Review Article

Airmass Trajectories and Long Range Transport of Pollutants: Review of Wet Deposition Scenario in South Asia

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Received 24 April 2014; Revised 6 June 2014; Accepted 9 June 2014; Published 12 August 2014

Academic Editor: M. Ángeles García

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This paper presents a review of airmass trajectories and their role in air pollution transport. It describes the concept, history, and basic calculation of air trajectories citing various trajectory models used worldwide. It highlights various areas of trajectory applications and errors associated with trajectory calculations. South Asian region receives airmasses from Europe, Middle East, Africa, and Indian Ocean, and so forth, depending upon the season. These airmasses are responsible for export and import of pollutants depositing in nearby states. Trajectory analysis revealed that soil is contributed by the dust storms coming from Oman through Gulf and Iran, while most of black carbon (BC) sources are located in India. A detailed review of trajectories associated with wet deposition events indicated that airmasses coming from Europe and Middle East carry high concentration of acidic pollutants which are deposited in Himalayan ranges. Similarly, trajectory analysis revealed that acidic pollutants from continental anthropogenic sources are transported to an ecosensitive site in Western Ghats in India and the outward fluxes of anthropogenic activities of Indo-Gangetic region are transported towards Bay of Bengal. Hence, transboundary and long range transport of pollutants are very important issues in South Asia which need immediate attention of scientists and policy makers.

1. Introduction

1.1. Concept of Trajectories. Air flow may be described in two different ways: (i) Eulerian (named after Swiss mathematician Leonhard Euler) and (ii) Lagrangian (named after French mathematician J L Lagrange) [1–3]. In the Eulerian approach, the air flows through the points fixed in the space whereas in the Lagrangian approach [3], individual air parcel is chosen and followed as it moves in time and space. Most of chemistry models are based on Eulerian approach as this is a useful tool to explain various chemical and physical processes. In the Eulerian model, chemical reactions are calculated based on the concentration of a pollutant diluted over the entire grid scale. Most of transport and dispersion models use Lagrangian approach due to some limitations in Eulerian model, for example, boundary layer in top entrainment and convective transport, and so forth [4, 5]. The advantage of Lagrangian approach is that it has minimum numerical diffusion [5]. Airmass trajectory is calculated to show the pathway of an infinitesimally air parcel through a centerline of an advected airmass having vertical and horizontal dispersion.

Tracing of the pathway followed by an air parcel upwind from the selected coordinates is termed as “backward air trajectory,” while calculation of best possible pathway to be followed downwind from the selected coordinates in due course of time is called “forward trajectory.” The calculation of backward air trajectory using Lagrangian approach is easier and computationally cheap as it excludes the influence of upwind on the receptor site. Lagrangian approach has also been applied in photochemical modeling [6, 7].

1.2. History of Air Trajectories. Trajectories were first computed by Petterssen in 1940 which were based upon graphic representation [8]. But with the advancement of computer in 1960s, people started isentropic analysis, trajectory calculations, and their graphic representation on computers [9]. Since then, trajectory calculations and their presentation have experienced gradual advancement in techniques and technologies. Trajectory calculations gained more importance when Rodhe plotted airmass trajectories and established that acid rain occurrence in Northern Europe was mainly due

to industrial sources located in the south and west [10]. In air pollution science, Fox and Ludwick also reported one of the pioneering studies using backward air trajectories which identified the source region of pollution [11]. Ashbaugh and coworkers used air trajectories for identifying the sources of sulphur [12]. They also used these trajectories to predict the airmass history and corresponding high levels of sulphates.

In due course of time, different workers started looking into various aspects of trajectory calculations such as errors, accuracy of calculation, and multiple presentations. Polissar and coworkers addressed various issues related to trajectory calculation such as vertical transport, phenomenon of sub-grid scale, convection, turbulences, and uncertainties about meteorological data [13]. Stohl and Fleming and coworkers have reported a comprehensive review of back trajectories [14, 15]. At present, the trajectories are more accurate with 3D presentation having several other computer driven features [16]. Simulations of multiple trajectories are also possible now. The advantage of multiple trajectories is that these provide measure of uncertainty in the pathways of airmass transport [17]. However, calculated trajectories always have some uncertainty and hence user should know the magnitude of such errors while interpreting the results. Assumptions regarding vertical transport, sparse meteorological data, numerical inaccuracies during computation, subgrid scale phenomenon, turbulence, convection, evaporation, and condensation contribute various errors in back trajectory calculations [12, 18, 19]. A review of the associated errors and probabilities within them related to back trajectories has been provided by Polissar and coworkers [13].

