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Identification of Noble Gases Sources using Atmospheric Trajectory Modelling

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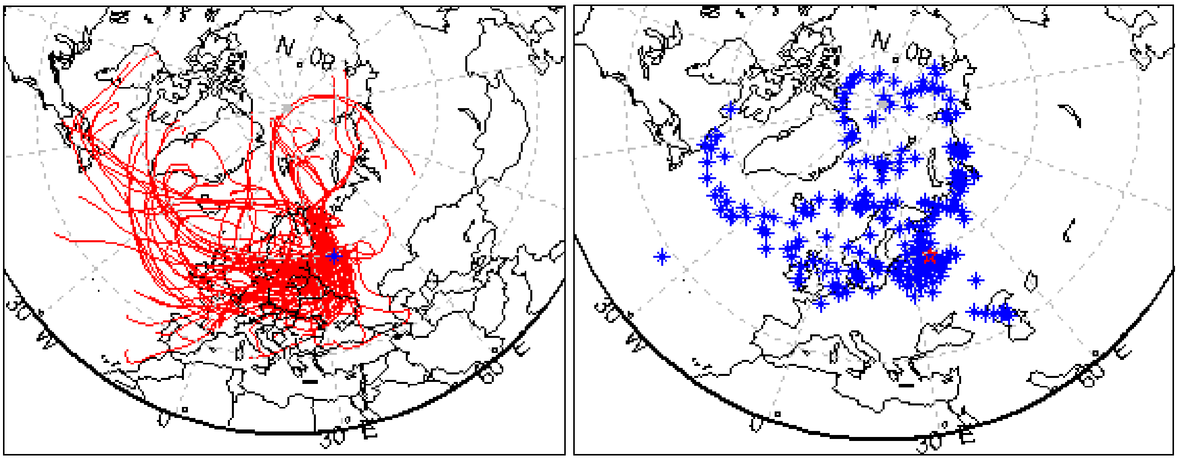

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Abstract

Studies connected with atmospheric transport, distribution, and variations of radioactive noble gases (^{85}Kr and ^{133}Xe) are of significant importance for the environment and population. Continuous accumulation of ^{85}Kr in the atmosphere and identification of accidental nuclear releases as well as monitoring of safe operation of nuclear power plants by evaluating ^{133}Xe atmospheric transport are challenging problems that are needed serious attention of the scientific community.

The main goal of this study was an estimation of atmospheric transport patterns for elevated and lowered concentrations (i.e. above/ below a range of accepted standards and mean values) of radioactive noble gases (RNG).

The RNG monitoring was performed during the period of two years (Aug 2006 – Jul 2008) by the V. G. Khlopin Radium Institute (St. Petersburg, Russia) at the Cherepovets measurement site (Vologda Region) considered as a background station. These measurements were performed within the framework of the international project «*Development of methodical bases and mobile equipment for monitoring of Xe and Kr radionuclides in the Northwest region of Russia*» (2005-2009).

At first, in this work the time-series of ^{85}Kr and ^{133}Xe measurements were analyzed. As a result, in total 28 (26) elevated and 17 (36) lowered episodes for krypton (xenon) were selected. At second, in order to estimate atmospheric transport of these radioactive gases as well as identify potential sources of their emissions the HYbrid Single-Particle Lagrangian Integrated Trajectory model was used. Almost 1100 individual backward trajectories were simulated for ^{85}Kr and ^{133}Xe selected episodes and analyzed in more details. At third, ensembles of trajectories related to selected extreme episodes for both gases as well as periods with elevated and lowered concentrations were analyzed. This allowed estimating general directions and sectors of contaminated air transport and identifying potential sources of emissions associated with these episodes as well as estimate spatial variability of airflow patterns. Finally, to derive mean atmospheric flow patterns and obtain relevant probabilities the cluster analysis technique was applied.

The dominating atmospheric transport associated with elevated concentrations of RNGs occurred from the western sector where a large number of nuclear power plants are situated and accounted 42.5% and 36.2% for krypton and xenon, correspondingly. Lowered concentrations of ^{85}Kr and ^{133}Xe were related to atmospheric transport from the northern sector accounting for 34.9% and 31.6%, correspondingly.

