. A study of long term trend in surface air temperatures in India by Hingane et al. (1985) indicated an increase in mean annual temperature of 0.4°C over the past century. A study of changes in air temperature of Qinghai-Xizang (Tibetan) Plateau showed a decreasing trend from 1950 to 70 and an increase after 1970 (Li and Tang, 1986).

The present study is the first to be based on surface temperature data from a network of stations in Nepal. Individual records dating back to the early 1960's were investigated to reveal temperature trends in various geographical settings and demographic situations. Based on this, a period representative of present temperature trends was selected. For the period selected, annual and seasonal patterns of maximum temperature trends in Nepal were investigated. Further, temperature trends from Kathmandu Valley, which has the longest record in Nepal (1921 to 1994), were compared to large scale (global) temperature trends.

DATA AND METHODS

Physiographically Nepal can be divided into 6 regions: Terai; Siwalik; Middle-Mountains; High Mountains; High Himalaya; and Tibetan Plateau (WECS, 1986). The elevation ranges from 65 m a.s.l. in the Terai to over 8000 m a.s.l. in the High Himalaya. About 90 percent of the country is characterized by mountainous terrain.

With the purpose of representing all parts of the country, 49 temperature stations were selected (Table II.1). Among them records from 33 stations begin in 1971, while records from 14 stations begin in the early to mid-1960's. The record from the Kathmandu Indian Embassy (Kathmandu IE) is the longest, dating back to 1921.

Data for all the stations were collected and published by the Department of Hydrology and Meteorology (DHM), His Majesty's Government (HMG) of Nepal . Data from 1971 to 1994 for most of the stations are published, while data after 1994 are still in the preliminary stage of publication and not in accessible formats (DHM, 1997). Temperature data are available as monthly means of daily maxima and minima. A preliminary analysis showed high year to year fluctuation and lack of significant trend in minimum temperature in most of the station records in Nepal. Furthermore, minimum temperatures have been found to be more affected by urbanization as compared to maximum temperature (Mitchell, 1961). Therefore we used maximum temperatures for our analysis, which are hereafter referred to simply as temperature.

We used the temperature anomaly for 24° to 40° North latitude as an indicator of large scale northern hemisphere mid-latitude temperature trends. This data set was obtained from Goddard Institute of Space Science (GISS) on-line data products (Hansen et al., 1996) and is hereafter referred to as the 24-40° North latitude temperature anomaly.

All the Nepal stations data were carefully analyzed for homogeneity and for missing data. We only selected stations where instrument locations were not significantly changed within the period of record. Further, the relative homogeneity test was conducted by graphical study of ratios of temperature records from neighboring stations (WMO, 1966a). Any inhomogeneity in the record is depicted by a significant departure of the ratio from the mean. About 15 stations were discarded as a result of this test. The stations selected for this study did not show such inhomogeneity and had few missing data. Missing data were filled by temporal and spatial interpolation whenever possible.

Annual and seasonal means were calculated for each year for all stations. The four seasons are divided as: winter (December of previous year, January and February), pre-monsoon (March-May), monsoon (June-September), and post monsoon (October and November). Annual mean is the average of January to December monthly means.

Seasonal and annual temperature trends were computed for all stations using linear regression. Spatial distributions of temperature trends were computed by interpolating the station trends to a rectangular block of 80.25° to 88.25° E and 26.25° to 30.25°N with the grid size of 0.25°x0.25°. The error associated with the use of rectangular block instead of spherical will be minimal due to small area considered in the study. The grid size was selected in such a way that close stations were individually represented. For the selection of the grid size interpolations were performed by gradually reducing the grid size until further reduction did not provide significant change in the results. An analysis and visualization program (Spyglass 3.01) was used for interpolation by Kriging. Kriging is superior to any other interpolation method because in this method every known and missing data values are assigned a variance and a variance matrix is generated. Kriging is considered to be the best interpolation algorithm when the known values are fewer than 100 and missing values are relatively more (Fortner, 1992). To test this algorithm, mean annual temperature maps of Nepal were prepared using several interpolation techniques with same number of stations. Kriging provided the best result as the map prepared by this technique clearly showed elevational and latitudinal gradient in temperature, much similar to maps prepared by the Department of Hydrology and Meteorology using much more station data.

Based on the annual and seasonal distributions of temperature trends, mean trends for different physiographical regions and for the whole country were calculated. Due to the low number of high elevation stations, we combined stations in the High Mountain and High Himalaya regions. The combined region is referred to as the Himalayan region (Fig. II.1). Annual mean station temperatures were spatially interpolated for each individual year from 1971 to 1994 and regional and all-Nepal mean temperatures were derived for that period. The Kathmandu IE record extends from 1921 to 1976. We used multiple linear regression between this station and four others in the Kathmandu Valley during the period of overlap (1971-1976) to create a single Kathmandu record through 1994. The extension of the Kathmandu IE record by this method allowed us to directly compare the extended record with records representing large scale trends. Nevertheless, it has to be noted here that due to short overlap period (6 years) it was not possible to test the values with actual observations.

RESULTS

Analysis of Individual Station Records

Records from all 14 stations which begin in the mid-1960's were individually analyzed. Mean annual maximum temperature plots of almost all of these records show one basic characteristic: decreasing or constant trends in the earlier part of the records and increasing trends in the latter part of the records (Fig. II.2a). The turning points (identified by arrowheads in Fig. II.2a) were determined visually and verified by fitting a fifth order polynomial functions. Interestingly, these points in most of these records occur around the mid-1970's. Exceptions are: Chisapani, which shows a decreasing trend throughout the record period, Chialsa, which shows an increasing trend throughout the record period, and Chainpur which does not show any distinct trend.

In contrast, minimum temperatures for these station show high inter annual variability, while they do not display any significant trends (Fig. II.2b)

Regional and All-Nepal Annual Temperature Trends

Since 11 out of 14 stations show two different trends over the period of record, it is not valid to consider the whole period (1971 to 1994) for calculating temperature trends. Only the latter parts of records (after turning points) are therefore considered representative of current temperature trends. The selection of a common period for all stations is complicated by the fact that the turning points, although close, do not occur in the same year in all stations. In addition, several stations were added in the mid-1970's. It was, therefore, necessary to select a period providing the best spatial coverage, as well as representing contemporary temperature trends of different regions and the country.

For this, spatial distribution of annual mean temperatures was calculated using Kriging for the period 1971 to 1994. Based on this distribution, temperature trends were derived for five different physiographical regions as well as for the whole country. Temperature trends in the five regions and the country were constant or decreasing before 1978, whereas after 1978 they were increasing (Fig. II.3). This result is consistent with the result obtained from analyses of individual stations. The period 1977 to 1994 was selected for further analysis.

Regional and all-Nepal temperature trends change from season to season. The greatest warming trend is observed in the post-monsoon season (Table II.2). According to seasonal as well as annual warming trends, the five regions may be divided into two groups. Trans-Himalayan, Himalayan and Middle Mountain regions fall in the first group with high warming rates, while Siwalik and Terai regions fall in the second group as they show low warming rates. However, for the post-monsoon season even the Siwalik and the Terai regions show high warming rates.

While simple linear regression is an easy technique for qualitative assessments of trends in climatological parameters, this technique has some limitation. Presence of outliers and non-randomness in the record limit the usefulness of linear regression. To check this possibility we carried out non-parametric tests (Mann-Kendall test and Spearman test) for trends in the temperature records (WMO, 1966a). Results of these tests support the results of the simple linear regression (Table II.3) . It is therefore suggested that the simple linear regression is a valid method for assessing trends in temperature records considered in this study.

Seasonal and Annual Temperature Trends

The spatial distributions of mean annual temperature trends over the period of 1977 to 1994 are presented in Figure II.4a. The mean annual maximum temperature trend distributions show warming in most parts of the country. High mean annual temperature increases (>0.06 °C yr^{-1}) occur in most of the northern belt (the Trans-Himalayan and Himalayan regions and central and western parts of the Middle-Mountains). Within the Middle Mountain region there are two pockets of anomalously high warming rates (≥0.12 °C yr^{-1}): the western middle mountain region and the Kathmandu Valley. Most of the Siwalik and the Terai regions show considerably low increasing trends (<0.03 °C yr^{-1}) or even decreasing trends (-0.03 °C yr^{-1}).

Winter, pre-monsoon and monsoon temperature trend distributions closely resemble the yearly temperature distribution, with the winter season showing the greatest, and the pre-monsoon season showing the lowest, rate of increase (Fig. II.4b and II.4c). As in the case of annual temperature trends distributions, most of the northern, the central, and the mid-western parts of the country show high rates of increase in temperature. The contrast in temperature trends between the northern and southern regions is much greater for the monsoon season (Fig. II.4d). The post-monsoon temperature trend distribution is strikingly different from all other distribution patterns (Fig. II.4e). Almost the entire country shows an increasing trend during this season.

Comparison with Northern Hemisphere Temperature Anomaly

To study relationships between temperature trends in Nepal and global scale trends, we compared Kathmandu annual temperature with 24-40° North latitude annual temperature anomaly after applying a 10-year running mean to both records (Fig. II.5). There are some striking similarities between these records. 24-40° North latitude temperature shows rising trend before 1940, decreasing trend from 1938 to 1972 and increasing trend again after 1972. There is a general trend of cooling from 1935 to 1974 in the Kathmandu record. This period, however, was interrupted by two episodes of warming (1948-1954 and 1964-1969) and subsequent cooling (1954-1964 and 1969-1974). The last warming trend in the Kathmandu record is the longest monotonic trend in the entire record. The range of cooling and warming in these records are of different magnitudes. The cooling of 1940 to 1970 lowered the 24-40° North latitude temperature by about 0.23 °C, while in the Kathmandu record, the drop in temperature from 1934 to 1975 is about 0.6 °C. Similarly, the warming after 1970 has raised the 24-40° North latitude temperature by about 0.5 °C, whereas the Kathmandu temperature has risen by 1 °C since 1975.

For comparison, the all-Nepal annual temperature record and annual temperature records of four Kathmandu Valley stations are also presented in Figure II.5. The all-Nepal temperature shows a rise in temperature of about 0.9°C since 1977.

DISCUSSION

The presence of general cooling or constant trends before 1970 and warming after the mid-1970's in most of the Nepal station records extending back to the early 1960's (Fig. II.2a), and similar warming in regional as well as all-Nepal temperature trends (Fig. II.3), demonstrates the spatial coherence of these records despite tremendous contrast in the physiographic and demographic characteristics among the stations. This in turn suggests that the Nepal meteorological station data presented here reflects regional climatic trends which predominate over the whole country. The relatively high rate of warming in the all-Nepal record after the mid-1970's is due predominantly to the high rates of warming in the high elevation areas of the Himalaya and Middle-Mountain regions. The mountainous regions of Nepal therefore appear to be amplifying the regional warming trend which supports the concept that alpine regions provide a sensitive indicator of climate change.

The recent reduction of snow and glacier cover in the Himalaya may also be contributing to the higher rates of warming observed in the higher elevations regions of

Nepal. A reduction in snow and glacier cover in the high elevation will change the surface albedo of the region, which inturn will increase the surface air temperature, thereby acting as positive feedback mechanism (e.g. Meehl, 1994). The importance of snow and glacier cover variations is manifested by the effect of the Eurasian snow cover variations on the regional climate, mainly the summer monsoon as suggested by several empirical as well as model studies (Dey and Bhanu Kumar, 1982; Dey and Bhanu Kumar, 1983; Khandelkar, 1991).

Seasonal temperature trend distributions appear to be influenced by summertime monsoon circulation. The increased warming rate in the highlands in the north and low warming or cooling in the plains in the south during the monsoon season (Fig. II.4d) is possibly due to the advection of moisture and associated increased cloudiness in the south, while comparatively dry and fair weather dominates in the north. This may also account for the lower rates of warming in Middle-Mountain and Siwalik regions. Siwalik (700 to 1500 m a.s.l.) is where the monsoon circulation encounters the first orographic barrier and the region receives a large amount of monsoon precipitation. The Middle-Mountain contains the Mahabharat range (1500 to 2700 m a.s.l.), the second but a higher barrier to monsoon circulation which results in several pockets of very high precipitation (Shankar and Shrestha, 1985). Similarly, the increased warming rate during the post-monsoon season is likely linked to the retreat of monsoon. The extent of cloud cover has been found to minimize the temperature difference between urban sites and rural sites by reducing the magnitude of urban heat islands (Sundborg, 1951). The actual mechanism that connects monsoon circulation and the observed differences in temperature trends is difficult to explain. It is possible that a mechanism similar to that operating in the case of urban heat islands is also responsible for the reduction of larger scale warming. Another possibility is a feedback mechanism in the form of strengthened monsoon due to warming in the Himalayas (and possibly in the Tibetan Plateau). A stronger monsoon is associated with increased cloudiness, hence reduces the warming rate.

The lack of cloud cover data precludes the possibility of investigating cloud cover variations in the region. A separate study on precipitation fluctuations in Nepal is in progress. Results of this study may help test the hypothesis presented here on the relationship between temperature trends and monsoon. Preliminary results of the study indicate some increases in monsoon precipitation in the country, which is in agreement with model prediction regarding increase in monsoon precipitation with global warming (e.g. Follarrd, 1990).

The temperature trends found in the Kathmandu record are similar to those in Tibetan Plateau, which showed decrease in temperature from the 1950's to the 1970's and warming thereafter (Li and Tang, 1986). Warming after 1965 was also observed in temperature records from the Ganges basin in India (Kothyari and Shing, 1996). The temperature trend of India as a whole, however, is different as it shows low and constant warming over the whole century, lacking the cooling around the 1940's and the distinct warming after the 1970's (Hingane et al., 1985). It is likely that the southern part of the Indian sub-continent is behaving climatologically differently than the Himalayan region.

Nearly synchronous cooling and warming trends in the Kathmandu and the 24-40° North latitude temperature anomaly records indicates links between temperature trends in Nepal and large scale trends (Fig. II.5). Despite the visual resemblance between the Kathmandu record and the 24-40° North latitude temperature records there is a large difference in the magnitude of warming. The Kathmandu Valley has experienced tremendous urbanization associated with remarkable population growth in the last two decades (Sharma, 1986; CBS, 1993; EPC, 1993). Nevertheless, it is unlikely that the warming trend in the Kathmandu record is due only to the urbanization, mainly because even the cooling rate between 1934 and 1975 in the Kathmandu record greatly exceeded the global cooling between 1940 and 1970 and this cannot be attributed to urban effects. Moreover, the urban effect of Kathmandu city should have been negligible in the 1940's. It is possible that the high warming rate observed in the record from Kathmandu is due to its physiographical characteristics, which may make the valley more sensitive to climatic change. The Kathmandu record may in general be reflecting the large scale Northern Hemisphere trend with some amplification, while the last two decades may have had the additional influence of urbanization.

Although the warming trend after the mid-1970's in the all-Nepal record is slightly lower than that in the Kathmandu record, it is still greater than the global trend. The all-Nepal temperature trend, which is a spatial average of all station records also includes station records that may have been influenced by urbanization. Nevertheless, the net effect of such stations is minimal because of their localized nature. A similar conclusion was also drawn by Hansen et al. (1996) regarding the influence of urban warming on the global temperature analysis. Furthermore, high warming trends are observed in the Himalayan and Trans-Himalayan region, which lack urban centers. It is therefore concluded that the all-Nepal trend is not biased by urban effects. The presence of trends in several stations with records dating back to the early-1960's, similar to that in the Kathmandu record supports that warming trends observed in the records from Nepal are not local phenomena but are occurring on a country-wide or regional scale.

Also associated with population growth are radical changes in land use and land cover over the country (Ekholm, 1975; Hrabovsky and Miyan, 1987; Karan, 1987). Several studies have suggested a link between changes in air temperature and changes in land use and land cover, especially tropical deforestation (Henderson-Sellers and Gornitz, 1984; Gash and Shuttleworth, 1991). Sinha and Swaminathan (1991) have suggested possible climate change and its impact on agriculture as a result of deforestation in India. Meher-Homji (1991) suggested the possible impact on the hydrology of India related to deforestation. If the warming in Nepal is due to local land use and land cover changes, the highest warming should have been observed in the Terai and the Siwalik regions, which have experienced the greatest amount of land use/land cover changes over the past few decades. A recent study comparing aerial photographs taken in late-1970's and satellite images taken in the early-1990's show deforestation in Terai at an average rate of 1.3% per year (FRSC, 1994). A similar study has just been initiated for the middle mountain regions. Preliminary results indicate that at the regional scale the forest cover has not changed significantly in last 10 to 12 years (per. comm., Sharma R.R., Forest Research and Survey Centre). Since the Terai region actually shows the least amount of temperature increase over the past three decades we suggest that the influence of land use and land cover change in the all-Nepal temperature is not significant, thus these analyses are representative of actual climatic temperature trends in Nepal.

Growing numbers of studies suggest increases in the atmospheric concentrations of greenhouse gases to cause global warming (e.g. IPCC, 1990).Near synchronous warming as well as cooling trends in 24-40° North temperature and Kathmandu between the period of 1920 to the mid-1970's and similar synchronous warming in these two records as well as in the all-Nepal record after the mid-1970's suggest that trends in all these records are due to some common forcing mechanism. Although it is not possible in the present study to suggest possible forcing mechanism for the observed trends in Nepal, increase in the concentrations of greenhouse gases could have played important role.

CONCLUSION

Spatial distributions of maximum temperature trends in Nepal show high warming trends in most of the Himalayan region and the Middle-Mountains, while low warming or even cooling trends are observed in most of the Terai and the Siwalik regions. Though the actual mechanisms are not well understood, monsoon circulation may play an important role in the distribution of seasonal temperatures as well as temperature trends. A study on trends of precipitation over Nepal currently in progress should provide additional information for understanding temperature trends found in the present study.

The longest temperature record from the country, the Kathmandu record, shows features similar to the 24-40° North latitude temperature record, namely the cooling trends from 1935 to 1975 and warming trends thereafter, thereby suggesting connections with northern hemisphere temperature trends. Characteristics in the Kathmandu record are also very similar to those found in China and to some extent to those in northern India. The relatively higher magnitude of cooling and warming in the Kathmandu record is attributed to the physiographical characteristics of the Kathmandu valley and its sensitivity to climatic changes. The warming after the mid-1970's may also be reflecting urbanization.

Several other locations in Nepal, which do not show overall growth rates comparable to Kathmandu, show warming trends of comparable or even higher rates than Kathmandu. The all-Nepal temperature record, a spatial average of 49 station records, also shows similar warming after the mid-1970's. We suggest that the warming is not due to urban effects or local land use/cover changes, but is a widespread trend and is representative of regional scale trends. It is the warming in the high elevation regions that contributes to the high warming rate in the all-Nepal record. We suggest that the Middle-Mountain and high Himalayan regions are more sensitive to climate change due to their physiographic characteristics.

ACKNOWLEDGMENTS

We thank Dr. B. Keim, Department of Geography, University of New Hampshire, Dr. S.P. Adhikary, Himalayan Climate Center, Nepal. Thanks are extended to the Central Data Processing Unit, Department of Hydrology and Meteorology, Nepal for proViding some data in digital format. We also thank two anonymous reviewers, whose suggestions on the earlier version of the text helped a lot to improve it.

TABLE II.1. Descriptions of stations included in this study

Station No.[†]	Station Name	Elevation (m)	Latitude (°N)	Longitude (°E)	Physiographic Regions[¥]
0203	Silgadi Doti	1360	29.27	80.98	MM
0207	Tikapur	140	28.50	80.95	TAR
0209	Dhangadi	1360	28.68	80.60	TAR
0215	Godavari (W)	288	28.87	80.63	TAR
0303	Jumla	2300	29.28	82.17	H
0401	Pusma Camp[$]	950	28.88	81.25	MM
0402	Dailekh[$]	1402	28.85	81.72	MM
0405	Chisapani[$]	225	28.65	81.27	SW
0406	Surkhet	720	28.60	81.62	SW
0409	Khajura	190	28.10	81.57	TAR
0416	Nepalgunj	144	28.07	81.62	TAR
0508	Tulsipur	725	28.13	82.30	SW
0604	Thakmarpha	2566	28.75	83.70	TH
0612	Mustang	3705	29.18	83.97	TH
0703	Butwal	205	27.70	83.47	SW
0705	Bhairahawa	110	27.52	83.43	TAR
0706	Dumkauli	154	27.68	84.22	SW
0707	Bhairahawa AG	120	27.53	83.47	TAR
0802	Khudi Bazar[$]	823	28.28	84.37	SW
0804	Pokhara Airport[$]	827	28.22	84.00	MM
0809	Gorkha[$]	1097	28.00	84.62	MM
0811	Malepatan Pokhara	856	28.22	83.95	MM
0814	Lumle	1642	28.30	83.80	MM
0815	Khairini Tar	190	28.03	84.10	MM
0902	Rampur[$]	256	27.62	84.42	SW
0905	Daman	2314	27.60	85.08	MM
0906	Hetaunda NFI	466	27.42	85.05	SW
0909	Simara Apt	130	27.17	84.98	TAR
0911	Parwanipur	115	27.07	84.97	TAR
1007	Kakani	2064	27.80	85.25	MM
1022	Godavari	1400	27.58	85.40	MM
1029	Khumaltar	1350	27.67	85.33	MM
1030	Kathmandu Airport[$]	1336	27.70	85.37	MM
1039	Panipokhari	1335	27.73	85.35	MM
1043	Nagarkot	2150	27.72	85.52	MM
1103	Jiri[$]	2003	27.63	86.23	H

TABLE 1. continued

Station No.[†]	Station Name	Elevation (m)	Latitude (°N)	Longitude (°E)	Physiographic Regions[¥]
1111	Janakpur Airport	90	26.72	85.97	TAR
1206	Okhaldunga[§]	1720	27.32	86.50	H
1209	Bhojpur IMD[§]	1595	27.18	87.05	MM
1220	Chialsa[§]	2770	27.52	86.62	H
1303	Chainpur[§]	1329	27.28	87.33	H
1304	Pakhribas	1677	27.05	87.28	MM
1307	Dhankuta	1160	26.98	87.35	MM
1319	Biratnagar Airport	72	26.48	87.27	TAR
1320	Tarahara	200	26.70	87.27	SW
1323	Dharan BC	400	26.78	87.28	SW
1405	Taplejung[§]	1732	27.35	87.67	H
1407	Ilam tea Estate	1300	26.92	87.90	SW
1416	Kanyam Tea Estate	1658	26.87	88.07	SW

† Station numbers according to Department of Hydrology and Meteorology station index numb Numbers increases from west to east and north to south.

¥ TAR stands for Terai; SW stands for Siwalik; MM represents Middle-Mountain; H stands for Himalayan; and TH stands for Trans-Himalayan regions

§ Stations with records starting from or before mid-1960's

TABLE II.2. Regional Mean Temperature Trends (°C per year)

Regions	Seasonal				Annual
	Winter Dec-Feb	Pre-monsoon Mar-May	Monsoon Jun-Sep	Post-monsoon Oct-Nov	Jan-Dec
Trans-Himalaya	0.124[‡]	0.005	0.109[¥]	0.099[*]	0.090[¥]
Himalaya	0.090[¥]	0.050	0.062[¥]	0.075[‡]	0.057[¥]
Middle Mountains	0.059[*]	0.050	0.055[¥]	0.094[¥]	0.075[¥]
Siwalik	0.015	0.010	0.021	0.077[‡]	0.041[‡]
Terai	0.006	-0.004	0.014	0.069[‡]	0.041[‡]
All-Nepal	0.061[‡]	0.032	0.051[‡]	0.081[¥]	0.059[¥]

[*] significant at 95% level

[‡] signifcant at 99% level

[¥] significant at 99.9% level

TABLE II.3. Non-parametric Tests for Trends

Regions	Mann-Kendall test	Spearman test	
	τ	r_s	t
1971-94 (N=24)	$\tau_t = \pm 0.29$ †	$t_{N-2;0.05} = \pm 2.074$ §	
	Annual		
All-Nepal	0.39	0.55	3.12
Tran-Himalaya	0.38	0.52	2.84
Himalaya	0.38	0.54	3.03
Middle-Mountain	0.49	0.67	4.24
Siwalik	0.27	0.40	2.04
Terai	0.28	0.36	1.83
1977-94 (N=18)	$\tau_t = \pm 0.33$ †	$t_{N-2;0.05} = \pm 2.120$ §	
	Annual		
All-Nepal	0.56	0.66	3.48
Tran-Himalaya	0.57	0.65	3.39
Himalaya	0.54	0.73	4.28
Middle-Mountain	0.66	0.61	3.07
Siwalik	0.40	0.27	1.12
Terai	0.46	0.49	2.23
	Winter		
All-Nepal	0.35	0.42	1.87
Tran-Himalaya	0.45	0.49	2.27
Himalaya	0.49	0.66	3.47
Middle-Mountain	0.32	0.25	1.02
Siwalik	0.07	-0.16	-0.65
Terai	0.06	-0.02	-0.09
	Premonsoon		
All-Nepal	0.12	0.06	0.25
Tran-Himalaya	0.05	-0.05	-0.20
Himalaya	0.24	0.34	1.46
Middle-Mountain	0.22	0.07	0.30
Siwalik	0.07	-0.20	-0.81
Terai	0.05	-0.06	-0.24
	Monsoon		
All-Nepal	0.46	0.51	2.35
Tran-Himalaya	0.52	0.61	3.10
Himalaya	0.54	0.67	3.62
Middle-Mountain	0.48	0.40	1.75
Siwalik	0.12	-0.13	-0.51
Terai	0.11	-0.01	-0.05
	Post-Monsoon		
All-Nepal	0.53	0.63	3.26
Tran-Himalaya	0.22	0.33	1.40
Himalaya	0.36	0.60	2.97
Middle-Mountain	0.58	0.54	2.60
Siwalik	0.45	0.35	1.50
Terai	0.42	0.48	2.16

† τ values beyod this range indicates trend significant at 95 %.

§ t values beyond this range indicates trend significant at 95%

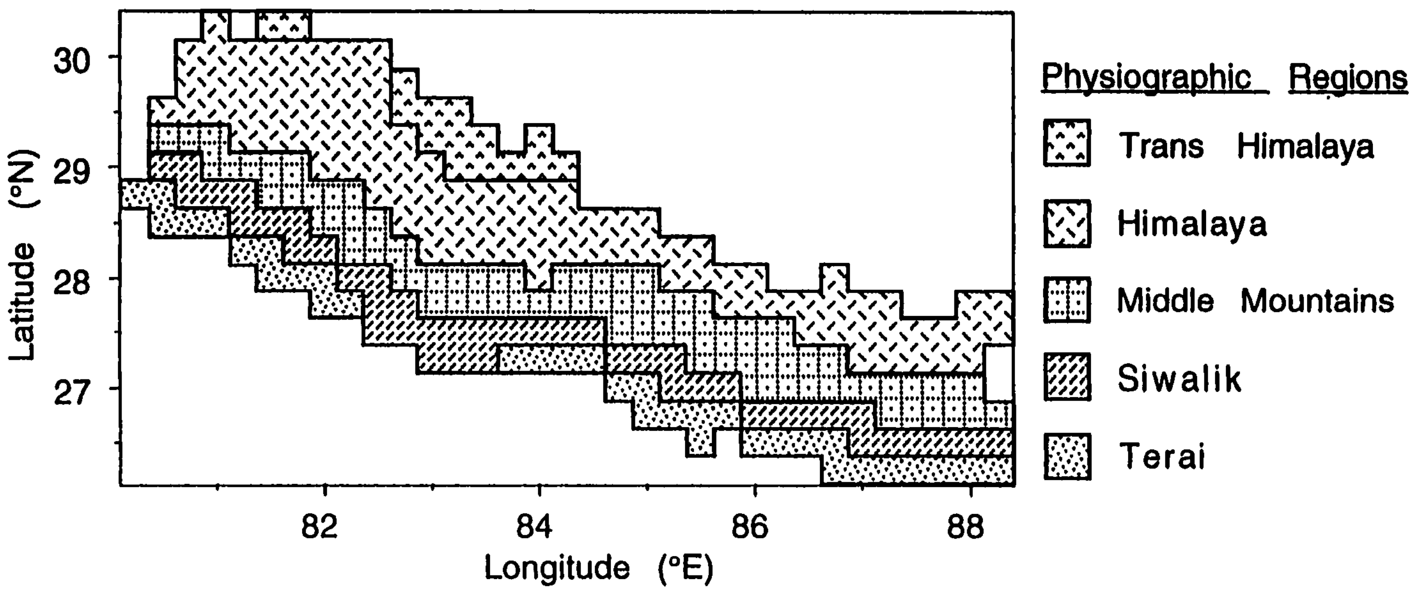


Figure II.1. Delineation of physiographic regions of Nepal. The grid size is 0.25°x0.25°.

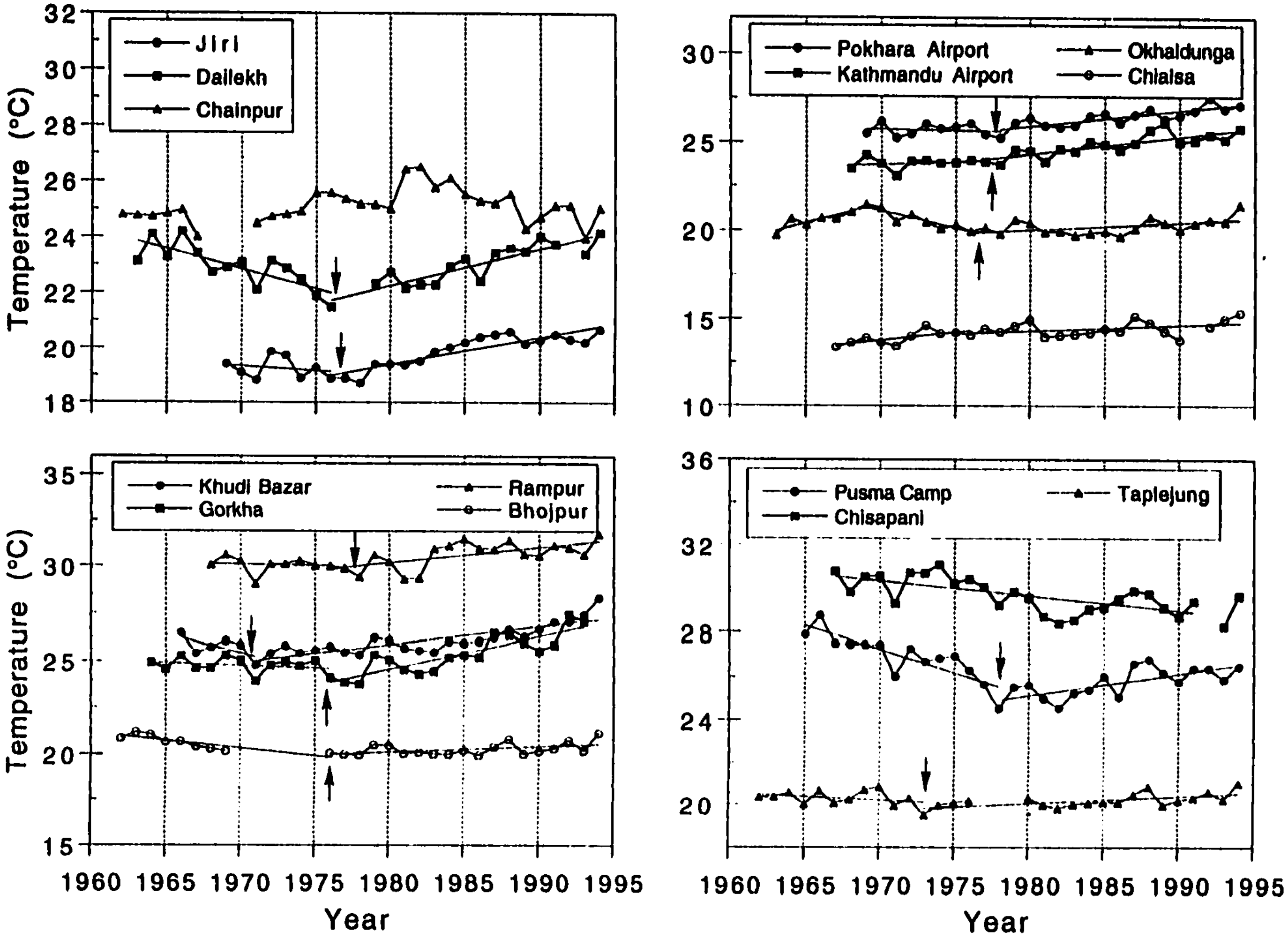


Figure II.2a. Maximum Temperature trends in 14 stations with records extending back to early to mid-1960's. Arrows indicate possible turning points in those records.

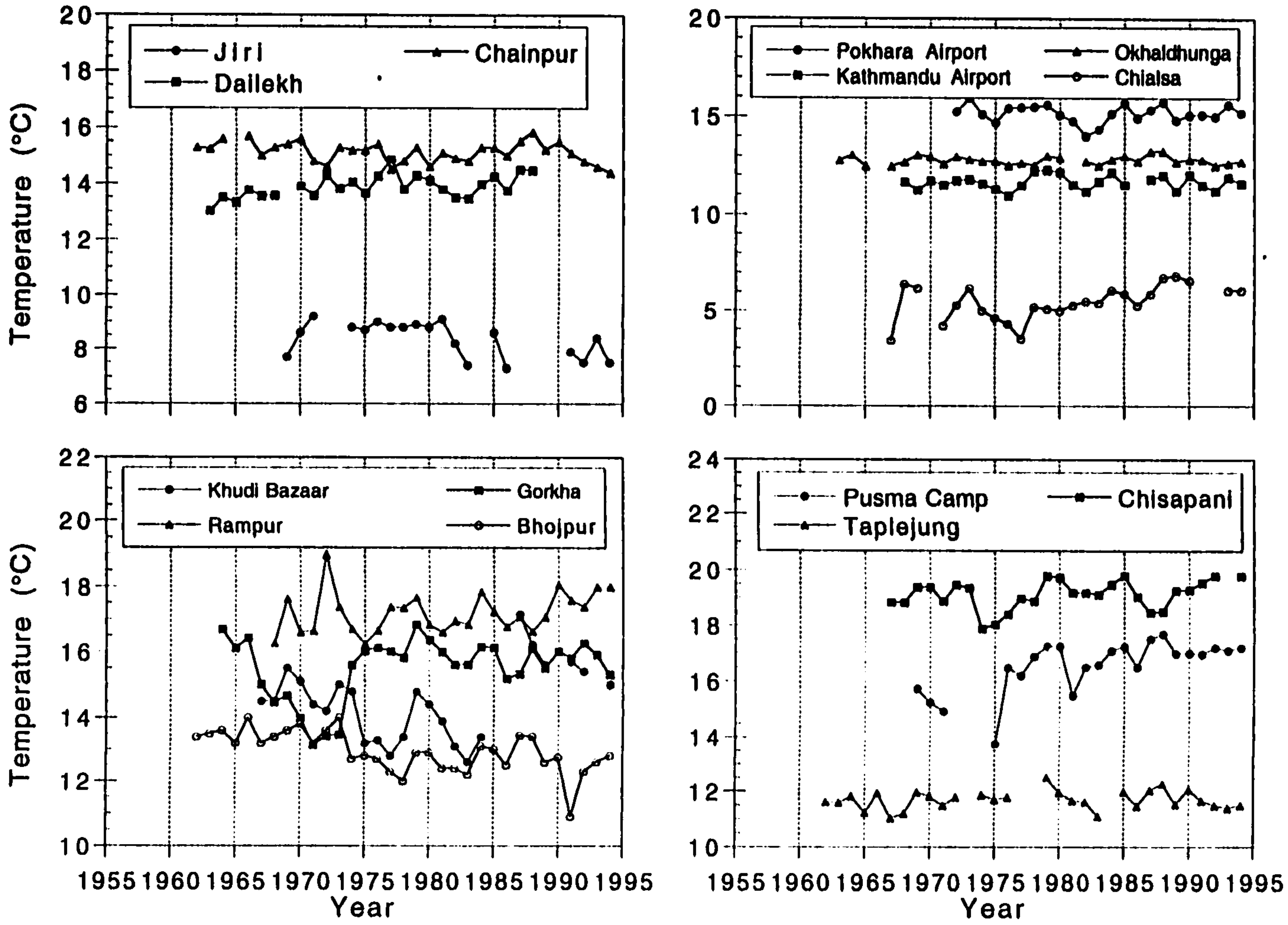


Figure II.2b. Minimum temperature trends in 14 stations with records extending back to early to mid-1960's.

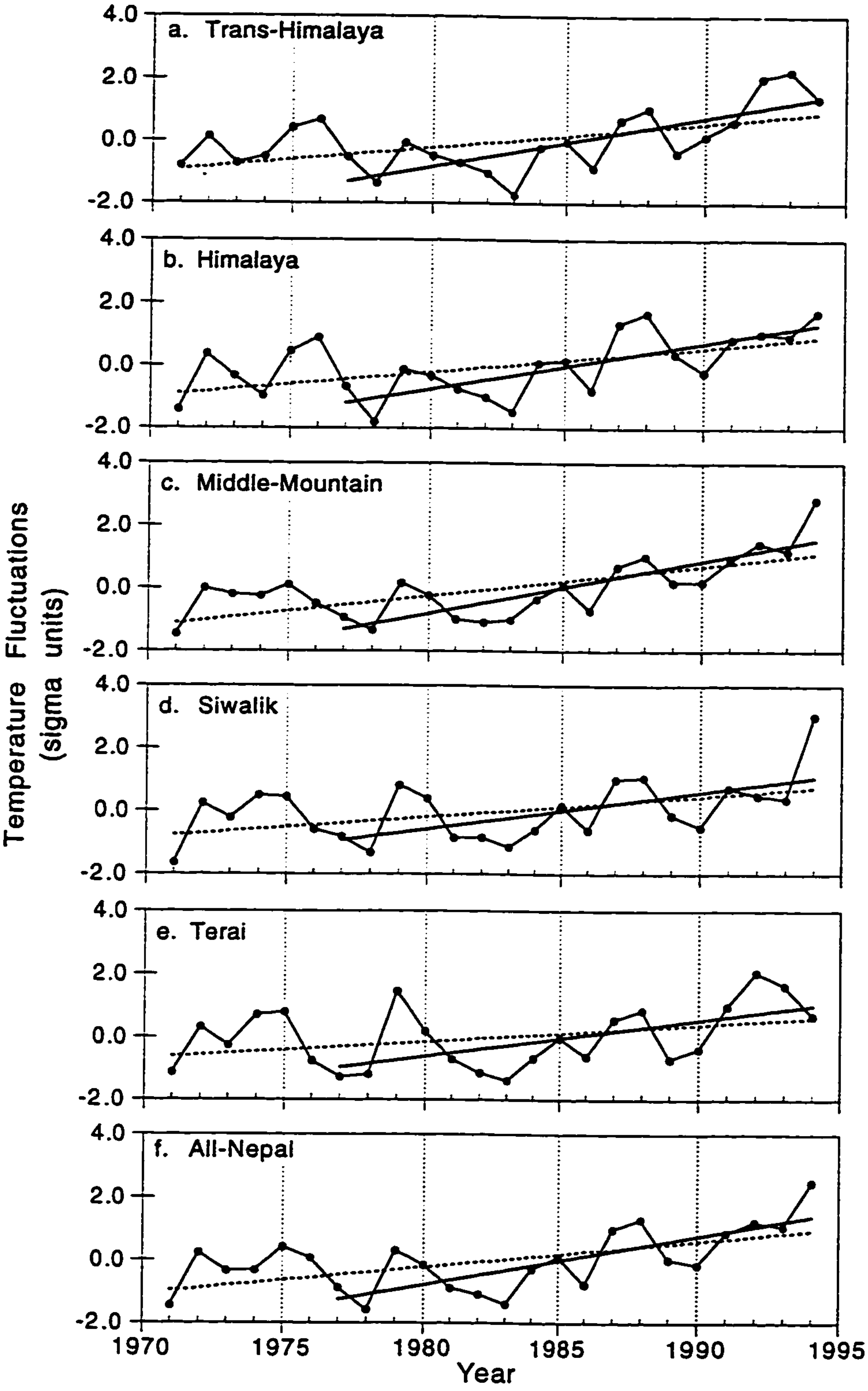


Figure II.3. Time series of mean maximum temperatures for: (a) Trans-Himalaya; (b) Himalaya; (c) Middle-Mountain; (d) Siwalik; and (e) Terai regions; and (f) all-Nepal.