1.3. Basic Calculation of Airmass Trajectories. A trajectory can be defined by a differential equation

$$\frac{dr}{dt} = v[r(t)], \quad (1)$$

where $r(t)$ is position vector at time t and v is the velocity field.

Assuming a two-dimensional flow, Seibert suggested the following numerical solution [20]:

$$\begin{aligned} x_1 &= x_0 + \Delta t u(x_0, y_0, t), \\ y_1 &= y_0 + \Delta t v(x_0, y_0, t), \\ x_n &= x_0 + 0.5\Delta t [u(x_0, y_0, t) + u(x_{n-1}, y_{n-1}, t + \Delta t)], \\ y_n &= y_0 + 0.5\Delta t [v(x_0, y_0, t) + v(x_{n-1}, y_{n-1}, t + \Delta t)], \end{aligned} \quad (2)$$

where (x_0, y_0) = starting position at time t , x_n, y_n = n th iteration of the position for the time $t + \Delta t$, u = function of space, and v = function of time.

Details of higher trajectory solutions can be found in Isaacson and Keller and Brumer [21, 22].

1.4. Trajectory Models. Table 1 gives the list of various models used for the calculation of the trajectories. According to the available reports worldwide, HYSPLIT (hybrid single particle Lagrangian integrated trajectory) model has been

TABLE 1: List of various airmass trajectory models used worldwide.

Trajectory models	References
NMC (US National Meteorological Centre)	[23]
Flexible Trajectories (FLEXTRA) model	[24]
TRADOS model (Finnish Meteorological Institute)	[25]
CMDC (Climate Monitoring and Diagnostics Laboratory)	[13]
UK Universities' Global Atmospheric Modelling Programme (UGAMP) model	[26]
Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model	[27]
NIES (Russian National Institute for Environmental Studies)	[28]
Centre for Air Pollution Impact and Trends Analysis CAPITA Monte Carlo (CMC) model	[29]
APTRA model	[30]
European Centre for Medium-Range Weather Forecasts (ECMWF) three-dimensional isentropic model	[31]
LAGRANTO model	[32]
COSMO TRAJ	[33]
CAABA/MJT model	[34]
COBRA	[35]
Stochastic Time-Inverted Lagrangian Transport (STILT) model	[36]

applied in most of the studies [27, 37–41]. The model has been developed by NASA. Besides airmass trajectories, the model also computes dispersion and disposition simulations. Very recently HYSPLIT model has been upgraded. However, FLEXTRA model has also been used by various workers worldwide [3, 14, 15]. Recently, web based system called READY (real-time environmental applications and display) system is developed by the Air Resources Laboratory (ARL). READY is also being used to run trajectory and dispersion model accessing and displaying meteorological data. Simultaneous application of dispersion models, graphical display programs, and textual forecast programs makes this system very useful for atmospheric scientists.

1.5. Applications of Trajectories. In the beginning, trajectories were used to find out the source region and transport processes of air pollution, but, with the advancement of knowledge, trajectories are found to be useful to address various aspects related to atmosphere and environment, as shown in Figure 1. Trajectories are applied in various fields such as climatology [42], meteorology [43], transport of pollutants [9, 14, 44, 45], residence time analysis [46], air quality [47, 48], source apportionment [49, 50], aerosol measurements [51–53], precipitation chemistry [54, 55], and policies [56].

Tarasick and coworkers have used backward and forward trajectories to produce ozone maps using North American ozonesonde data linking it to the grid points of the trajectory pathways [57]. This approach was found to be

FIGURE 1: Applications of air mass trajectories in various fields.

very successful in providing trajectory mapped ozone values having reasonable agreement with the actual soundings. Trajectories have been used to isolate periods of stratosphere-troposphere (S-T) exchange at several rural sites in the United States by Lefohn and coworkers [58]. Trajectory models have been used to study circulation of pollutants [59] including dust storm trajectory simulations. Ashrafi and coworkers attempted such simulations over Iran using dust module of HYSPLIT model which is primarily based on the PM_{10} dust emissions for desert areas [60]. According to their findings dust motion simulations obtained from the model were similar to the MODIS images.