1. Introduction

The discovery of ozone hole (1980s), the use of chemical and biological agents in World Wars I and II, and of course the intensive nuclear tests realization started in 1940s as well as Chernobyl accident (1986) played a crucial role in research activity in the area of atmospheric transport and dispersion of air particles and gases. Research organizations in different countries launched their research programmes in order to study nuclear releases, fallout and perform source estimations. Following the Comprehensive Nuclear-Test-Ban Treaty (CTBT) (since 10 Sep 1996) all nuclear explosions in all environments are strictly prohibited now, but the routine operation of nuclear power plants (NPP) still causes radioactive gases being released into the atmosphere. Among a variety of radioactive gases being originated during the nuclear fuel reprocessing processes there are noble gases that are less harmful for people, but very important for environment radiation condition control (ERCC).

One of significant steps in realization of ERCC is an estimation of atmospheric transport of radionuclides. Solving problems of contaminated air mass transport both macro-meteorological conditions (i.e. meteorological conditions determined by geographical areas) and small-scale spatial and temporal variations of meteorological parameters should be taken into account. A number of complex atmospheric processes and factors - like turbulence, cyclonic and anticyclonic activity caused by advection and dynamical factors, convection (although its influence in comparison with horizontal movement is rather small), common movement of the atmosphere due to the Earth rotation, etc. - determine pollution transport processes (dispersion, deposition, chemical transformation as well as radioactive decay in case of radionuclides) in the atmosphere. A majority of these factors are taken into account in numerical simulation models which allow determining individual and multiple atmospheric transport paths, spatial distribution of air masses, etc. depending on the methods applied.

For estimation of possible influence of radiation risk objects by means of atmospheric transport the trajectory modelling approach can be used (*Mahura et al., 1999; Mahura & Baklanov, 2002*). This approach is widely used in different research areas like, for example, radioactive accidents studies (*Baklanov et al., 2002*), aerosols (*Mukai & Suzuki, 1996*) or airborne pollen dispersion and elevated birch episodes (*Šaulienė et al., 2006; Mahura et al., 2007*), ozone (*Guangfeng Jiang et al., 2003*) studies, and others.

In this study on the basis of the measured ^{85}Kr and ^{133}Xe volumetric activities (obtained by the V. G. Khlopin Radium Institute at the Cherepovets measurement site (59,11°N; 37,90°E, Vologda region, Russia)) for the period of two years (Aug 2006 – Jul 2008) the trajectory modelling in combination with cluster analysis technique was applied. From a point of view of environmental and radiation safety of the North-West Russia (and Cherepovets, in particular) the backward trajectory calculation technique was of the largest interest. A level of potential contamination by radioactive noble gases (RNGs) in the observed region, as well as possible source areas that could be reasons for episodes with elevated and lowered volumetric concentrations of krypton and xenon, and potential sources of these noble gases emissions are presented and discussed.

In particular, the preliminary estimation for several episodes with krypton (^{85}Kr) and xenon (^{133}Xe) elevated concentrations for the North-Western region of Russia (St. Petersburg and Cherepovets) was performed by *Petrova et al. (2008ab; 2009).* Here, a more detailed analysis is presented for the Cherepovets measurement site considered as the background station. Moreover, similar approach of trajectory calculation author also used to link trajectories with elevated ozone levels (*Mahura et al., 2010*).

2. Methodology

2.1 Noble Gases – Characteristics, Measurements, and Sources

Radioactive noble gases are emitted into the environment mainly during routine operation of the nuclear power plants and nuclear fuel reprocessing. In this section the main peculiarities of ^{85}Kr and ^{133}Xe gases (§2.1.1, §2.1.2), monitoring, measuring technique and sampling (§.2.1.3, §2.1.4, §2.1.5) as well as potential sources of these gases (§2.1.6) will be discussed.