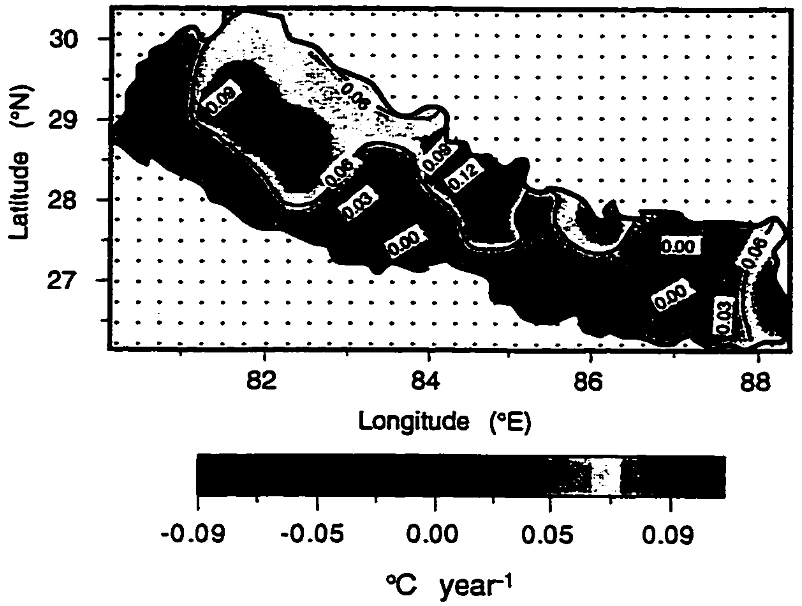


Figure II.4a. Spatial distributions of annual mean temperature trends for the period 1977 to 1994

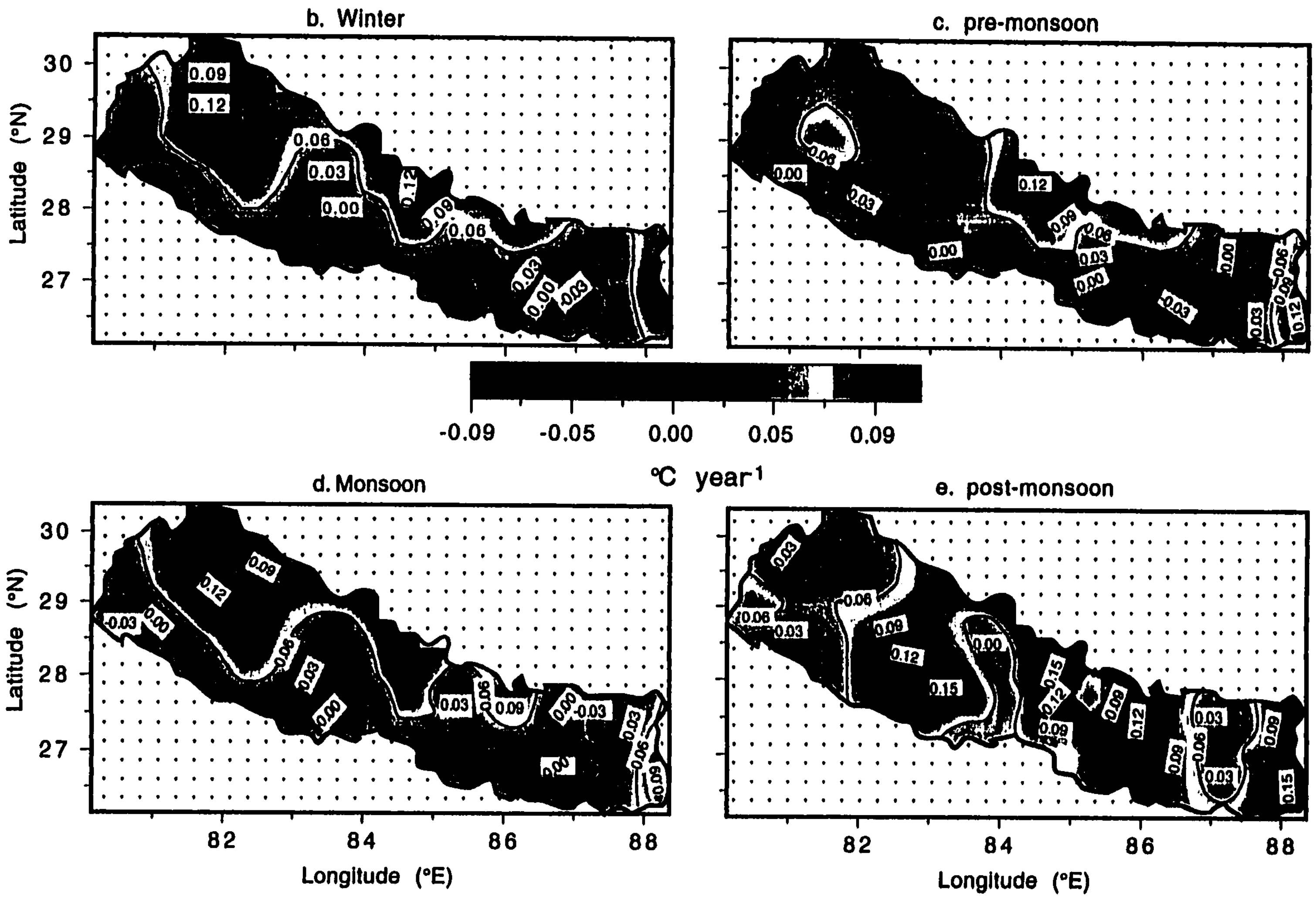


Figure II.4b-c. Spatial distributions of mean temperature trends for the period 1977 to 1994 for: (b) Winter; (c) pre-monsoon; (d) monsoon; and (e) post-monsoon seasons.

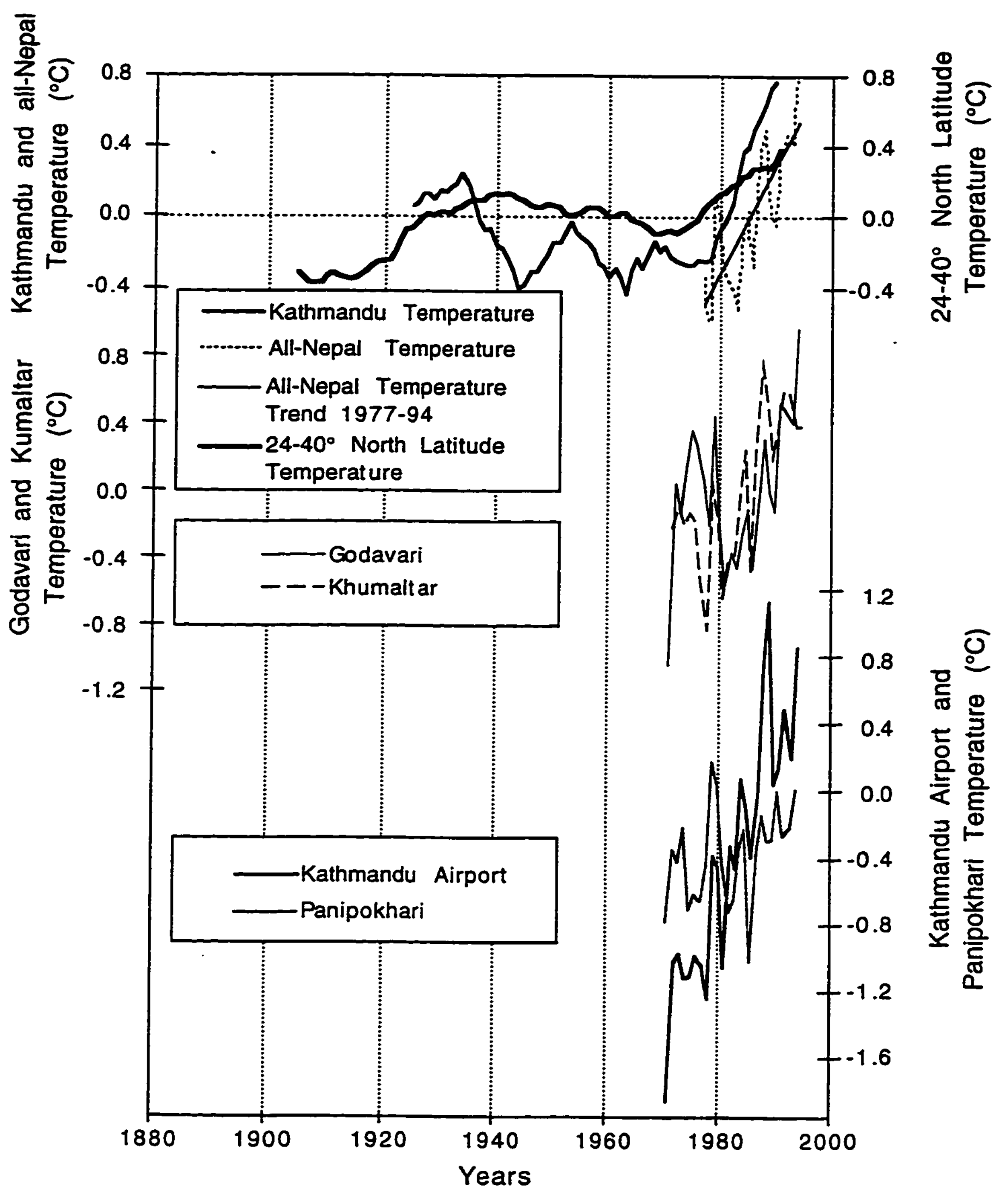


Figure II.5.Comparison between 10-year running means of Kathmandu record and 24-40° latitude temperature. For comparision all-Nepal annual temperature series and 1977-94 trend are also presented here. Also four Kathmandu valley stations' records, used to construct the 1978-94 part in the Kathmandu temperature record are included in this figure.

CHAPTER THREE

PRECIPITATION FLUCTUATIONS IN THE HIMALAYA AND ITS VICINITY AND RELATIONSHIP WITH SOME LARGE SCALE CLIMATOLOGICAL PARAMETERS.

This chapter has been submitted to the International Journal of Climatology for publication (Sept. 28, 1998)

III. PRECIPITATION FLUCTUATIONS IN THE HIMALAYA AND ITS VICINITY AND RELATIONSHIP WITH SOME LARGE SCALE CLIMATOLOGICAL PARAMETERS.

ABSTRACT

Precipitation records from 78 stations distributed across Nepal were analyzed and all-Nepal (1948-1994) and sub-regional records (1959-1994) were developed. The all-Nepal and regional precipitation series showed significant variability on annual and decadal time scales. Distinct long-term trends were not found in these precipitation records. The all-Nepal record agrees well with precipitation records from northern India and decadal scale variations in the eastern Himalaya series are consistent with the precipitation record from Lhasa. In contrast, precipitation in Nepal does not compare well with the all-India precipitation record.

The all-Nepal monsoon record is highly correlated with the Southern Oscillation Index (SOI) series and the decadal scale fluctuation in the all-Nepal record correlates well with the sunspot record with a lag of about four years. The exceptionally dry year in 1992 recorded in Nepal coincides with the elongated El Niño of 1992-93 and the Mt. Pinatubo eruption. A remarkable cooling in the region covering the Tibetan Plateau also occurred in 1992, suggesting a major role of Pinatubo aerosol in the drought of that particular year in Nepal. In other years, the correlation between precipitation record from Nepal and the temperature of the Tibetan Plateau is not significant, while there is a stronger correlation with temperature over the Indian Ocean and southern India. This provides further support for the dominant role of El Niño/Southern Oscillation on the precipitation fluctuation in Nepal. Strong relationships between precipitation in Nepal and SOI, sunspot record, temperature over the Tibetan Plateau, and temperature over the Indian Ocean/India suggest the influence of multiple large scale forcings on the precipitation fluctuation in the Himalaya. Further investigations may enable prediction of monsoon precipitation in the Himalaya and its vicinity using those parameters.

KEY WORDS: monsoon; El Niño; climate change; Nepal; south Asia; solar cycle.

INTRODUCTION

Surface air temperature is commonly used to study the state of climate systems and numerous studies have investigated trends in global, hemispheric, and zonal mean temperatures (e.g., Angell and Korshover, 1978; Jones et al., 1986a; Jones et al., 1986b; Jones et al., 1986c; Hansen and Lebedeff, 1987; Hansen and Lebedeff, 1988; Hansen et al., 1996). In addition, several model studies have tried to calculate changes in temperature due to increases in greenhouse gases (e.g., Mitchell et al., 1990; IPCC, 1996). However, while temperature is a good indicator of climate change, precipitation may be of equal or greater importance in terms of monitoring global change in low and mid-latitude regions due to their vulnerability to water shortages and water quality (Farmer and Kelly, 1989; Mirza and Dixit, 1997). Analyses of global precipitation variations reveal marked trends in recent decades. For example, rain and snowfall amounts over the middle and high latitudes rose steadily over the past decades, whereas a pronounced decreasing trend occurred in the sub-tropics (Bradley et al., 1987; Diaz et al., 1989). This trend in the sub-tropics is largely accounted for by droughts in parts of sub-Saharan and Sahelian Africa (Bradley et al., 1987; Follard et al., 1990).

South Asia is a region influenced by the summer monsoon and the agricultural subsistence of people in the region depends greatly on monsoon rains. While several studies on monsoon and annual precipitation in India have not identified any long term trends (Parthasarthy and Mooley, 1978; Hakkarinen and Landsberg, 1981; Mooley and

Parthasarthy, 1983; Parthasarthy et al., 1992), Kothyari and Singh (1996) suggest a slight decrease in precipitation over the past few decades.

The climate of South Asia is dominated by the monsoon circulation system. The summer monsoon dominates the climate from May to September and westerly circulation dominates from November to March. The influence of these two circulation systems is not distributed evenly over the Himalaya, with summer rainfall greater in the southeast and westerly derived winter precipitation greatest in the north-west (Nayava, 1980; Mani, 1981).

While abundant work has been done on monsoon and annual precipitation in India, there is only limited knowledge concerning precipitation variations in the foothills and mountains of Central Asia. Recently, we studied temperature trends over Nepal and found broad agreement with temperature trends in the Northern Hemisphere (NH) and the Tibetan Plateau, while Nepal temperature trends differ from trends found in the Indian plains (Chapter II).

In this paper, we analyze precipitation records from stations distributed across Nepal for the longest possible period, 1948-1994. Based on these records, for the first time we develop an all-Nepal (1948-1994) and, within Nepal, regional (1959-1994) precipitation series to study annual and seasonal precipitation changes. We compare and contrast these records with precipitation records from India and Lhasa, temperature records from neighboring regions to north and south of the Himalaya, as well as other records of large-scale climatological parameters.

DATA AND METHODS

Precipitation data were obtained from the Department of Hydrology and Meteorology, Nepal (DHM, 1966-97). Precipitation records from 89 stations with data beginning prior to 1976 and continuing until the present were examined. In 1948, the number of precipitation stations in Nepal was only 14, but by 1959 this number increased to 83. The year 1959 is also the earliest that provides adequate length of record (~40 years ; WMO, 1966a) and includes enough stations for a fairly widespread geographic coverage. These 83 stations have relatively few missing data (less than 3% of the total number of annual values). Due to great changes in elevation within relatively short distance, there are great differences in precipitation within relatively small aerial distances. Hence it is not possible to fill missing data solely by spatial interpolation (Hormann, 1994). Therefore, major portions of missing data sets were filled using temporal interpolation (monthly value as an average of the same month for a period between +2 and -2 years; WMO, 1966a).

The 83 stations selected were tested for homogeneity, normality, and the presence of persistence using rigorous statistical examinations. The homogeneity of the series was tested by Swed and Eisenhart's run test (run above and below median) (WMO, 1966a; WMO, 1966b). Only two stations failed the test as their numbers of runs were above the limit at the 5% significance level. The chi-square test was performed to test the normality of each station record (Crow et al., 1960). Only four stations showed frequency distributions other than Gaussian. To examine the presence of Markov type persistence in the time series, auto-correlation up to 10 lags (10 years) were examined. Only three stations showed significant auto-correlation. Altogether, 5 station records were discarded after these statistical tests, reducing the number of stations used for further investigations to 78 (Fig. III. 1).

Precipitation data used in this study are published as monthly precipitation values (DHM, 1966-97). From the monthly precipitation, annual precipitation values were calculated as the sum of January to December precipitation. Further, seasonal precipitation values were derived as: Winter (December of previous year to February); Pre-monsoon (March to May); Monsoon (June to September); and Post-monsoon (October and November).

All-Nepal precipitation series were developed using the method described in Hansen and Lebedeff (1988). The advantage of this method over simple area weighted averaging is the possibility of using a variable number of stations for different time-periods. In this method, initially a relatively long record is chosen. The second record (generally shorter) is linearly shifted by a value equal to the difference between the averages of the two records, for the overlapping period (sub-period). For that sub-period, the annual precipitation values of the first record are substituting by averages of the original first record and the transformed record for respective years. This process is repeated with the transformed first record and remaining records. The time series developed by this method can be used to analyze trends and variability; however, absolute precipitation values cannot be obtained.

The regional precipitation series were only derived for the period after 1959, when all 78 stations were operational. In this analysis, the country was divided into three regions: Terai and Siwalik; Middle Mountain; and Himalaya. These divisions are based primarily on the representation of each region by the maximum possible and uniformly distributed number of stations. It is fairly common to combine Terai and Siwalik regions as one climatic region (Chalise, 1986). Due to the narrow elongated nature of these three regions and the uneven zonal dominance of the summer monsoon and westerly circulation over these regions, they were further divided into western and eastern parts (Fig. III.1). For these six regions, precipitation series were developed by arithmetically averaging the station records. To remove the inter-annual variability and to investigate if trends are present in the record, the all-Nepal and 6 regional precipitation series were smoothed using a robust spline with 65% smoothing (Meeker et al., 1995). All precipitation series are standardized by subtracting the mean and dividing by the standard deviations.

To investigate if the all-Nepal record had periodicities common to large-scale climatological phenomena, the all-Nepal precipitation series was subjected to spectral analysis. The spectral analysis was performed by fast Fourier transform (FFT) using a split cosine window with 25% tapers on both sides but without any padding (Meeker et al., 1995).

RESULTS

All-Nepal precipitation series

The standardized all-Nepal monsoon precipitation series shows great interannual variability in the amount of precipitation (Fig. III.2a). The spline curve shows there is also a significant decadal scale variability with distinct peaks in 1962, 1973, and 1984.

Oscillatory characteristics, visible in the series, are supported by spectral analyses which show significant peaks (at 95% confidence level) at periods close to 2.5 and 11 years in both the all-Nepal annual and the monsoon precipitation series (Fig. III.3a,b). Similarly, the winter, pre-monsoon, and post-monsoon series also contain significant peaks at periods close to 2.5 years, although the 11-year peak is not present (figures not shown).

After 1990 the all-Nepal series shows a downward trend, the timing of which is consistent with the near 11-year periodicity, although the duration of the drop is longer than would be expected from the 11-year cycle alone. This is possibly due to end effects inherent in the smoothing technique. Note that 1992 is the driest year in the entire record (more than two standard deviations below normal).

Regional precipitation series

The 2 to 5 year oscillatory characteristics in the all-Nepal series are also visible in regional series (Fig. III.2b-g). Further, most of the regional series display oscillations similar to the 11-year oscillation in the all-Nepal series, although the phasing and amplitudes of these oscillations differ by region.

As in the spline of the all-Nepal record, all of the smoothed regional series show a drop in precipitation in the early-1990s and 1992 is markedly dry in all of the regional series.

DISCUSSION

Model studies suggest an increase (5-15%) in monsoon precipitation with greenhouse induced global temperature increases (e.g., Follard et al., 1990; Meehl, 1993; Meehl, 1996). This is consistent with the basic principle of the monsoon - increase in land-ocean thermal contrast due to global warming, resulting in intensification of monsoon circulation (e.g., Meehl, 1994). Preliminary model results on changes in precipitation in Nepal due to an increase in atmospheric greenhouse gases (GHG) also predict precipitation increases, especially in the western part of the country (per. comm., Dr. M.L. Shrestha, DHM), although model studies that include sulfate aerosol forcing predict monsoon precipitation to decrease (Lal et al., 1994; Lal et al., 1995; Mudur, 1995). However, there is no indication of an overall trend in precipitation in Nepal. An increasing trend is apparent between 1967 and 1990 in the all-Nepal and the three western regions' records, although the magnitude is low compared to the annual and decadal variability. The lack of a distinct long term trend in precipitation in Nepal may be the result of Nepal's location between two large emission sources in Asia, i.e., China and India (Foell, 1995; Arndt and Carmichael, 1998). Recent studies show that sulfate aerosol burden in the atmosphere in Nepal is largely of Indian origin (Arndt and Carmichael, 1998). It is likely that the increase in atmospheric sulfate aerosol has already begun affecting the monsoon in the Himalaya, offsetting the increasing trend in monsoon precipitation that would have been caused by the increase in atmospheric GHG alone.

Despite the lack of long term trends, the presence of characteristics common to all Nepal series and regional precipitation series indicates homogeneity in precipitation on annual and seasonal time scales, despite great orographic disparity. The presence of 2.5 year and 11 year periodicities in the all-Nepal annual and seasonal series suggests that the records could be related to large scale climatological parameters such as the quasi-biennial oscillation (QBO) and sunspot cycles, respectively. A direct comparison between the all Nepal precipitation record and the QBO record (equatorial wind speed at 50 mb; NOAA, Climate Prediction Center; http://nic.fb4.noaa.gov/data/cddb/) shows that the relationship is not consistent as the two records are in-phase during some period, while out of phase in other, suggesting that the QBO is not responsible for the 2.5 year periodicity in the all-Nepal records. The all-Nepal precipitation record was also compared with the Zürich sunspot number (NOAA, National Geophysical Data Center; http://www.ngdc.noaa.gov/stp/SOLAR/SSN/SSN.html). The 5 year running mean of all-Nepal data shows a good correlation with the sunspot record with a 3 to 5 year lag (Fig. III.4a). A positive correlation between monsoon precipitation and sunspot numbers can be explained by the overall increase of solar output with an increase in sunspot number (Hartmann, 1994), resulting in an increase in the land-ocean thermal contrast, which in turn enhances the intensity of the south-west monsoon. However, the rather long time lag between the peaks in the sunspot and in the smoothed all-Nepal records suggests an indirect link not possible to reveal based on available data.

Several studies on monsoon precipitation in India have discovered a close relationship between the monsoon and ENSO (e.g., Mooley and Parthasarathy, 1983; Khandekar and Neralla, 1984; Khandekar, 1991; Barnett, 1991). El Niño warm phases are associated with warmer sea surface temperature (SST) in the equatorial Pacific as well as in the Indian Ocean, resulting in decreased land-ocean thermal contrast, thus reducing the strength of the monsoon. A spectral analysis of the monthly southern oscillation index (SOI) record (NOAA, National Geophysical Data Center) for the period 1967-1994 (overlapping the length of the all-Nepal record) shows the presence of significant peaks at 2.5 and 4.7 years (Fig. III.3c). Furthermore, direct comparison between the all-Nepal precipitation series and the SOI shows strong agreement between these records, i.e., low all-Nepal precipitation during El Niño warm phase periods (low SOI)(Fig. III.4c). The all-Nepal monsoon precipitation also contains a periodicity close to the 4.7-year periodicity in SOI (i.e., 4.6 years), although significant only at 90% (Fig. III.3b), suggesting that the 2.5 year periodicity in the ENSO has a dominant affect on the all-Nepal monsoon record. The relationship between the all-Nepal record and SOI is much better than that between the all-India precipitation and SOI, especially for the period after 1970. The correlation coefficient between the all-Nepal monsoon record and annual averaged SOI for the period 1970-1994 is 0.65 ($N=24$; $p<0.05$).

A comparison between precipitation fluctuations over Nepal and over India does not show good agreement. Although a dry (wet) year in the all-India record (Sontakke et al., 1993; Indian Institute of Tropical Meteorology, Pune, India) is generally a dry (wet) year in the all-Nepal record, the magnitudes are considerably different (Fig. III.4c). The year 1992 was the driest in the all-Nepal as well as in most of the regional records (about 2 standard deviations below normal). In the same year, the all-India record precipitation was also below normal, but not as low as in Nepal (Fig. III.4c). On the other hand, the weighted mean precipitation from five northern Indian climatological divisions exhibit a comparable drop in precipitation from 1990 to 1992 (Fig. III.4c). Variations thereafter are also comparable to those in the all-Nepal record. This result asserts that the unique climate episode of 1991-92 prevailed in the northern part of the sub-continent, but was absent in the southern regions. We suggest that the precipitation climatology in the northern part of the sub-continent (including the Himalayan region) is different from the rest of the sub-continent and the precipitation record from India as a whole (and generally excluding the Himalayan region), is not always a suitable representation for the region.

The exceptionally dry year of 1992 coincides with two climatologically prominent incidents: an elongated warm phase El Niño of 1991-1993 and the Mt. Pinatubo eruption of June 1991. Both of these events could have negative impacts on monsoon precipitation

(Nicholls, 1983; Handler, 1986; Barnett et al., 1991; Meehl, 1994). Widespread cooling has been recorded due to the Mt. Pinatubo eruption (Hansen et al., 1996; WMO, 1995), although such cooling was not observed in Nepal (Chapter II). However, a considerable drop in temperature occurred in the region to the north of the Himalaya (Box 21 mean temperature record developed by Hansen et al. (1988; 1996)) including the Tibetan Plateau (Fig. III.5a). For the whole length of the record, there is a significant positive correlation between the all-Nepal precipitation and the Tibetan Plateau temperature records ($r^2=0.40$; $p=0.05$), whereas the correlation is not significant if the 1992 data is not included ($r^2=0.30$; $p>0.05$). A similar comparison with Box 36 (including India and the Indian Ocean) shows a better negative correlation ($r^2=0.45$; $p=0.01$; analysis includes 1992 data)(Fig. III.5b). We suggest the anti-correlation between India/Indian Ocean temperature and the all-Nepal precipitation is the manifestation of the link between El Niño events and the monsoon precipitation over the Himalaya. The year 1992 is unique in the sense that there are synchronous drops in the Tibetan Plateau temperature and all-Nepal precipitation records, while there is no reciprocal rise in the India/Indian Ocean temperature (Fig. III.5a,b). The decrease in Tibetan Plateau temperature for the year 1992 cannot be associated with the El Niño event as a decrease in land temperature (in Central Asia) due to El Niño events has not been observed so far. This result asserts that the Pinatubo aerosol is responsible for the temperature decrease over the Tibetan Plateau, which in turn resulted in the decrease in monsoon precipitation in Nepal in that particular year. We also investigated the potential influence of two other major volcanic eruptions within the period of precipitation records from Nepal: Agung (March 1963) and El Chichon (April 1982). The precipitation record from Nepal and the Tibetan Plateau temperature record do not show a drop in 1963 similar to that in 1992. This is most likely a result of significantly lower aerosol loading from the Agung eruption (McMormick et al., 1995), combined with the fact that the aerosol loading was concentrated more in the southern hemisphere (Sato et al., 1993). While the eruption of El Chichon coincides with a significant drop in precipitation in Nepal (and in India) and a slight drop in the Tibetan Plateau temperature record, 1982 also experienced a strong El Niño. Furthermore, the amount of stratospheric aerosol produced by the El Chichon eruption was much smaller (McCormick et al., 1995), suggesting that the 1982 El Niño event was more significant for precipitation in Nepal.

We also investigated the influence of Eurasian Snow Cover (ESC) (NOAA, Climate Prediction Center; http://nic.fb4.noaa.gov/data/cddb/) on monsoon precipitation in Nepal. Previous work identified an inverse relationship between all-Indian monsoon precipitation and ESC (e.g., Dey and Bhanu Kumar, 1982; Khandekar, 1991), although this relationship has recently been questioned (per. comm., C. Duncan, University of Massachusetts). A comparison of ESC and the all-Nepal precipitation record showed only a weak correlation (r=-0.01).

SUMMARY AND CONCLUSION

Precipitation data from Nepal over the past three decades show large inter-annual and decadal variability in the all-Nepal as well as regional (within Nepal) precipitation records. The lack of a long-term increasing trend in the precipitation records, despite the fact that climatic models predict an increase in monsoon precipitation due to greenhouse gas induced warming, could be an indication of the countering effects of the recent increases of atmospheric sulfate aerosol due to combustion of fossil fuels in Asia.

We found indications of an association between precipitation in Nepal with the sunspot cycle and ENSO events. Further, precipitation records from Nepal compare much better with the precipitation from the northern part of India compared to the aggregated record for all of India. This suggests that the precipitation climatology of the Himalaya and its vicinity behave differently from the southern part of the sub-continent and the all-India precipitation record does not provide a valid representation of the entire subcontinent.

The exceptionally dry year in 1992 coincided with two important climatic events: an elongated El Niño event and the eruption of Mt. Pinatubo. A drop in the Tibetan Plateau temperature record, similar to that in the all-Nepal precipitation record for 1992, supports a strong role for the Pinatubo aerosol in the observed dry event. However, over the whole period of record, a strong anti-correlation between the all-Nepal precipitation record and temperature over the Indian Ocean and India, the lack of significant correlation between the all-Nepal precipitation record and the temperature record from the Tibetan Plateau, and lack of significant anti-correlation between the Eurasian snow cover and the all-Nepal precipitation record supports the dominating affect of ENSO events on the precipitation over the Himalaya. Relationship between precipitation in Nepal and sunspot numbers, ENSO and tropical volcanic events suggests the possibility of using these to predict precipitation in the Himalaya and adjoining regions in the future.

ACKNOWLEDGMENTS

We thank Dr. R.W. Talbot, Earth Sciences Department, Dr. B.D. Keim, Department of Geography, University of New Hampshire, Mr. P.J. Krusic, Columbia University, and, Dr. S.P. Adhikary, Himalayan Climate Center, Nepal for detailed review of this paper. Thanks are extended to the Central Data Processing Unit, Department of Hydrology and Meteorology, Nepal for providing some data in digital format. START Secretariat provided partial support to conduct this study.

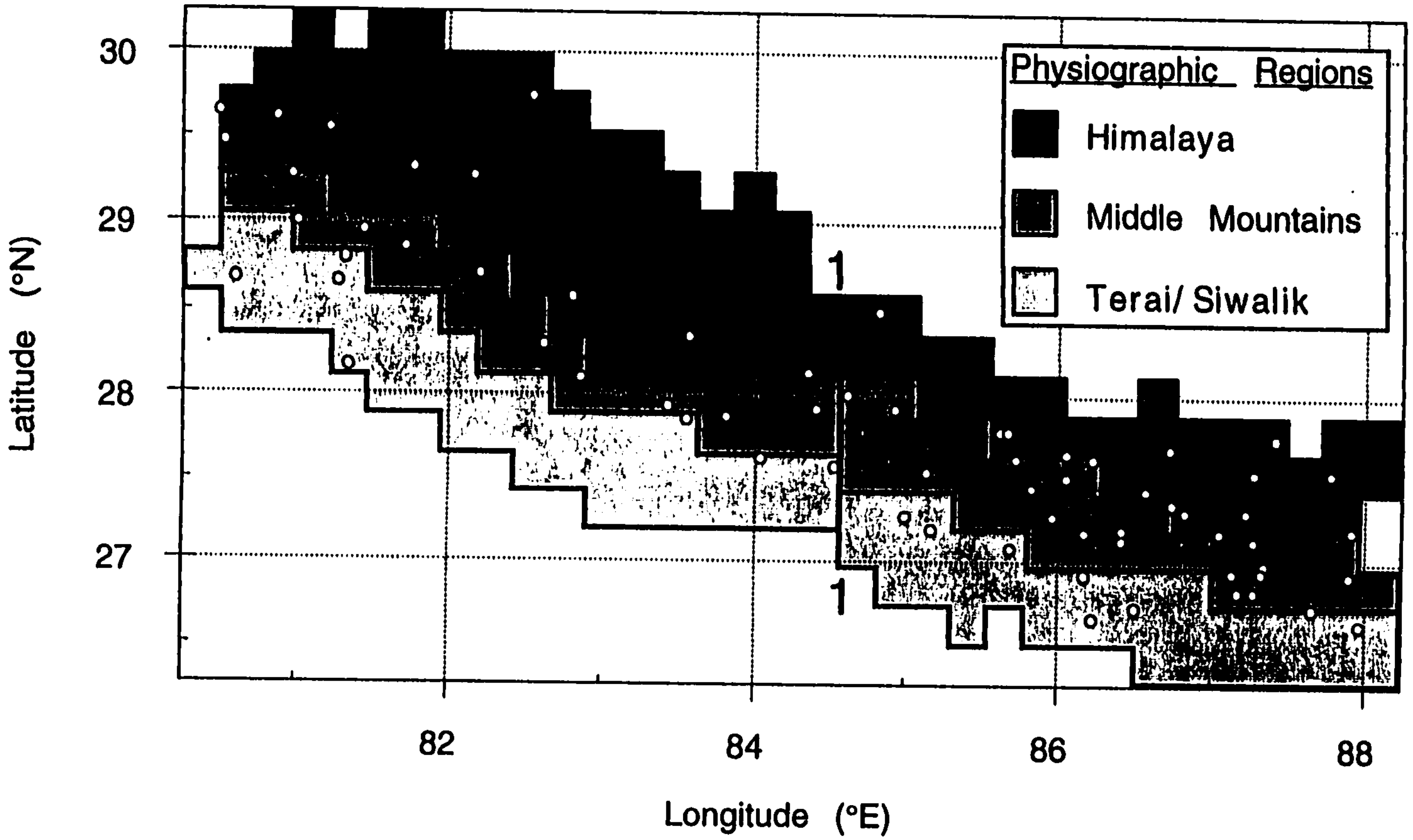


Figure III.1. Station location map and delineation of six sub-regions. The vertical line 1-1 separates eastern and western sub-regions. Open circles identify station locations for precipitation records used in this study.

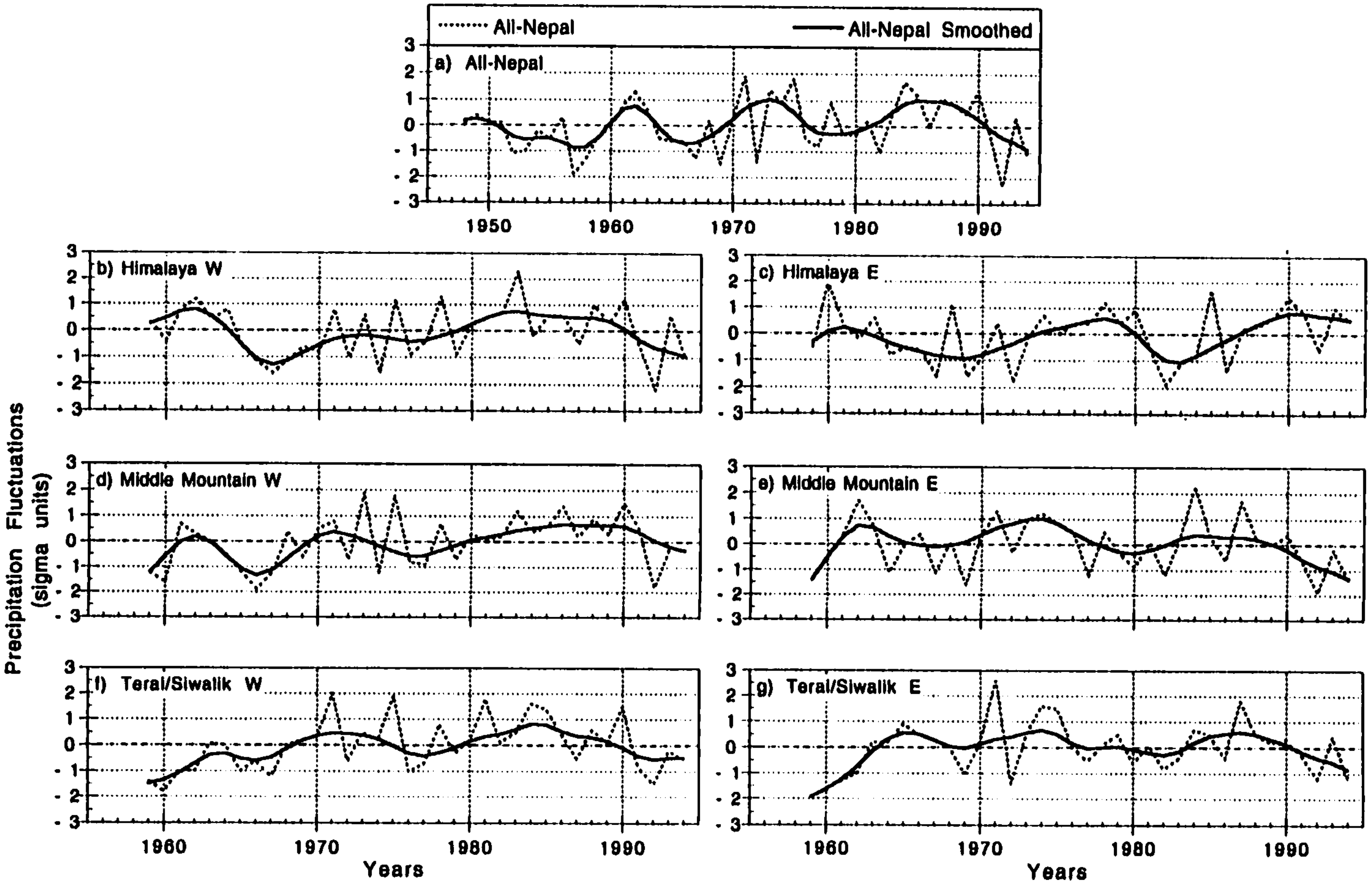


Figure III.2. Standardized monsoon precipitation series (dotted lines) and their spline curves (solid lines)for: a. all-Nepal; b. Himalaya-West; c. Himalaya-East; d. Middle-Mountain-West; e. Middle-Mountain-East; f. Terai/Siwalik-West; and g. Terai/Siwalik-East sub-regions.