1.6. Trajectories and Air Pollution Transport in South Asia.

A number of studies have been reported on air pollution transport using air mass trajectories in South Asia. Table 2 gives the list of most of the studies on air mass trajectory calculations to demonstrate transboundary and long range transport of air pollution in South Asian countries. Because of large volume of work reported on various aspects of trajectories, it is not possible to describe all these studies in this review. However, we have tried to highlight major pathways of air pollution transport and their wet deposition in South Asian region by describing few important studies in various parts of South Asian region.

1.6.1. Bangladesh. Transboundary air pollution in South Asian countries, namely, Bangladesh, India, Pakistan, and Sri Lanka, has been reported by Brumer to find out the source areas that are primarily responsible for the transport of pollutants [22]. During this study, simultaneous sampling of particulate matter using similar methodology was carried out in Bangladesh, India, Pakistan, and Sri Lanka. According to their report, BC rich smoke is contributed by agricultural waste burning sources located mainly in Northern India (Figure 2). Fine soil is contributed by the dust storms due to which particulate matter levels are enhanced in the air. Based upon the trajectories, they observed that long range transport of the desert dust from Oman through the Gulf and Iran is transported to Pakistan and other South Asian countries.

Saadat and coworkers reported that transboundary transport of pollutants has significant impact on air quality of Bangladesh due to the industrial activities of neighboring

countries India and China [39]. The levels of SO_x and NO_x were recorded to be higher whenever air mass movement was noticed through India (North and North West). However, the levels of these gases were recorded significantly to be lower when the airmasses passed through India but spent enough time over Bay of Bengal. Using HySPLIT model and 10-day backward trajectories, Iqbal and Oanh observed that majority of airmasses arriving to Bangladesh were originated from the west [62]. However, according to their inventory report, 70% of SO_2 emissions in Dhaka division were attributed to local brick kilns.

1.6.2. Indian Ocean. There are a very limited number of wet deposition studies using trajectories which have been reported over Indian Ocean. However, studies on aerosol transport over Indian Ocean have been reported by several workers (Table 2). Granat and coworkers (2002) have reported rainwater chemistry in Indian Ocean region during the Indian Ocean Experiment (INDOEX) campaign held in January–March 1999 [54]. Samples collected onboard the research vessels Ronald H. Brown and Sagar Kanya were analyzed for major ions and some trace metals. They calculated air trajectories arriving at the location of the ship (at 950 ha) with the McGrath trajectory model using wind and mass fields from ECMWF. Five patterns of trajectories around the Indian Ocean have been reported in this paper. Air mass trajectories analysis (Figure 3) revealed that very high concentrations of non-seasalt SO_4^{2-} , NO_3^- , NH_4^+ , $nss-K^+$, and non-seasalt Ca^{2+} in rainwater over the Indian Ocean were due to the influence of pollution and soil sources in Asia.

Long range transport of dust and other pollutants over Indian Ocean has been reported by couple of workers. Most of these studies have been part of INDOEX and focused upon the aerosol optical depth (AOD) or the monsoon circulations. Krishnamurti and coworkers have reported the possibility of long range transport of dust from Arabian Desert to the central Indian Ocean region with a transit time of 2-3 days [66]. Li and Ramanathan presumed that long range transport of dust from the Horns of Africa over the Arabian Sea and mid tropospheric transport of dust from the Arabian Peninsula can affect the summer monsoon and can lead to higher aerosol loadings in south of the equator [65]. According to them, long range transports of emissions from Indonesia forest fires in 1997 were responsible for increase in AODs over most of the equatorial Indian Ocean. Verma and coworkers have used a hybrid approach to find out the pathways of pollutant transport over Indian Ocean during Intensive Field Phase of INDOEX during January–March 1999 [67]. They used an Eulerian forward transport calculation in a general circulation model (GCM) with region-tagged emissions along with an analysis of Lagrangian back trajectories and emission inventory for overlapping time periods. They found that, during the early part of the cruise when ship was moving over Indian Ocean, airmasses from the Indo-Gangetic Plain, Central India, or South India transported sulphate aerosols and organic matter over to Indian Ocean. But during late February–early March when ship was moving in the Arabian Sea, dust species dominated

FIGURE 2: Air parcel back trajectories showing the likely source areas for smoke in India, Bangladesh, Pakistan, and Sri Lanka [38].