2.1.1 Krypton

After successful development of nuclear power engineering and industry in the second part of the XXth century the potential environmental risk of nuclear tests and nuclear power plants exploitation has been recognized (*Burlakova et al., 1998; Bogatov et al., 2006*). One of such risks appears to be accumulation in the atmosphere of radioactive gas called krypton-85 with the half-life time $T_{1/2}$ = 10.76 years.
This radionuclide can be of natural (radiogenic and cosmogenic) as well as induced (technogenic) origin. The amount of natural ^{85}Kr remains almost the same with a time while a great amount of induced origin is routinely formed during the nuclear fuel reprocessing processes and nuclear power plants (NPP) operation. That is why the amount of technogenic krypton in the atmosphere is continuously increasing.
Substantial growth of ^{85}Kr content in the atmosphere determined by intensive realization of nuclear tests during 1945 - 1956 and 1961 - 1963 as it was registered at the beginning of observations. Nowadays typical content of ^{85}Kr for the middle latitudes' surface air of the Northern Hemisphere is equal to 1.5 Bq/m^3 that exceeds the initial value (before the nuclear era) at almost million times (*Pahkomov, 2009*). Other conducted observations also show significant increase of this radionuclide concentrations during the periods of 1950-1977 in the Northern hemisphere (*Rozanski, 2003*), during 1993–2004 in the atmosphere over the Eastern Europe (*Winger et al., 2005*) as well as during 1995 – 2001 in Japan (*Hirota, 2004*). The continuous development of nuclear industry even without its military applications will inevitably lead to further growth of krypton concentration in the atmosphere that may cause worsening of population health and pollution of the environment as well as change of atmosphere electrical conductivity and greenhouse effect formation (*Harrison & ApSimon, 1994*).

2.1.2 Xenon

Like all noble gases placed in the 18th group of the Mendeleev's periodic table xenon (Xe) at normal conditions is odourless, colourless gas that is generally nonreactive. It can be formed by natural sources as well as during the radioactive decay of such elements like iodine, uranium and plutonium. The main radioactive isotopes of Xe generated as a result of nuclear fission reaction are ^{133}Xe ($T_{1/2}$=5,245 days), ^{135}Xe ($T_{1/2}$=9,1 hours), ^{133m}Xe ($T_{1/2}$=2,19 days), and ^{131m}Xe ($T_{1/2}$=11,9 days), emitted along with ^{85}Kr in enormous amounts in the ambient atmosphere as a result of nuclear explosions and accidents (*Shultis and Faw, 2002*).
The highest yield among the xenon isotopes has ^{133}Xe. Having a relatively large half-life time of more than five days this radionuclide can be measured at considerable distances from the emission sources. The main sources of ^{133}Xe are nuclear plants, medical radioisotope production facilities and hospitals with nuclear medicine departments and research laboratories. The contribution of the last is extremely weak in comparison with the two previous potential sources. Elevation of concentration of short-lived xenon can be used as an indicator of breaking down of gas purification operation system on NPPs, emission of xenon by medical radiopharmaceutical enterprises or for identifi-

cation of non-announced nuclear accidents.
In contrast to krypton, xenon is not accumulated in the atmosphere but it is also of importance for radiation condition control, and this gas is also routinely monitored by research organizations and special production factories.