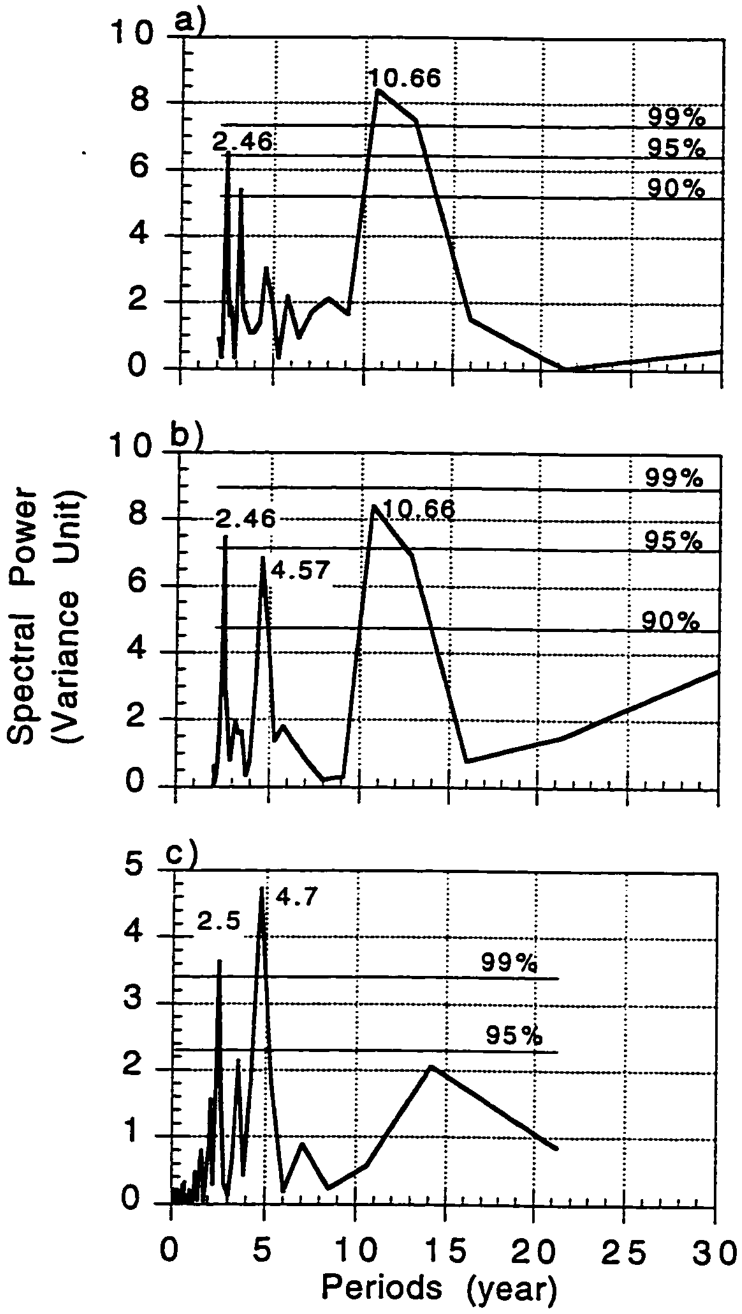


Figure III.3. Power spectra of all-Nepal precipitation series for: a. annual; b. monsoon season; and for c. SOI series.

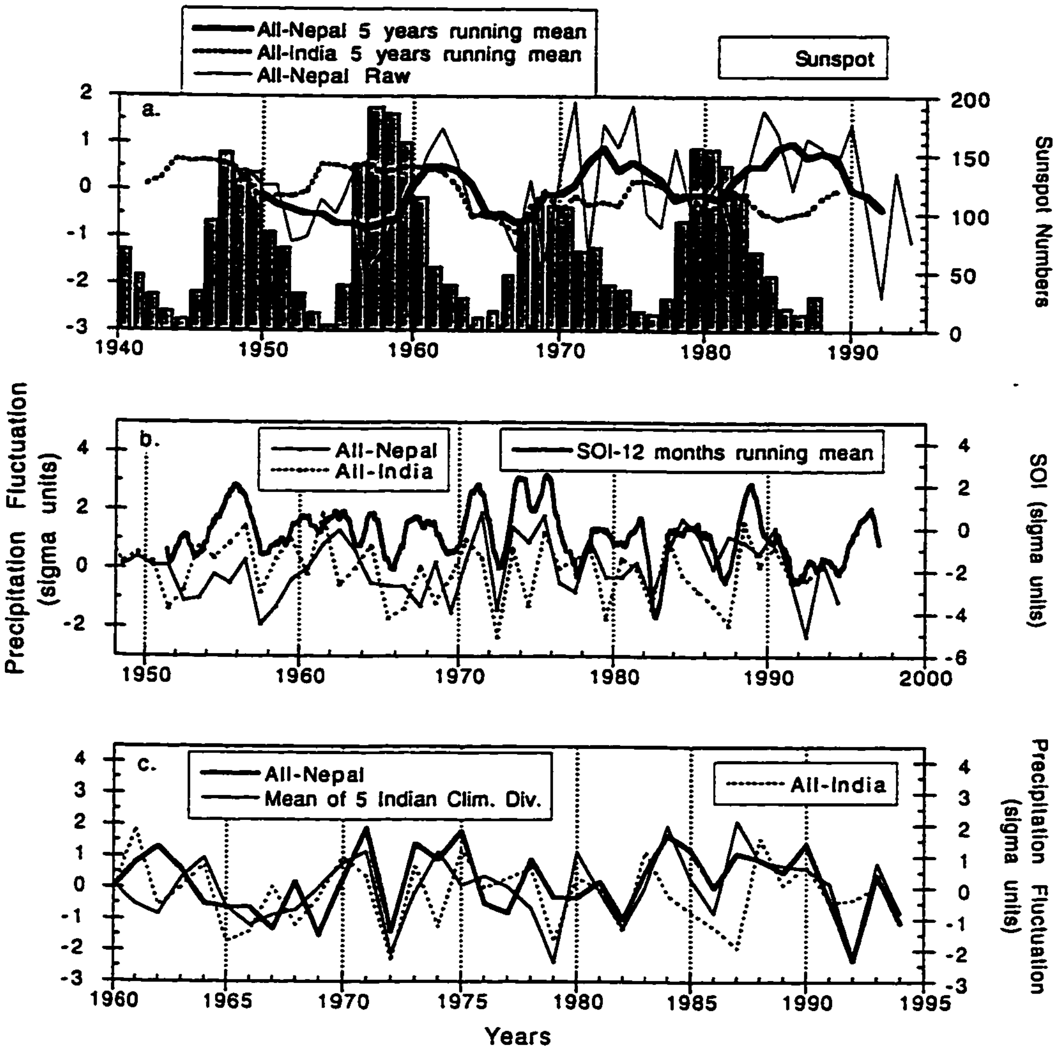


Figure III. 4. Comparison between: a. Zürich sunspot numbers, all-Nepal precipitation series (raw and 5 years running mean) and 5 year running mean of all-India precipitation series; b. all-Nepal precipitation, all-India precipitation, and SOI; c. all-Nepal precipitation, all-India precipitation, and weighted mean precipitation series of 5 northern Indian climatological divisions

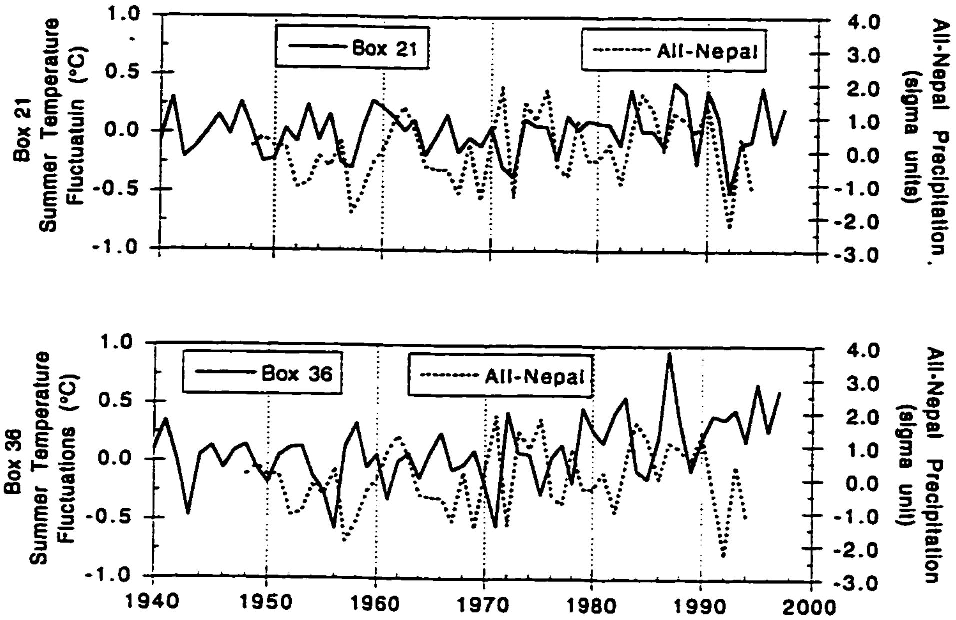


Figure III.5. Comparision of all-Nepal monsoon precipitation with Box 21 (top) and box 36 (bottom) temperatures. See text for explanations of box temperatures.

CHAPTER FOUR

CHEMICAL COMPOSITION OF AEROSOL AND SNOW IN THE HIGH HIMALAYA DURING THE SUMMER MONSOON SEASON

This chapter has been published in the Atmospheric Environment
(Vol. 31, No.17, pp. 2815-2826, 1997)

IV. CHEMICAL COMPOSITION OF AEROSOL AND SNOW IN THE HIGH HIMALAYA DURING THE SUMMER MONSOON SEASON

ABSTRACT

Aerosol and surface snow samples were collected from Hidden Valley in the Dhaulagiri region of western Nepal during the summer monsoon of 1994. Temporal variations of major ion (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-}) concentrations in the aerosol samples are clearly related to the influx of monsoon air masses. Snow was enriched in NH_4^+, and NO_3^-, while Na^+/Cl^- ratios were lower in the snow compared to the aerosol. A large part of this is explained by the difference in the air masses represented by aerosol and snow chemistry. Snow chemistry in general represented stronger southerly monsoon circulation, which resulted in precipitation events in Hidden Valley, whereas aerosol chemistry represented weaker monsoon or local circulation as the sampling was not conducted during rainy and foggy weather. Enrichment of NH_4^+ and NO_3^- in snow is attributed to their biogenic and agricultural sources from villages to the south and east of Hidden Valley. In addition, scavenging of HNO_3 present in the air could also have contributed to the enrichment of NO_3^- in the snow. A lower Na^+/Cl^- ratio in snow is attributed to scavenging of HCl present locally and/or due to less fractionation of monsoon air masses during more intense circulation and shorter travel time. The observed differences in the chemistry of the two media due to the influence of monsoon verses local air masses supports the concept of using glaciochemical records from that region to interpret monsoon variations in the past.

Although the aerosol samples show excess cations, our data suggests the presence of acidic gases in the air locally. The overall major soluble ion concentrations of the aerosol

are comparable or lower than those measured at several other remote tropospheric sites. Our results further support the concept that high elevation mountain sites in the Himalaya can be used to investigate the composition and the evolution of the remote continental troposphere.

INTRODUCTION

Investigation of aerosol and precipitation chemistry in the Himalayas to date has been limited in both temporal and spatial scales, mainly due to difficult logistics. However, as the Himalayas are far removed from highly industrialized centers they provide suitable locations to monitor the chemistry of the remote troposphere and to study the evolution of atmospheric composition (e.g. Cunningham and Zoller, 1981). A limited number of studies conducted in the past, including short-term aerosol chemistry investigations on the southern slopes of Nepal Himalaya (Ikegami et al., 1978; Davidson, 1981; Wake et al., 1994), in north-western India (Kapoor and Paul, 1980), as well as surface snow studies (Mayewski et al., 1983; Wake, 1989), have indeed shown that the concentrations of pollution related species such as nitrate and sulfate are comparable to, or lower than, those measured in several other remote regions of the world.

Glaciochemical records have proven to be an excellent tool to investigate the atmospheric composition of the past (e.g. Mayewski et al., 1983; Mayewski et al., 1990; Wake et al., 1990). The Himalayan region contains a plethora of sub-tropical high elevation glaciers from which ice cores records spanning decades to several centuries of atmospheric chemistry variations could be collected (e.g. Mayewski et al., 1984; Lyons et al., 1991; Wake and Mayewski, 1993; Wake et al., 1993). Due to its high elevation, the Himalayan range acts as a boundary limiting the northern extent of the Indian summer monsoon, and therefore ice core records from such sites could provide insight into the variations of the monsoon in the past. However, understanding of the chemical composition of the air, its temporal and spatial variability, and the relationships between the composition of snow and the air it forms in are essential for an improved interpretation of glaciochemical records. The main purpose of this study was to obtain an improved perception of aerosol composition in the high Himalayan region and investigate relationships between aerosol and snow chemistry. A secondary goal was investigating the influence of atmospheric circulation patterns on aerosol and snow chemistry. Hidden Valley in western Nepal was selected as the site for the study of aerosol and snow chemistry mainly because it is located to the north of main Himalayan range, almost at the edge of monsoon influence. Since only stronger monsoon circulation can overcome the orographic barrier and reach the site, relatively small variations in monsoon circulation could have significant impact on the chemistry of local atmosphere.

Aerosol samples were collected at 5050 m in Hidden Valley, during the summer monsoon season of 1994. These samples were analyzed for the water soluble major inorganic ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, SO_4^{2-}). Similarly, fresh and surface snow samples collected from different locations in the valley were also analyzed for major ions. Although sampling only covered 13 days, this study is the most detailed of its kind in this remote region.

THE SITE

The site selected for this study is located at 28°50' N latitude and 83°35' E longitude in Hidden Valley, immediately north of main crest of the Himalaya in western Nepal (Fig. IV.1). The elevation of the valley floor is 5050 m a.s.l. Hidden Valley is surrounded by mountains ranging from 6000 m to over 8000 m a.s.l. including Dhaulagiri (8167 m). Hidden Valley possesses unique physiographical and climatological characteristics different from the rest of the country, mainly because the main crest of Himalayan range restricts the entry of the summer monsoon into these areas. The main pathways for monsoon moisture to Hidden Valley are Dhampus pass in the south-east and French pass in the south of the valley (Shrestha et al., 1976). The valley floor is covered by light vegetation, whereas the moderately sloped mountain sides are not vegetated and are severely weathered.

EXPERIMENTAL

Aerosol Samples

Aerosols samples were collected over a 13 day period (from 14 to the 27 of August, 1994) on 2 µm pore size, 90 mm diameter Zefluor™ Teflon filters (Gelman Sciences) mounted in a protective polyethylene cover.

The air was drawn by a 24 V high-volume pump powered by a combination of photovoltaic cells and batteries. The volume of air sampled was measured by an in-line flow meter. Corrections for ambient temperature and pressure allowed conversion of the measured volumes to standard cubic meters (scm). The mean flow rate was 4.35 scm hr^{-1} and the mean volume of samples was 41.2 scm. Zefluor™ filters have been found to have minimum positive artifact with regard to sorption of gaseous species like HNO_3 and SO_2 (Spicer and Schumacher, 1979; Appel et al., 1984). In contrast there might have been some evaporation of NO_3^- due to extended sampling, therefore the NO_3^- values reported here should be regarded as lower limit of true airborne concentration. For the mean flow rate of 4.35 scm hr^{-1} the velocity at the face of the filter was sufficiently high such that the efficiency of collection for particles as small as 0.3 µm is greater than 99% (Liu et al., 1984). Aerosol samples were collected in filters facing downwards, placed in a cylindrical protective cover. The flow velocity at the opening of the protective cover was calculated to be 0.04 m sec^{-1}, according to which the cut-off for large particle, as given by sedimentation velocity of particles is estimated at about 10 µm (Davidson, 1987; Warneck, 1988). Filters were changed twice daily representing approximately daytime and night time samples, but samples were not collected during rainy/foggy periods.

Care was taken to minimize contamination both in the laboratory and in the field. The filter cartridges were loaded in a Class 100 clean lab, packed and transported in clean plastic bags. After sampling the filters, still in their cartridges, were returned to the original clean plastic bags. Loading and unloading of samples was carried out wearing a non-particulating clean suit, hood, face mask and plastic gloves. Blank filters were handled in the same manner as the samples.

In order to analyze the major ion concentration the sample and blank filters were wetted with 0.5 ml ultra pure methanol. The soluble components were then extracted with three 5 ml aliquots of deionized Milli-Q water. Major ion concentrations in the aqueous extracts were determined by ion chromatography using Dionex model 4000 ion chromatograph. Cations were analyzed using a CS12 column, 22 mM MSA eluent and CSRS suppresser and anions were analyzed using an AS11 column, 6.6 mM NaOH eluent and MMS suppresser. The sample loop was 250 μl and the sample running time was approximately 9 minutes.

Five field blanks and two lab blanks were collected. We define the detection limit for major inorganic ions as the standard deviation of all the blanks divided by the mean volume of all the samples (after Talbot et al. 1986). The detection limits for aerosol species were (in neq scm^{-1}): Na^+ (0.143), NH_4^+ (0.028), K^+ (0.033), Mg^{2+} (0.003), Ca^{2+} (0.018), Cl^- (0.048), NO_3^- (0.025), SO_4^{2-} (0.035). Blank values were variable, especially for Na^+, K^+ and Cl^-. Mean blank values were subtracted from the sample concentrations, which resulted in below detection limits (bd) values for Na^+, K^+ and Cl^- in some of the samples.

The uncertainty in aerosol major inorganic ion concentrations associated with variability in the blank values and precision of ion chromatograph analysis were determined based on the propagation of errors (Miller and Miller, 1988). The overall mean uncertainty for samples were (in neq scm^{-1}): Na^+ (0.153), NH_4^+ (0.048), K^+ (0.034), Mg^{2+} (0.004), Ca^{2+} (0.038), Cl^- (0.050), NO_3^- (0.065), SO_4^{2-} (0.080).

Snow Samples

Surface snow samples were collected in Hidden Valley during the summer monsoon of 1994. Since post-depositional processes might alter the composition of snow chemistry, only fresh surface snow samples (28 samples) collected in different parts of the valley during the period of aerosol sampling are included in this study. All snow samples were collected within 12 hours of deposition. Table IV.2 and Figure IV.1 provide information on the site, date, and number of snow samples collected. Samples were collected using pre-cleaned plastic scrapers and were placed in sealed plastic 'Whirl-Pak' bags for melting. Snow samples were allowed to melt at the base camp and were poured into laboratory cleaned vials. Care was taken during sample collection and transfers to avoid outside contamination. Samples were kept frozen in the field using a gas powered freezer, but thawed during transport and were refrozen only when they arrived in New Hampshire. The samples were analyzed for major inorganic ions using Dionex model 2010i ion chromatograph.

Blanks were prepared by filling Whirl-Pak bags from the same lot as those used for samples, which had been transported to the site and back, with 50 ml of Milli-Q water. They were sealed and agitated for about 15 seconds. The bags were then cut open by a razor and Milli-Q water was transferred to laboratory cleaned 40 ml vials.

The blank values were variable for Na^+ and NH_4^+, but were relatively constant for other major inorganic ions. Detection limits of snow samples were taken as one standard deviation of all blanks. The detection limits of species in snow were in (µeq kg^{-1}): Na^+ (0.139), NH_4^+ (0.198), K^+ (0.054), Mg^{2+} (0.017), Ca^{2+} (0.040), Cl^- (0.062), NO_3^- (0.006, SO_4^{2-} (0.021).

Meteorological Data

A temporary meteorological station was established in the valley at an elevation of 5050 m a.s.l. Temperature and relative humidity were measured continuously by a thermohygraph, which had been set up in a Stevenson screen. Precipitation was measured by a Hellmann type rain gauge daily at 8:00 AM. Barometric pressure was measured every 4 hours using an altimeter-barometer.

RESULTS

Water soluble inorganic ions in the aerosol

Concentrations of water soluble inorganic ions in the aerosol (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^- and SO_4^{2-}) in neq scm^{-1} are presented in Table IV.2. Although several of the samples showed Na^+, K^+, and Cl^- values below detection limits, all the values are presented to show general trend within the sampling period. The average total aerosol load (total anion + total cation) was 7.36 neq scm^{-1}. SO_4^{2-}, NH_4^+, and Ca^{2+} are the dominant ions with concentrations above 1 neq scm^{-1} and account on average for more than 80% of the total ion burden. The sum of cations generally exceeded the sum of anions in our samples. The ion-budget was in the range of -0.32 to +1.54 with a mean value of +0.75 (negative sign indicates excess anion and positive sign indicates excess cation). Note that H^+, CO_3^{2-} and HCO_3^- were not measured in our samples.

So far only one study conducted in the Nepal Himalaya reported the same suite of chemical species as this study. Figure IV.2 compares the average concentrations of major inorganic ions in the aerosol in our samples with samples from Nguzompa Glacier, Khumbu Himal, eastern Nepal (Wake et al., 1994). The concentrations of aerosol related to secondary aerosol such as SO_4^{2-} in Hidden Valley are comparable to those in Nguzompa, whereas NO_3^- and NH_4^+ are even lower in Hidden Valley. Wake et al. (1994) pointed out that NO_3^- and SO_4^{2-}concentrations in Nguzompa samples were comparable to concentrations measured in the remote free troposphere. On the other hand, Na^+, Cl^- and Ca^{2+} are higher in Hidden Valley.

All the water soluble inorganic ions, in general, show similar temporal variation. Nevertheless there are certain differences, which facilitates dividing them into three groups: (1) Na^+ and Cl^-, (2) Ca^{2+}, Mg^{2+} and NO_3^-, and (3) SO_4^{2-} and NH_4^+, presented in Figure IV.3a, b, and c respectively. For comparison, precipitation and pressure anomalies are presented in Figure IV.3d. The concentrations of all the ions are low in the beginning of the sampling period. Due to large detection limits inter-sample variability in the first group (Na^+ and Cl^-) cannot be resolved with much certainty, nevertheless, general trend in concentrations over the sampling period is obvious. For example, peaks on August 20 and 22 are obvious in Figure IV.3a. The temporal variations in the second group (Ca^{2+}, Mg^{2+} and NO_3^-) show three peaks. An abrupt increase occurred on 16/17 August, after which the concentrations gradually decreased, but peaked again on August 20. The third peak occurred around 23 August. Species in the third group (SO_4^{2-} and NH_4^+) display temporal variations similar to those for species in the second group, although they lack the peak on 20. In this group the 23 August peak was greatly subordinate to the one on 16-17 August (Fig. IV.3c). The temporal distribution of K^+ is somewhat different from others and follows partly Na^+ and Cl^- (after August 20), and partly the second group (before August 20). Concentrations of all the ions were low at the end of the sampling period (after August 25).

Linear regression on all water soluble species in the aerosol showed that Na^+ and Cl^- are highly correlated (r=0.91). Similarly, Ca^{2+} correlates well with Mg^{2+} (r=0.80), while SO_4^{2-} is highly correlated with NH_4^+ (r=0.99). In addition, Ca^{2+} correlates well with NO_3^- (r=0.90) and SO_4^{2-} (r=0.74). All correlation coefficients are significant at p=0.01. Correlation coefficients between all species are shown in Table IV.3.

During the first half of the sampling period, the temporal variations in the aerosol ion concentrations show a relationship with meteorological conditions, especially precipitation. The concentration levels were low during the precipitation of 14 to 17 August, and sharply increased as the intensity of precipitation diminished. However, the scenario is different during the second precipitation event of 23-27 August as concentrations of the aerosol species, especially Ca^{2+} remained moderately high for the first two days of precipitation. By the night of 24, all species showed low concentrations.

Despite the high correlation between Na^+ and Cl^-, there is a deficiency in Cl^- in the aerosol compared to sea-water Na^+/Cl^- ratio (Table IV.2). Cl^--deficit shows an inverse relationship with SO_4^{2-} in our samples (with the exception of the August 22 sample). This relationship improves when the amount of SO_4^{2-} necessary to fully neutralize NH_4^+ is deducted from the total SO_4^{2-} (Fig. IV.4). The correlation coefficient between Cl^--deficit and SO_4^{2-} minus NH_4^+ is -0.56 (excluding 22 Aug. sample), which is statistically significant ($p=0.05$).

The 22 August sample shows the highest Cl^--deficit (-0.71, Fig. IV.4), but also the highest Na^+ and second highest Cl^- concentrations of all the samples (Table IV.2). The high value of Na^+ in this sample, responsible for the high Cl^- deficit (negative excess), is difficult to explain, as the Na^+ was not likely of crustal origin since concentrations of all other crustal species were low in this sample.

Another interesting result is the striking inverse relationship between Ca^{2+}/SO_4^{2-} ratio and NH_4^+/SO_4^{2-} ratio in our samples (Fig. IV.5). The correlation between these ratios is 0.75 ($p=0.01$). For comparison $(Ca^{2+}+NH_4^+)/SO_4^{2-}$ ratio is also plotted in Figure IV.5. Seven samples show the ratio greater than one. The same number of samples have the ratio less than one, while three samples show the ratio close to unity.

It has been suggested that acidic aerosols can be contaminated by ammonia in ambient air while processing filter in laboratory (Hayes et al., 1980; Silvente and Legrand, 1993). The ammonium concentrations can be suspect if the high correlation between SO_4^{2-} and NH_4^+ is due to laboratory contamination of our samples. It is unlikely that such reaction took place in our samples, as SO_4^{2-} is not fully neutralized in any of our samples, since NH_4^+ to SO_4^{2-} equivalence ratios are less than 1 (Table IV.3). Furthermore, the study of Hayes et al. (1980) included highly acidic stratospheric aerosol samples, whereas our samples are generally basic or only slightly acidic.

Snow Chemistry

Samples of surface snow chemistry collected during the aerosol sampling program were compared with aerosol chemistry. Although the number of snow samples collected within the period of aerosol sampling is limited, they facilitate comparison of overall trends in these two media. Snow samples were not collected on August 23 and 24, the last two days of the four-day precipitation event.

It has to be noted that snow samples included in this study were collected from several different locations in the valley. It is therefore possible that the temporal variations observed in the snow chemistry could be largely due to variations between the sampling locations. In addition the variability within the events was large. Logistic considerations resulted in the collection of fresh snow at more than one location only for the 23 August event. For this one case samples were collected from one site in Rika Samba glacier and at North Glacier (Table IV.1). The Student's t-test shows that for that event the mean concentrations of all soluble ions were statistically not different ($p=0.05$) at the two sites. Assuming this result is valid for other events as well, we suggest that the variability in snow chemistry is dominated by temporal variability rather than variability between sites.

Crustal species, SO_4^{2-}, and to some extent sea salt species, in the snow show temporal variations in general similar to those in the aerosol (Fig. IV.6). Other species on the other hand do not show such similarity. The most remarkable difference is observed in the temporal variations of NH_4^+ and NO_3^- in the aerosol and in the snow (Fig. IV.6). In addition, unlike in the aerosol, NH_4^+ in the snow is not correlated with SO_4^{2-}. The NH_4^+/SO_4^{2-} ratio in the aerosol is never higher than 1, whereas this ratio in snow in all with the exception of one event, is always higher than 1 and even exceeds 25 on 21 August

(Fig. IV.7a). On the other hand the ratio $NH_4^+/(SO_4^{2-}+NO_3^-)$ in snow is much lower, ranging from 0.5 to 1.5.

Similarly, temporal patterns of ratios of Na^+ and Cl^- also display conspicuous contrasts in the two media. The Na^+/Cl^- ratio is much lower in snow compared to aerosol (Fig. IV.7b). The Na^+/Cl^- ratio in the aerosol ranges from 1.25 to 7.24 (i.e. Cl- deficit) whereas, the ratio in the snow ranges from 0.41 to 1.16 indicating Cl^- excess in most (5 out of 6) samples.

DISCUSSION

Temporal Variations in the Aerosol and its relationship with Meteorological Condition

Wet deposition is an effective mechanism for the removal of aerosols, especially in the size range of 0.1 to 10 µm (Barrie, 1985; Warneck, 1988; McGann and Jennings, 1991). It is therefore expected that low concentration of atmospheric constituents will occur during and following precipitation events. This simple relation is not always observed in our study. During the 14-16 August precipitation event, a high amount of precipitation fell on the first day but diminished rapidly in the following days (Fig. IV.3d). In contrast, during the 23-27 August event precipitation was maintained at a moderate rate for 4 days. Study of upper air synoptic maps at 500 mb level shows that the precipitation of August 14-16 and 23-27 had a similar cause: formation of high pressure cell over Bay of Bengal. The difference between these two events was that the high pressure cell in the first event was of higher magnitude (592 mb) but lasted for only short period, whereas in the second event the magnitude was not as high (~588 mb) although it lasted for longer period.

The patterns shown by synoptic maps are consistent with the results of the atmospheric pressure measurements made in Hidden Valley (Fig. IV.3d). The first precipitation event coincides with sharp but short drop in pressure, whereas pressure pattern during the second event shows more gradual but long lasting decrease. Consequently precipitation in the first event was of high intensity in the first day, which efficiently scavenged the atmospheric constituents. The circulation in the following days (August 15, 16, and 17) was not as strong, the precipitation was also low, which possibly was not efficient to scavenge atmospheric constituents. Hence the levels of most species increased considerably during those days. The circulation in the second event was of moderate strength resulting in moderate intensity precipitation, which likely was not as effective in removing atmospheric constituents. In addition, the continued moderate circulation brought in atmospheric constituents in the following days. The dry period between event one and two was relatively free from monsoon circulation and more dominated by local circulation, therefore concentrations of sea salt species were low and concentrations of crustal species were high. K^+ is better correlated with Mg^{2+} and Ca^{2+} during this period. Conversely during periods of stronger monsoonal influence such as after August 20, K^+ correlates better with sea salt species. Although airborne concentration depends on many processes including upwind chemical processes and variability in source regions, our results suggest an important role of circulation system and precipitation intensity.

Relationship between Snow and Aerosol Chemistry

It is common to observe enrichment of SO_4^{2-} relative to NH_4^+ in the snow compared to the aerosol due to SO_2 oxidation in clouds (e.g. Calvert et al., 1985). On the contrary, our samples display huge enrichment of NH_4^+ relative to SO_4^{2-} in the snow. NH_4^+/SO_4^{2-} ratio shows that only a small fraction of NH_4^+ in snow is neutralized by SO_4^{2-} (Fig. IV.7a). Unlike the aerosol, a large fraction of NH_4^+ is balanced by NO_3^- in snow. Fractionation of NH_3 cannot explain such a large difference as the NH_4^+/SO_4^{2-} ratio in the air is always less than 1 (Table IV.2). The only plausible reason behind such a contrast between aerosol and snow chemistry is that different air masses are represented by the aerosol and snow samples. The aerosol sampling was not conducted during intensive precipitation and fog, therefore the filter samples were strongly biased towards air masses representing weaker monsoon circulation or local circulation. On the other hand, fresh snow chemistry reflects explicitly monsoon air mass with more southerly origin. The monsoon air mass, before entering Hidden Valley, travels over the low lying valley and mountain slopes dominated by cultivated land, forests, and vegetation. There are also several small villages, where animal husbandry and fertilization of fields with manure is a common practice. The high proportion of NH_4^+ in snow is therefore suggested to be due to enrichment of NH_3 while the air mass traveled over these areas. As there are no industrial activities within several tens of kilometers and firewood is main source of domestic energy, SO_4^{2-} enrichment is low compared to NH_3. As a result NH_3 in these air masses was not fully neutralized by SO_4^{2-}, it was therefore available to react with HNO_3 resulting in higher proportion of NO_3^- in snow. However, some of the samples show $NH_4^+/(SO_4^{2-}+NO_3^-)$ ratio less than 1 suggesting additional scavenging of HNO_3 in the air by snow.

Much lower Na^+/Cl^- ratio in the snow compared to the aerosol (Fig. IV.7b) could be due to two factors: scavenging of gaseous HCl present in the air, or relatively low fractionation of sea salt particle in the air mass represented by the snow. Since the snow chemistry represents stronger monsoon circulation with shorter travel time, it is possible that the fractionation of Cl^- with respect to Na^+ is less in such air masses compared to air masses representing weaker circulation. However, most of the snow events show excess-Cl^-, which cannot be achieved by this process alone. Therefore scavenging of HCl is possibly the major cause of the observed excess-Cl^- in snow.

The difference in NH_4^+ and the ratio of sea salt species in air and snow indicates that the sensitivity towards monsoon strength is well characterized by snow chemistry. Only stronger monsoonal circulation can overcome the large orographic barrier and reach the site, while weaker circulation cannot, so local circulation dominates during such periods. Such sensitivity towards monsoon strength suggest that the ice core chemistry can provide a strong record of monsoon variations in the past.

Water soluble inorganic ions in the aerosol and their sources

The concentration distributions during the measurement period were distinct for sea-salt aerosols (Na^+ and Cl^-), continental aerosols (Ca^{2+} and Mg^{2+}) and secondary aerosols (SO_4^{2-} and NH_4^+). High correlation between Na^+ and Cl^- suggests their common origin. Despite great distance from ocean Na^+ and Cl^- were likely of marine origin, mainly because there are no significant evaporite deposits in the surrounding regions. In addition Na^+ and Cl^- are the most conservative of all measured species and yield the greatest values of non-seasalt fractions when used as reference species. Crustal origin of these species can also be precluded due to their poor correlation with Ca^{2+} and Mg^{2+}. The deficiency in Cl^- (~30%) in the aerosol compared to $Na^+/Cl-$ ratio in seawater, and the inverse relationship between excess-Cl^- and SO_4^{2-} (Fig. IV.4), suggests that Cl^- was depleted due to reaction between sea salt particle and H_2SO_4 in the air (Hitchcock, 1980; Ohta and Okita, 1990). This result further suggests that the enrichment of Cl^- in the snow was partially due to scavenging of HCl in the air.

The fairly high correlation between crustal species and SO_4^{2-} and NO_3^- and the inverse relationship between Ca^{2+}/SO_4^{2-} and NH_4^+/SO_4^{2-} ratios (Fig. IV.5) can also be explained by the affinity of H_2SO_4 and HNO_3 gases to be absorbed on surfaces of mineral particles and reacting to form salt (Wolff, 1984; Mamane and Gottlieb 1989; Mamane and Gottlieb, 1992; Wu and Okada, 1994). The samples with $(Ca^{2+}+NH_4^+)/SO_4^{2-}$ ratio less than one suggest the existence of gas phase H_2SO_4 in air masses sampled. The H_2SO_4 in the ambient air not neutralized by NH_3 would be available to react with the surface of crustal particle, possibly $CaCO_3$ releasing CO_2. The covariance between crustal species and NO_3^- in our samples is also believed to be due to reaction between NO_2 and HNO_3 and crustal particle after all the SO_4^{2-} was neutralized.

The excellent correlation between NH_4^+ and SO_4^{2-} suggests that these species are present in the same particles. The mean value of NH_4^+/SO_4^{2-} equivalence ratio is 0.69 (Table IV.2), nearly in the middle of the range 0.5 and 1 corresponding to NH_4HSO_4 and $(NH_4)_2SO_4$ respectively. Sulfate, on the other hand, was also correlated with crustal species and Na^+, indicating its association with these species. We suggest that the NH_4^+ existed mainly as NH_4HSO_4, while SO_4^{2-} existed also in the form of Na_2SO_4 and $CaSO_4$

Earlier studies suggested that SO_4^{2-} in the Himalayan region did not represent anthropogenic emissions from more southerly location but reflected the influx of crustal species in the form of $CaSO_4$ (Mayewski et al., 1983). This study was conducted in the western Himalaya, where the influence of dust from Thar desert in north-west India is significantly greater compared to eastern Himalaya (Wake et al., 1993). In our samples from Hidden Valley evidence of reactions between acidic gases and sea salt, as well as crustal particles occurring locally, suggests the presence of acidic gases (although quantitatively low) such as H_2SO_4 , HNO_3, and HCl in the air despite excess cations observed in the aerosol in this part of Himalaya.Widespread biomass burning (firewood) for domestic energy in Nepal cannot be ignored as source of $NO_3^-$and SO_4^{2-} (Davidson et al., 1986). Nevertheless, since the sample site is seperated from the nearest settlement by several kilometers, long distance transport rather than local pollution is possibly their source.While gaseous H_2SO_4 has a very short lifetime against condensation onto existing aerosol or formation of new aerosol, SO_2 can enter aqueous aerosols or films and oxidize to SO_3^- and then H_2SO_4 .

The comparable or lower concentrations of chemical species in the aerosol in Hidden Valley with respect to Nguzompa glacier (Fig. IV.2) further supports the possibility of using high Himalayan sites to monitor the global remote troposphere. Relatively higher amount of sea salt species in Hidden Valley is likely due to marine influence on air mass during the monsoon season, although the air has traveled a considerable distance from marine environments to reach the site, while the samples from Ngozumpa glaciers were collected during the post-monsoon season. Higher concentrations of crustal species suggests local generation of dust. There are more local sources of dust in Hidden Valley, which has more weathered, moderately sloped terrain compared to steep unweathered rock faces above treeline or vegetation covered slopes below treeline in Ngozumpa. In addition Hidden Valley receives much less precipitation than Ngozumpa glacier.

CONCLUSIONS

Samples collected from Hidden Valley during the monsoon of 1994 show that short-term variation in monsoonal circulation was reflected in aerosol chemistry such that temporal variations in the concentrations of aerosol species were related to the pattern and intensity of atmospheric circulations. Differences between the temporal variations in chemical species in the air and in the snow is likely due to different air masses represented by these media. A higher proportion of NH_4^+ in snow indicates that snow chemistry reflects stronger monsoon circulation, advecting air masses rich in NH_3 of agricultural and biogenic origin in low lying areas in the vicinity of Hidden Valley. Aerosol sampling biased against periods of precipitation and fog conditions, and therefore, reflect weaker circulation and the dominance of local air masses. This finding suggests that isolated high Himalayan sites are sensitive towards monsoon strength and this sensitivity is translated to aerosol and snow chemistry. Glaciochemical records from these sites should therefore provide a valuable record of monsoon variations of the past.

Our study shows the presence of gaseous acidic species in air such as H_2SO_4, HNO_3 and HCl. The result of this study provides the most detailed data so far on soluble species in atmospheric aerosol in the high Himalaya during the summer monsoon season.

This data series is longer, and therefore adds confidence to the results of Wake et al. (1994), reinforcing the possibility of using high elevation sites in the Himalaya as a platform to investigate the chemistry of the remote troposphere.

The variations in the composition of snow and aerosols, besides changes in atmospheric circulation, depend on many processes within and before the sampling region. Therefore the concept of chemical variation due to changes in atmospheric circulation should be put to more rigorous test by extending the sampling to longer period covering different seasons and conducting synchronous sampling of air and snow media. Parallel sampling at stations in progressively lower elevation extending to the southern, lower elevation regions of the country will enhance the knowledge of variation in chemical composition of air masses as they move northwards.