FIGURE 3: Measured concentration ($\mu\text{mol m}^{-3}$) of nss-Ca^{2+} , NH_4^+ , and nss-SO_4^{2-} and associated 10-day backwards trajectories for each rain sample; dots show the point of sample collection along the cruise tracks of ships during INDOEX 1999 [54].

which were transported by the airmasses from Africa, West Asia, or Northwest India.

Generally, higher pollution is reported in the north of equator in Indian Ocean [54] as most of the emission sources are located in northern hemisphere. Relatively pristine atmosphere is observed in Southern Indian Ocean due to negligible human perturbations. However, the search operation for missing Malaysian airlines flight MH 370 is expected to emit large amount of aerosols and gaseous pollutants in Southern Indian Ocean due to fuel burning by a number of searching aircrafts and ships. More than a month long operation had used around 20 aircrafts and 20 ships. According to BBC (<http://www.bbc.com/news/world-asia-26514556>), this has been the largest search operation which covered 7.68 million sq km area which is equivalent to 11% of the Indian Ocean and 1.5% of the surface of the Earth. In this relation, the calculations of airmass trajectories can be

of great help to find out the pathways of transport of air pollutants in Southern Indian Ocean. Scavenging of various aerosols and gaseous pollutants can modify the clouds in south of intertropical convergence zone (ITCZ) which can further affect monsoon process.

1.6.3. Bay of Bengal and Arabian Sea. Deposition of acidic components has been reported over Bay of Bengal and Arabian Sea by Reddy and coworkers [52]. This study was carried out during Integrated Campaign on Aerosol and trace gas Radiative Forcing (ICARB). They observed that the air over Bay of Bengal and Arabian Sea was influenced by the airmasses coming from four sectors: (i) passing through Indian land, (ii) from Indian Ocean region, (iii) from Northern Arabian Sea and Middle East, and (iv) from African continent. Figure 4 shows the origin of airmasses reaching various cruise points over Bay of Bengal and Arabian Sea. The highest nss-SO_4^{2-} was observed during airmasses coming from the Indian land while the lowest concentrations were observed when the airmasses were coming from oceanic region. However, total fluxes of pollutants contributed by the airmasses of different direction are not estimated in any of the studies reported in this region.

1.6.4. Maldives. Most of the studies reported from Maldives are part of Atmospheric Brown Cloud (ABC) project as MCOH Climate Observatory is located at Hanimaadhoo Island. Transport of pollutants up to MCOH has been reported by Das and coworkers [68]. They have used airmass trajectories to interpret the water soluble inorganic components in rain deposited at MCOH to find out seasonality and possible source regions. The concentrations of non-seasalt SO_4^{2-} , NH_4^+ , and NO_3^- were observed to be higher by a factor of 4 when airmasses arrived from India as compared to the marine airmasses. Such effect resulted in very low pH of rainwater (4.7), which was significantly lower than the pH values (6.0) contributed by the marine airmasses. Figure 5 shows the trajectories arriving at MCOH (Hanimaadhoo).

TABLE 2: Summary of trajectory studies reported in South Asia.