ACKNOWLEDGMENTS

The authors thank David S. Averill for collecting the aerosol samples and providing the snow chemistry data, and Sallie Whitlow for analyzing the aerosol samples. Thanks are also due to Department of Hydrology and Meteorology, Nepal for providing meteorological instruments and synoptic maps. We also wish to thank Mr. Sher Bahadur for collecting the meteorological data and Mr. Dawa Nurbu for helping with logistical support.

Table IV.1. Site and number of snow samples included in this study

Date in August 1994	Sites	Number of Samples
14	Base Camp (BC)	1
16	Rika Samba II (RS II)	10
17	Rika Samba IV (RS IV)	2
21	Rika Samba III (RS III)	3
23	Rika Samba I (RS I)	5
	North Glacier (NG)	4
24	Rika Samba I (RS I)	3

Table IV.2. Water soluble ions in aerosol (neq scm^{-1}) at Hidden Valley measured between 14 and 27 August 1994.

Date	Cations					Anions			Ion			Excess-
	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	Balance[b]	NH_4^+/SO_4^2	Na^+/Cl^-	Cl^-
14	bd	0.90	0.02	0.41	0.79	bd	0.37	1.22	0.52	0.74	----	----
14/15[a]	bd	0.32	0.02	0.19	0.74	bd	0.15	0.69	0.43	0.46	----	----
15	ni	bd	0.07	0.61	0.57	0.24	0.10	0.49	0.43	----	----	----
16/17[a]	0.27	9.56	0.22	0.91	3.24	0.04	1.24	13.24	-0.32	0.72	6.65	-0.27
17/18[a]	0.27	7.64	0.19	0.55	2.42	0.05	0.73	8.96	1.31	0.85	5.01	-0.26
18	0.32	3.24	0.18	0.30	1.18	0.13	0.30	4.63	0.16	0.70	2.45	-0.24
18/19[a]	0.06	2.85	0.04	0.14	0.60	bd	0.21	3.57	-0.09	0.80		
19	0.48	3.36	0.18	0.27	1.05	0.14	0.43	4.42	0.35	0.76	3.35	-0.42
19/20[a]	0.20	2.74	0.13	0.26	1.18	0.08	0.28	3.82	0.34	0.72	2.52	-0.16
20	0.77	3.47	0.33	0.46	2.45	0.57	0.71	4.65	1.54	0.74	1.37	-0.34
21	0.28	0.68	0.05	0.11	0.55	0.22	0.14	0.83	0.48	0.83	1.25	-0.10
22	1.06	0.50	0.33	0.09	0.22	0.52	0.15	0.51	1.01	0.97	2.02	-0.71
23	0.50	1.01	0.11	0.46	2.64	0.34	1.52	1.93	0.92	0.52	1.47	-0.24
23/24[a]	bd	0.66	bd	0.28	1.29	bd	0.35	1.24	0.64	0.53		
24	0.37	1.28	0.15	0.51	2.27	0.19	1.34	2.29	0.76	0.56	1.98	-0.24
24/25[a]	0.08	0.07	bd	0.09	0.31	0.03	0.09	0.25	0.19	0.28	3.12	-0.07
25	bd	0.22	bd	0.19	0.25	bd	0.03	0.40	0.23	0.55	----	----
26	bd	bd	bd	bd	bd	bd	0.01	bd	-0.01	----	----	----
27	0.25	1.09	0.03	0.13	0.48	0.03	0.10	1.11	0.75	0.98	7.24	-0.26
Mean	0.38	2.33	0.14	0.33	1.24	0.20	0.44	3.01	0.51	0.69	3.20	-0.27
Min	0.06	0.07	0.02	0.09	0.22	0.03	0.01	0.25	-0.32	0.28	1.25	-0.71
Max	1.06	9.56	0.33	0.91	3.24	0.57	1.52	13.24	1.54	0.98	7.24	-0.07
Std. Dev.	0.28	2.66	0.10	0.22	0.94	0.18	0.46	3.40	0.47	0.18	2.04	0.17

Note. Here, blank rows indicate gaps in sample collection due to rainy/foggy weather condition.

[a] Indicates night time samples.

[b] represents sum of cations minus sum of anions, bd stands for samples below detection limit, and ni indicates samples not included due to contamination.

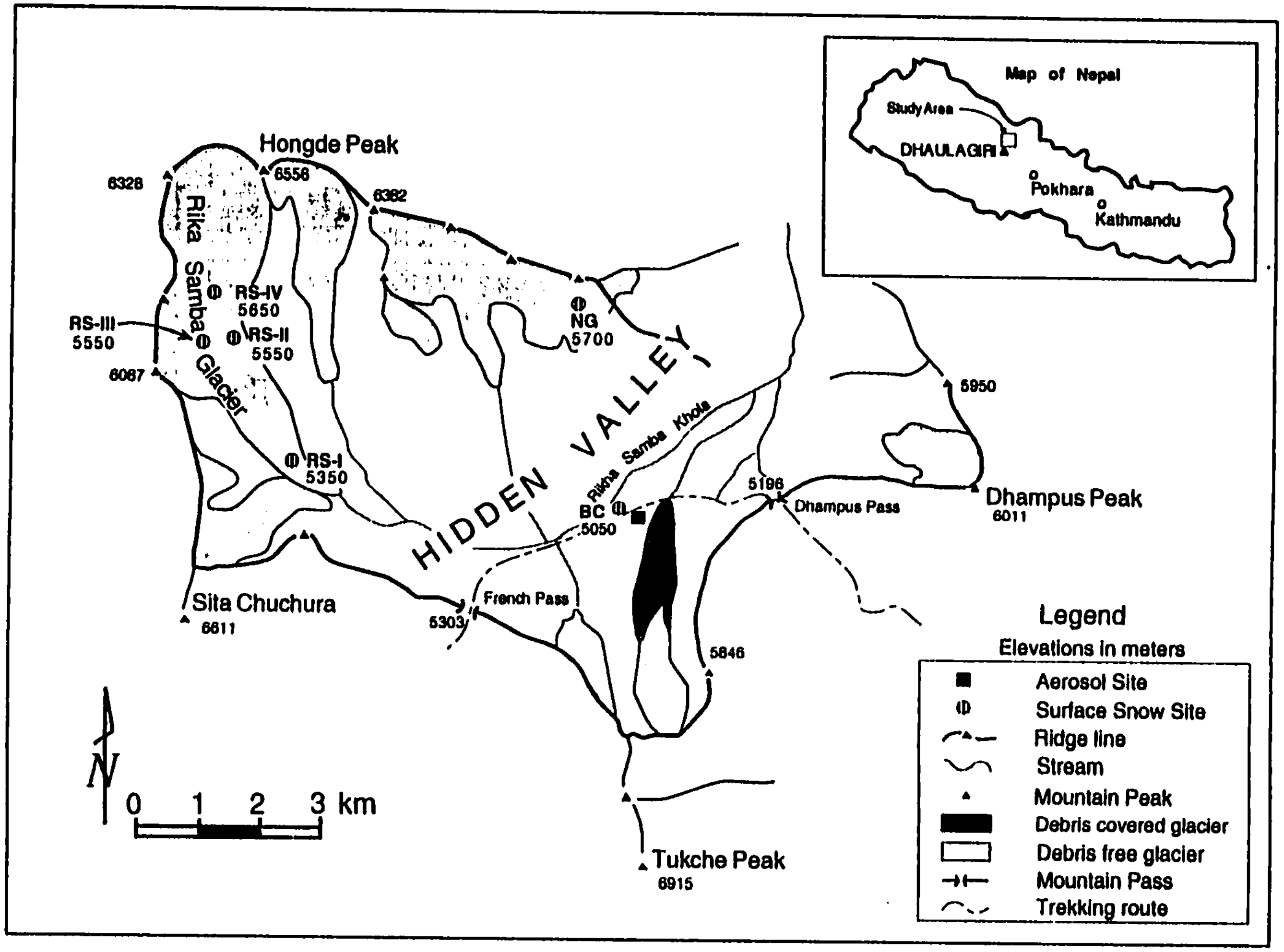


Figure IV.1. Location map of aerosol and fresh snow sampling sites in Hidden Valley western Nepal. Major mountain peaks and glaciers are also shown in the map. Map modified after Nakawo *et al.*, (1976)

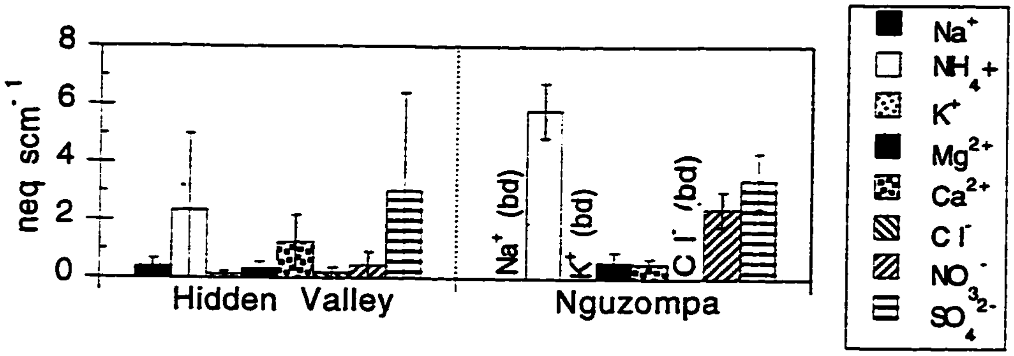


Figure IV. 2. Comparison between mean aerosol concentration in Hidden Valley from this work and Nguzompa from Wake et al. (1994). The error bars represent standard deviations of concentrations within sampling period.

Table IV.3. Inter-correlation between the water soluble ions in air measured in Hidden Valley from 14-27 August, 1994.

	Na^+ (13)	NH_4^+ (17)	K^+ (16)	Mg^{2+} (18)	Ca^{2+} (18)	Cl^- (13)	NO_3^- (19)	SO_4^{2-} (18)
Na^+	1.00							
NH_4^+	-0.16	1.00						
K^+	0.80[a]	0.37	1.00					
Mg^{2+}	0.00	0.77[a]	0.23	1.00				
Ca^{2+}	0.07	0.69[a]	0.41	0.80[a]	1.00			
Cl^-	0.91[a]	-0.30	0.59	-0.09	0.03	1.00		
NO_3^-	0.12	0.45	0.28	0.70[a]	0.91[a]	0.09	1.00	
SO_4^{2-}	-0.16	0.99[a]	0.40	0.70[a]	0.74[a]	-0.30	0.52	1.00

Note. Number of samples are shown in parentheses

[a] Indicates correlation significant at p=0.01.

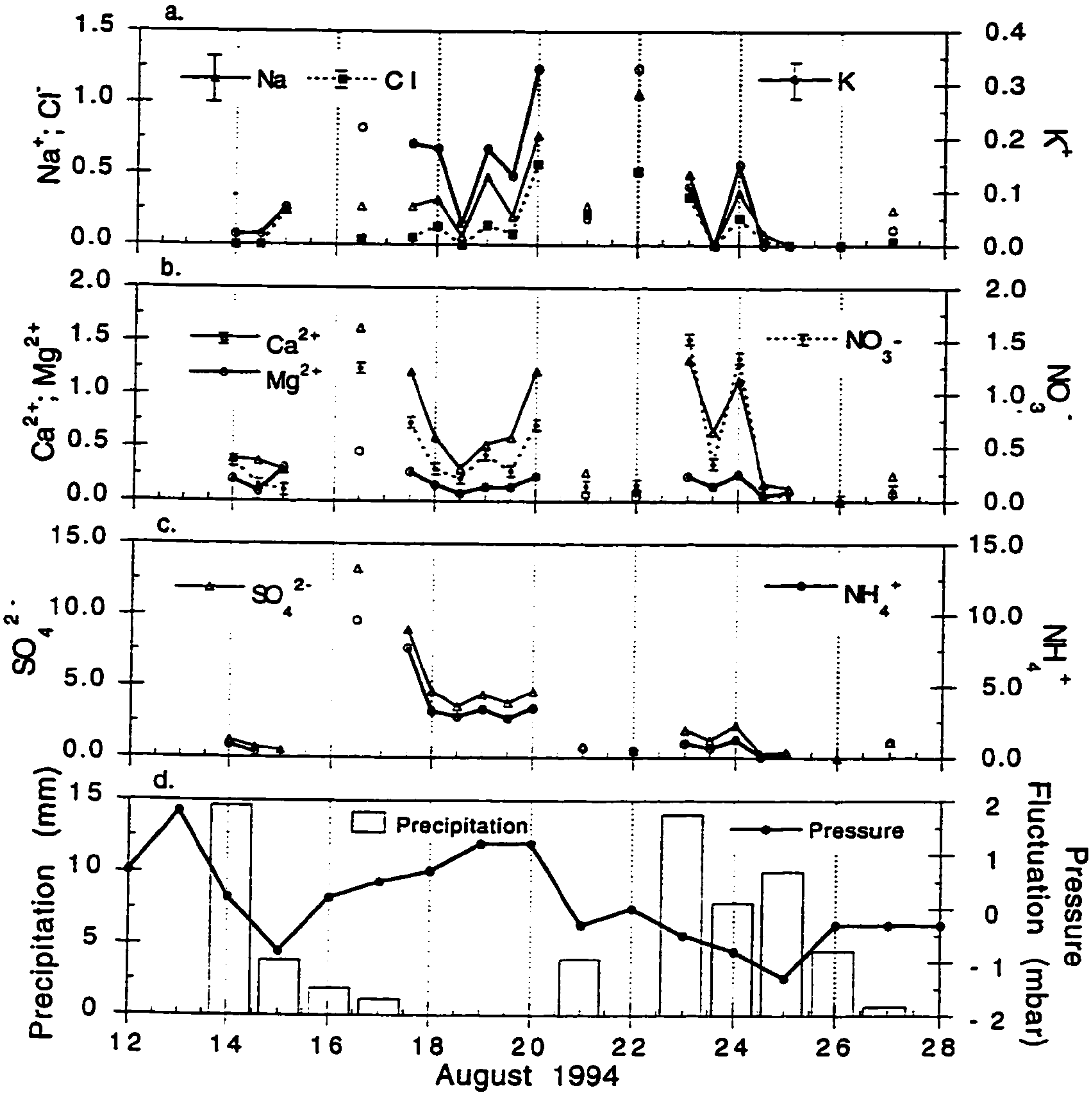


Figure IV.3. Time series of a.Na$^+$, K$^+$, and Cl$^-$; b. Ca^{2+}, Mg^{2+}, and NO$_3^-$; c. NH$_4^+$ and SO$_4^{2-}$; and d. daily precipitation and pressure fluctuations measured in Hidden valley. Concentrations are plotted centered at the middle of the sampling period and vertical bars represent detetcion limits.Detection limits for Mg^{2+}, SO$_4^{2-}$, and NH$_4^+$ are not shown as thery are insignificant. Gaps in the series indicates periods when sampling was not conducted due to rainy or foggy weather conditions.

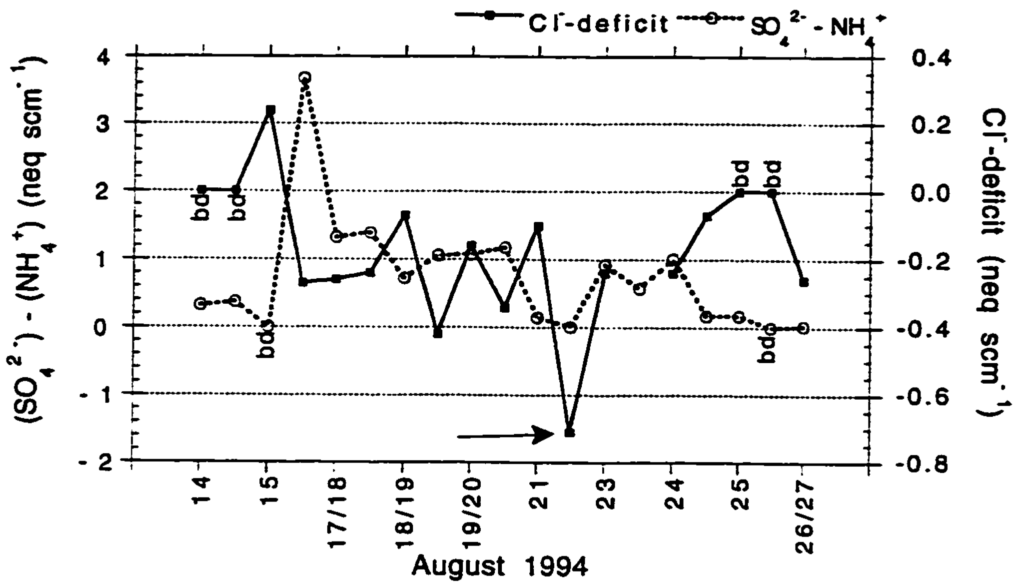


Figure IV.4. Relationship between excess-Cl$^-$ and SO_4^{2-} in the aerosol. Here SO_4^{2-}-NH_4^+ indicates SO_4^{2-} after deducting the amount required to totally neutralize NH_4^+. The arrow indicates an anomalous data point, which does not follow the general inverse relationship.

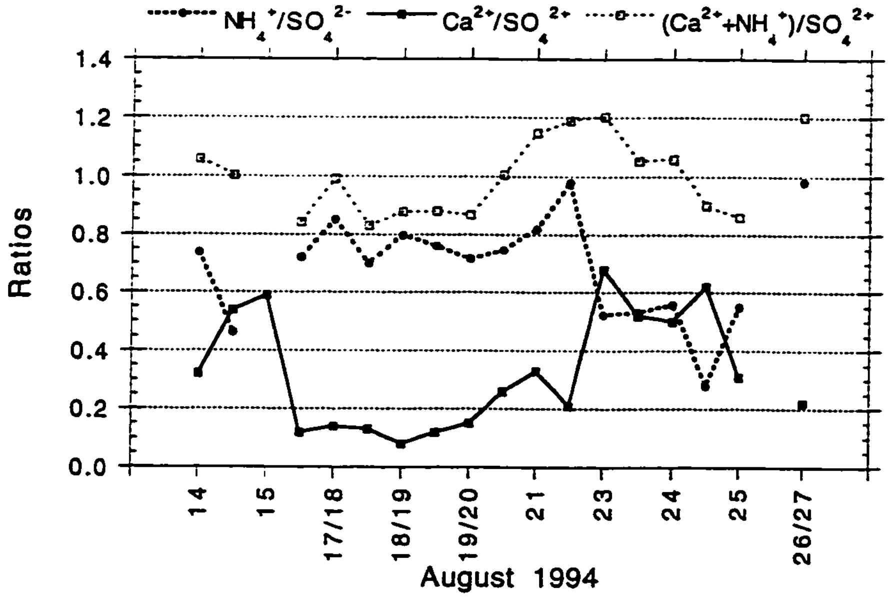


Figure IV.5. Relationship between NH_4^+/SO_4^{2-} and Ca^{2+}/SO_4^{2-} ratios in aerosol. The correlation coefficient between the two ratios is 0.75.

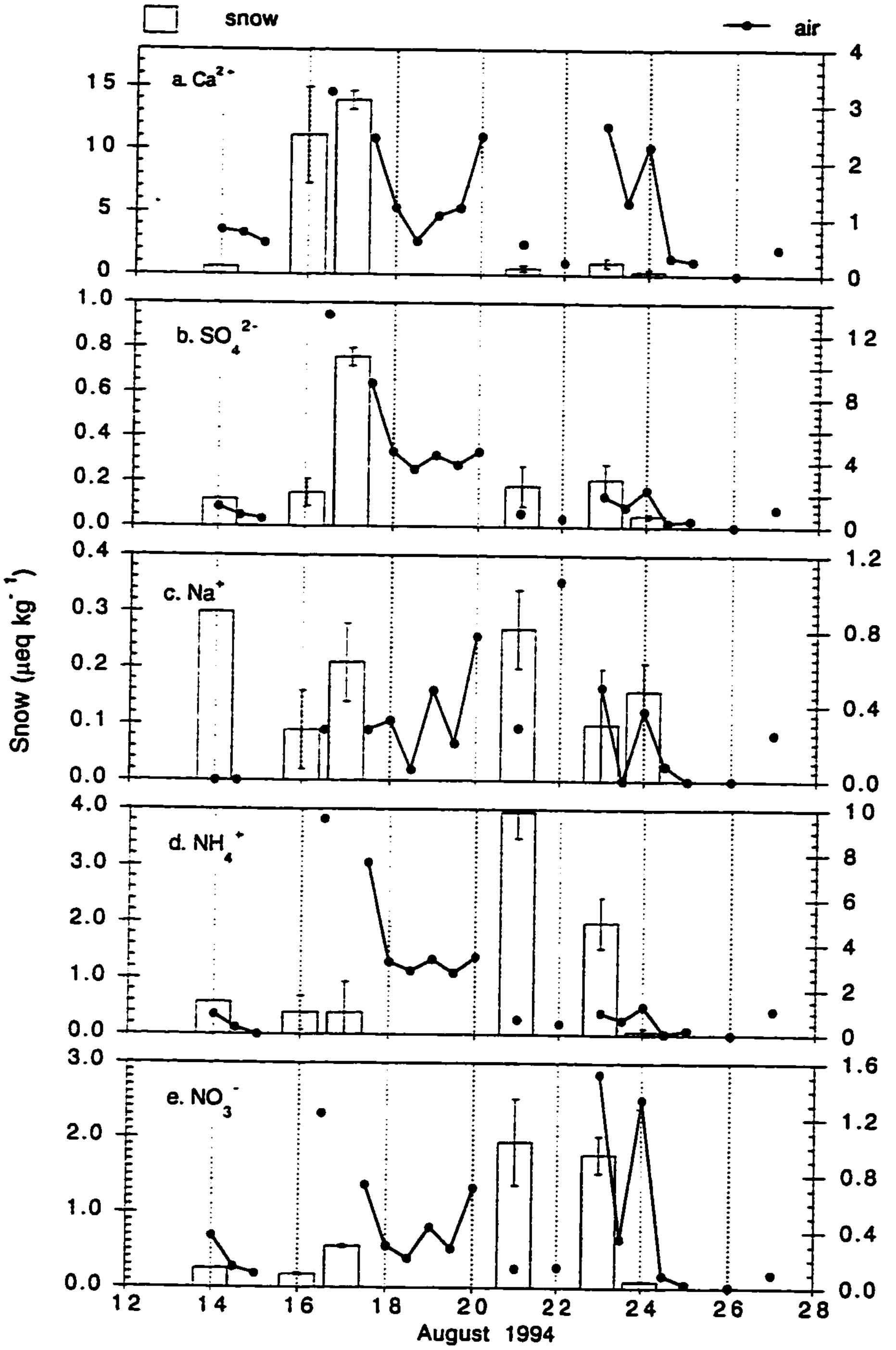


Figure IV.6. Comparison between temporal variations in a. Ca^{2+}, b. SO_4^{2-}, c. Na^+, d. NH_4^+, and e. NO_3^-concentrations in snow and aerosol in Hidden Valley. All data points are assumed to represent 12 hour periods (day time and night time) and are plotted centered at the middle of the periods. Gaps in the series indicates periods when sampling was not conducted due to rainy or foggy weather conditions. Error bars on the snow data represents standard deviations of samples within the same event.

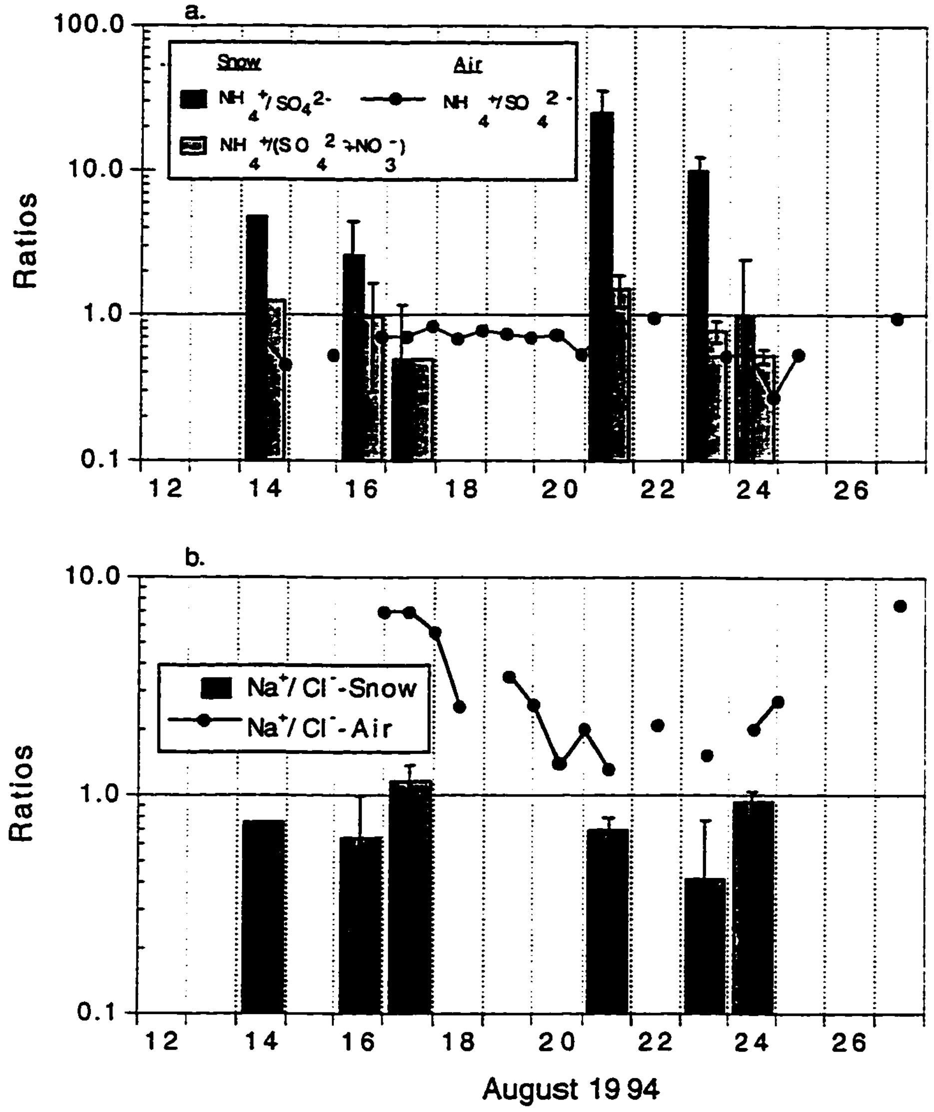


Figure IV.7. Comparison between a. the NH_4^+/SO_4^{2-} ratio in the aerosol and NH_4^+/SO_4^{2-} and $NH_4^+/(SO_4^{2-}+NO_3^-)$ in the snow and b. Na^+/Cl^- ratios in the aerosol and the snow.

CHAPTER FIVE

SEASONAL VARIATIONS IN AEROSOL CONCENTRATIONS AND COMPOSITIONS IN THE NEPAL HIMALAYA.

This chapter will be submitted to the Atmospheric Environments for publication

IV. SEASONAL VARIATIONS IN AEROSOL CONCENTRATIONS AND COMPOSITIONS IN THE NEPAL HIMALAYA.

ABSTRACT

This paper summarizes a 15 month long study (Sep. 1996 to Nov. 1997) on water soluble components (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, and SO_4^{2-}) in atmospheric aerosols conducted at a remote Himalayan site and a rural Middle-Mountain site in Nepal. Most of the species sampled behave similarly in terms of seasonal and short-term variations. The aerosol concentrations were low during the second half of the monsoon and post-monsoon seasons and gradually increased during the winter season. Aerosol concentrations in some samples were up to 20 times higher during the pre-monsoon and early-monsoon seasons than they were in post-monsoon. The seasonal variations are clearly related to variations in atmospheric circulation. The high concentrations during the pre-monsoon season are attributed to regional scale valley wind systems and the maximum concentrations at both sites coincide with maxima in the local wind speed record. Occasional shifts in the large scale circulation resulted in dramatic fluctuations in aerosol concentrations during the pre-monsoon and early monsoon seasons. In the middle of May the large scale circulation changed from westerly to south-easterly monsoon in the Himalaya. The summer monsoon circulation brought considerable amounts of pollution to the sites from south-east. Precipitation played an important role in short-term (few samples, e.g., during the late winter at the Himalayan site) to longer term (several samples, e.g., after mid-June at both sites) reduction in the aerosol concentrations, while the lack of precipitation in the pre-monsoon and early monsoon supported a gradual build-up of pollutants in the atmosphere. During the pre-monsoon and early monsoon, when concentrations were elevated at both sites, they were often higher at the remote Himalayan

site than at the middle mountain site. A pollution layer in the monsoon circulation, as a result of inefficient precipitation scavenging of pollutants above a certain altitude could be responsible for such peaks. Alternatively, dry convective rise of pollutants at distant sources and horizontal upper air transport by monsoon flow could also have caused these elevated concentrations.

INTRODUCTION

The Himalayas and the Tibetan Plateau play a central role in regional scale atmospheric circulation patterns over Asia. Intense solar radiation in the highlands creates high temperatures and reinforces the low pressure center over northern India (Flohn, 1968), which drives the summer monsoon circulation over the Indian sub-continent. The extensive Himalayan range stretches about 2500 km from west to east along the southern margin of the Tibetan Plateau, thereby forming a barrier between tropical and polar air masses (Nieuwolt, 1977). During the winter, the Himalaya and Tibetan Plateau deflect strong westerly winds along their northern and southern margins, while in summer, the Himalaya restrict the penetration of south-easterly monsoon air masses into the interior regions of the Plateau. In this sense, the atmospheric composition over the Himalayas should reflect interactions of these major circulation systems.

On a more regional scale, the circulation system in the Himalayas is also characterized by valley wind systems (Barry, 1981). Due to high radiation and intense heating of the high elevation valleys, a regional scale up valley wind (valley wind) develops during the day time, whereas during the night time cool and dense air masses move down slope and down valley (mountain wind) (e.g., Hindman, 1995b; Hindman et al., 1996). Because of the proximity to the ground surface, this circulation is likely to pick-up considerable amounts of surface derived materials and transport them on a daily basis to the interiors of the Himalaya (Nakajima, 1976). The intensity of this circulation system changes with season, which should have an important effect on atmospheric composition.

The composition of the atmosphere over the region, however, is poorly known because of the limited study outside of major cities. Ideal sites for monitoring atmospheric composition and its evolution should be free from local pollution and representative of the remote troposphere (e.g., Adams et al., 1977; Huebert and Lazrus, 1980). Short-term (days to a few weeks) aerosol sampling programs by Ikegami et al. (1978); Wake et al. (1994); and Shrestha et al. (1997) have indeed shown low pollutant concentrations in the Himalayan atmosphere suggesting representation of the remote troposphere by these sites. However, these studies did not provide information on the seasonal variations in atmospheric composition that are expected due to the dramatic changes in atmospheric circulation over these longer time scales. Furthermore, continued population growth and increasing industrialization of Asia is expected to result in large increases in anthropogenic emissions and air pollution (Foell et al., 1995; Zhao and Sun, 1986; Galloway et al., 1987; Carmichael et al., 1995). Systematic monitoring of atmospheric composition in the remote Himalayas will serve as a base line to monitor the influence of anthropogenic activities on this climatologically important region.

An aerosol sampling program was conducted simultaneously at a remote Himalayan site and a rural Middle-Mountain site between September 1996 and November 1997. Sulfur-dioxide was also measured at these sites using passive diffusion samplers. The resulting data base provides the first year-long investigation of aerosol and SO_2 chemistry at remote Himalayan sites and provides the basis to study seasonal variations in the concentrations of atmospheric constituents, transport mechanisms, and chemical associations.

SAMPLING SITES

Phortse: One sampling site was located near Phortse, a remote village (4100 m a.s.l.; 27°51' N 86°45' E) in the Himalayan region of eastern Nepal (Fig. V.1). The site was situated 350 m above the village close to where a northwest to southeast trending ridge merges with the mountain slope. Although the predominant circulation over the valley is south-westerly, observations of the local wind pattern reveal that wind blows from the south-south-east (across the ridge) and the sampling site is rarely, if ever, affected by pollution from the village. The village of Phortse consists of about 60 permanent houses. There are small patches of farm-land around the village, where potato and buckwheat are cultivated between April and September. There are also some forested areas dominated by juniper and different varieties of pine. A small airstrip at Syangboche is located at a distance of about 4 hours walk (5 km aerial distance) while a more regular airport at Lukla is 3 days walk from the Phortse village. The nearest road head (Jiri) is at a distance of 7 days walk (ca. 70 km aerial distance).

Jiri: The second sampling site was located at Jiri (1900 m a.s.l.; 27°38' N 86°14' E), a small town at the upper limit of Middle-Mountain region of Nepal (Fig. V.1). The sampling station was established on the east bank of a wide river valley, upwind from the major settlement to eliminate the direct influence of local emissions. The town is connected with Kathmandu by a motorable road, although the number of vehicles arriving at Jiri is less than 20 per day. A livestock farm is located to the west and slightly north from the site. Data from an existing meteorological station close to the sampling site indicates that wind is predominantly southerly at the site, hence emissions from the farm should not directly influence the aerosol sampling. A dense mixed forest is located to the east and cultivated fields and pasture surrounds the site on all other sides.

METHODS

Aerosol Samples

Aerosol samples were collected from September 24, 1996 to November 18, 1997 at Phortse and from September 8, 1996 to October 27, 1997 at Jiri. The major part of the samples represent 5 day sampling intervals, though at Phortse three intensive sampling programs were conducted, during which 6 to 12 hourly samples were collected. The purpose of the intensive sampling was to study diurnal variations in concentrations and their relationship with meteorological parameters and precipitation chemistry. These intensive samples are also presented in this paper as 5 day samples by taking their weighted mean, while detailed results based on the intensive samples appear in Chapter VI. There were some periods when the sampling equipment (pump) failed. Due to limited communication and poor accessibility, some data gaps occurred at both sites. At Phortse gaps occurred during Dec. 14-17, 1996; Jul. 1-Aug. 8, 1997 and Sep. 29-Oct. 9, 1997, and at Jiri there is no data for Dec. 18, 1996-Jan. 1, 1997; Jan. 5-Feb. 2, 1997; Mar. 16-Apr. 12, 1997; and Apr. 27-May 15. The data gaps comprise 15% and 20% of the total sampling period at Phortse and Jiri, respectively .

Aerosol samples were collected on 2 µm pore size, 25 mm diameter (90 mm for intensive sampling) Zefluor™ Teflon filters (Gelman Sciences). The filters were placed facing downwards in a cylindrical polyethylene protective cover. The air was drawn by 24 Volt miniature vacuum pumps (Fürgut GmbH, Germany), powered by a combination of photovoltaic cells and batteries. The volume of air sampled was measured by an in-line flow meter. Corrections for ambient temperature and pressure allowed conversion of the measured volumes to standard cubic meters (scm). For the mean flow rate of 0.40 scm hr^{-1} the velocity at the face of the filter was sufficiently high that the efficiency of collection for particles as small as 0.3 µm is greater than 99% (Liu et al., 1984). The flow velocity at the opening of the protective cover was calculated to be 3 cm sec^{-1}, according to which the cut-off for large particles, as given by the sedimentation velocity of particles, is estimated at 8 µm (Barrie, 1985; Warneck, 1988; Davidson, 1989).

Zefluor™ filters have been found to have a minimum positive artifact with regard to sorption of gaseous species like HNO_3 and SO_2 (Spicer and Schumacher, 1979; Appel et al., 1984). In contrast, there might have been some evaporation of NO_3^- due to extended sampling, therefore the NO_3^- values reported here should be regarded as a lower limit of true airborne concentrations, since the samples are generally slightly acidic.

Care was taken to minimize contamination both in the laboratory and in the field. The filter cartridges were loaded and packed in clean plastic bags in a Class 100 clean lab at the University of New Hampshire. The 25 mm cartridges in plastic bags were placed and transported in air-tight glass bottles. The glass bottles were opened only immediately before sampling and the plastic bags were cut open with clean surgical blades. After sampling, the filters, still in their cartridges, were returned to the original clean plastic bags and sealed in the glass bottles. The 90 mm cartridges were packed in clean plastic bags and transported in air-tight containers. After sampling, the filters were removed from the cartridges and placed in pre-cleaned air-tight glass bottles. Loading and unloading of samples was carried out wearing a non-particulating clean suit, hood, face mask, and plastic gloves. Blank filters were handled in the same manner as the samples. All aerosol samples were transported to the University of New Hampshire, where laboratory analyses were conducted.

To analyze for major ion concentrations, the sample and blank filters were wetted with 0.075 ml ultra pure methanol (0.5 ml for 90 mm filters). The soluble components were then extracted with two 5 ml aliquots of deionized Milli-Q water. Major ion concentrations in the aqueous extracts were determined by ion chromatography using a Dionex model 4000 ion chromatograph. Cations were analyzed using a CS12 column, 22 mM MSA eluent and CSRS suppresser and anions were analyzed using an AS11 column, 6.6 mM NaOH eluent and ASRS suppresser. The sample loop was 100 µl and the sample running time was approximately 10 minutes.

Mean blank values were deducted from the sample values. Detection limits are taken as two standard deviations of blank values divided by the mean volume of samples. The detection limits are presented in Table V.1.

Sulfur-dioxide Samples

Passive diffusion samplers were used to determine SO_2 concentrations in the air. The samplers were mounted under a rain shield, 2 m above ground surface. Filters used at Jiri were exposed for one month and those used at Phortse were exposed for two months. The exposure time was longer at Phortse due to expected lower SO_2 concentrations. Sulfur-dioxide samples were then shipped to the Swedish Environmental Research Institute (IVL) for laboratory analyses. Passive diffusion samplers are inexpensive and effective devices to study several gaseous species in the air, including SO_2. These types of samplers were found successful in Europe and recently in several parts of Asia. (Carmichael et al., 1995). Details on the method of SO_2 monitoring using passive samplers are given in Carmichael et al. (1995), Ferm and Rodhe (1997), and WMO/GAW (1997).

Meteorological Data

Meteorological observations directly at Phortse were limited to the periods of the three intensive sampling programs. Therefore, to infer the meteorological condition throughout the sampling period we used three reference stations: Syangboche (27°48'N, 86°43'E; 3800 m a.s.l), Dingboche (27°53'N, 86°50'E; 4200 m a.s.l), and Lobuche (27°57'N, 86°48'E; 4900 m a.s.l.). All three reference stations are in the Khumbu region, within a radius of 10 km of Phortse. Continuous data from whichever of these three stations showed the best relationship with the measurements taken at Phortse during the intensive sampling programs is used in this study. Radiation data was found to be spatially homogeneous over the entire region, therefore we used Lobuche data, which is the most complete. We used air temperature, relative humidity, and wind speed data from Dingboche as they showed better covariance with data from Phortse. The precipitation data collected at Phortse during September-October and February-March intensive samplings were found to be linearly related to Syangboche precipitation data for the respective periods. A linear regression equation was developed and used to transform the Syangboche precipitation record to represent precipitation at Phortse.