Location	Country/region	Latitude and longitude	Trajectory model used	Components measured	References
Arabian Sea	Arabian Sea	9.0°N to 22°N 58°E to 77.3°E	CGER METEX programme	Major ions (PM10 aerosols)	[52]
INDOEX	Arabian Sea and Indian Ocean	Multiple points	Wind data (NCEP)	AOD	[61]
Dhaka (AECD)	Bangladesh	23.73°N, 90.40°E	HYSPLIT	BC, aerosol	[38]
Sathkhira	Bangladesh	22.18°N 89.02°E	HYSPLIT	SOx, NOx	[39]
Dhaka	Bangladesh	23.73°N, 90.40°E	HYSPLIT	SOx, NOx, CO, NMVOC, and PM	[62]
Bay of Bengal	Bay of Bengal	20.5°N to 5.6°N, 80.4°E to 93.4°E	CGER METEX programme	Major ions (PM10 aerosols)	[52]
Navi Mumbai	India	19.07°N, 72.97°E	HYSPLIT	BC, aerosol	[38]
Hudegade, Western Ghats	India	14.36°N, 74.54°E	HYSPLIT	Major ions (rain)	[55]
Sinhagad	India	18°21'N, 73°45'E	HYSPLIT	Major ions (cloud water)	[41]
Sinhagad	India	18°21'N, 73°45'E	HYSPLIT	Major ions (rain water)	[40]
Sinhagad	India	18°21'N-73°45'E	McGrath (1989) model with ECMWF	pH and major ions (rain water)	[63]
Pune	India	18°32'N-73°51'E	McGrath (1989) model with ECMWF	pH and major ions (rain water)	[63]
Goa	India	15°45'N- 73°85'E	McGrath (1989) model with ECMWF	pH and major ions (rain water)	[63]
Bhubaneswar	India	20°15'N-85°52'E	McGrath (1989) model with ECMWF	pH and major ions (rain water)	[63]
Manali	India	32.31°N, 77.20°E	HYSPLIT	pH and major ions (snowfall)	[64]
Anantpur	India	14.62°N, 77.65°E	HYSPLIT	O3	[37]
INDOEX	Indian Ocean	30°N to ~35°S, 40°E to ~100°E)	No (wind data from NCEP/NCAR)	AOD	[65]
INDOEX	Indian Ocean	Multiple points	No	AOD	[66]
INDOEX	Indian Ocean	Multiple points	McGrath model with ECMWF	pH and major ions (rain water)	[54]
INDOEX	Indian Ocean	20°S to 17.2°N and 57.5°E to 77°E	HYSPLIT (hybrid approach)	BC, sulphate (aerosol)	[67]
MCOH	Maldives	6°46'N and 73°11'E	HYSPLIT	pH and major ions (rain water)	[68]
Nilore	Pakistan	33.37°N, 73.06°E	HYSPLIT	BC, aerosol	[38]
Multiple sites	Pakistan	Multiple points	HYSPLIT	AOD	[69]
Colombo	Sri Lanka	6.933°N, 79.833°E	HYSPLIT	BC, aerosol	[38]
Delhi	India	28.38°N, 77.12°E	HYSPLIT	Rain chemistry	[70]
New Delhi	India	28.6°N, 77.2°E	HYSPLIT	AOD	[71]

1.6.5. Nepal. Trajectory analysis for wet deposition in Nepal has not been reported. However, according to MISU data collected for rain chemistry at ABC observational site Godavari indicated that airmasses from sectors are received at the observatory. These included (i) from Arabian Sea passing over India, (ii) from Bay of Bengal passing through Bangladesh, (iii) from Middle East, and (iv) from Europe [67]. Sometimes, airmasses were coming from Southeast Asia through Myanmar, Bangladesh, and India. However, our recent observations on snow chemistry at Manali in Himachal Pradesh revealed that airmasses coming from Nepal were found to be responsible for air pollution transport

to India. Such trajectories have been discussed in the last section of this paper.

1.6.6. India. India has the highest landmass, population, and natural resources, as well as the largest industrial setup among all the South Asian countries. Hence, it can be termed as the biggest pollution exporter to the nearby countries and oceanic regions. At the same time India imports huge amount of pollution through long range transport from Europe, Middle East, Africa, Indian Ocean, and nearby countries. However, the amount of such import and export of pollution

FIGURE 4: Airmass trajectories over Bay of Bengal and Arabian Sea during ICARB [52].

from different sectors has not been estimated due to absence of regional emission regulatory body. 1979 Geneva Convention on Long-range Transboundary Air Pollution (CLRTAP) is one such ideal example which aims to limit and, as far as possible, gradually reduce and prevent air pollution including long-range transboundary air pollution by each participating country [72]. CLRTAP has 51 parties so far.

Most of the studies given in Table 2 have reported the origin of airmasses and the type of pollutants contributed by these airmasses. The reported information using trajectories is highly useful in order to understand the sources and pathways of pollution transport to various sites in India. Satyanarayana and co-workers noticed that the long range transport of pollution from various sectors significantly affected the pH and chemistry of rain water at Hudegadde, a rural site located in an ecological sensitive area of Western Ghats [55]. They observed that Hudegadde received the airmasses from five different sectors, namely, central part of Indian Ocean, Northwest Indian Ocean, Southwest Indian Ocean, and Gulf and airmasses passing through Northwest Indian Ocean and south continental part of India. Airmass trajectory analysis suggested a significant influence of local and intercontinental anthropogenic and natural sources. Sources located in the African and Gulf regions had influenced the acidity at the site. They found that urban activities located in southwest of Indian continent (north of sampling site) and the sources in Gulf region were responsible for acid rain in Western Ghats (Table 3).