The Jiri sampling station was established close to a permanent meteorological station operated by the Department of Hydrology and Meteorology, Nepal (DHM). The parameters measured at the station include minimum and maximum temperatures, wet and dry bulb temperatures, wind speed, wind direction, and precipitation. The measurements are taken at World Meteorological Organization standard meteorological observation times for Nepal: 0845 and 1745 local time. The data presented here are daily means calculated as averages of the two observations.

RESULTS

Seasonal Variations

Statistical details of aerosol concentrations at Phortse and Jiri by season are presented in Table V.2. Geometric and arithmetic mean concentrations are significantly different for all species at Phortse during the pre-monsoon (March-May) and monsoon (June-September) seasons, while the two means are not considerably different at Jiri. The differences between the two means at Phortse is clearly due to high concentration variations during the pre-monsoon and monsoon seasons (Fig. V.2). During these two seasons aerosol concentrations at Phortse fluctuate from values close to detection limit to concentrations up to 10 times greater than the seasonal mean concentration within a few sample intervals. Sulfate and NH_4^+ are the dominant ions in the Phortse aerosol for all seasons followed by Ca^{2+}, NO_3^-, and K^+. Sulfate and NH_4^+ are also dominant species at Jiri. The concentration order of these major ionic species changed slightly with season.

Mean concentration (both geometric and arithmetic) of all species at Phortse are highest during the pre-monsoon. However, the second rank in the order of seasonal concentrations is variable for different species. Generally, geometric mean winter (December-February) concentration for most species rank second, whereas the arithmetic mean concentrations during the monsoon are higher than during winter. Higher arithmetic mean monsoon concentrations are due to the influence of outliers (high concentrations) present in the earlier part of monsoon. The order of seasonal mean concentration at Jiri are independent from methods of their calculation, but varies with species. Pre-monsoon mean concentrations are highest for Na^+, K^+, Mg^{2+}, and Ca^{2+}, followed by winter mean concentrations. Conversely NH_4^+, Cl^-, NO_3^-, and SO_4^{2-} show highest concentrations in winter followed by pre-monsoon.

Inter-site comparison

One surprising result apparent from the statistical details is that the arithmetic mean concentrations of NH_4^+, K^+, Cl^-, NO_3^-, and SO_4^{2-} during the pre-monsoon season are significantly greater at Phortse compared to those at Jiri (1.3 to 3.6 times greater except for Na^+ and Ca^{2+}), despite the fact that Phortse is located 2300 m higher and further removed from the dominant (southerly) sources of pollution (Table V.2). On the other hand, during other seasons the concentrations at Jiri are significantly higher than those at Phortse for most species. Further, when geometric mean concentrations during pre-monsoon are compared, concentrations at Phortse is higher compared to Jiri only for Cl^-, NO_3^-, and SO_4^{2-}. The five isolated samples with high mean concentrations of aerosol species during pre-monsoon and monsoon seasons are responsible for the higher arithmetic means at Phortse. Average concentrations (both geometric and arithmetic) at Phortse, derived without including the five high concentrations are comparable or lower than those at Jiri for most species (except SO_4^{2-}) for the pre-monsoon season. The five high concentration samples from Phortse during the pre-monsoon and the early monsoon seasons are unique and interesting and are examined in detail in the discussion section.

Inter-species Correlation

Time series of aerosol data from Jiri show that temporal variations of some major ionic species are different in the period from August to March, whereas they are more similar after that (Fig. V.2). Therefore, inter-species correlation coefficients were derived separately for these two periods (Table V.3). The correlation coefficients at Jiri are considerably different for the two periods, improving significantly in the second period. Based on the inter-species correlation several groups of species can be envisaged for Jiri. They are: for the first period 1. Na^+-K^+-Mg^{2+}-Ca^{2+}; 2. NH_4^+-NO_3^- ; 3. Cl^--NO_3^- and for the second period 1. Na^+-Mg^{2+}-Ca^{2+}-SO_4^{2-}; 2. NH_4^+-SO_4^{2-}; and 3. Cl^--NO_3^- -SO_4^{2-}. At Phortse, all species except Na^+ tended to covary over the whole sampling period (Fig. V.2). Therefore correlation coefficients for Phortse were derived for the whole sampling period. Inter-species correlation coefficients are generally high for Phortse except for Na^+ and NH_4^+ (Table V.3).

Sulfur-dioxide concentrations

Sulfur-dioxide concentrations at Phortse are extremely low and are close to the detection limit, whereas at Jiri the concentrations are comparable to values found in rural and semi-urban sites of Nepal (Carmichael et al., 1995) (Figure V.3). Sulfur-dioxide measurements taken earlier at several places of Nepal showed distinct seasonal changes with maximum in pre-monsoon and minimum during monsoon seasons (Carmichael et al., 1995). The SO_2 record from Jiri shows a similar pattern, while low concentrations at Phortse mask any clear seasonal pattern.

DISCUSSION

Concentration Levels: Local Emission Versus Long Range Transport

Earlier aerosol measurements made in the Nepal Himalayas during monsoon and post-monsoon showed low concentrations of all species. Secondary aerosols such as SO_4^{2-}, NH_4^+ and NO_3^- were found in concentrations comparable to, or even lower than, those at several other remote continental sites (Wake et al., 1994; Shrestha et al. 1997). While concentrations at Phortse measured in this study during the late monsoon and post monsoon are similar to those at Nguzompa glacier in post-monsoon (Wake et al., 1994) and in Hidden Valley in monsoon seasons (Shrestha et al. 1997), the present study also shows that aerosol concentrations during pre-monsoon and early monsoon reach remarkably high levels for such remote sites. Concentrations of most species (Table V.2) during winter and pre-monsoon seasons at Phortse are comparable to concentrations at several urban sites in Europe and North America (e.g. Stelson and Seinfeld, 1981; Rahn and Lowenthal, 1985; Kaneyasu et al., 1995).

The elevation of the Phortse site is lower than Hidden Valley and Nguzompa sites. Further, due to proximity to the village, local emissions from the village may be transported to the sampling site by the vertical mixing of air (Reiter et al., 1976). Davidson (1981) observed the influence of indoor pollution (cooking stoves) on the chemical composition of air and snow in the vicinity of a village in the Khumbu. However, several lines of evidence suggest that the high concentrations during the pre-monsoon and the early monsoon are not due to the influence of local emissions. 1) The aerosol measurement conducted by Davidson (1981) included a few samples for NO_3^- and SO_4^{2-}, collected to investigate the influence of local pollution on the ambient atmosphere. Seasonal concentrations at Phortse are lower than that measured by Davidson (1981) for all species except SO_4^{2-} for the pre-monsoon and five high peaks. 2) The seasonal variations in the aerosol concentrations do not agree with seasonal changes in local emissions. Emissions from Phortse and other villages in the neighborhood are likely at a maximum in winter when the houses are heated by burning firewood and yak dung and at a minimum during the entire monsoon, when villages are nearly deserted and people migrate with their cattle to higher elevation pastures. 3) The low concentrations of SO_2 at Phortse suggest that SO_4^{2-} in aerosol is not from local emissions. The two month long samples of SO_2 may not be affected by isolated short episodes of high concentration that occurred during pre-monsoon and the early monsoon. However, besides the isolated peaks the concentrations of SO_4^{2-} during pre-monsoon is generally high (20 nmol scm^{-1}), while SO_2 concentrations were close to detection (max. 3 nmol scm^{-1} at Phortse; Fig. V.3). Sulfur-dioxide concentrations were found to be generally low at non-urban sites in Nepal. Carmichael et al (1995) found that the highest SO_2 concentration at Godavari, a site south of Kathmandu (within the same valley) was lower than 45 nmol scm^{-1}. Comparable SO_2 concentrations were found in other rural and semi-urban sites in Nepal, suggesting that there are no large sources of SO_2 in Nepal outside of major urban cities. 4) The eight major ionic species measured in this study show high inter-species correlations. Such high inter-species correlations at isolated Arctic and Antarctic sites have been suggested to be indicative of thoroughly mixed air masses due to long travel distances (e.g., Heidam, 1984; Mayewski et al., 1994). There could be some emission sources for some or all of the eight species in the Khumbu valley (firewood burning for NH_4^+, K^+, and to some extent SO_4^{2-}; application of manure in the field for NH_4^+ and NO_3^-; agriculture activities, movement of people and cattle for Ca^{2+}, Mg^{2+}; etc). The rather long sampling intervals of our samples (5 d.) may be influenced by different air-masses, incorporating pollutants from different source regions causing the observed high inter-species correlation. Nevertheless, intensive sampling (12 h.) also show similar inter-species relationship (Chapter VI) suggesting that high correlations are likely due to long distance transport. 5) The observation of wind direction at Phortse revealed that the sampling site is rarely influenced by wind flowing directly over the village. The 12-hourly wind data taken during the March intensive sampling program shows that the wind direction was close to 240° (direct from village) only three times (Fig. V.4). Further there is no direct relationship between the wind direction and concentration levels (Fig. V.4). 6) The high episodes in aerosol concentrations at Phortse, as is subsequently discussed, coincide with unique periods of changes in atmospheric circulation and meteorological characteristics with the potential for transporting pollutants from distant sources such as populated and industrialized regions of Northern India. Long distance transport of pollutant to Nepal is supported by the study of Arndt et al. (1998) , who suggest that 70% of sulfur deposition in Nepal is of Indian origin.

We therefore suggest that as a result of careful site selection at Phortse with respect to valley orientation, predominant wind directions and local pollution sources (e.g., biomass (fire-wood) burning, agricultural dust, manure etc.) we were able to avoid the affect of local sources of pollution on the measurements. Ammonium and NO_3^- at Jiri may have been partly affected by local sources because of the proximity of the sampling site to agricultural lands, forest, and animal husbandry (Harrison and Pio, 1983 and references therein). Even here, SO_2 concentrations are relatively low (Fig. V.3b), confirming that SO_4^{2-} concentrations at Jiri are relatively unaffected by local sources.

Seasonal Variations in Aerosol Concentrations

The seasonal variations in aerosol concentrations observed in Phortse and Jiri are quite similar to those in SO_2 concentrations at different sites in Nepal (Carmichael et al.,

1995) and total suspended particle (TSP) concentrations in Kathmandu (DHM, unpublished), suggesting that the seasonal variations in atmospheric composition are governed by factors operating on regional scales. The chemical species measured in this study represent a wide variety of sources, including crustal, biogenic, anthropogenic, and marine. Further, since seasonal as well as inter-sample variations for most species are similar, there is likely a common transport mechanism that brings all these species to the sampling sites. We suggest that a combination of physical processes in the atmosphere combined with variations in source strengths can account for the high seasonal variability in aerosol concentrations in the Himalaya.

The large scale circulation during post-monsoon and winter is generally westerly in the upper atmosphere. Pollutants transported by westerly flow (sub-tropical jet) from Europe have been observed at distant sites, although mostly in gas phase (e.g., Talbot et al., 1997). Our sampling sites are 15-20° south of the industrialized region of Europe and there are no large industrialized and populated regions directly upwind during these seasons. Westerly air-masses are therefore relatively depleted of pollutants, especially particulates (e.g., Mayewski et al., 1983; Wake et al., 1993). Occasional shifts in the atmospheric circulation cause the westerly flow to take a southward dip (discussed in detail in following paragraph), which could bring pollutants from Europe. In contrast, such events resulted in lower aerosol concentrations at our sites. Therefore, it is unlikely that westerly circulation is responsible for the high aerosol concentrations. Hindman, (1995b), Hindman et al. (1996), and Hindman and Upadhyay (1997) found that sub-micron particles can be transported into the Himalaya from places as far south as the Terai region and even from India, driven by thermal valley wind systems, while Ciccioli et al. (1993) found similar transport of gas phase organic compounds. The presence of valley winds in the Khumbu Valley is supported by the wind data, which shows a gradual increase in wind speed between November and March (Fig. V.5c) and gradual change in the wind direction from south-southeasterly to south-southwesterly direction (Fig. V.5d). The difference between the day-time and the night-time wind directions are greater in winter, implying large diurnal change in the local circulation is weakest during June/July, and therefore local circulation is dominated by regional scale southerly flow. The peak in wind speed is synchronous with peaks in aerosol concentrations (April in Phortse and Feb./Mar. in Jiri). The increase in wind speed is accompanied (and possibly caused) by increases in air temperature and radiation (Fig. 5a, b). The build-up of pollution in Phortse was visually observed during the March intensive sampling. During the middle of the sampling program, low level haze appeared at the end of the river valley. The haze gradually rose and moved up-valley on a daily basis. The haze must have reached the sampling site a few days after the intensive sampling was completed. It is therefore suggested that regional scale thermally driven valley winds were responsible for the transport of pollutants to the sites that resulted in high aerosol concentrations during the pre-monsoon.

While the valley wind can gradually bring in pollutants from southerly locations, it is unlikely to cause large and relatively rapid fluctuations in the concentrations, as seen in our data. Since the valley wind is generally limited to lower levels (Nakajima, 1976) it should affect the Jiri site more than the Phortse site. However, the data shows that during several occasions Phortse experienced a higher degree of pollution than Jiri (e.g. early April, end of May, and middle of June). Analyses of synoptic maps show that each of these peaks in concentrations are associated with significant shifts in large scale flow. During winter and pre-monsoon the wind flow is westerly in the upper air (i.e., 500 mb) as well as in lower levels (e.g., > 850 mb). However, the occurrence of troughs (lows) and wedges (highs) upwind from the sampling sites cause the westerly flow to curl and change to north-westerly or south westerly direction. It is observed that a north-westerly flow resulted in low concentrations at Phortse (generally close to detection limits) while a south-westerly flow resulted in dramatic increase in major ion concentrations. Such change in circulation took occurred during the early March peak (Fig. V.6a,b). The change in aerosol concentrations associated with large scale flow is best demonstrated by samples from early

April. (Fig. V.2).The flow was north-westerly at the end of March (Fig. V.6c). On April 2, the flow changed abruptly to a south-westerly direction, which could have brought considerable amount of pollutants from the populated and industrialized middle-Indian states and Kathmandu, as well as crustal species from the great Indian (Thar) desert (Fig. V.6d) and caused the large peaks in aerosol concentrations (V.2(left)). After 7 April, the circulation re-established the north-westerly direction at which time the polluted air masses were replaced by cleaner ones (Fig. V.2 (left)).

By the middle of May, low pressure cells started forming at the surface over the Indian sub-continent and the atmospheric circulation changes from westerly to south-easterly over the Nepal Himalayas. There are also differences between upper air flow and the lower level flow patterns during this period. The large scale flow at 500 mb was slightly north-westerly between May 22 and 27 (Fig. V.6e). The flow changed to south-westerly at the end of May (Fig. V.6f). The peaks at the end of May and early June at both sites occurred when southerly circulation of a monsoonal nature had already begun. At lower levels (i.e., 700 mb) the monsoon trough started forming and the atmospheric circulation was southerly (figure not shown). Nevertheless, rainfall was scarce in Phortse and only low intensity precipitation occurred in Jiri during this period. Similar sequences of changes in atmospheric circulation explain the high concentration event in the middle of June (Fig. V.6e,f; Fig. V.2), suggesting that the early monsoon circulation is responsible for the high concentrations at Phortse and Jiri after the end of May.

The changes in large scale circulation generally caused changes in the precipitation regime as well. Wet scavenging is an effective mechanism for the removal of aerosols, especially in the size range of 0.2 to 10 μm (Barrie, 1985; McGann and Jennings, 1991). The lack of precipitation between November and mid-January likely supported the build-up of pollution, especially apparent in the record from Phortse. The precipitation between mid-January and March could have reduced the build up of pollutants to some extent (Fig. V.2 (left); Fig. V.5e). This precipitation was caused by westerly disturbances (DHM, 1997).

Precipitation during this period was rare in Jiri (Fig. V.5i), which explains the earlier rise in aerosol concentrations (Fig. V.2 (right)). Significant amounts of precipitation occurred in Phortse right after the peak in concentrations in early-April, which resulted in the cleansing of the atmosphere; the concentrations of all species dropped and remained low until the end of May (Fig. V.2(left); Fig. V.5e). Precipitation of considerable intensity also occurred in Jiri at the end of April and early-May (Fig. V.5i). Apparently, it was less effective in cleansing the air of pollutants. This could be because of differences in the physical processes of rain formation between the two sites and the efficiency with which they can incorporate pollutants in clouds (Barrie, 1985; Davidson, 1989). The abrupt drop in concentrations, both at Phortse and Jiri, coincides with the start of persistent monsoon precipitation associated with the surface level (1013 mb) depression formed over the Bay of Bengal (DHM, 1997).

It is surprising that the peaks in concentrations at Phortse (although limited to 4 samples) are considerably higher than those at Jiri. There are some data gaps in records from both sites. However, only the April peak at Phortse coincides with a data gap at Jiri, while major ion concentrations for most species at Jiri were significantly lower during other peaks at Phortse ($p=0.05$) with the exception of Na^{+}, Mg^{2+}, and Ca^{2+}. Since a local source of pollution at Phortse is not responsible for these peaks, the high concentrations at Phortse are likely reflecting a pollution layer that is restricted to a certain altitude range higher than the elevation of Jiri. The elevation of Phortse falls within the elevation range typical for long distance transport of pollution (e.g., Talbot et al., 1986; Andreae et al., 1988). Monsoon air-masses reaching Nepal originate in the Bay of Bengal (Nayava, 1980; Singh, 1985) and may incorporate a large amount of pollutants enroute, mostly from the Indian plains (e.g., Valsecchi et al., in review). As the air-masses move inland in a north-westerly direction, precipitation occurs and pollutants below the cloud and within certain height of the cloud are scavenged. The elevation of Jiri site is close to the precipitation maximum elevation in the Himalaya (2000 m a.s.l.) (Bruijnzeel and Bremmer, 1989), while Phortse site is well above that elevation . The airmasses above this elevation are possibly subjected to less precipitation scavenging resulting in the formation of a pollution layer. The large peaks could be the result of intrusions of such a pollution layer into the interiors of the Himalaya causing large peaks in aerosol concentrations. Another possibility is that polluted airmasses are vertically raised by convective motion. Note the inter-tropical convergence zone (ITCZ) is aligned at a rather northerly position during the monsoon season. The elevated polluted airmasses are likely to be transported horizontally by the upper air southerly monsoon flow to the Himalaya. This is possible only if the convective motions are not accompanied by precipitation as precipitation will result in scavenging of atmospheric particles. The high concentration peaks observed in our records occurred during generally dry periods (i.e., pre-monsoon and the early monsoon) and the convective processes were likely dry. Backward trajectory analyses for airmasses arriving at the Phortse site during the monsoon of 1998 (http://www.arl.noaa.gov/) showed that the movement of airmasses at 500-600 mb levels were generally horizontal. This observation suggests the stratification and transportation of pollution occurs without vertical mixing and supports the second alternative for the pollutant transport to the site. We did not have access to the data necessary to conduct trajectory analyses for the period of our sampling, nevertheless it is likely that similar transport pattern prevailed during our sampling period. The two mechanisms presented here based on the available data are suggestive rather than conclusive. Future studies needs to be designed to address this issue in detail.

Seasonal variations in aerosol composition and aerosol sources

Similar to the results of earlier studies in the Himalayas, aerosol NH_4^+ and SO_4^{2-} at Phortse are highly correlated (Chapter IV). Ammonium/SO_4^{2-} and NH_4^+/NO_3^- ratios at Phortse showed a clear difference before and after April (Fig V.7a-b). Based on these ratios, it is suggested that the predominant compound was a combination of $(NH_4)_2SO_4$ and NH_4NO_3 during October, November and early December (NH_4^+/SO_4^{2-} ratio around

1.0; Fig.V.7a) while the association changed to NH_4HSO_4 thereafter until the end of first period (NH_4^+/SO_4^{2-} ratio around 0.5; Fig.V.7a). This suggests that NH_4^+ decreased relative to SO_4^{2-} and NO_3^-. When inter-species correlations were derived for the period before April, NH_4^+ showed high correlations with K^+ and NO_3^- ($r^2= 0.96$ and 0.90 respectively; $p=0.01$). This relationship is characteristics of a biomass burning signal (Andreae et al., 1988; Legrand et al., 1992; Dibb et al., 1996). The filters used during this period were dark after sampling indicating the presence of soot, most likely from biomass burning emission. This type of relationship was absent during the second period, reflecting the fact that biomass burning in Nepal consists mainly of burning stubble, widespread after the harvesting season (i.e., September-October) and ceases with the approach of the cultivation season (i.e., March-April). Forest burning was widespread in Nepal after the 1950's, but has declined sharply in recent decades, whereas slash and burn shift agriculture is still practiced in western Nepal (e.g., Tucker, 1987). Uniform NH_4^+/K^+ ratios (of about 5) for the period before April indicates that both NH_4^+ and K^+ originated from the same source, likely biomass burning. Lack of a biomass burning signal after April is supported by lack of such uniformity in the NH_4^+/K^+ ratio (Fig. V.7c).

The NH_4^+/SO_4^{2-} and NH_4^+/NO_3^- ratios at Jiri (Fig. V.7e,f) suggest that the association between NH_4^+, SO_4^{2-}, and NO_3^- is a combination of NH_4HSO_4 and $(NH_4)_2SO_4$ for the period between September and December, while NH_4NO_3 dominated the period thereafter until the end of the first period. This can be explained by the southerly location, lower elevation, and proximity of the sampling site to forests, agricultural lands, and animal husbandry. Lack of a biomass burning signal (i.e., strong NH_4^+-K^+ relationship) at Jiri may be because a significant portion of NH_4^+ comes predominantly from animal husbandry (Harrison, 1983 and references therein) despite the fact that burning of agricultural remains is much more extensive near Jiri than Phortse.

High correlation coefficients ($r^2>0.9$) of SO_4^{2-} and NO_3^- with crustal species (Ca^{2+}, Mg^{2+} and K^+; Table V.3) and lack of uniformity in the ratios NH_4^+/SO_4^{2-} and

NH_4^+/NO_3^- at Phortse after April (Fig. V.7a, b) suggest chemical associations of SO_4^{2-} and NO_3^- with alkaline earth elements. Despite the good correlation of SO_4^{2-} and NO_3^- with crustal species the $Ca^{2+}/(SO_4^{2-}+NO_3^-)$ ratios are generally below 0.5 (except some samples), suggesting that the association is due to external mixture of these species likely due to common transport from distant source. The NH_4^+ excess over $(SO_4^{2-}+NO_3^-)$ in the period after April was possibly introduced at intermediate locations likely from animal husbandry, agriculture, and vegetation in eastern Nepal and/or northern India. One such period with high NH_4^+/SO_4^{2-} ratios between September and November, 1997 is characterized by low concentrations of both species, while the similar period in June is characterized by high concentrations. The June episode shows high cation excess and NH_4^+ during this period was likely largely balanced by organic anions, not included in our chemical analysis. The filter extracts for this period were pale, characteristic of organic content. Size fractionated analysis of aerosol in future investigations will facilitate partitioning SO_4^{2-} associated with crustal species and with NH_4^+, whereas analysis for organic species will help investigate sources of NH_4^+ in the aerosol in the Himalaya.

The influence of crustal sources is evident in the Jiri aerosol by the presence of the groups Na^+-Mg^{2+}-Ca^{2+} in the first period and Na^+-K^+-Mg^{2+}-Ca^{2+}-SO_4^{2-} in the second period. Sulfate is introduced in these groups only in the second period. Similar to the measurement made in Hidden Valley, there is no correlation between Na^+ and Cl^- at either site (Table V.3). Further, Na^+ is greatly in excess over Cl^- compared to sea water ratio (Fig. V.7d). This could mean that either the transport of marine aerosol to the sites is insignificant or that the marine aerosol species are highly fractionated due to the long travel distance. On the other hand, Na^+ was found to correlate well with most species in the period before April, namely: Mg^{2+}, SO_4^{2-}, NH_4^+, Ca^{2+} suggesting the importance of a crustal source for Na^+ in this period.

SUMMARY AND CONCLUSIONS

More than one year long measurement of aerosol at two sites in Nepal, one in a remote High Himalayan location (Phortse) and another in the Middle-Mountain region (Jiri), provide important information on concentration levels and seasonal variations of water soluble inorganic ions. While the concentrations during monsoon and post-monsoon were low and comparable to earlier studies in Nepal Himalayas, concentrations increased dramatically in winter, pre-monsoon, and the early monsoon at both sites.

The seasonal variations in the aerosol concentrations at Phortse and Jiri are attributed to changes in atmospheric circulation. During winter and pre-monsoon large scale circulation patterns (predominantly westerly) transport air-masses over the Himalaya. These air masses were not responsible for bringing pollutants to the Himalaya, possibly because they are long traveled from urban and industrialized regions and are significantly depleted of pollutants compared to southerly airmasses. Thermally driven valley wind system operating during this period on a regional scale transport pollutants to the High Himalayan site. Nevertheless, the valley wind system cannot be responsible for the high peaks at Phortse as the more southerly and low elevation site, Jiri does not experience such high aerosol concentrations. The peaks in major ion concentrations during this period are attributed to changes in large scale circulation. After the end of May monsoon circulation was the dominant mechanism of pollutant transport. Both the valley wind system and monsoon circulation have likely brought in pollutants from distant sources as far south as the Indian plains.

The changes in large scale circulation patterns were associated with changes in the precipitation regimes. Precipitation played an important role in cleansing the atmosphere and the onset of the active monsoon period resulted in a drastic reduction of aerosol concentrations at both sites. A lack of precipitation in the pre-monsoon and early monsoon was generally characterized by a gradual build up of pollutants. Precipitation scavenging of pollutants below a certain height of clouds and survival of others may be responsible for the higher concentrations at Phortse during the pre-monsoon and early-monsoon periods, compared to the much lower site at Jiri. Alternatively, dry convective rise of polluted airmasses to high elevation and horizontal transport of those airmasses in the upper air by the monsoon flow could have a similar affect in aerosol concentrations. Airborne measurement of atmospheric constituents, planned to be carried out in near future in the Nepal Himalaya (per. comm., Prof. E. Hindman, City College of New York) may quantify the extent and elevation range of this pollution layer during the monsoon. The large peaks in the aerosol concentration show that even remote isolated places in the Himalaya are occasionally affected by a high degree of pollution. This in turn highlights the extent of pollution in the southern region from where the pollutants must have originated.

Aerosol composition also show temporal variations at both sites. Biomass burning chemical signals were present in the Phortse data in the first period, while they were absent in the second period, reflecting changes in source strength. Long traveled crustal species together with SO_4^{2-} and NO_3^- were observed in the aerosol while some portion of NH_4^+ is suggested to have been introduced at intermediate locations. Marine aerosols are not an important contributor to the aerosol at either sites.

Although this study provides important insights into seasonal variations in aerosol chemistry, the temporal and spatial coverage is not sufficient to confirm that the observations are representative of the larger Himalayan region. Further, the presence of data gaps lowers the overall confidence in our interpretation. Continued studies on aerosol chemistry with more extensive spatial coverage are necessary to better understand the issues discussed in the present study.

ACKNOWLEDGMENT

We thank villagers of Phortse and residents of Jiri town for their support of the sampling program. We highly appreciate the Livestock farm in Jiri for allowing us to operate the sampling station in their compound. Dr. R.W. Talbot and Dr. B.D. Keim provided detailed reviews of this chapter. We also thank DHM for providing meteorological equipment and other logistical support. Most of all, we are thankful to our observers, Karna Jirel and Dorzee Sherpa for their hard work. This study was partially funded by the Global Change System for Analysis Research and Training (START).

Table V.1. Detection limits (neq scm^{-1}) of aerosol samples from Phortse and Jiri

	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
Phortse	0.02	0.02	0.01	0.01	0.03	0.01	0.00	0.01
Jiri	0.03	0.03	0.02	0.02	0.05	0.01	0.00	0.02

Table V.2. Seasonal geometric and arithmetic mean concentrations and standard deviations (neq scm^{-1}) of water soluble inorganic species in aerosol at Phortse and Jiri aerosol

	Phortse				Jiri			
	Post-mon. n=17	Winter n=14	Pre-mon. n=17	Monsoon n=9	Post-mon. n=17	Winter n=10	Pre-mon. n=13	Monsoon n=26
Na^+	0.22	0.27	1.22	0.61	0.49	1.45	3.67	1.04
	0.25	0.46	1.79	1.16	0.48	1.64	3.23	1.12
	(0.13)	(0.57)	(1.40)	(1.57)	(0.33)	(1.26)	(1.65)	(0.78)
NH_4^+	9.17	6.20	15.50	2.08	12.47	29.11	19.71	8.62
	10.67	8.32	86.30	14.45	11.17	34.26	21.20	8.00
	(7.34)	(7.31)	(143.41)	(40.51)	(6.34)	(32.02)	(14.02)	(4.80)
K^+	1.27	1.51	3.95	0.41	3.20	6.44	6.48	2.52
	1.43	1.87	12.79	3.73	2.68	5.45	6.56	2.08
	(0.88)	(1.11)	(19.99)	(12.92)	(1.06)	(1.97)	(4.13)	(0.60)
Mg^{2+}	0.29	0.59	2.24	0.15	1.09	2.58	6.48	1.30
	0.32	0.79	8.34	1.09	0.96	2.48	5.47	1.19
	(0.18)	(0.68)	(13.28)	(3.35)	(0.50)	(1.43)	(2.01)	(0.63)
Ca^{2+}	1.44	3.38	5.77	0.65	3.16	10.68	27.55	2.00
	1.76	4.92	19.40	2.63	3.52	11.78	23.30	2.25
	(1.16)	(4.07)	(27.40)	(6.46)	(3.04)	(8.35)	(8.61)	(2.57)
Cl^-	0.04	0.17	0.39	0.34	0.05	0.48	0.31	0.08
	0.06	0.19	1.31	0.48	0.04	0.60	0.33	0.07
	(0.06)	(0.12)	(2.23)	(0.89)	(0.04)	(0.62)	(0.22)	(0.05)
NO_3^-	1.18	2.74	11.11	0.25	0.73	9.42	7.67	0.33
	1.43	3.78	23.66	5.48	0.99	13.81	7.34	0.40
	(0.94)	(2.93)	(40.74)	(19.77)	(1.16)	(14.04)	(5.17)	(0.46)
SO_4^{2-}	7.95	11.58	40.98	1.77	18.95	39.57	16.90	5.38
	8.43	15.13	78.52	14.10	16.66	40.24	39.97	9.93
	(4.20)	(11.24)	(115.06)	(43.66)	(8.09)	(23.67)	(25.10)	(7.24)

The three values presented in the table are geometric mean, arithmetic mean, and arithmetic standard deviations (in parantheses). Post-monsoon, winter, pre-monsoon, and monsoon seasons represent samples taken in Oct. to Nov., Dec. to Feb., Mar. to Jun. 20, and Jun. 21 to Sep. respectively.

Table V.3. Inter-species correlation coefficients

	Phortse								Jiri							
	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
	All samples								Sep., 1996-Mar., 1997 samples							
Na^+	1.00	0.30	0.21	0.21	0.23	0.14	0.15	0.28	1.00	0.58	0.75	0.94	0.94	0.67	0.71	0.60
NH_4^+		1.00	0.89	0.91	0.86	0.81	0.86	0.93		1.00	0.68	0.57	0.50	0.87	0.95	0.67
K^+			1.00	0.94	0.92	0.91	0.97	0.97			1.00	0.79	0.66	0.61	0.68	0.25
Mg^{2+}				1.00	0.98	0.95	0.97	0.97				1.00	0.97	0.69	0.69	0.48
Ca^{2+}					1.00	0.94	0.96	0.96					1.00	0.65	0.65	0.56
Cl^-						1.00	0.97	0.90						1.00	0.94	0.72
NO_3^-							1.00	0.96							1.00	0.73
SO_4^{2-}								1.00								1.00
									Apr. 1997-Nov., 1997 samples							
Na^+									1.00	0.68	0.81	0.92	0.83	0.68	0.81	0.81
NH_4^+										1.00	0.72	0.78	0.71	0.57	0.72	0.83
K^+											1.00	0.93	0.93	0.56	0.91	0.89
Mg^{2+}												1.00	0.96	0.73	0.94	0.92
Ca^{2+}													1.00	0.74	0.96	0.92
Cl^-														1.00	0.77	0.68
NO_3^-															1.00	0.90
SO_4^{2-}																1.00

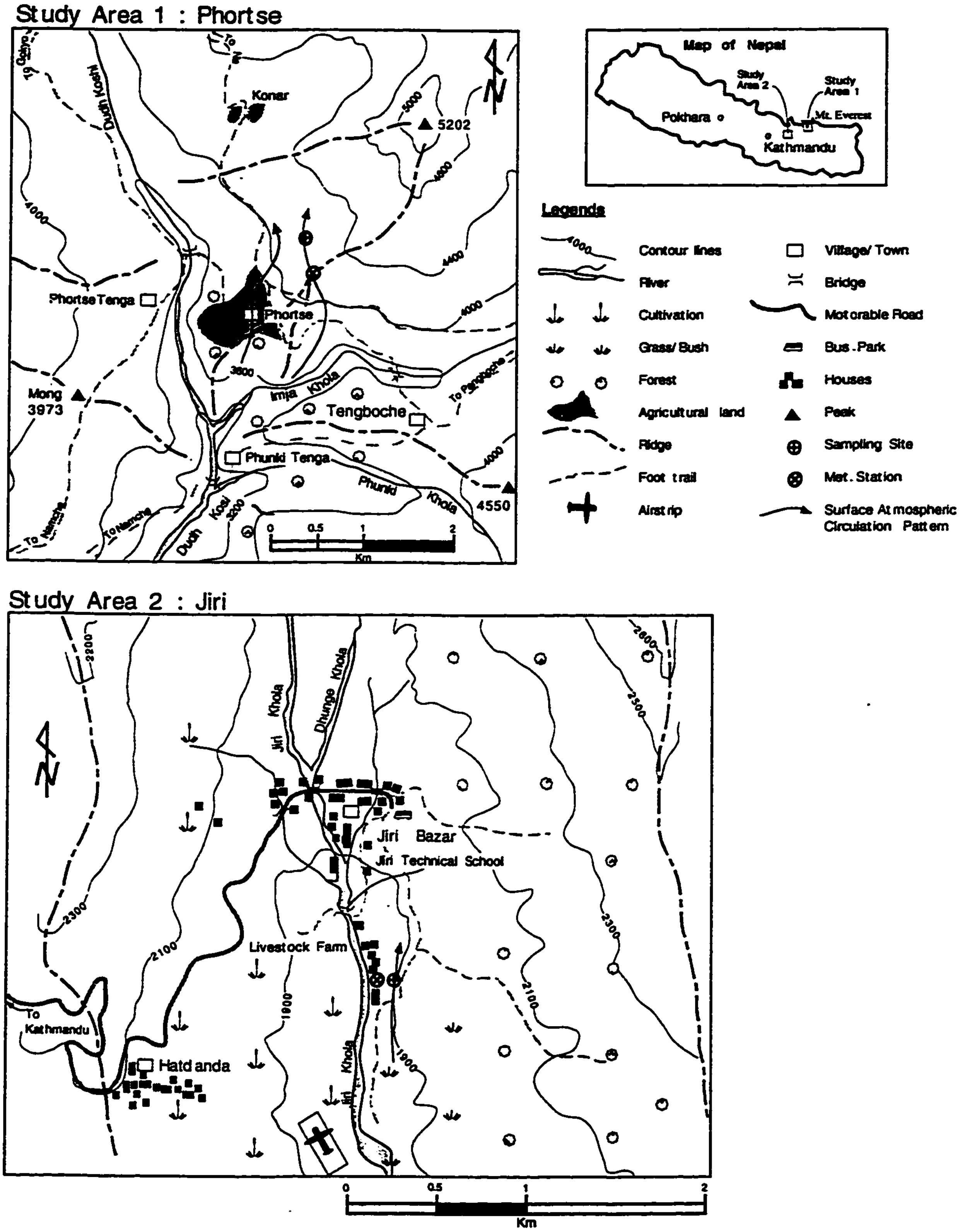


Figure V.1. Location map of Phortse and Jiri sampling stations. Arrows indicate predominant surface wind flow near the sampling stations

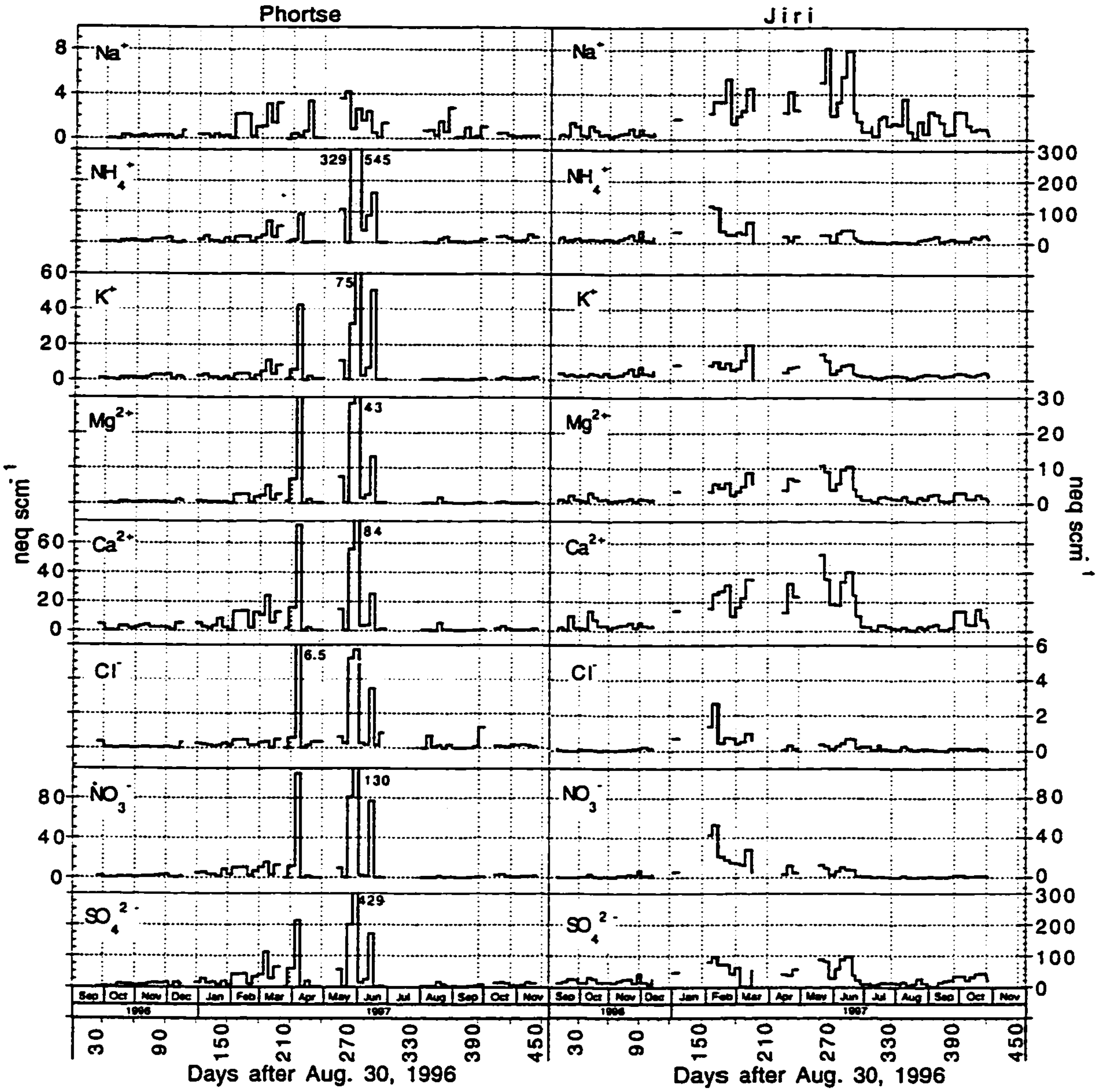


Fig. V.2 Time series of water soluble inorganic ions in aerosol measured in Phortse(left) and Jiri (right).

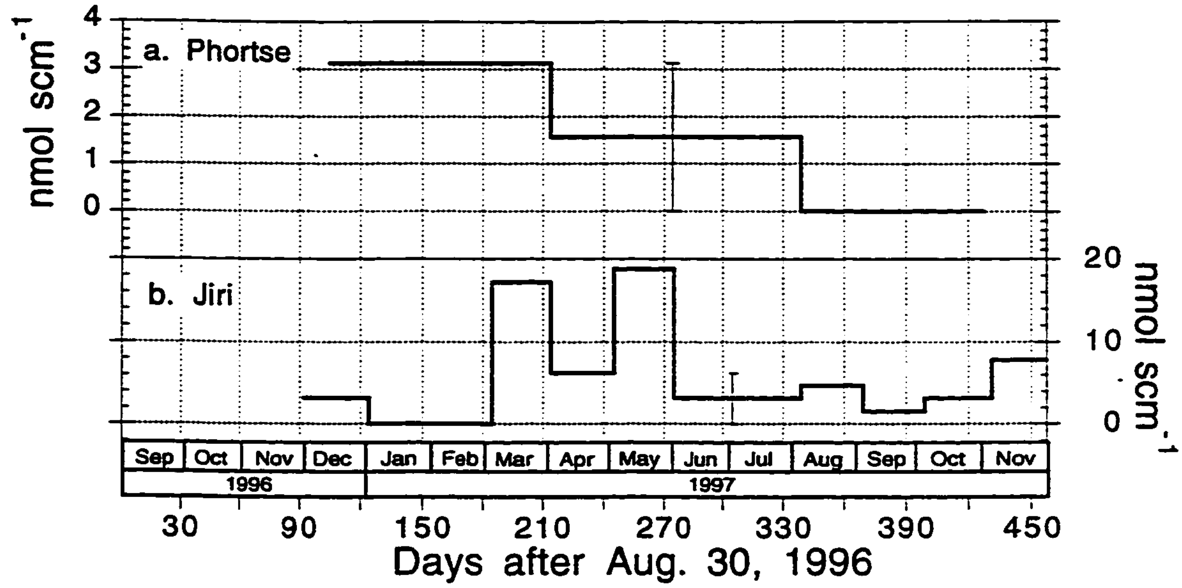


Figure V.3. SO_2 concentrations at: a) Phortse and b) Jiri.

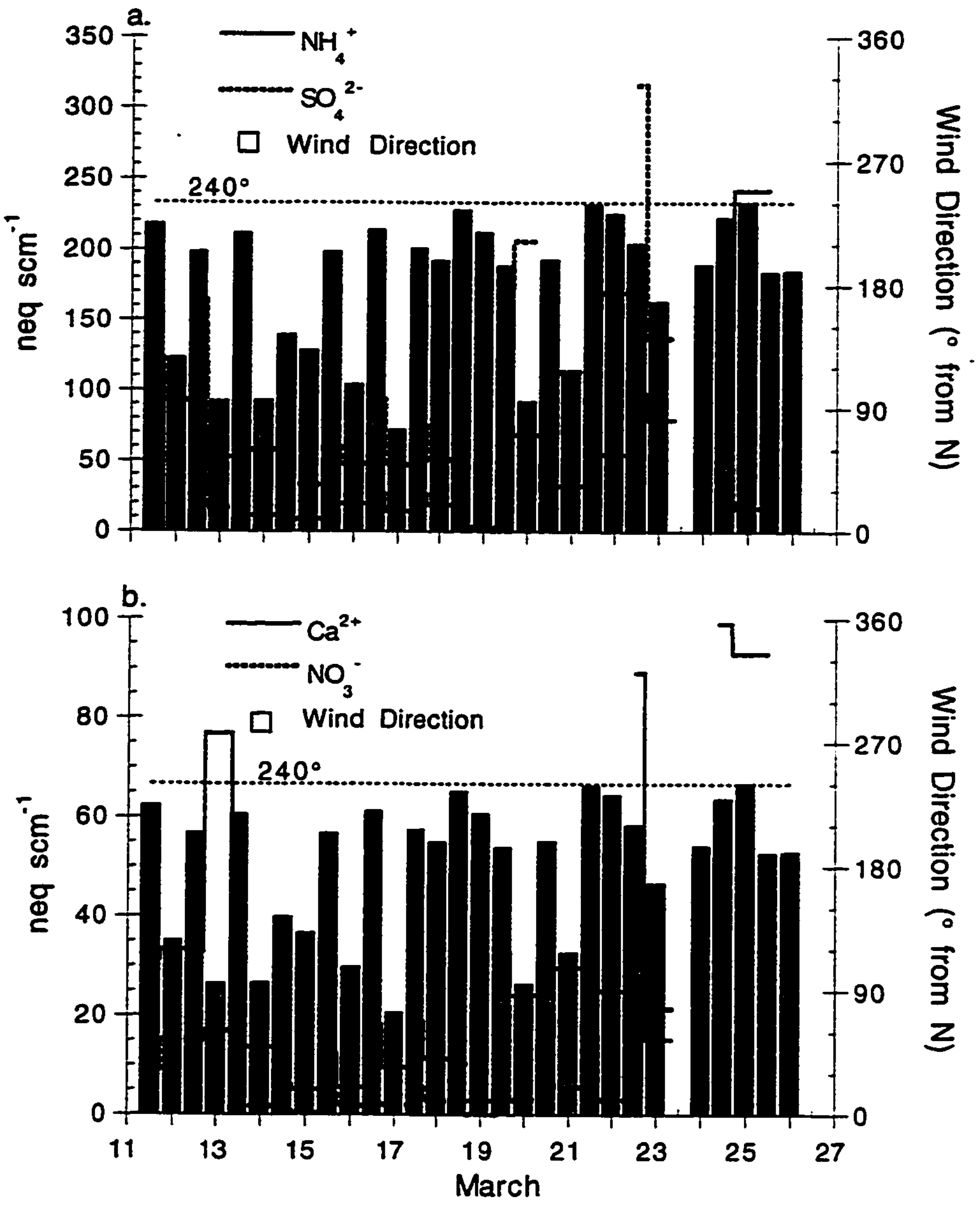


Figure V.4. Twelve hourly wind direction data (bars) and major ions concentrations of selected species in aerosol (lines) measured at Phortse during the March, 1997 intensive sampling program. Major ticks indicate 00:00 hours and minor ticks indicate 12:00 hours.

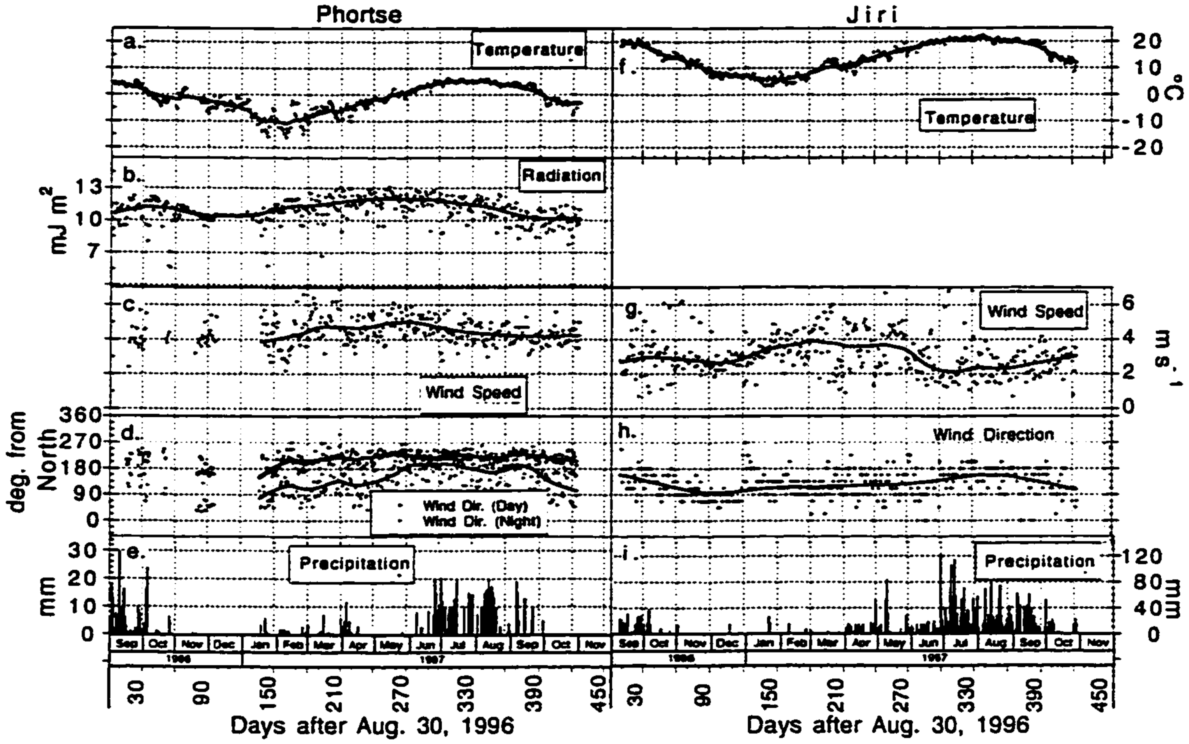


Figure V.5. Meteorological data from Phortse reference stations: a) daily mean temperature at Dingboche; b) solar radiation at Lobuche; c) daily mean wind speed at Dingboche; d) day time (6:00-17:00) and night time (18:00-5:00) mean wind directions at Dingboche; and e) precipitation at Phortse and from Jiri: f) daily mean temperature; g) daily mean wind speed; h) daily mean wind directions; and i) daily precipitation. Note different vertical scales for precipitation at Phortse and Jiri.

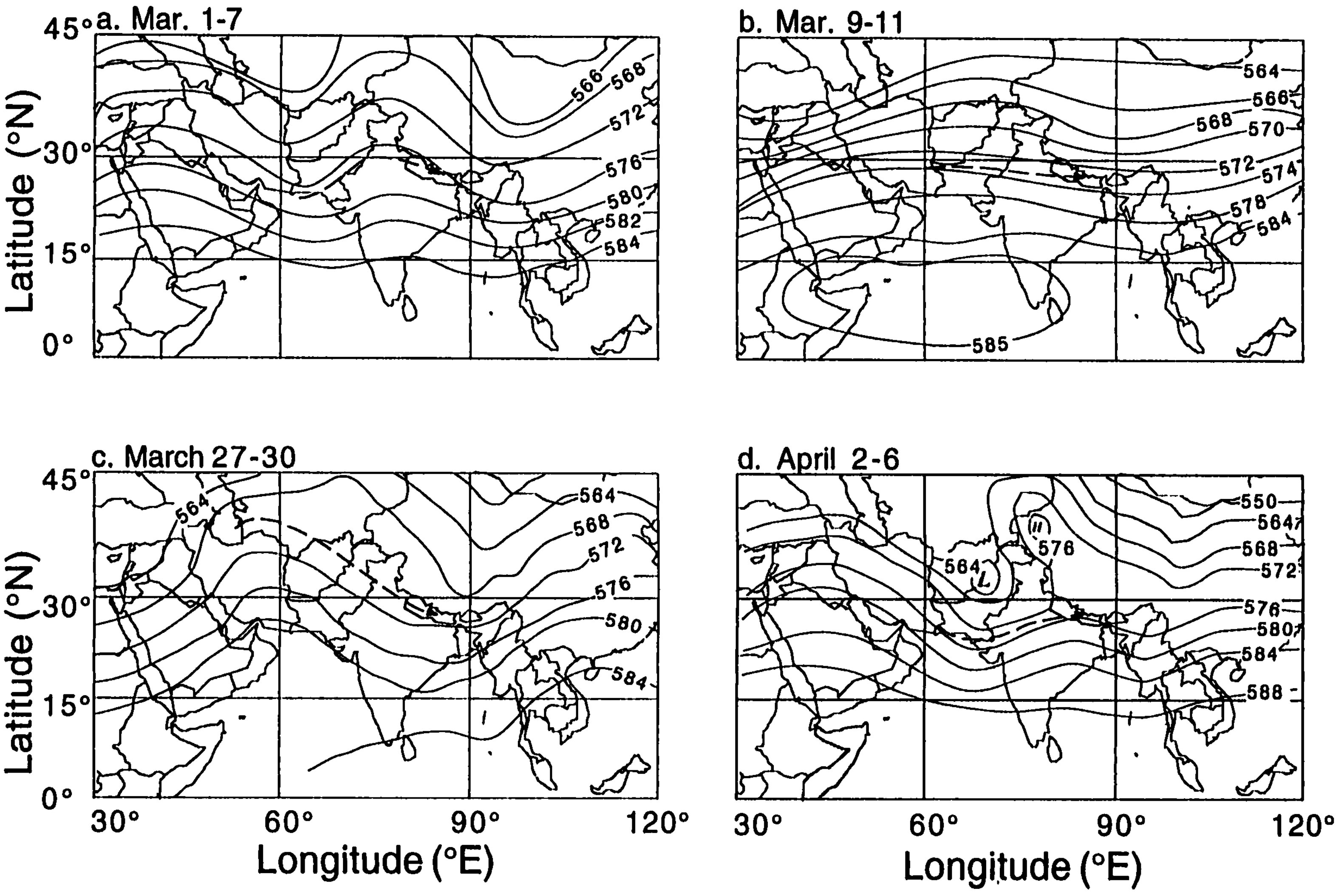


Figure V.6. Geopotential heights (x 10 m) at 500 mb level . The arrows indicate tentative trajectory of airmasses arriving at sampling sites

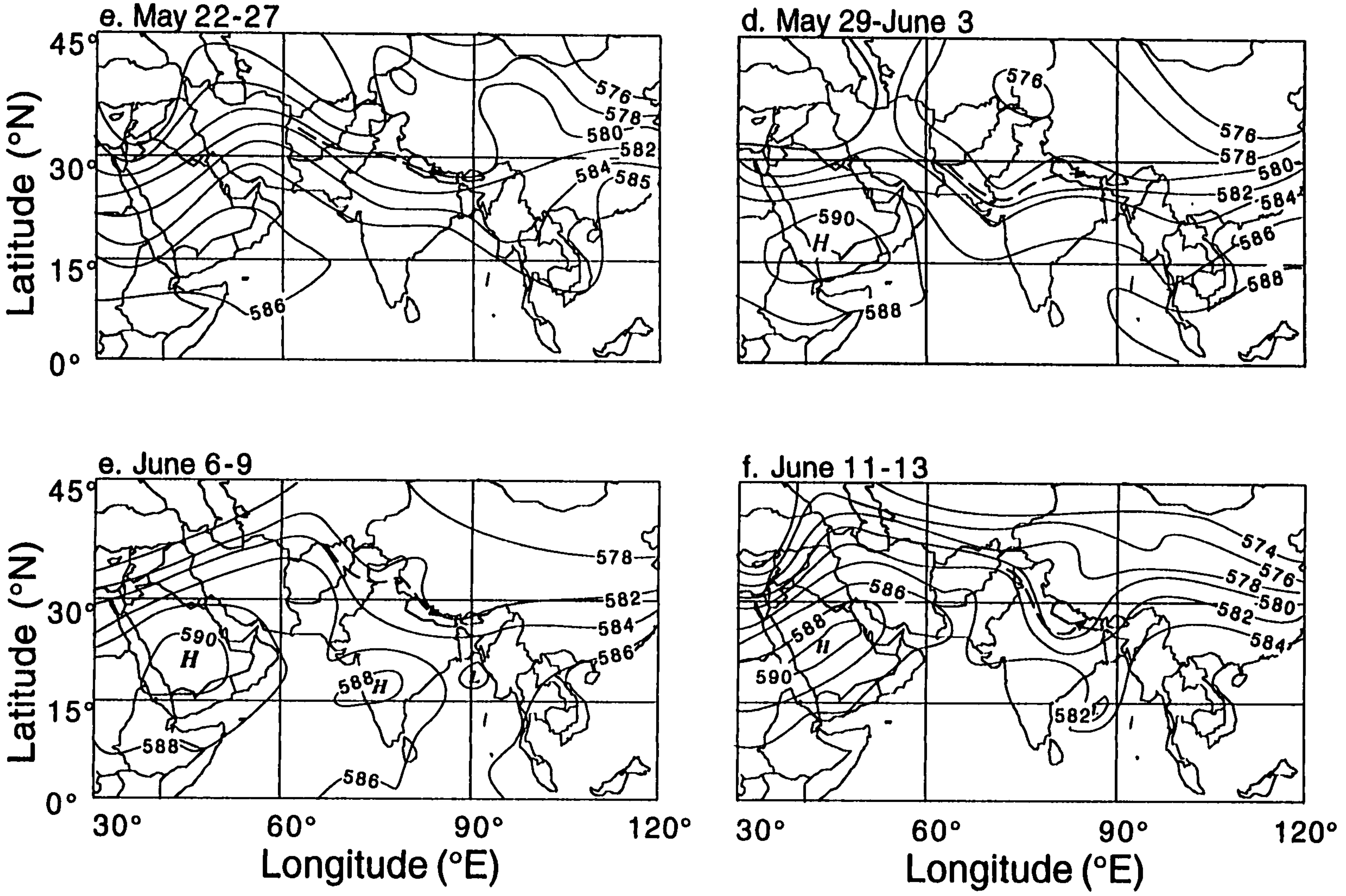


Figure V.6. Continued

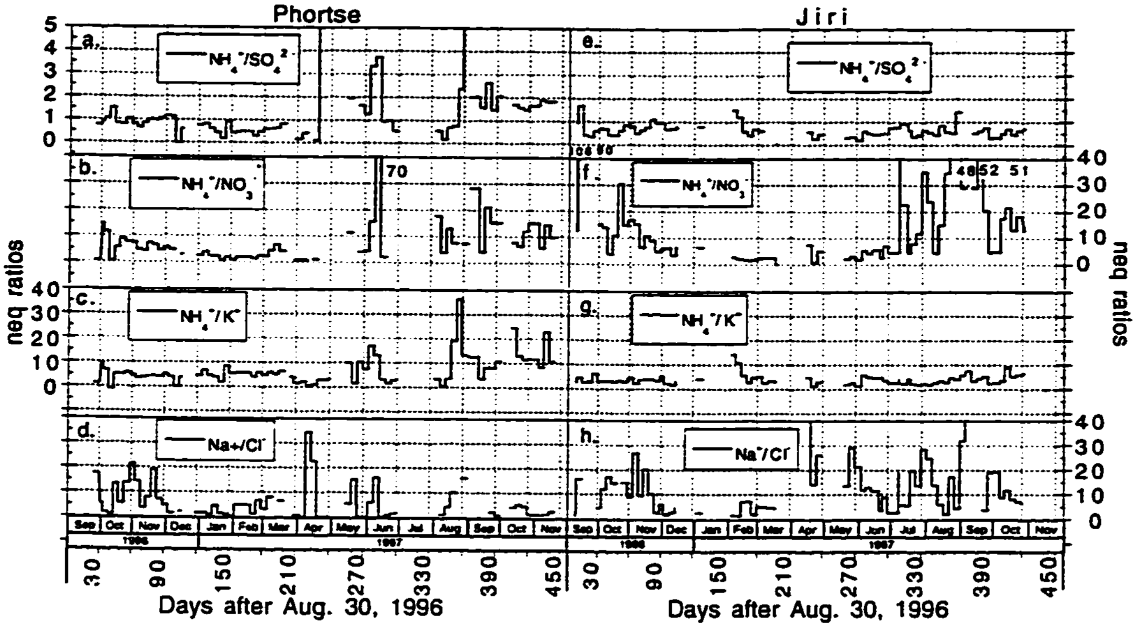


Figure V.7. Ratios between aerosol species: a) NH_4^+/SO_4^{2-} ; b) NH_4^+/NO_3^-; c) NH_4^+/K^+; and d) Na^+/Cl^- at Phortse and e) NH_4^+/SO_4^{2-}; f) NH_4^+/NO_3^- g) NH_4^+/K^+; and h) Na^+/Cl^- at Jiri.

CHAPTER SIX

AEROSOL AND PRECIPITATION CHEMISTRY AT A REMOTE HIMALAYAN SITE IN NEPAL

This chapter will be submitted to Tellus for publication

VI. AEROSOL AND PRECIPITATION CHEMISTRY AT A REMOTE HIMALAYAN SITE IN NEPAL

ABSTRACT

As a part of a year long aerosol measurement program in the Nepal Himalayas, a series of 12 hour samples of aerosol and event based samples of precipitation were collected in the post-monsoon, 1996; pre-monsoon, 1997; and monsoon, 1997. Results show that diurnal variations in the local valley wind system are responsible for variations in the major ion (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, and SO_4^{2-}) concentrations in the aerosol. On time scales greater than a day, changes in atmospheric circulation and associated changes in the precipitation regime have an important effect on aerosol ion concentrations.

Periods of high aerosol concentration generally coincide with similar periods in precipitation chemistry, although a linear relationship between concentrations in these two media is not robust due to limited data. High scavenging ratio values are found for all species except SO_4^{2-}. Nitrate, Cl^- and NH_4^+ are enriched in the precipitation compared to the aerosol, implying the presence of gaseous acidic species (HNO_3 and HCl) and gaseous NH_3 in the air. Ammonium is also enriched with respect to SO_4^{2-} in aerosol during a dry episode in the monsoon season. This may represent a relatively close source of NH_3 (from neighboring villages), that survived scavenging due to lack of precipitation.

Empirical Orthogonal Function (EOF) analysis clearly shows the presence of two dominant pollutant transport mechanisms for the pre-monsoon and monsoon seasons (i.e., valley wind system and the monsoon circulation). Although physically different, these two transport mechanisms follow similar transport pathways, bringing air into the Himalaya from similar source regions. Further, EOF analysis shows a southerly maritime signal in

the aerosol during monsoon and a more distant westerly maritime signal during pre-monsoon. Our results support the potentials for using glaciochemical records from the Himalaya to investigate variations in the strength of monsoon circulation and westerly disturbances in the past.

INTRODUCTION

Ice cores retrieved from Greenland and Antarctica contain atmospheric chemistry records ranging from seasons to hundreds of thousand of years, providing insight into climatic variations over these periods (De Angelis et al., 1987; Delmas and Legrand, 1989; Jouzel et al., 1990; Mayewski et al., 1993; 1994; and 1996). Tropical and sub-tropical glaciers do not contain records as far back in the past, however they may provide information concerning past climatic changes in regions where such information is virtually non existent (e.g., Thompson et al., 1984; Lyons et al., 1991). The vast glaciated regions in the Himalayas contain valuable information on monsoon variations over the past several thousand years. Quantitative estimates of past atmospheric chemical composition based on glaciochemical data depends greatly on the knowledge of the relationship between chemistry of the precipitation and that of the air-mass within which it forms. These relationships are site specific, so an interpretation of glaciochemical data from the Himalayas requires knowledge of the relationship between air and precipitation chemistry in the region.

Studies on the chemical composition of air and precipitation in the Himalaya have been limited to date on both temporal and spatial scales. A short-term sampling program in the Nguzompa glacier basin near Mt. Everest showed low ion burdens in both air and snow (Wake et al., 1994). A two week sampling project in Hidden Valley in the Himalayas of western Nepal showed that temporal variations in aerosol concentrations were related to the influx of air-masses by monsoon circulation (Chapter IV). Recently, a year long sampling of atmospheric aerosol (with sampling intervals of 5 days) was conducted at a remote Himalayan site and rural Middle-Mountain site in Nepal (Chapter V). This study showed marked seasonal variations in the aerosol concentrations due to changes in circulation and associated precipitation scavenging. As part of the same program, aerosol samples were collected over 12 h sample intervals at the Himalayan site during three different seasons, during which precipitation samples were also collected. This paper investigates the variations in aerosol concentrations during the intensive sampling programs and their connection with meteorological variables and transport mechanisms. We also examine the relationship between aerosol concentrations and concentrations in the precipitation.

THE SITE

The sampling site was located near the remote village of Phortse in the Khumbu region of the Himalayas (Fig. V.1). The sampling site stands at an elevation of 4100 m a.s.l. at 27°51' N latitude and 86°45' E longitude and is situated about 350 m above the village, close to where a northwest to southeast trending ridge merges with the mountain slope. The ridge continues down to the confluence of Imja Khola and Dudh Kosi rivers, at an elevation of 3550 m a.s.l. A meteorological station was established 50 m below the aerosol sampling site, on the same ridge. Although the predominant wind direction over the valley is south-westerly, observations of the local wind reveal that wind near the sampling station blows predominantly from south-south-east direction (across the ridge). Therefore the sampling site is well protected from sources of pollution in the village. The village of Phortse consists of about 60 permanent houses. There are small patches of farm-land around the village, where potato and buckwheat are cultivated between April and September. There are also some forested areas dominated by juniper and different varieties of pine. A small airstrip at Syangboche is located at a distance of about 4 hours walk (5 km aerial distance) and a slightly larger airport, Lukla, is 3 days walk from the Phortse village. The nearest road head, Jiri, is at a distance of 7 days walk (ca. 70 km aerial distance).

EXPERIMENTAL

Aerosol Samples

The sampling described in this paper was part of a year-long aerosol sampling program at Phortse (details are provided in Chapter V). During three different periods, regular 5 day integrated sampling was replaced by intensive sampling programs during which samples were collected over time intervals of 6 to 12 hours. These intensive sampling programs were conducted between Sept. 24 to 29 ,1996; Mar. 11 to 29, 1997; and Aug. 11 to 31, 1997. The first sampling was relatively short due to a pump breakdown. Therefore only data for the pre-monsoon and monsoon samples are discussed in detail.

The sampling method was the same as that detailed in Chapter IV, therefore only a brief description is presented here. The air was drawn by a 24 V DC high-volume, constant speed pump powered by a combination of photovoltaic cells and batteries. The volume of air sampled was measured with an in-line flow meter/totalizer. Corrections for ambient temperature and pressure allowed conversion of the measured volumes to standard cubic meters (scm).

After sampling, filters were brought to Kathmandu, where they were stored for several days. The filters were then shipped to the Climate Change Research Center (CCRC) at the University of New Hampshire by air cargo for laboratory analysis. Earlier sampling in the Himalayas showed an abundance of NH_4^+ in the aerosol and it was thought that post-sampling reaction with NH_3 in the ambient air might have created a positive artifact for NH_4^+ (Haagenson and Shapiro, 1979; Hayes et al., 1980; Silvente and Legrand, 1993).

To avoid contamination by ambient air as well as evaporation of sampled species during the transport and storage, filters were removed from their holders and were individually placed in pre-cleaned air-tight glass bottles immediately after sampling at Phortse. The glass bottles were then placed in air-tight dark containers. The filters were removed from glass bottles just prior to chemical analysis. The filters were analyzed for major water soluble ionic species at the CCRC laboratory using a Dionex ion chromatograph. The detection limits for each species, determined as described in Chapter IV, are presented in Table VI.1.

Precipitation Samples

Precipitation samples were collected in 500 and 1000 ml polypropylene jars with an orifice diameter of 150 mm (larger jars were used for collecting solid precipitation). The pre-cleaned jars were packed and sealed in clean bags. Prior to exposure, the jars with caps on, were removed from plastic bags and mounted on a 1.5 m high stand. Next, the cap was removed from the jar and placed in the original plastic bag. After sample collection, the jar was sealed tightly by the cap and was placed in air-tight dark containers. Between 6 to 12 hours after sampling, the precipitation samples were transferred to 30 ml polypropylene bottles. Depending on the amount of precipitation two to four aliquots were prepared for each precipitation event. To avoid changes in concentrations due to bioconsumption, half of the aliquots were treated with ultra pure chloroform (1% of the aliquot volume; Galloway, 1978). The polypropylene bottles were then placed in dark air-tight containers.

Precipitation samples were also analyzed at the CCRC laboratory using a Dionex ion chromatograph. Ammonium concentrations were determined as average values of aliquots treated with chloroform, while Cl^- values were determined from untreated samples. For other species, average values of all samples from each precipitation event were used. Standard deviations of multiple samples representing same precipitation event were adequately low (<5% of the mean).The amount of precipitation was determined by the readings from a rain gauge. Detection limits, determined as described in Chapter IV are presented in Table VI.1. The first precipitation sample from the pre-monsoon sampling program is suspect because of unrealistically high (>200 times the average) concentration for all species except NH_4^+ and NO_3^- and is therefore not included in further analyses.

Meteorological Data

A meteorological station established and operated in conjunction with the aerosol sampling program included a thermohygrograph, an actinograph, an anemometer, a rain gauge, and an altimeter barometer. General weather condition at the site such as cloud cover and movement, visibility, fog, mist etc. were observed and noted twice daily at standard meteorological observation times for Nepal (0845 and 1745 h) during the sampling program. Synoptic weather maps prepared by the Department of Hydrology and Meteorology, Nepal were acquired to obtain information on synoptic scale circulation during the sampling programs. Wind direction data was not obtained during the 1997 monsoon due to instrument malfunction. For that period, data from a permanent meteorological station at Dingboche (about 5 km to the north-east from Phortse) is used. The wind data from Dingboche was well correlated with wind data from Phortse during other sampling periods.

RESULTS AND DISCUSSION

Role of Precipitation and Circulation Variations on Aerosol Concentrations

Diurnal Variation

The mountainous terrain is characterized by distinct diurnal variations in the local wind pattern. Generally the wind is up-slope (valley wind) during the day-time and down-slope (mountain wind) during the night-time (Nakajima, 1976; Hindman, 1995a; Hindman, 1995b). The timing of the diurnal wind variation depends on the geometry of the valley and season. Time series of water soluble ionic concentrations in aerosol measured during the pre-monsoon and the monsoon sampling programs are presented in Figure VI.1. Initially, three samples were taken daily in the beginning of the post-monsoon sampling at 0800 to 1400, 1400 to 1800 and 1800 to 0800 hours. The two day-time samples were similar for most species, except for SO_4^{2-}. Afternoon samples show even higher concentrations of SO_4^{2-} compared to morning samples (figure not shown). The afternoon samples should show lower concentration if the reversal of local wind took place much before 1800 hours. The lack of significant and consistent differences between the two day-time samples combined with the study of local wind patterns, indicates that the mountain wind dominates only after 1700 h and the day-time samples were affected primarily by valley wind. In the sampling programs thereafter, only one day-time (0800-1800 h) and one night-time sample (1800-0800 h) were collected daily.

The night-time samples generally show lower concentrations than day-time samples. We selected events for NH_4^+, NO_3^-, and SO_4^{2-}, three dominant ionic species in aerosol, when consecutive day-time and night-time samples were taken (Fig. VI.2). There are 19 such events with conspicuous differences in day-time and night-time concentrations. Among these, day-time samples show higher concentration than night-time for 12 events while night-time samples show higher for the remaining 7 events. In the pre-monsoon sampling program only night-time samples collected between March 19/20 and 21/22 show considerably higher concentrations of SO_4^{2-} compared to day-time samples (Fig. VI.2 right).

In general, a close relationship existed between variations in aerosol concentrations and precipitation events (Fig. VI.1; Fig. VI.3). Precipitation was generally nocturnal in Phortse, which could have resulted in observed diurnal variations in aerosol concentration (Fig. VI.2). However, there were several instances when nocturnal precipitation did not occur and night-time concentrations are still reduced (e.g., March 15, 17 and 21). Besides precipitation, the diurnal variation in regional scale wind systems (Chapter V) could also effect the aerosol concentrations. Pollutants transported to the site by regional scale valley wind systems during the day-time is replaced by relatively cleaner air form aloft by mountain wind system in the night-time. The north-easterly wind direction during the night on March 15 and 20 indicates the presence of mountain wind at the sampling site (Fig. VI.3). The differences in day-time and night-time samples, even when there was no precipitation event, suggests that the role of diurnal changes in the wind pattern is the dominant control on the diurnal fluctuation of aerosol concentrations. Coincidentally, after March 23 in the pre-monsoon samples, there was no precipitation and the mountain wind system also ceased (Fig. VI.3).

In contrast, night-time concentrations are substantially higher than day-time from Aug. 22 to 24 and on Aug. 27 for most species (Fig. VI.2). Wind direction data from Dingboche shows the presence of mountain winds during this period (Fig. VI.3). Mountain winds occur over a wide area in the Khumbu region (Hindman et al., 1996) and the observation of low level cloud movement at Phortse did show diurnal changes in the local winds. Occasionally, during the early-monsoon, the Phortse station experienced higher levels of pollution compared to a lower elevation station at Jiri, which was suggested to be due to influence of a polluted monsoon layer in the atmosphere above the Phortse site (Chapter V). It is likely that the site was under the influence of the pollution layer during the later part of the monsoon intensive sampling program and the nocturnal mountain wind could readily transport polluted air-masses to the Phortse site causing night-time rises in major ion concentrations during this dry period. An airborne measurement of aerosol species and meteorological parameters is planned in the near future in the Nepal Himalaya, which would provide valuable insight into the existence of a pollution layer overriding relatively clean air-masses (per. comm., E. Hindman, City College of New York).

Alternatively, night-time mountain wind can also bring pollutants from larger villages (larger pollution sources) up-valley. During night-time flows move along mountain slopes (as revealed by cloud movement), merge at valley bottoms or in the river valley and flow down valley. This pollutant transport mechanism is not likely to influence the Phortse site as it is situated on the valley wall and not in the valley bottom. An aerosol measurement program was conducted near Dingboche in November, 1995 at a site slightly higher and located to the south-west of the village itself. The measurement showed low concentrations of major ionic species, although down-valley wind occurred during the sampling period (Table VI.2), This suggests that the influence of local emission due to down-valley wind is negligible even on such short distance due to the elevated location of the sampling site with respect to the village, therefore its influence on Phortse samples is not likely.

Aerosol Concentration Variations at Time Scales of Multiple Samples.

Pre-monsoon concentrations of NH_4^+, K^+, Mg^{2+}, Ca^{2+}, NO_3^-, and SO_4^2 are 2 to 10 times greater than the monsoon concentrations (Fig. VI.1).The differences in aerosol concentrations between the two sampling periods (pre-monsoon, and monsoon) are a part of even larger seasonal variations. The seasonal variations are discussed in detail in Chapter V. The discussion herein focuses on concentration variations within each sampling season.

Concentration variations at time scales of several samples are characteristic of the pre-monsoon and monsoon concentration series (Fig. VI.1). At these time scales, precipitation events were generally associated with decreases in aerosol concentrations. For example, during the pre-monsoon sampling program concentrations of most ionic species decreased dramatically following precipitation events in March (i.e., March 13 to 14, 17/18 and 19/20). However, the precipitation of March 23 was of low intensity and apparently did not significantly alter aerosol concentrations. The large scale circulation at 500 mb was north-westerly during this period (Fig. VI.4a). Unlike during the monsoon, air-masses arriving at upper levels are relatively clean and this circulation pattern does not support advective transport of pollutants from the south (Chapter V). The high concentration levels between March 20 and March 25 coincide with a change in upper air flow to a south-westerly direction (Fig. VI.4b). This shift in large scale circulation likely incorporated more pollutants from distant southerly and south-westerly sources. After March 25, the 500 mb circulation re-established the pattern similar to that at the beginning of the month (figure not shown).

The monsoon samples also display significant variations in major ion concentrations on time scales of a week. The monsoon samples can be divided into two parts, August 8 - 20 and August 20 - 28. The first period is characterized by low concentrations, close to or less than 1 neq scm^{-1} for all species except Na^+ and Cl^- (Fig. VI.1 right). The sample of August 19/20 (night) shows a large increase in concentrations of all species except Cl^-, although all ion concentrations remain significantly lower than those during the pre-monsoon period. These high concentrations continued almost until the end of the sampling period, when they returned to the lower levels, characteristic of the first half of the monsoon sampling (Aug. 8 - 20). The sequence of high and low concentrations of atmospheric aerosol on weekly time scales in the monsoon sampling period matches similar sequence of variations in amount of precipitation. Precipitation record for the monsoon sampling program shows two distinct periods: a wet period between Aug. 8 and Aug. 17 and a substantially drier period between Aug. 18 and Aug. 27. The last days of the monsoon sampling program (Aug. 28-30) shift back into a wet period. This sequence of wet and dry periods may be related to the quasi 10-day oscillation in the monsoon circulation associated with cycles of depressions over the Bay of Bengal and the Tibetan upper level high (Yasunari, 1976). Analyses of synoptic weather maps shows that the monsoon trough shifted southward during that period (Fig. VI. 4 c and d). This caused air-masses reaching the site to change trajectories from south-easterly to easterly. Such a shift elongated the travel distance from the Bay of Bengal and likely led to the depletion of moisture in the earlier part of the path. Moisture depleted air masses likely picked-up pollutants on the pathway and caused high concentration in aerosol between Aug. 18 and 27.

This analysis indicates that changes in precipitation regime influence variations in aerosol concentrations (during both pre-monsoon and monsoon sampling programs). However, in several cases changes in precipitation regime were associated with changes in large scale circulation. The decrease in the major ion concentrations followed by precipitation events could be due to incorporation of pollutants from different source regions. It is not possible to distinguish the relative importance of these two processes on the aerosol chemistry from the available data.

Air-precipitation Relationship

Concentrations of chemical constituents in precipitation collected during pre-monsoon and monsoon intensive sampling programs are shown in Figure VI.1. As with the aerosol samples, the concentrations of major ions in precipitation were high during the pre-monsoon season as well as in the two isolated precipitation samples of Aug. 21 and 21/22, and low in the first half of the monsoon sampling. In addition, the concentrations of water soluble species in precipitation in the post-monsoon sampling is similar to that during the first half of the monsoon sampling. Figure VI.1 shows that a period with high aerosol concentrations generally corresponds to similar periods in precipitation concentration.
As a preliminary step to quantify the relationship between concentrations in the precipitation and in the air, scavenging ratios were calculated according to:

$$W=\rho_a C_s/C_a$$

where C_s=concentrations in precipitation (ng g^{-1}); C_a=concentration in air (ng m^{-3}); and ρ_a=density of air (764 g m^{-3} after correcting for the standard temperature and pressure) (Davidson et al., 1993).

The scavenging ratios for most species were significantly higher than those found in Greenland (e.g., Davidson et al., 1993) or in the Swiss Alps (Baltensperger et al.,1993) (Table VI.4). The scavenging ratio for SO_4^{2-}, although relatively low compared to scavenging ratios of other species, is comparable to values found in Greenland. The scavenging ratios are highly variable for all species (Table VI.4). The variability may be attributed to differences in precipitation formation, atmospheric concentrations, or the extent of below cloud scavenging between individual events, as samples were collected during different weather conditions and seasons. Our data set does not allow assessment of the relative importance of the different processes controlling relationships between air and precipitation composition.