Chemical composition of precipitation during different trajectory classes has been reported by Norman and coworkers (2001) at Bhubaneswar, Goa, Sinhad, and Pune [63]. They compared the rain chemistry results with trajectories and precipitation fields. As shown in Table 4, a systematic change in chemical composition and pH of rain water was noticed in accordance with the origin of airmasses and the amount of rainfall along the trajectory. Bhubaneswar which is located in Eastern India received airmasses from Arabian Sea, Bay of Bengal, and continental sources. Different trajectories arriving to Bhubaneswar have been shown in Figure 6.

TABLE 3: Concentrations ($\mu\text{eq L}^{-1}$) of major ions in different air-masses at the site (source: [55]).

	C.I.O.	N.W.I.O.	S.W.I.O.	Gulf	N.W.I.O. + S.W.I.C.
Cl^-	12	40	64	0.3	0.3
NO_3^-	4	29	9	11	13
nss SO_4^{2-}	11	35	35	12	16
Na^+	16	45	59	7	3
NH_4^+	5	28	2	2	4
K^+	5	3	9	16	2
nss Ca^{2+}	38	71	68	21	3

C.I.O.: Central Indian Ocean; N.W.I.O.: Northwest Indian Ocean; S.W.I.O.: Southwest Indian Ocean; N.W.I.O. + S.W.I.C.: Northwest Indian Ocean + Southwest Indian Continent.

From West India, most of wet deposition studies have been reported for Pune region. Using airmass trajectories, Begum and coworkers (2011) observed that the long range of transport of air pollutants and dust was responsible for elevated levels of selected ions in cloud water at Sinhad which is a high altitude site near Pune [38]. In another study at Sinhad, Reddy and coworkers (2010) observed that monsoon and postmonsoon airmasses had their origin from different directions affecting rain chemistry accordingly [37]. As shown in Figure 7, during monsoon season, air moved from African region. But during postmonsoon season, air parcel is seen originating from Middle East and Europe travelling through North India and finally reaching Sinhad. Authors have reported that, due to this reason, the concentrations of NO_3^- , NH_4^+ , nss-SO_4^{2-} , and nss-K^+ were observed to be higher in postmonsoon samples as compared to monsoon samples.

1.6.7. Long Range Transport up to Western Himalayan Ranges.

In order to observe the airmass movement in North India up to the Western Himalaya, we carried out snow chemistry study coupled with trajectory analysis. In our study, the samples of snowfall were collected at a high altitude site Kothi (32.31°N , 77.20°E) located in Kullu district of Himachal Pradesh state of India, which were processed for chemical analysis. Five-day back trajectories at 5000 m altitude were calculated for these samples using HYSPLIT model from NOAA [27]. Airmass trajectories revealed that Kothi was receiving snowfall through the airmasses originating from six major sectors which are termed as (1) North Atlantic Ocean origin (NAO), (2) African origin (Af), (3) Middle East origin (ME), (4) European origin (Eu), (5) West India origin (WI), and (6) Nepal origin (Np). Figures 8(a)–8(f) show typical examples of these trajectories.

North Atlantic Ocean Origin (NAO). Airmasses originating from North Atlantic Ocean and passing over Europe to Afghanistan to Jammu and Kashmir and reaching the sampling site, that is, Kothi, have been referred to as NAO (Figure 8(a)). This cluster represented the highest number of events (40%).

African Origin (Af). Airmasses originating from African continent and passing over Middle East/Gulf to Pakistan

FIGURE 5: Classification of trajectories arriving at MCOH, (a) Marine, (b) Arabian Sea, (c) Indian, and (d) mixed [68].

TABLE 4: Median concentrations (mmol L^{-1}) in rain samples at Bhubaneswar for different trajectory groups and frequency of rain along the trajectory ([63]).