The high variability in scavenging ratios shows that aerosol concentrations are not reflected in corresponding precipitation events in sample-by-sample basis. The linear regression between aerosol concentrations and corresponding concentrations in precipitation is not significant (figure not shown). Nevertheless, major ion concentrations for most species in aerosol covary with those in precipitation when concentrations are averaged over episodes of high and low concentrations and over the two sampling seasons (pre-monsoon and monsoon) (Table VI.3). While these results show that the concentration increase (decrease) in one medium is reflected by a similar increase (decrease) in the other, the magnitude of change is not consistent and the relationship is not linear. Since the relationship significantly improved by averaging over longer time periods, it is likely that the relationship will further improve and approach a linear relationship as more data are collected in the future. This would provide greater confidence when using glaciochemical data for interpreting atmospheric composition of the past.

The mean scavenging ratio of NO_3^- is relatively high. In the aerosol, NO_3^- is equivalent to only one third of NH_4^+ concentrations, while in precipitation NO_3^- concentrations are greater than NH_4^+ (Figure VI.6a-b). The relative increase in NO_3^- concentrations is greater in the period of higher aerosol concentrations (e.g., during pre-monsoon). This suggests the presence of gaseous HNO_3 in the air (largely unsampled on our filters), especially when the air is more polluted. Relatively high scavenging ratios for NO_3^- could result from scavenging of gas phase HNO_3 by precipitation. Similarly, our results show enhancement of Cl^- with respect to Na^+ in the precipitation compared to aerosol. Chloride was in significant deficit relative to Na^+ and there was no correlation between these species in aerosol (Fig. VI.5c). In contrast, Na^+ and Cl^- were not only linearly related in the precipitation, Cl^- concentrations are increased relative to Na^+ (Fig. VI.5d). The best fit line shows that Cl^-:Na^+ in precipitation is close to the mean sea-water ratio (1.165; Drever, 1988). Such increase in Cl^- concentrations with respect to Na^+ in precipitation could be due to two factors. 1) Presence and scavenging of gaseous HCl in air can cause enrichment of Cl^- in the precipitation. The Cl^-/Na^+ ratio in the precipitation will approach the average sea water ratio if both of these components originated as marine aerosol and underwent chemical fractionation by the reaction with, for example, H_2SO_4 as they traveled to the site. Evidence of fractionation of sea salt particle by H_2SO_4 was found in Hidden Valley, where similar enhancement of Cl^- in the precipitation was observed (Shrestha et al., 1997; Chapter IV). 2) Air masses transported to the site by more vigorous circulation are likely to be less fractionated due to shorter travel time. Since airmasses causing precipitation represent stronger circulation, enrichment of Cl^- in the precipitation could be because of the bias of sampling such air masses. When only the aerosol data during the precipitation events in Phortse were examined, the relationship between Cl^- and Na^+ improved remarkably (r=0.83) and the ratio between Cl^- and Na^+ is 0.6. Although this ratio is only half of the mean sea-water ratio, it is significantly higher than when all aerosol data is considered. Further, when only aerosol data during monsoon precipitation events (excluding the two samples of Aug. 22 and 22/23 with high concentrations) are considered, a similar ratio is obtained, while the correlation is slightly lower (r=0.72).

It is common to observe enrichment of SO_4^{2-} relative to NH_4^+ in precipitation due to SO_2 oxidation in clouds (e.g., Calvert et al., 1985). On the contrary, our samples display relative enrichment of NH_4^+ in the precipitation with respect to SO_4^{2-} (Fig. VI.5e and f; Fig. VI.6a; Fig. VI 6b (Aug. 9-13)). Similar enrichment was also observed in Hidden Valley , although enrichment of NH_4^+ occurred to a much higher extent (Shrestha et al., 1997). Poor scavenging of SO_4^{2-} could have been responsible for the relative enrichment of NH_4^+. Low scavenging ratio of SO_4^{2-} compared to all other species in our samples supports this. However, different scavenging rates should not occur if SO_4^{2-} and NH_4^+ are internally mixed. Southerly air-masses that bring in precipitation travel over populated areas, forests, and cultivated lands in the lower mountain and plains in Nepal before reaching the Phortse site. These air-masses could accumulate additional gaseous NH_3 emitted from animal manure and fertilizers. As there are no industrial activities in this intermediate region (ca. 50 km from the site), additional sources of SO_4^{2-} are insignificant. Low SO_2 concentrations measured in rural and semi-urban sites supports that there are no significant SO_2 (and SO_4^{2-}) sources in Nepal (e.g., Carmichael et al., 1995). While concentrations of all species are elevated in the period between Aug. 20 and 28, this period is also characterized by enrichment of NH_4^+ with respect to SO_4^{2-} in the aerosol (Fig. VI.6b) supporting the presence of NH_3 in the ambient air.

Inter-species Relationships, Sources and Transport

Multivariate analysis was performed on the aerosol data to investigate inter-species relations with respect to sources and transport mechanisms. We used multivariate empirical orthogonal function (EOF) analysis, rather than repeated simple linear regression or the multiple linear regression, because of its ability to assess joint behavior of several species (variates). EOF decomposition provides objective representations of multivariate data by analyzing the covariance structure of its variates (Pixoto and Oort, 1992; Mayewski et al., 1997; Meeker et al., 1997). This analysis was performed on the pre-monsoon samples and the monsoon samples. Aerosol concentrations below detection were represented by the detection limits.

The signs and magnitudes of variance explained by EOF1 shows that 6 of the major ions (excluding Na^+ and Cl^-) are strongly loaded in both pre-monsoon and monsoon samples (Table VI.4). Chloride shows negative loading both in pre-monsoon and monsoon samples. The first EOF is interpreted to represent well mixed air-masses. High loading of six species indicate long travel distance from source regions (Heidam, 1984; Mayewski et al., 1997; Meeker et al., 1997). Thermal advection represents the dominant transport mechanism during the pre-monsoon season, while monsoon air-masses carry pollutants to the site during the monsoon season (Chapter V). While these two transport mechanisms are driven by separate processes, their transport pathways are similar, in the sense that both flow from the south. This implies that the valley wind system, due to thermal advection, is relatively extensive, and the source regions are similar to those of large scale monsoon circulation, hence it causes similar variations in aerosol concentrations.

Sodium and Cl^- dominate the second EOF for both seasons suggesting that sources/transport of Na^+ and Cl^- are different from those of other species (Table VI.4). EOF1 explains 18% of variance in Na^+, while a majority of Na^+ variance is explained by EOF2 (54%) . This implies that Na^+ has dual sources. The loading on EOF1 may support an association with a crustal source. The crustal source of Na^+ is insignificant during the monsoon season (2%; Table VI.4). High loading of Na^+ and Cl^- on EOF2 for both seasons is a surprising result. Joint loading of these two species is indicative of marine aerosols. It is natural to expect some influence of marine aerosol in the monsoon season. Crustal sources of Na^+ and Cl^- (evaporites) from arid regions of central Asia transported by the westerly circulation is precluded because of the lack of Ca^{2+} and Mg^{2+} loading in this EOF. The occasional precipitation during winter and pre-monsoon has its moisture source at the Mediterranean Sea (westerly disturbance; Mani, 1981; Nayava, 1980). It is likely that the EOF2 during pre-monsoon is reflecting the distant maritime origin of moisture transported by westerly flow. Although Mg^{2+} is also a major component of sea salt it is absent in the second EOF possibly because its crustal fraction is much larger than the sea salt fraction.

Unlike EOF1 and EOF2, EOF3 is different for the two seasons and shows mixed loading both in sign and magnitude (Table VI.4). Loading of NH_4^+ and K^+ may represent a biomass burning signal. The pre-monsoon is a season when biomass burning is common in the low-lands and hills of Nepal (Chapter V). Similar loading of Cl^- could be because a fraction of NH_4^+ is associated with Cl^-. This can happen if NH_4^+ is in excess after completely neutralizing NO_3^- and SO_4^{2-} and there is gaseous HCl present in the ambient air (Harrison and Pio, 1983).

SUMMARY AND CONCLUSIONS

Intensive aerosol sampling during post-monsoon, pre-monsoon and monsoon seasons at Phortse in the Nepal Himalayas showed diurnal variations and variations at the time scale of several samples (i.e., week) in the concentrations of water soluble inorganic chemical species. Generally, higher concentrations in the day-time and lower concentrations in the night-time samples reflect the diurnal variation in regional scale valley wind systems and also the influence of scavenging of particles by predominantly night-time precipitation events, although the influence of the former is dominant. More distinct variations in aerosol concentrations occurring at weekly time scales is related to precipitation events. At this time scale cleansing of atmospheric constituents by precipitation resulted in decreased concentrations. However, these precipitation events also coincided with considerable changes in circulation; changes in the source regions associated with the changes in the circulation may therefore also be affecting the aerosol concentrations.

Event based precipitation sampling showed that precipitation chemistry in general mimics variability in aerosol chemistry. While the aerosol concentrations are not reflected in corresponding precipitation events on a sample-by sample basis, the relationship between the ionic concentrations in the two media improve as the samples are averaged over several days to a season, supporting the potentials for using glaciochemical time series from the Himalaya to infer the atmospheric composition of the past in that region. Scavenging ratios were found to be highly variable and the mean values were relatively high for most species except SO_4^{2-}. High scavenging ratios for NO_3^- suggest enrichment in precipitation due to the presence of gaseous HNO_3 in air. Similarly, Cl^- is also enhanced with respect to Na^+ in precipitation compared to air, and in precipitation it shows a close relationship with Na^+. This is attributed to the scavenging of HCl present in the ambient air, likely of same source as Na^+ (i.e., sea salt particles fractionated due to reaction with H_2SO_4). In addition, low fractionation of precipitating air masses due to vigorous circulation is also likely responsible for the apparent enhancement of Cl^- with respect to Na^+ in precipitation compared to aerosol. Enrichment of NH_4^+ in the precipitation with respect to SO_4^{2-} is attributed to higher solubility of gaseous NH_3 compared to SO_4^{2-}. Furthermore, similar enrichment in the aerosol during a dry episode in the monsoon sampling is attributed to presence of NH_3 in the ambient air, from areas in the neighborhood of the sampling site. The present data sets are too limited to suggest the observed relationship as conclusive. Continued observations of synchronous aerosol and precipitation chemistry in the future may substantiate such relationships enabling us to better interpret the glaciochemical data from the Himalayas.

EOF analysis shows that there is a dominant transport mechanism controlling aerosol concentrations of all species except Na^+ and Cl^-, both during pre-monsoon and monsoon seasons. While large scale thermal winds and monsoon circulation are the predominant transport mechanism operating in the pre-monsoon and monsoon seasons respectively, remarkable similarities between EOF1 for both seasons suggests that both transport mechanism have similar pathways and incorporate pollutants from similar source regions. EOF2 during the monsoon represents marine contributions to the aerosol, transported by south-easterly monsoon circulation, while during pre-monsoon EOF2 likely depicts distant maritime sources transported by westerly disturbances. These results are promising concerning the use of glaciochemical data from the Himalaya to infer variations in continental contribution to the atmospheric composition of the regions and variation in relative strengths of monsoon and westerly circulation.

ACKNOWLEDGMENT

We thank villagers of Phortse and residents of Jiri town for their support to the sampling program. Dr. R.W. Talbot and Dr. B.D. Keim provided a detailed review on this chapter. We also thank DHM for providing meteorological equipment and other logistical support. Most of all, we are thankful to our Dorzee Sherpa, Phortse for his hard work. This study was partially funded by the Global Change System for Analysis Research and Training (START).

Table VI.1. Detection limits of aerosol and precipitation samples from Phortse.

Period	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
				Aerosol (neq scm^{-1})				
Sep. 1996	2.20	0.03	0.18	0.20	1.08	0.49	0.62	0.45
Mar. 1997	0.22	0.95	0.14	0.07	0.35	0.19	0.15	1.00
Aug. 1997	0.21	0.41	0.02	0.05	0.46	0.08	0.00	0.07
				Precipitation (µeq kg^{-1})				
Sep. 1996	0.04	0.02	0.00	0.03	0.04	0.17	0.00	0.00
Mar. 1997	0.00	0.00	0.00	0.01	0.07	0.01	0.00	0.00
Aug. 1997	0.00	0.04	0.02	0.00	0.05	0.00	0.00	0.00

Table VI.2. Comparision between aerosol concentrations at Phortse and Dinboche

	Phortse (August, 1997)			Dingboche (November, 1995)		
	Geometric Mean	Arithmetic Mean	Standard Deviation	Geometric Mean	Arithmetic Mean	Standard Deviation
Na^+	0.65	1.45	1.74	0.85	1.73	2.37
NH_4^+	5.45	6.03	2.94	2.18	8.07	13.05
K^+	0.41	0.60	0.59	0.24	0.38	0.36
Mg^{2+}	0.15	0.23	0.22	0.15	0.47	0.77
Ca^{2+}	1.04	1.41	1.19	0.60	1.85	3.26
Cl^-	0.18	0.26	0.27	0.30	0.56	0.94
NO_3^-	1.06	1.48	1.28	0.71	0.31	0.71
SO_4^{2-}	3.29	4.59	4.19	1.41	5.14	8.75

Table VI.3. Scavenging ratios for water soluble major ions.

	Na$^+$	NH$_4^+$	K$^+$	Mg^{2+}	Ca^{2+}	Cl$^-$	NO$_3^-$	SO$_4^{2-}$
Mean	1393	1811	2436	2580	1805	2582	2502	487
S. Dev.	1593	2351	2092	2277	1873	1870	2007	326

Table VI.4. Event Based Geometric Means of Selected Major Ionic Species in Aerosol and Precipitation.

Events	NH$_4^+$		SO$_4^{2-}$		Ca^{2+}	
	Aerosol	Precipitation	Aerosol	Precipitation	Aerosol	Precipitation
Mar. 13-19	15.56 (4)	12.92 (5)	53.35 (3)	5.91 (4)	9.67 (4)	21.27 (4)
Mar. 19-23	58.05 (2)	21.10 (2)	152.79 (1)	33.22 (1)	29.70 (2)	92.93 (2)
Mar. 24-25	241.77 (1)	nd	33.93 (4)	nd	186.01 (1)	nd
Pre-monsoon Average	38.68 (3)	15.21 (4)	73.74 (2)	10.51 (2)	22.01 (3)	34.78 (3)
Aug. 8-19	0.35 (8)	0.44 (8)	0.80 (8)	0.40 (8)	0.31 (8)	0.15 (8)
Aug. 20-28	7.18 (5)	20.66 (3)	3.40 (5)	6.49 (3)	1.13 (5)	20.67 (5)
Aug. 29-29	6.36 (6)	1.00 (6)	1.20 (7)	0.42 (7)	0.60 (7)	0.55 (6)
Monsoon Average	1.72 (7)	0.61 (7)	1.41 (6)	0.60 (6)	0.60 (6)	0.35 (7)

Here, events are based on periods of distinct high and low aerosol concentrations. Values in parantheses indicate the rank of the mean concentration. There was no precipitation during the Mar. 24-25 event and is indicated by 'nd'. Ranks for precipitation concentrations were assumed to be same as for aerosol for this event.

Table VI.5. Result of EOF analysis.

Species	Variance Explained (%)					
	Pre-monsoon			Monsoon		
	EOF1 (63%)	EOF2 (17%)	EOF3 (10%)	EOF1 (54%)	EOF2 (17%)	EOF3 (13%)
Na^+	21	-55	-10	0	-60	7
NH_4^+	80	0	8	52	7	36
K^+	72	3	14	73	0	21
Mg^{2+}	93	0	0	90	-2	-3
Ca^{2+}	82	0	0	66	-1	-15
Cl^-	-7	-69	17	0	-63	1
NO_3^-	75	0	0	60	-2	-17
SO_4^{2-}	90	0	0	90	2	0

Values in parenthesis indicate variance explained by respective EOFs. Negative signs indicate that the specis is loaded upposite in the EOF.

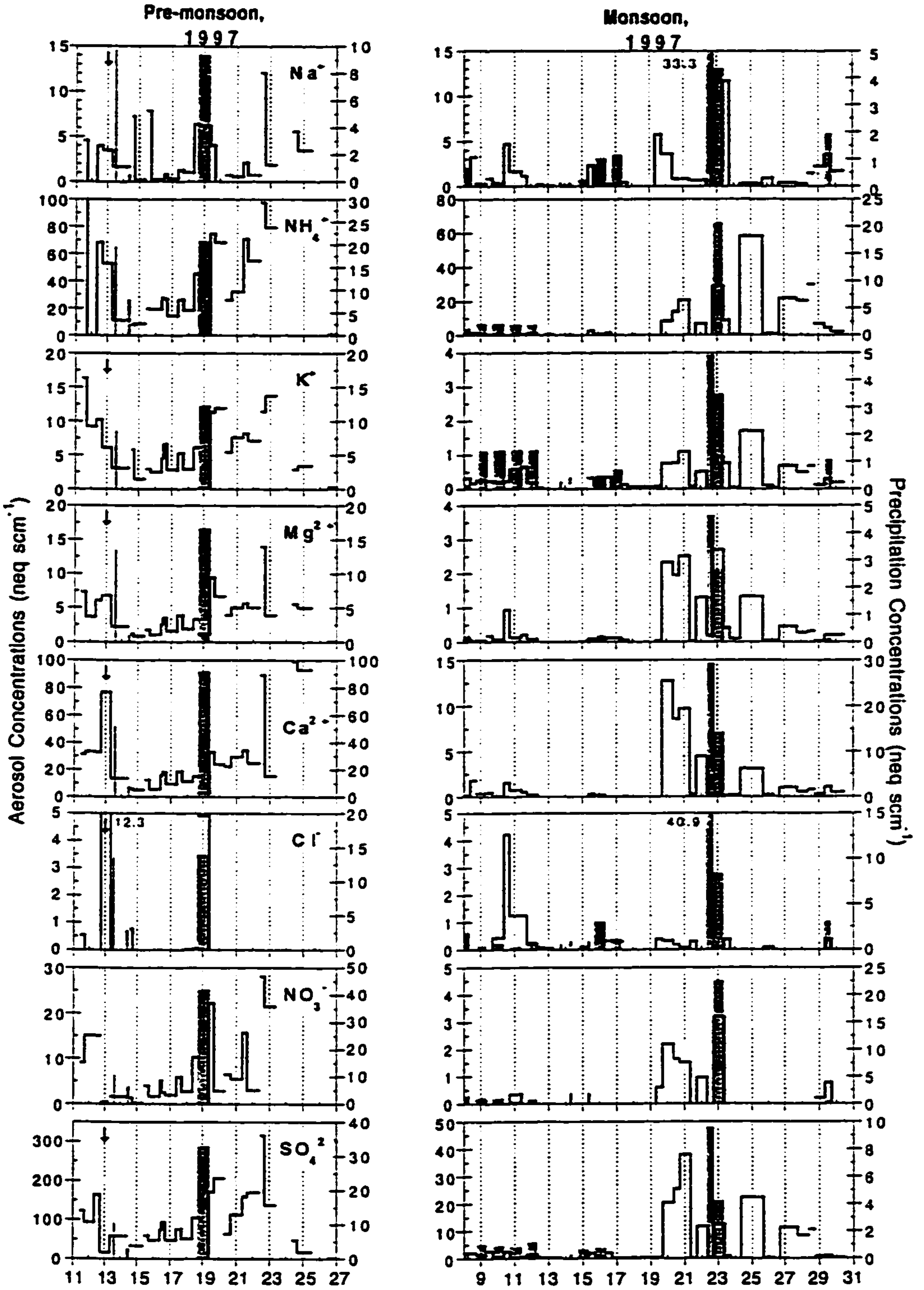


Figure VI.1. Time series of water soluble major ions in the aerosol (solid lines) and precipitation (stippled bars) measured at Phortse during , pre-monsoon, 1997 (left), and monsoon, 1997 (right) seasons. Vertical arrow indicate bad data in precipitation chemistry. Note the scale difference.

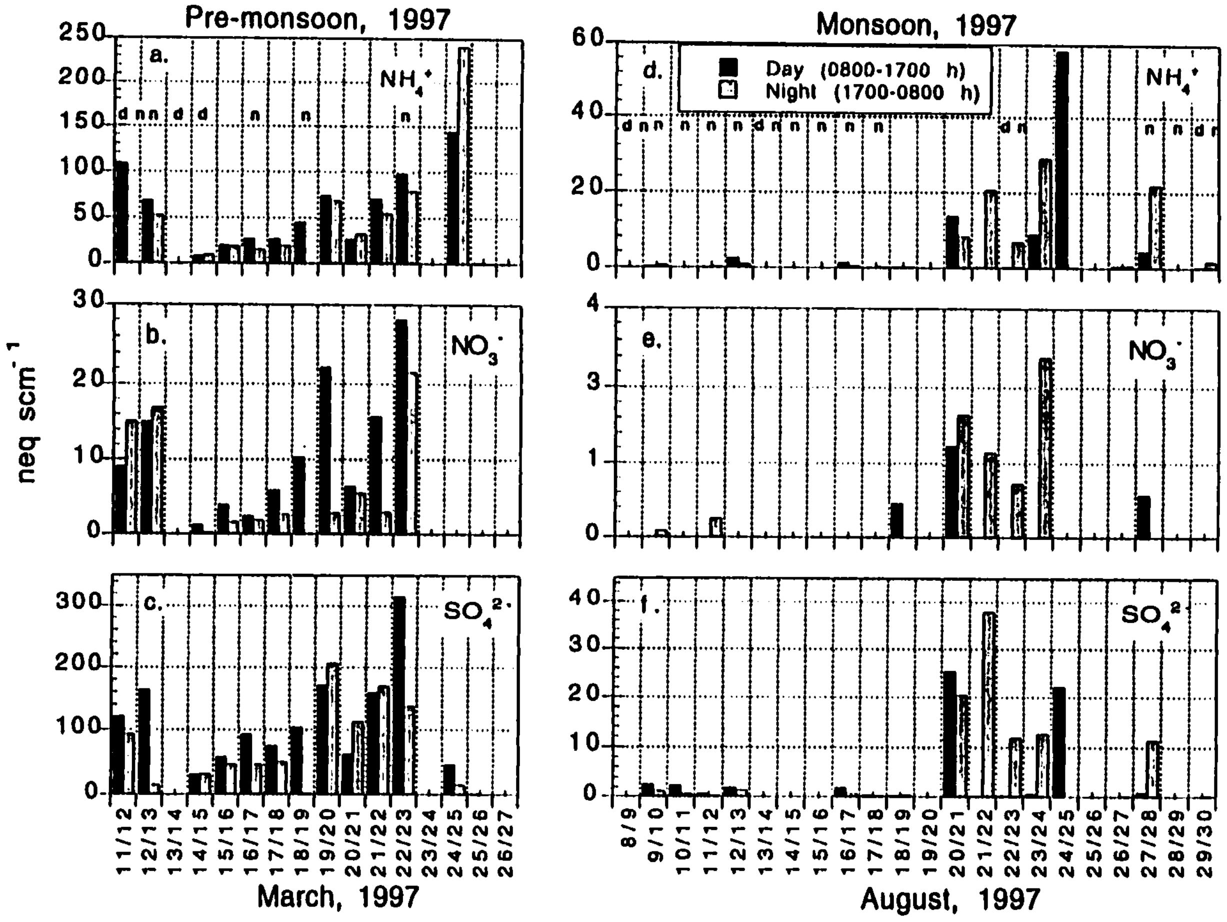


Figure VI.2. Daytime and nighttime concentrations of major ionic species in aerosol samples collected at Phortse. Days without consecutive day and night samples are not presented in the figure. Daytime and nighttime precipitation events are indicated by 'd' and 'n' in Figures a. and d., whereas 'dn' indicates precipitation events covering both the daytime and the following night. Note the difference in vertical scales.

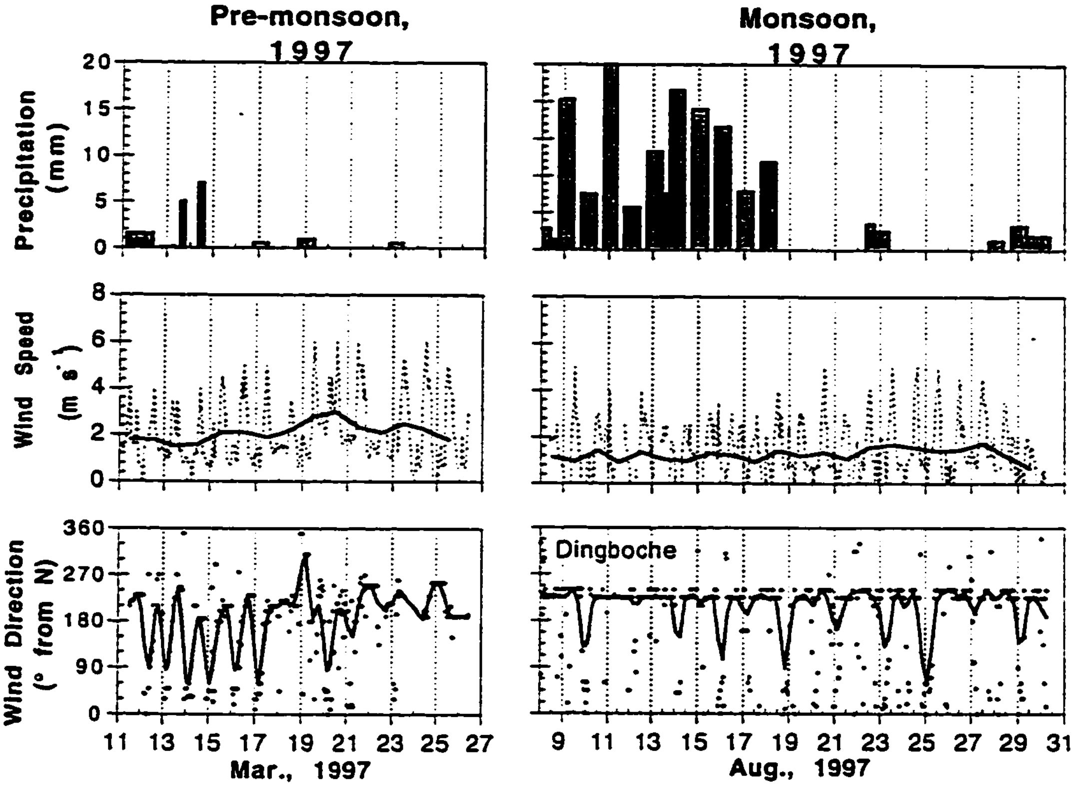


Figure VI.3. Meteorological data collected in Phortse during pre-monsoon, 1997 (left), and monsoon, 1997 (right). Here we use wind direction data from Dingboche for monsoon, 1997 as there is no wind direction data from Phortse for that period.

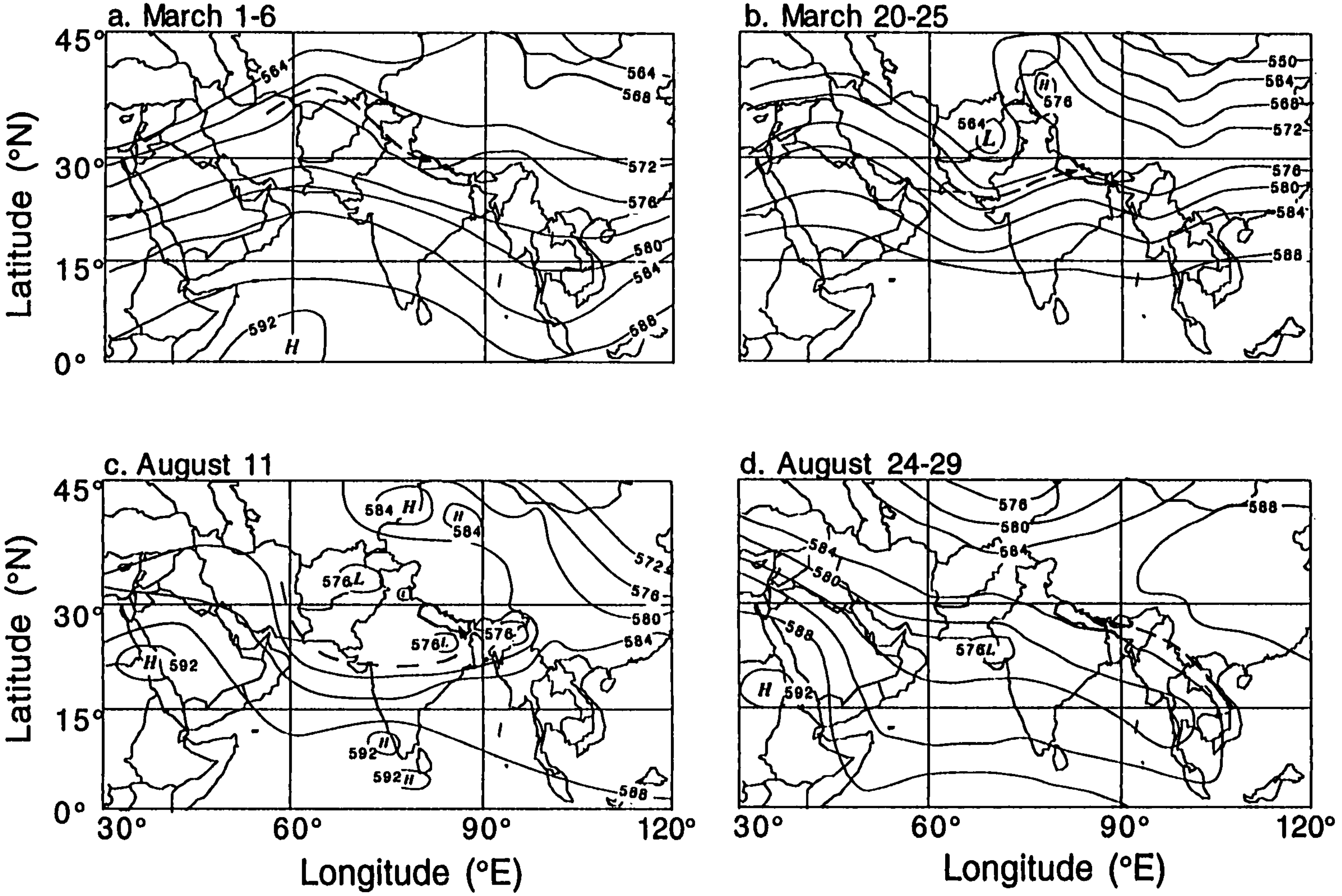


Figure VI.4. Geopotential heights at 500 mb level. The dashed arrow indicates tentative isobaric trajectory of air-masses arriving at the site

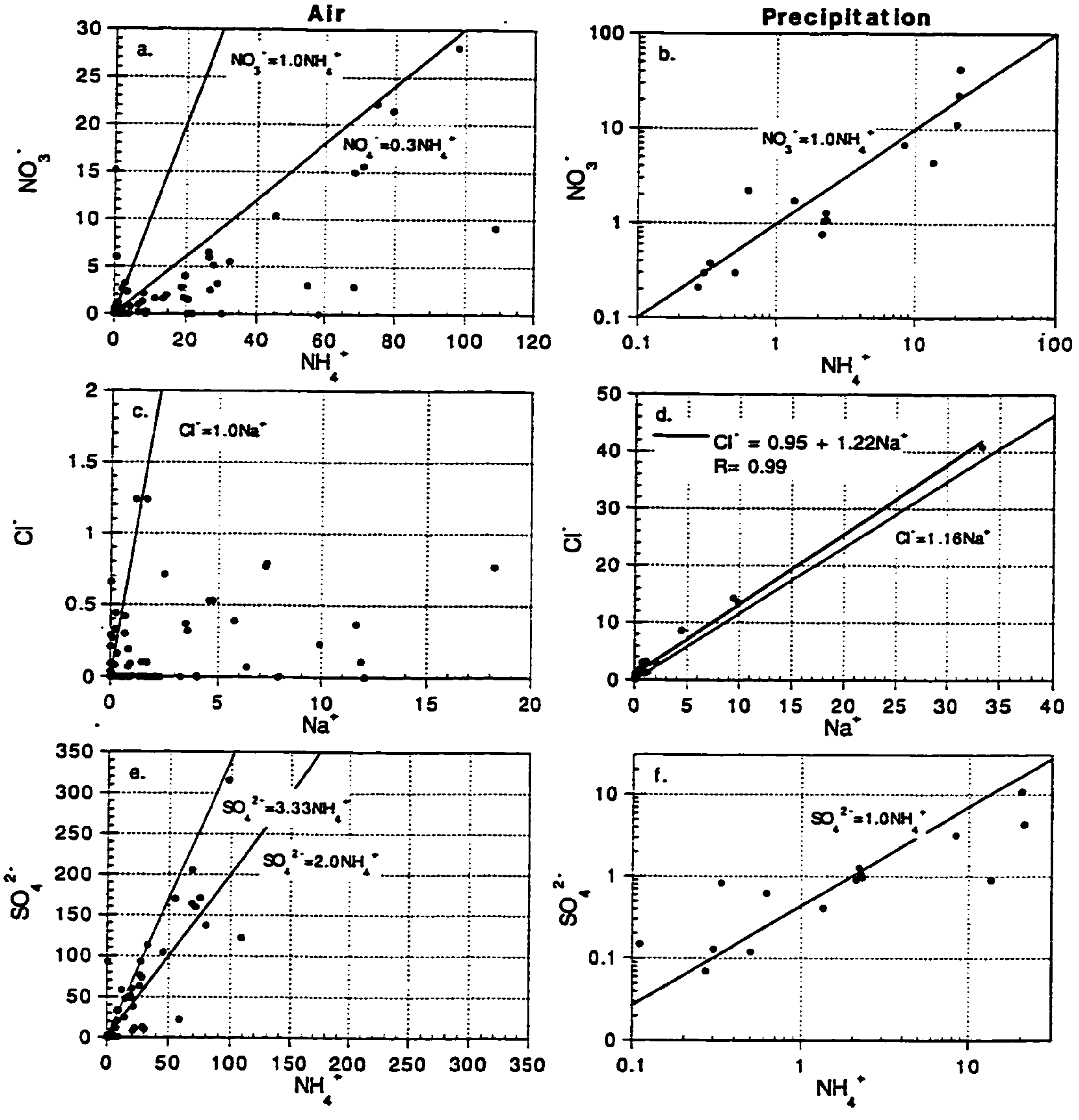
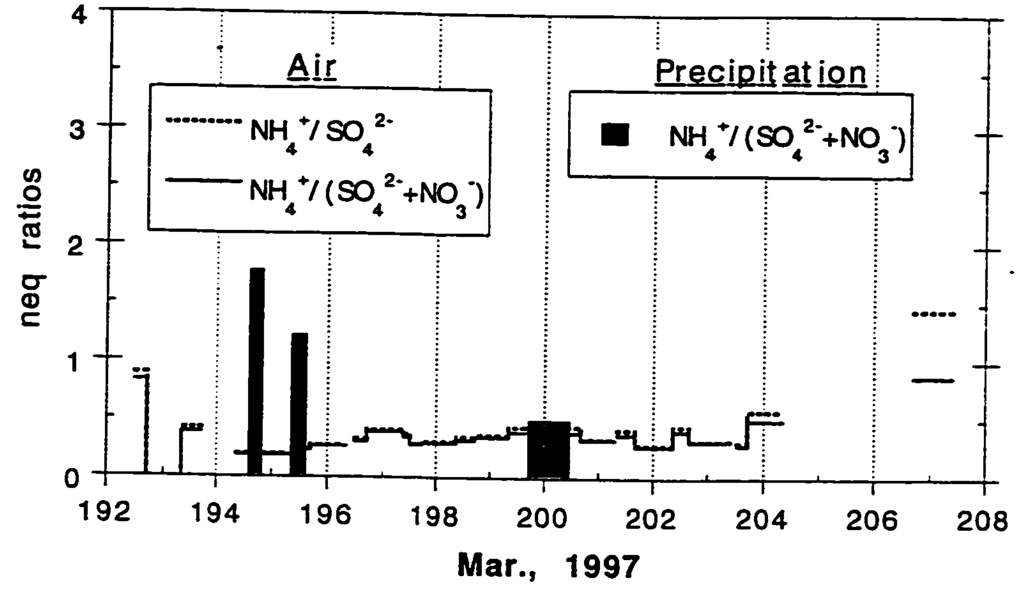


Figure VI.5. Relationships between water soluble major ions in air (left) and precipitation (right)



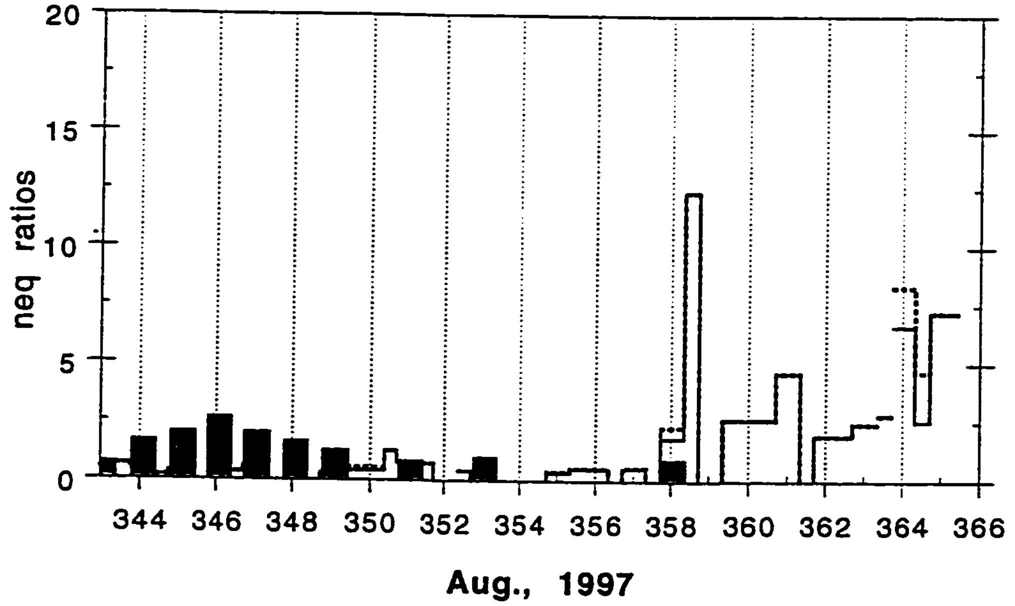


Figure VI.6. NH_4^+/SO_4^{2-} and $NH_4^+/(SO_4^{2-}+NO_3^-)$ ratios in the air and NH_4^+/SO_4^{2-} ratios in the precipitation for the pre-monsoon sampling (top) and the monsoon sampling (bottom).

CHAPTER SEVEN

CONCLUDING REMARKS

VII. CONCLUDING REMARKS

The studies on the temperature and precipitation trends in the Himalaya, which make up Chapters II and III of this dissertation are the first to be based on the instrumental records from Nepal. The temperature records from Nepal show significant rise after the mid-1970's, coinciding with similar trends in global and hemispheric temperature records. The temperature record from Kathmandu, dating back to 1921 and the longest from Nepal shows trends similar to those in the global temperature record, i.e., rising trend before the 1940's and decreasing trend between 1940 and the mid-1970's. These similarities between temperature in Nepal with global scale temperature trends indicate that the global changes in past decades could be responsible for the climate change in the Himalayas. One important aspect of Himalayan environmental change are potential changes in the hydrological regime of mountainous watershed due to global climate change (e.g., Mirza and Dixit, 1997). In recent decades, glacier lake outburst floods due to glacier retreat and glacier lake level rise have been an important natural hazard in the region (e.g., Chikita et al., 1997). Global change could be driving the climate change in the region and therefore investigations and mitigation of such problems should be a global effort.

As with the temperature record, precipitation records from Nepal show links with several large scale climatological records, such as sunspot numbers and the southern oscillation index (SOI) and temperature over the Tibetan Plateau and over India/Indian Ocean. Such relationships between precipitation in Nepal and several large scale climatological parameters suggest that these parameters may prove useful for forecasting monsoon precipitation in the Himalaya. The lack of a distinct long term trend in precipitation in Nepal, despite increasing trends in temperatures, may be the result of Nepal's location between two large emission sources in Asia, i.e., China and India (Foell, 1995; Arndt and Carmichael, 1998). The increase in atmospheric SO_4^{2-} aerosol burden due to emission from these countries may already be influencing the monsoon circulation.

These studies clearly illustrate that while the climate over the Himalaya is linked to several large scale climatological parameters, it differs significantly from the climate of the adjacent plains of the Indian sub-continent. These finding suggest that the general practice of using the all-India precipitation record to relate the climate of the physiographically diverse and vast area of the sub-continent to external forcing factors is not a valid approach.

These studies on physical climate in the Himalaya, however, are unable to provide any measure of climate change over the last century due to limited record length. It is important to continue the investigation as meteorological data from the region accumulate. An emphasis has to be made on the study of meteorological records from high elevation stations recently established in the Nepal Himalaya (Grabs and Pokhrel, 1992). Information from an ice core recently recovered from the northern side of Mt. Everest, in addition to the planned recovery of an ice core from the Nepal Himalaya by the Climate Change Research Center, University of New Hampshire in near future will provide valuable insight concerning monsoon fluctuations in past centuries and millennium. In addition, several tree ring data sets have been collected from sites across Nepal by the Tree Ring Laboratory at Columbia University (per. comm. Dr. E. Cook, LDEO). The all-Nepal and regional temperature and precipitation series developed in this dissertation research will provide a valuable tool for the calibration of the ice core and the tree-ring records. Combined, these records could provide information on the spatial variability of climate along the stretch of the Himalaya and across the Himalayan transect, and thereby represent a key contribution to the study of global environmental change in the region.

The studies on atmospheric composition in the Nepal Himalaya, comprising chapters IV, V, and VI are the most detailed so far on the chemical climate of the Himalaya. The short term study in the western Nepal Himalaya and the one year long investigation of major ionic species in the atmospheric aerosol at two sites in the eastern Nepal Himalaya and in the Eastern Middle-Mountain shows that species measured are long traveled. The high concentrations during pre-monsoon and early monsoon seasons are due to transport from distant sources, likely India. The concentrations of the ionic species clearly reflect changes in atmospheric circulation. Regional scale valley wind systems are responsible for the transport of pollutants to the Himalaya during the pre-monsoon, while monsoon circulation dominates transport mechanisms during the monsoon season. These findings regarding long distance transport of pollutants are in agreement with the chemical source-receptor relationship found in Asia under the RAINS-ASIA project (Streets, 1995; Arndt et al., 1998).

The intensive aerosol and precipitation sampling during pre-monsoon and monsoon seasons in the Himalayan site suggest broad agreement between concentrations of major ionic species in the air and in precipitation on seasonal time scales. The aerosol variations at daily time scales are related to diurnal variations in local wind patterns and precipitation, while much larger variations in ionic concentrations at weekly scales are clearly related to atmospheric circulation. This investigation suggests that the regional scale valley wind system and monsoon circulation responsible for transporting pollutants from distant sources during pre-monsoon and monsoon seasons, respectively, although being physically different, are similar in terms of incorporating a similar suit of pollutant species and transporting them to the interiors of the Himalaya. There are indications of the presence of two maritime signals in the aerosol data- the summertime monsoon signal and the pre-monsoon westerly disturbance signals.

Combined, the three studies on atmospheric composition in the Nepal Himalaya have significantly improved our understanding of concerning aerosol loading in the region and its temporal variation. The results of these studies should represent a valuable contribution to ongoing US and international efforts to improve understanding of tropospheric chemistry (UCAR, 1986; IGBP, 1990). These data may serve as model verifications for future efforts that will focus on relationships between aerosol and climate in the Indian sub-continent, such as the Indian Ocean Experiment (INDOEX)(SASCOM/CGC, 1998). Data collected will substantially supplement the atmospheric composition data base of the Background Air Pollution Monitoring program (BAPMon) and UNDP/WMO sponsored Global Atmospheric Watch (GAW)(WMO, 1990; WMO, 1997).

Although the studies discussed here provide important insight into aerosol concentration and composition variation and their connection with atmospheric circulation and meteorological parameters, there are several issues that need to be addressed in future research projects. It would be particularly interesting to know how the composition of air-masses changes as the monsoon circulation transports them inland and orographic barriers force them to rise across the Himalaya. The temporal and spatial coverage of observations that present studies are based on is insufficient to confirm that the observations are representing the large Himalayan region. Further, the presence of data gaps lowers the overall confidence in our interpretation. Continued studies on aerosol chemistry with more extensive spatial coverage are necessary to better understand the issues observed in this study. A larger network of sampling stations aligned along a north-south transect across the country will help gain insight into these issues. In addition, the existence of the pollution layer in monsoon air-masses, reaching the interiors of the Himalaya needs to be investigated in detail. Airborne measurement of atmospheric constituents, planned to be carried out in near future in the Nepal Himalaya (per. comm., Prof. E. Hindman, City College of New York) may quantify the extent and elevation range of this pollution layer during the monsoon. We are now planning a two year long sampling of aerosol at two sites in the Nepal Himalaya. The concentrations of soluble ionic species, organic carbon and elemental carbon in aerosol will be determined on a continuos basis (at a nominal 2 d intervals). The aerosol sampling will be complemented by continuos multiwavelength aerosol optical depth measurement at both sites. The optical measurement will enable us to ascertain the relationship between surface level aerosol burdens and the aerosol burden in the overlying column. These measurements my be helpful in inferring radiative forcing of tropospheric aerosol in the region.

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APPENDICES

Appendix A. Aerosol Chemsitry Data from Phortse for the Continious (5 day) Sampling Program.

Sampling Detail											
Date On	Time On	Date Off	Time Off	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
Oct 01	10:50	Oct 03	08:30	0.04	2.63	0.28	0.09	0.21	0.01	0.18	2.87
Oct 03	08:35	Oct 08	08:00	0.04	3.11	0.46	0.10	0.34	0.03	0.27	2.90
Oct 08	08:05	Oct 13	07:50	bd	bd	bd	bd	bd	bd	bd	bd
Oct 13	08:00	Oct 18	08:30	0.32	7.73	1.52	0.63	3.20	0.02	1.40	9.68
Oct 18	08:35	Oct 23	08:00	0.29	6.65	1.32	0.49	2.81	0.05	0.75	8.23
Oct 23	08:05	Oct 28	07:55	0.16	8.37	1.42	0.36	1.30	0.01	1.09	8.04
Oct 28	08:05	Nov 02	07:55	0.17	4.18	0.91	0.31	2.10	0.01	0.56	5.21
Nov 02	08:05	Nov 07	08:00	0.28	3.58	1.08	0.45	3.24	0.02	0.76	5.44
Nov 07	08:05	Nov 12	08:00	0.17	6.02	1.57	0.45	4.12	0.05	1.39	6.95
Nov 12	08:05	Nov 17	09:00	0.24	11.62	2.69	0.42	2.06	0.03	1.62	11.51
Nov 17	09:05	Nov 22	08:35	0.29	12.42	2.89	0.44	2.17	0.01	1.91	12.72
Nov 22	08:40	Nov 27	07:45	0.28	11.54	2.79	0.51	2.57	0.04	2.57	10.82
Nov 27	07:50	Dec 02	08:55	0.31	17.16	3.35	0.47	2.04	0.07	3.05	14.90
Dec 02	09:05	Dec 07	08:35	0.04	2.15	0.56	0.16	0.79	0.03	0.46	1.91
Dec 07	08:45	Dec 12	08:05	0.19	4.98	3.62	0.90	6.42	0.14	2.37	6.56
Dec 12	08:10	Dec 14	08:55	0.19	bd	2.21	1.15	5.24	bd	0.67	15.40
Dec 27	14:35	Jan 02	09:00	0.71	5.18	1.33	0.70	4.80	0.32	1.75	8.65
Jan 02	09:10	Jan 07	09:55	0.36	10.89	2.39	0.72	5.11	0.26	4.59	14.26
Jan 07	09:00	Jan 12	08:55	0.38	22.28	3.30	0.60	2.40	0.22	5.67	27.15
Jan 12	09:00	Jan 17	09:00	0.14	7.15	1.52	0.38	1.65	0.16	3.03	11.61
Jan 17	09:10	Jan 22	10:45	0.41	6.69	1.71	0.58	3.61	0.09	3.02	15.28
Jan 22	10:55	Jan 27	09:45	0.20	1.07	0.58	0.41	8.46	0.15	1.48	6.67
Jan 27	09:55	Feb 01	09:00	0.27	15.37	1.82	0.46	2.10	0.27	8.34	16.97
Feb 01	09:10	Feb 06	09:30	0.06	2.40	0.45	0.17	0.46	0.17	2.73	5.57
Feb 16	07:30	Feb 21	09:40	2.20	19.66	3.42	2.55	13.36	0.47	10.16	40.94
Pre-monsoon Intensive Sampling											
Mar 26	11:35	Apr 02	10:30	bd	2.85	0.68	0.67	1.89	bd	bd	bd
Apr 02	09:25	Apr 07	07:35	0.41	9.63	5.79	6.83	15.75	0.59	11.65	59.50
Apr 07	08:30	Apr 12	08:00	0.19	92.27	42.17	30.88	72.31	6.39	104.69	213.51
Apr 12	10:35	Apr 17	08:45	0.65	0.03	0.09	0.08	0.19	0.02	bd	bd
Apr 17	07:40	Apr 22	08:10	3.36	2.13	2.17	1.13	2.23	0.15	1.89	19.23
Apr 27	08:40	May 02	10:40	0.07	1.89	0.63	0.22	0.46	0.36	bd	0.37
May 17	08:30	May 22	08:30	3.57	106.9	10.74	7.47	14.68	0.64	9.40	55.10
May 22	08:35	May 27	07:55	4.19	0.42	0.29	0.16	0.24	0.27	bd	bd
May 27	08:00	Jun 01	08:55	0.83	329.08	31.84	28.31	55.98	5.22	81.44	201.28
Jun 01	08:35	Jun 06	08:50	2.60	545.16	73.38	43.06	86.81	5.73	124.55	429.53
Jun 06	08:00	Jun 11	08:30	1.60	39.51	2.33	1.45	3.27	0.27	2.53	11.88
Jun 11	08:00	Jun 16	08:25	2.41	87.14	6.72	2.41	3.53	0.15	1.21	23.30
Jun 16	08:00	Jun 21	07:55	0.50	159.58	50.41	13.12	25.31	3.44	76.94	171.27
Jun 21	08:40	Jun 26	08:00	0.13	0.19	0.12	0.05	0.24	0.14	bd	0.19
Jun 26	08:30	Jul 01	07:55	1.36	0.53	0.18	0.22	1.06	0.85	bd	1.00
Aug 05	10:15	Aug 08	10:30	0.68	0.92	0.26	0.06	0.48	bd	0.05	1.69

Appendix A. Continued

Sampling Detail											
Date On	Time On	Date Off	Time Off	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
Month [illegible] Sampling											
Aug 31	15:15	Sep 09	7:55	0.15	3.63	0.29	0.08	0.13	bd	0.13	1.80
Sep 09	08:05	Sep 14	7:30	0.98	0.18	0.05	0.02	0.14	bd	0.04	0.11
Sep 14	08:00	Sep 19	9:55	0.07	1.42	0.17	0.03	0.06	bd	0.07	0.54
Sep 19	07:40	Sep 24	7:55	0.23	3.31	0.41	0.15	0.45	0.10	0.22	2.29
Sep 24	10:00	Sep 29	7:55	1.06	11.39	1.06	0.23	0.76	1.19	0.75	5.55
Oct 09	8:00	Oct 14	7:50	0.51	20.17	0.82	0.29	1.49	0.12	2.59	11.90
Oct 14	8:40	Oct 19	9:15	0.53	21.61	1.65	0.48	3.04	0.10	3.39	13.95
Oct 19	8:00	Oct 24	7:55	0.31	14.30	1.26	0.24	1.26	0.07	1.18	9.76
Oct 24	9:20	Nov 03	8:55	0.22	6.90	0.59	0.12	0.51	0.17	0.45	4.16
Nov 03	8:00	Nov 08	7:55	0.26	9.94	1.18	0.17	0.72	0.21	1.72	5.19
Nov 08	9:00	Nov 13	9:05	0.23	27.28	1.19	0.15	0.68	0.15	1.89	14.92
Nov 13	8:05	Nov 18	9:05	0.27	18.26	1.71	0.22	1.26	0.10	1.84	9.89

Here, bd indicates concentrations below detection and blank rows indicate data gap due to instrument failure. Concentrations are in neq scm^{-1}

Appendix B. Aerosol Chemsitry Data from Phortse for the Intensive (12 hour) Sampling Program.

Sampling Detail											
Date On	Time On	Date Off	Time Off	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
Post-monsoon, 1996 Intensive Sampling											
Sep 24	13:00	Sep 25	09:30	9.90	3.46	0.49	0.07	0.95	0.23	2.36	5.47
Sep 25	10:35	Sep 25	13:50	bd	0.79	0.67	bd	bd	0.66	bd	0.25
Sep 26	14:15	Sep 26	17:05	bd	0.57	0.39	bd	bd	0.09	1.00	0.70
Sep 26	17:15	Sep 27	9:15	2.49	0.91	0.65	0.03	0.97	0.71	1.22	1.19
Sep 27	9:35	Sep 27	14:05	1.41	2.31	0.41	0.02	10.40	0.10	0.57	2.14
Sep 27	14:50	Sep 27	17:35	7.28	2.25	2.13	0.07	64.08	0.79	2.63	2.21
Sep 27	17:45	Sep 28	8:10	18.18	0.43	4.32	0.03	5.57	0.77	6.03	0.53
Sep 28	8:40	Sep 28	13:50	11.86	2.83	3.11	0.26	6.14	0.11	3.21	2.51
Sep 28	14:15	Sep 28	17:30	0.06	3.81	0.18	0.10	bd	0.03	bd	3.71
Sep 29	14:10	Sep 29	17:30	4.73	1.12	1.06	0.03	18.43	0.53	1.13	0.93
Sep 29	17:45	Sep 30	13:45	0.64	0.78	0.57	0.07	26.97	0.42	0.34	0.50
Pre-monsoon, 1997 Intensive Sampling											
Mar 11	11:45	Mar 11	17:15	4.55	108.50	16.28	3.64	31.30	0.53	9.12	122.13
Mar 11	17:20	Mar 12	08:20	0.00	0.00	9.18	1.81	33.21	bd	15.13	93.01
Mar 12	08:30	Mar 12	17:20	3.98	68.34	10.19	3.03	32.79	0.01	15.02	164.04
Mar 12	17:25	Mar 13	08:45	3.48	52.85	6.07	6.72	153.67	12.30	16.87	32.94
Mar 13	08:50	Mar 14	08:20	1.66	11.37	3.09	1.11	13.70	bd	1.65	58.17
Mar 14	13:50	Mar 14	17:50	7.21	7.88	5.80	0.60	5.90	0.77	1.33	32.90
Mar 14	18:00	Mar 15	08:20	0.24	8.82	1.49	0.43	5.05	bd	0.30	32.74
Mar 15	11:40	Mar 15	17:05	7.82	19.66	2.91	0.89	12.30	bd	4.02	59.60
Mar 15	17:10	Mar 16	09:00	0.24	19.36	2.47	0.52	5.52	bd	1.70	47.98
Mar 16	09:05	Mar 16	12:30	0.08	27.56	4.51	1.26	15.64	bd	5.14	73.58
Mar 16	12:40	Mar 16	17:20	0.83	26.70	6.54	1.74	18.18	bd	2.53	93.25
Mar 16	17:30	Mar 17	09:05	0.33	14.50	2.79	0.80	9.54	bd	1.96	47.51
Mar 17	09:10	Mar 17	17:20	1.30	26.35	5.29	1.95	18.81	bd	5.95	76.27
Mar 17	17:30	Mar 18	08:15	1.00	18.72	2.94	0.96	11.41	0.01	2.79	51.10
Mar 18	08:20	Mar 18	16:40	6.32	45.22	6.09	1.66	15.00	0.07	10.35	104.45
Mar 18	16:45	Mar 19	09:25	6.18	bd	0.13	0.57	2.86	4.90	0.09	2.60
Mar 19	09:30	Mar 19	16:40	4.01	74.58	11.34	4.70	32.80	bd	22.20	171.14
Mar 19	16:45	Mar 20	07:50	bd	68.31	11.89	3.33	24.08	bd	2.92	205.85
Mar 20	07:55	Mar 20	16:45	0.72	26.17	5.43	1.96	22.28	bd	6.49	63.41
Mar 20	16:50	Mar 21	09:10	0.55	32.24	7.62	2.50	29.59	bd	5.53	113.17
Mar 21	09:25	Mar 21	16:40	2.11	70.89	8.21	2.82	34.34	bd	15.70	159.73
Mar 21	16:50	Mar 22	09:00	0.71	54.69	7.13	2.52	24.96	bd	3.05	169.63
Mar 22	12:20	Mar 22	17:00	12.06	97.59	11.42	6.98	89.20	bd	28.08	316.09
Mar 22	17:05	Mar 23	08:00	1.86	79.23	13.75	1.94	15.21	bd	21.47	137.15
Mar 24	09:15	Mar 24	16:25	5.54	144.42	2.89	5.51	198.00	bd	bd	95.27
Mar 24	16:35	Mar 25	11:35	3.46	241.77	3.41	4.99	186.01	bd	bd	33.93
Mar 25	16:55	Mar 26	10:45	bd	bd	bd	bd	bd	0.04	bd	bd
Monsoon, 1997 Intensive Sampling											
Aug 08	14:10	Aug 09	09:05	0.10	0.99	0.31	0.04	0.10	bd	bd	1.98
Aug 09	09:15	Aug 09	17:50	3.24	1.32	0.17	0.02	1.80	bd	bd	2.02
Aug 09	17:55	Aug 10	08:55	0.28	0.71	0.25	0.02	0.35	bd	0.12	1.26
Aug 10	09:15	Aug 10	17:30	0.87	0.43	0.24	0.09	0.43	bd	bd	2.69
Aug 10	17:35	Aug 11	09:05	0.24	0.19	0.20	0.03	0.06	0.44	bd	0.57
Aug 11	09:10	Aug 11	17:30	4.67	0.00	0.22	0.47	1.53	4.21	bd	2.48

Appendix B. Continued.

Sampling Detail											
Date On	Time On	Date Off	Time Off	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
Aug 11	17:35	Aug 12	09:00	1.65	bd	0.58	0.07	0.73	1.24	0.35	0.29
Aug 12	09:05	Aug 12	17:20	1.12	0.22	0.64	0.10	0.49	1.24	bd	0.64
Aug 12	17:35	Aug 13	08:35	bd	0.88	0.18	0.04	0.22	0.21	bd	1.47
Aug 13	08:40	Aug 13	17:25	0.18	bd	0.03	bd	bd	0.08	bd	0.43
Aug 13	17:30	Aug 14	17:05	bd	bd	bd	bd	bd	bd	bd	0.20
Aug 14	17:10	Aug 15	07:25	0.30	0.09	0.18	0.00	bd	0.16	bd	0.28
Aug 15	07:30	Aug 16	09:00	0.10	0.59	0.31	0.01	bd	0.27	0.34	1.13
Aug 16	09:05	Aug 16	17:30	2.28	2.48	0.32	0.05	0.30	bd	bd	1.98
Aug 16	17:35	Aug 17	08:55	bd	0.51	0.09	0.01	bd	bd	bd	0.71
Aug 17	09:00	Aug 17	17:25	0.22	1.41	0.35	0.05	bd	0.33	bd	1.98
Aug 17	17:30	Aug 18	08:55	bd	bd	0.06	0.06	bd	0.29	bd	0.23
Aug 18	09:00	Aug 18	17:30	0.40	0.16	0.13	0.03	0.04	bd	bd	0.38
Aug 18	17:35	Aug 20	08:25	bd	bd	0.06	bd	bd	bd	bd	0.37
Aug 20	08:30	Aug 20	17:30	5.75	bd	0.08	0.02	bd	0.39	0.61	0.34
Aug 20	17:35	Aug 21	08:25	3.58	8.22	0.75	1.16	12.74	0.32	2.19	20.40
Aug 21	08:30	Aug 21	17:30	0.80	13.69	0.76	0.97	8.54	0.19	1.64	25.37
Aug 21	17:35	Aug 22	08:25	0.80	20.58	1.09	1.25	9.69	0.07	1.52	37.92
Aug 22	08:30	Aug 22	17:25	0.64	bd	0.07	0.01	0.30	0.30	bd	0.10
Aug 22	17:30	Aug 23	08:30	0.67	6.66	0.50	0.65	4.47	bd	0.96	11.75
Aug 23	08:35	Aug 23	17:30	bd	bd	bd	0.09	0.33	bd	bd	0.35
Aug 23	17:35	Aug 24	08:35	1.69	28.74	0.93	1.34	bd	0.10	3.19	12.54
Aug 24	08:40	Aug 24	17:25	11.64	8.76	0.75	0.20	0.15	0.37	bd	0.71
Aug 24	17:30	Aug 25	08:25	bd	0.32	0.04	0.05	0.19	bd	bd	bd
Aug 26	08:30	Aug 26	17:30	0.29	58.21	1.70	0.66	3.08	bd	bd	22.21
Aug 26	17:35	Aug 27	08:30	0.89	0.74	0.10	0.02	0.19	0.09	bd	0.16
Aug 27	08:35	Aug 27	17:30	bd	0.80	0.03	0.02	bd	bd	bd	bd
Aug 27	17:35	Aug 28	17:30	0.39	21.78	0.68	0.23	1.01	bd	bd	11.27
Aug 28	17:35	Aug 29	08:55	0.24	20.49	0.49	0.14	0.54	bd	bd	8.38
Aug 29	09:05	Aug 29	17:55	1.45	30.02	0.67	0.16	0.80	bd	bd	10.60
Aug 29	18:00	Aug 30	08:30	2.22	6.79	0.12	0.02	0.30	bd	0.21	0.82
Aug 30	08:35	Aug 30	17:40	3.49	4.36	0.30	0.10	1.07	0.37	0.77	0.94
Aug 30	17:45	Aug 30	10:40	1.66	1.84	0.19	0.10	0.49	bd	bd	0.26

Here, bd indicates concentrations below detection and blank rows indicate data gap due to bad weathe and low battery voltage. Concentrations are in neq scm^{-1}

Appendix C. Aerosol Chemsitry Data from Jiri for the Continious (5 day) Sampling Program.

Sampling Detail											
Date On	Time On	Date Off	Time Off	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
Sep 08	13:05	Sep 09	09:05	0.10	7.99	2.97	0.63	0.37	0.09	0.63	8.35
Sep 09	09:30	Sep 14	10:20	0.29	14.12	3.04	0.91	1.63	0.02	0.13	8.39
Sep 14	10:30	Sep 19	10:20	0.19	5.16	1.98	0.47	0.50	bd	0.11	13.28
Sep 19	09:10	Sep 24	09:05	1.12	6.99	2.47	1.75	7.81	bd	0.08	20.31
Sep 24	09:10	Sep 29	09:05	0.86	11.11	1.74	0.82	1.42	bd	0.06	18.37
Sep 29	09:05	Oct 03	09:00	0.25	5.87	1.99	0.60	1.20	0.05	0.39	8.47
Oct 03	09:20	Oct 08	09:20	0.16	5.92	1.85	0.43	0.95	0.01	0.43	8.64
Oct 08	09:22	Oct 13	09:20	0.89	9.38	2.94	2.24	10.50	0.05	2.38	22.68
Oct 13	09:40	Oct 18	09:15	0.57	7.69	2.66	1.40	5.84	0.04	0.71	19.01
Oct 18	09:22	Oct 23	09:20	0.26	6.23	1.85	0.76	2.50	bd	0.20	10.23
Oct 23	09:40	Oct 28	09:10	0.32	8.96	3.13	0.93	2.69	0.02	0.60	10.63
Oct 28	09:15	Nov 02	09:35	0.14	8.68	1.85	0.41	1.07	0.02	0.51	10.91
Nov 02	09:45	Nov 07	09:15	0.24	4.19	1.84	0.61	2.01	0.01	0.27	8.58
Nov 07	09:20	Nov 12	09:40	0.37	8.93	2.33	0.69	2.82	0.04	1.40	14.22
Nov 12	09:45	Nov 17	09:30	0.39	14.04	3.57	0.80	2.87	0.02	1.30	18.28
Nov 17	09:35	Nov 22	09:50	0.70	20.62	5.16	1.06	4.24	0.07	2.28	18.77
Nov 22	09:55	Nov 27	10:00	0.22	7.76	2.29	0.47	1.57	0.08	1.57	8.39
Nov 27	10:05	Dec 01	09:20	0.66	31.18	5.86	1.13	4.13	0.11	5.42	32.55
Dec 01	09:30	Dec 07	10:10	0.32	6.88	2.91	0.83	2.49	0.16	1.12	10.27
Dec 07	10:15	Dec 12	09:45	0.22	3.93	2.36	0.63	1.80	0.08	1.14	5.57
Dec 12	09:50	Dec 18	10:05	0.43	14.50	4.23	0.92	2.75	0.09	2.19	17.92
Jan 01	10:50	Jan 02	11:20	1.40	28.48	6.83	2.72	10.93	0.56	4.40	36.00
Feb 02	12:30	Feb 06	15:00	1.81	94.79	6.64	2.64	12.22	1.10	34.08	63.25
Feb 06	15:05	Feb 11	11:35	2.65	90.19	8.36	4.44	20.13	2.12	42.08	75.94
Feb 11	11:40	Feb 16	11:10	2.62	31.25	5.59	3.51	21.81	0.37	16.64	56.67
Feb 16	11:15	Feb 21	10:30	4.24	21.81	7.66	4.62	24.88	0.60	13.75	55.65
Feb 21	10:35	Feb 26	10:50	1.08	22.28	4.68	1.87	7.84	0.56	11.69	33.02
Feb 26	10:55	Mar 03	08:50	1.60	28.49	5.28	2.65	12.94	0.32	11.04	48.08
Mar 03	08:52	Mar 08	09:30	1.99	24.00	9.03	3.80	17.90	0.40	9.69	0.04
Mar 08	09:35	Mar 13	09:50	3.58	53.67	16.05	6.93	27.97	0.78	22.13	0.01
Mar 14	17:00	Mar 15		2.05	1.53	0.52	4.42	27.44	0.49	4.25	41.47
Apr 12	10:10	Apr 17	09:45	1.83	16.89	3.49	2.96	9.76	0.04	2.25	30.56
Apr 17	09:50	Apr 22	10:05	3.35	6.69	5.60	5.61	25.69	0.24	9.34	28.68
Apr 22	10:10	Apr 27	09:55	2.04	19.21	6.37	5.14	18.60	0.08	3.84	43.72
May 17	08:40	May 22	07:35	3.98	20.69	11.61	8.50	41.34	0.30	9.87	69.55
May 22	07:40	May 27	10:00	6.42	21.57	8.69	7.04	27.86	0.22	7.28	63.73
May 27	10:05	Jun 02	11:00	1.64	3.99	2.91	3.04	14.46	0.08	2.06	21.50
Jun 02	09:10	Jun 06	10:00	2.61	26.27	4.53	4.53	13.93	0.22	4.89	44.93
Jun 06	10:05	Jun 11	09:00	4.41	33.48	6.60	7.56	26.66	0.34	7.96	69.72
Jun 11	09:05	Jun 18	08:30	6.23	33.55	7.02	8.33	31.83	0.54	5.99	76.45
Jun 18	08:35	Jun 21	09:40	1.85	14.01	2.88	3.21	19.45	0.52	5.86	29.24
Jun 21	09:45	Jun 26	09:05	1.25	8.12	2.30	1.69	8.17	0.14	1.15	16.54
Jun 26	09:10	Jul 02	09:00	0.54	5.00	1.91	0.92	2.31	0.21	1.10	6.67
Jul 06	09:45	Jul 06	10:20	0.91	7.76	1.85	0.81	1.08	0.05	0.15	8.93
Jul 06	10:25	Jul 13	11:10	0.21	2.69	1.22	0.37	0.30	0.04	0.12	2.95
Jul 13	11:15	Jul 16	10:05	1.47	3.88	0.89	1.38	3.29	0.25	0.86	6.18

Appendix C. Continued

Sampling Detail											
Date On	Time On	Date Off	Time Off	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
Jul 16	10:10	Jul 21	09:20	1.63	3.55	1.60	1.64	3.16	0.08	0.45	10.12
Jul 21	09:25	Jul 26	09:40	0.95	3.68	2.01	1.37	2.27	0.07	0.31	8.50
Jul 26	09:45	Jul 31	09:20	1.10	5.66	2.35	1.14	1.44	0.04	0.16	9.12
Jul 31	09:25	Aug 05	10:25	1.03	3.37	1.96	0.90	1.05	0.04	0.14	6.32
Aug 05	10:30	Aug 10	09:20	2.86	4.65	1.79	1.65	2.04	0.21	1.06	10.90
Aug 10	12:40	Aug 15	11:00	0.52	2.96	0.85	0.56	0.65	0.09	0.20	3.52
Aug 15	11:05	Aug 20	10:25	0.08	3.70	1.31	0.39	0.48	0.04	0.11	6.67
Aug 20	10:30	Aug 25	10:10	1.26	9.88	2.14	1.37	2.18	0.07	0.21	19.41
Aug 25	10:15	Aug 30	10:05	0.39	11.02	2.76	0.98	1.35	0.09	0.21	7.71
Aug 30	10:10	Sep 04	09:35	1.93	14.28	2.51	1.91	2.71	0.06	0.49	0.02
Sep 04	10:35	Sep 09	11:10	1.72	19.04	2.54	2.08	3.53	0.01	0.07	0.03
Sep 09	11:20	Sep 14	10:10	1.17	5.24	1.57	0.65	0.71	bd	0.18	10.39
Sep 14	10:15	Sep 19	12:30	0.32	8.55	1.90	0.49	0.74	bd	0.17	14.10
Sep 19	12:35	Sep 24	09:50	0.41	12.07	2.27	0.75	1.67	0.11	0.59	16.07
Sep 24	09:55	Oct 05	10:00	1.92	7.93	3.19	2.40	10.66	0.10	1.70	26.28
Oct 05	10:00	Oct 09	11:00	0.96	8.17	2.30	1.13	3.59	0.11	0.46	16.39
Oct 09	11:05	Oct 14	09:00	0.61	16.72	1.85	0.98	3.31	0.05	0.77	24.76
Oct 14	09:05	Oct 19	10:40	0.75	14.40	2.71	2.00	11.64	0.09	1.10	32.42
Oct 19	10:45	Oct 24	11:00	0.71	20.75	3.42	1.31	6.24	0.10	1.13	33.29
Oct 24	10:05	Oct 27	11:00	0.27	11.95	1.78	0.57	2.15	bd	0.93	16.26

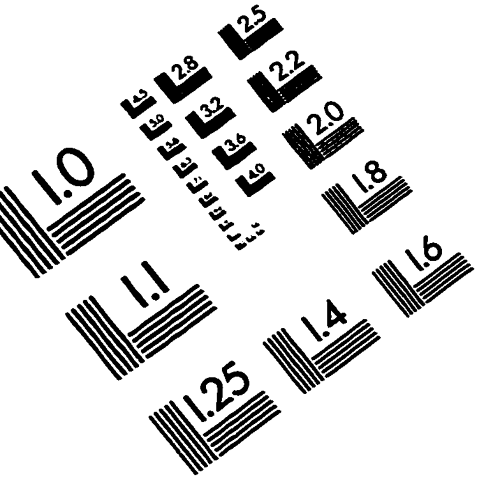
Here, bd indicates concentrations below detection and blank rows indicate data gap due to instrument failure. Concentrations are in neq scm^{-1}

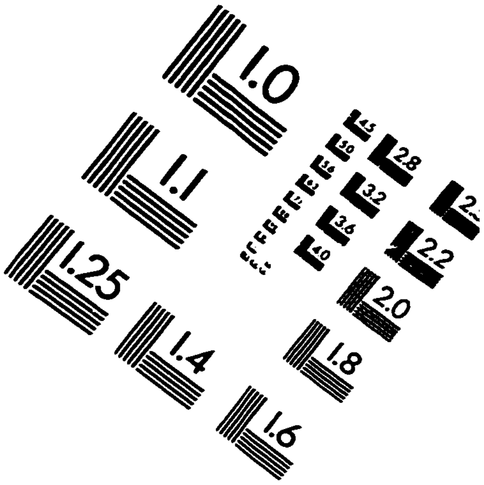
Appendix D. Precipitation Chemistry Data from Phortse

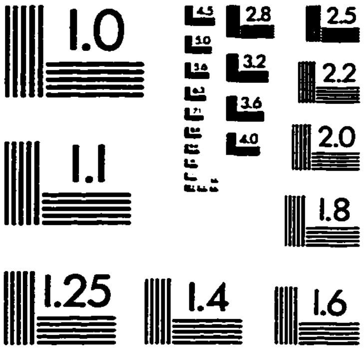
Date	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
Post-monsoon, 1996								
Sep 25/26^{n}	0.67	13.50	1.00	0.75	3.38	1.16	4.50	0.91
Sep 26/27^{n}	0.54	0.62	0.50	0.15	0.39	1.88	2.22	0.62
Sep 27/28^{n}	0.28	15.11	0.55	0.37	1.64	0.22	0.85	0.29
Pre-monsoon, 1997								
Mar 12/13^{n}	ni	bd	ni	ni	ni	ni	1.69	ni
Mar 13	9.82	20.04	8.91	13.62	52.66	13.56	11.29	10.92
Mar 13/14^{n}	0.74	8.33	0.60	1.22	8.59	3.06	6.78	3.20
Mar 18/19^{n}	9.38	21.10	12.51	16.70	92.93	14.24	42.63	33.22
Monsoon, 1997								
Aug 8/9^{n}	0.66	1.33	0.21	0.27	0.97	2.03	1.73	0.41
Aug 9/10^{n}	0.06	2.23	1.40	0.09	0.02	0.57	1.30	1.06
Aug 10/11^{n}	0.24	2.27	1.47	0.19	0.42	0.86	1.09	0.97
Aug 11/12^{n}	0.06	2.09	1.45	0.14	0.23	0.48	0.77	0.91
Aug 12/13^{n}	0.08	2.17	1.46	0.13	bd	1.14	1.08	1.26
Aug 13/14^{n}	0.13	0.50	0.02	0.04	0.14	0.46	0.30	0.12
Aug 14/15^{n}	0.10	0.27	0.02	0.02	0.04	0.15	0.21	0.07
Aug 15/16^{n}	0.27	0.00	0.04	0.11	0.23	0.65	0.32	0.72
Aug 16/17^{n}	1.08	0.33	0.50	0.28	1.01	3.14	0.38	0.83
Aug 17/18^{n}	1.18	0.11	0.72	0.16	0.20	1.32	0.00	0.15
Aug 18/19^{n}	0.04	0.30	0.00	0.04	0.01	0.17	0.30	0.13
Aug 22	33.34	bd	62.22	4.66	29.64	40.91	0.16	9.72
Aug 23/24^{n}	4.42	20.66	3.56	2.70	14.42	8.59	22.73	4.33
Aug 30	1.99	bd	1.10	0.19	0.55	3.13	0.80	0.42

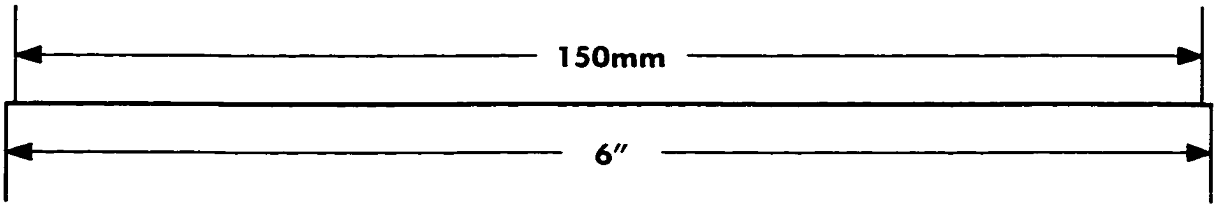
Here, ^{n} means night-time samples, bd indicates concentrations below detection and ni indicates samples not included due to erroneous values. Concentrations are in μeq kg^{-1}. Each data values are based on arithmetic averages of multiple samples representing the same precipitation event. Standard deviations of these multiple samples were adequate low (<5% of the mean).

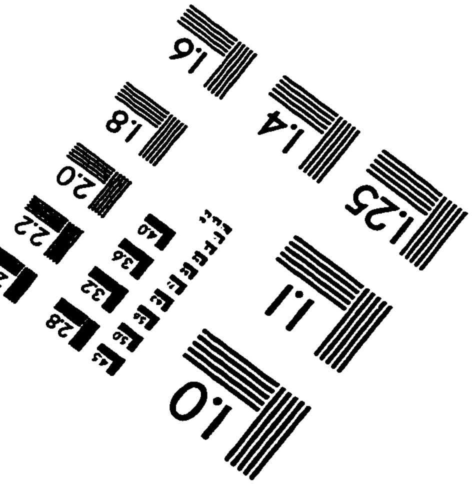
IMAGE EVALUATION TEST TARGET (QA-3)

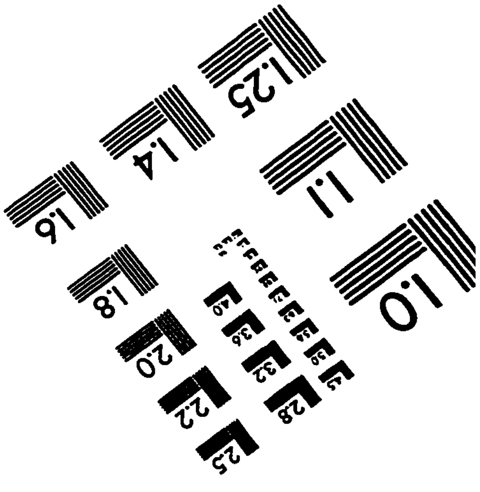












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