Sample group	pH	Na ⁺	NH ₄ ⁺	K ⁺	Mg ²⁺	Ca ²⁺	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	nss-SO ₄ ²⁻	Ca ²⁺ /Na ⁺	Number of samples
High rain frequency												
C	5.48	6.3	22.2	1.2	1.3	3.1	6.5	9.4	6.8	6.5	0.66	18
CL	4.84	28.2	73.5	9.4	6.9	28.1	34.7	53.8	37	35.3	0.79	2
B	5.54	6.1	9.1	0.5	0.7	1.5	7.5	2.5	3.1	2.8	0.25	14
M	5.82	11.9	20.7	1.1	11.6	7.2	12.6	10.9	8.9	6.7	0.53	23
Low rain frequency												
C	5.61	16.9	44.7	3.7	3.8	10.5	23	29	21.2	19.6	0.58	21
CL	5.03	18.2	52.6	4.5	5	28.4	20	44	40.2	39.1	2.33	2
B	5.28	31.8	81.3	7.9	6.3	17.7	34.5	49	36.7	35.8	0.8	6
M	5.77	12.4	11.3	0.6	2.4	5	14.5	9.4	9.2	8.4	0.56	8

Note: (a) continental (C), (b) continental local (CL), (c) monsoon (M), and (d) Bay of Bengal (B).

before reaching the sampling site have been termed as Af. This cluster represented 23% of snowfall events. Figure 8(b) is a typical example of such airmasses.

Middle East Origin (ME). Airmasses originating from Middle East and reaching the sampling site via Pakistan and Haryana have been termed as ME. This group represented 10% of the total snowfall events. Figure 8(c) is a typical example of ME airmasses.

European Origin (Eu). Airmasses originating from continental Europe and reaching the sampling site via Afghanistan and Jammu and Kashmir have been referred to as Eu. This class represented the least number (6%) of snowfall events. Figure 8(d) is a typical example of such airmasses.

Western India Origin (InW). Airmasses originating in Western India in the state of Rajasthan and transported over

FIGURE 6: Trajectories to Bhubaneswar, sorted into different trajectory groups during January 1997–October 1998. (a) Continental (C), (b) continental local (CL), (c) monsoon (M), and (d) Bay of Bengal (B) [63].

Punjab and Haryana and, finally, reaching the sampling site have been termed as InW. This group represented 13% of the total snowfall events. Figure 8(e) is a typical example of this type of airmasses.

Nepal Origin (Np). Airmasses originating from Nepal side reaching the sampling site via Uttarakhand have been categorized as Np. This cluster represented the second lowest number of events (8%). Figure 8(f) is a typical example of this type of airmasses.

In general, pH of the samples varied from 4.75 to 6.98 with an average value of 5.69 indicating slight alkaline nature of snow samples. Slight alkaline precipitation has been reported at higher altitude in Indian region [40, 41, 73–75]. Figure 9 shows the percent frequency distribution of pH of snowfall samples. Maximum number of samples had pH between 6.21 and 6.60 (29%) followed by a range of 5.81–6.20 (25%), 6.61–7.00 (17%), 5.41–5.80 (17%), 4.61–5.00 (10%), and 5.01–5.40 (4%). The high pH ($\text{pH} > 5.6$) has been reported in

global precipitation samples due to the influence of soil derived particulate matter [40, 68, 76–80]. Out of total, 83% of samples showed pH of snow above 5.6, while 17% of samples only were found to be acidic at Kothi. Acidic snow samples were found at this site in airmasses originated from Middle East Countries and Europe mainly. Acidic pH has been reported in precipitation samples in airmasses originated from Europe [80] and Gulf countries [55]. 43% of rain samples were observed to be acidic in nature at state botanical garden in Bhubaneswar, India [81]. 18% occurrence of acid rain has been reported at Delhi, India [76]. pH value ranged between 3.98 and 8.01 with 20% acidic precipitation samples having been reported at Montseny in Northeast Spain, Europe [80]. Out of total precipitation samples, 21% of samples were found to be acidic in nature at Sinhagad, Pune, India [82]. 79.3% of the total precipitation has a pH value less than 5.6 at Jinhua in Southeastern China's province [83].

Table 5 gives average concentration of chemical species in snowfall samples. On average, equivalent concentration of ionic species followed the following order: $\text{Ca}^{2+} > \text{Cl}^- >$

FIGURE 7: Seven-day back trajectories at Sinhgad during the (a) monsoon and (b) postmonsoon seasons of 2005 [40].

TABLE 5: Descriptive statistics of pH and major ions ($\mu\text{eq L}^{-1}$) in snowmelt.

	pH	Na^+	NH_4^+	K^+	Ca^{2+}	Mg^{2+}	F^-	Cl^-	NO_3^-	SO_4^{2-}	HCO_3^-
Average	5.69	43.69	18.46	10.84	47.13	17.04	3.02	45.99	17.42	27.45	27.40
s.d.	0.6	23.5	10.8	5.7	21.1	7.9	2.3	25.3	7.3	8.3	17.7
Min.	4.75	7.51	4.23	2.99	17.65	0.10	0.08	10.75	5.43	11.95	1.72
Max.	6.98	93.56	39.41	21.55	96.44	35.31	7.02	106.51	31.11	46.73	71.91

$\text{Na}^+ > \text{SO}_4^{2-} > \text{HCO}_3^- > \text{NH}_4^+ > \text{NO}_3^- > \text{Mg}^{2+} > \text{K}^+ > \text{F}^-$. Very high concentration of Ca^{2+} among all ions at this site indicated the crustal dominance in snowfall samples at Kothi. The average concentration of Ca^{2+} was compared with others of snow and found to be higher than Mount Everest [84], Mt. Logan Massif [85], Khumbu-Himal, Nepal [86], but lower than Yulong snow Mt. [87] and Tien Shan [88]. In Indian region, many researchers have reported high concentration of Ca due to suspended soil dust in precipitation samples [76, 89, 90]. Since the site is situated at high altitude with less eroded soil [73], Ca^{2+} is not expected to be contributed by local soil significantly. As discussed above in the same section, the contribution of transported dust from African region, West India, and Nepal was significantly high at this site. Very high concentration of SO_4^{2-} and NO_3^- might be due to its transport through Af, ME, InW, and Np airmasses. The concentration of SO_4^{2-} in snow samples at this site was lower than surface/fresh snow samples of Yulong snow Mt. [91], ice core samples of Dasuopu [92], and East Rongbuk [93], but lower than snow samples of Mt. Everest [84], Khumbu-Himal, Nepal [86], and Mt. Logan Massif [85]. Very similar range of concentration of SO_4^{2-} has been reported by many workers in global precipitation due to fossil fuel combustion mainly [55, 73, 80, 82, 89]. NO_3^- concentration was found to be very high, which might be due to transport of $\text{NO}_x/\text{NO}_3^-$ from various airmasses as discussed above in the same section. Some contribution of NO_3^- from biomass burning during winter season to produce heat by local people cannot be ruled out. After Ca^{2+} , very high concentration of Cl^- and Na^+ among all ions indicated strong influences of marine components at this site and it is very clear from the above discussion. The high concentration of NH_4^+ in snowfall samples might be due to its transport from African continents

and Nepal mainly. Similarly, high concentration of K might be due to its transport of airmasses from Af, InW, and Np mainly.

2. Conclusion

Presently, computation of trajectories is highly advanced. Multiple and 3D trajectories are possible with numerous additional features, which make computation of backward, forward, and vertical motion very simple. However, there are uncertainties associated with the calculated trajectories and hence precaution of such errors is necessary for their meaningful interpretation. Nevertheless, the airmass trajectories are powerful tools for tracing pollution sources and the routes of transport. Transboundary pollution is becoming a serious issue in South Asia. Transport of pollutants from nearby countries as well as from far Europe, Middle East, and African continental regions to South Asia needs immediate attention of scientists and policy makers. Studies indicate that the net import and export of pollutants vary depending upon the season as the origin and path of airmasses change during different seasons and so the deposition load of different types of pollutants at the receptor sites. Hence, similar to CLRTAP, South Asian region also needs to develop a network to monitor the nature and type of pollutants and their quantification of annual import and export, followed by appropriate methods of assessment in relation to damage to human health, forests, and various agroecosystems. Airmass trajectories revealed that acidic pollutants from Europe and Middle East regions are deposited up to the remote Himalayan ranges via long range transport. Trajectory calculations provided very vital information about the continental transported emissions which were responsible for the elevated level of

FIGURE 8: Typical examples of six different airmass trajectories arriving at the site. NAO = North Atlantic Ocean origin, Af = African origin, ME = Middle East origin, Eu = European origin (Eu), InW = Western India origin, and Np = Nepal origin.

FIGURE 9: Percent frequency distribution of pH of snowmelt for whole dataset.

acidity of rain water at an ecosensitive site in Western Ghats in Southwest India.

Conflict of Interests

The authors declare that they have no conflict of interests regarding the publication of this paper.

Acknowledgments

Support of DST-PURSE and UGC India is gratefully acknowledged.

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