2.1.3 Monitoring of noble gases

Obviously, the knowledge and development of nuclear engineering could not progress without routine tests and accidental releases. The total number of nuclear tests for the period of 1945-1998 implemented by 7 countries (USA, USSR, Great Britain, France, China, India, and Pakistan) amounts 2053 (*http://ctbto.org*). The intensive realisations of the tests in all environments (atmosphere, hydrosphere, and lithosphere) was followed by strong emissions of radioactive gases including noble ones, and have become the main reason for substantial growth of krypton content in the Earth's atmosphere.
Hence, the monitoring of radioactive noble gases now is of great interest due to the possibility to detect if some accidents (or even not announced test explosions) could take place. The leading world organization that implements monitoring of noble gases is the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) established in 1996 in Vienna, Austria. In particular, the minimum requirements specify that the four isotopes of noble gas xenon - ^{131m}Xe, ^{133m}Xe, ^{133}Xe and ^{135}Xe - are measured now by the CTBTO monitoring stations, although there are other noble gases which are of potential relevance for the CTBTO (*Noble gas handbook, 2003)*.
Noble gas of krypton is not monitored yet by the CTBTO. Nevertheless, measurements of this gas are performed by scientists from countries involved in the International Science and Technology Center (ISTC) project. It is also planning to expand the number of krypton monitoring sites. This will allow creating a global monitoring system for this radionuclide which concentration is continuously growing.
The radioactive noble gases can be used in radiation condition control, i.e. serve as indicators of underground nuclear explosions as well as nuclear power plants operation safety. The basic concept of such control is a monitoring conducting of gaseous radionuclides content in the atmosphere including isotopes of these radioactive noble gases. Measurements of atmospheric concentration of ^{85}Kr and ^{133}Xe have been made also, for example, by *Ferber et al., (1977); Telegadas et al., (1978); Fontaine et al.,(2004); Smith et al.,(2005)*. Systematic measurements of its content are carried out by the V. G. Khlopin Radium Institute (St. Petersburg) (*Dubasov et al., 2007*).

2.1.4 Technique for measurements of noble gases

At present the measuring of krypton and xenon concentrations is usually implemented on cryogenic factories dealing with generation of liquid air or nitrogen and oxygen. However, due to increasing number of nuclear reactors, nuclear plants, and nuclear fuel reprocessing plants the mobile equipment for Kr and Xe sampling has been designed in order to study RNGs emissions and their spatial and temporal distribution in the air
Normally the noble gases are extracted from the residual krypton-xenon mixture. Two main stages of these gases extraction from the sample are the following: on the first stage so-called “poor concentrate” of Kr and Xe is generated using refrigerating plant; on the second stage the samples obtained are enriched using adsorptive and chromatographic plants in which they are brought to concentration that can be measured by making use of radiometric methods (*Styro & Butkus, 1988*).
Such method with some modifications has been used by the Radium Institute specialists to analyze samples obtained at the Cherepovets measurement site.

2.1.5 Sampling of ^{85}Kr and ^{133}Xe in this study

Monitoring of radioactive noble gases was performed by the V. G. Khlopin Radium Institute specialists during Sep 2006 – Jul 2008 within the framework of the international project «*Development of methodical bases and mobile equipment for monitoring of Xe and Kr radionuclides in the Northwest region of Russia*» (2005-2009). Scientists from Russia, Canada, Sweden, Germany and Denmark have been involved in this project.

According to *Dubasov (2009)*, beginning 1 Aug 2006, Xe and Kr radionuclides monitoring was started in Cherepovets city (Vologda region). The measurement site was located far from potential nuclear risk objects. Xenon and krypton fraction extracts were sampled and analyzed on one of the factories dealing with oxygen production. The separation of krypton and xenon was performed with the use of low-temperature sorption on the charcoal SKT-3 in the laboratory with its further reduction in the form of spectrometric preparations. Gamma spectrometers with germanium detector were used as a tool for measuring the ^{85}Kr. Each sample was measured 3-5 times and the measuring duration was no less than two hours. The volume of ^{85}Kr in the measuring ampoule comprised 500-1380 sm^3 that was equivalent to 450-1200 m^{3}of atmospheric air.

Xenon fraction extracted from the xenon and krypton mixture was sorbed on a charcoal in an ampoule which was after that placed in a germanium detector. The sampling was performed once-twice per month.

The measurements obtained could be used for the improvement of radiation situation control at NPPs and adjacent areas. From these measurements the periods with elevated/ lowered concentrations above/ below mean values were identified. In total these accounted 45 and 62 episodes for Kr and Xe, correspondingly.

2.1.6 Nuclear power plants as sources of noble gases

Among all possible RNG sources the nuclear power plants (NPPs) were of interest in this study. A list of NPPs considered as potential sources is shown in the Figure 2.1. Their names and abbreviations, corresponding geographical coordinates and country of their location are presented in the Table 2.1

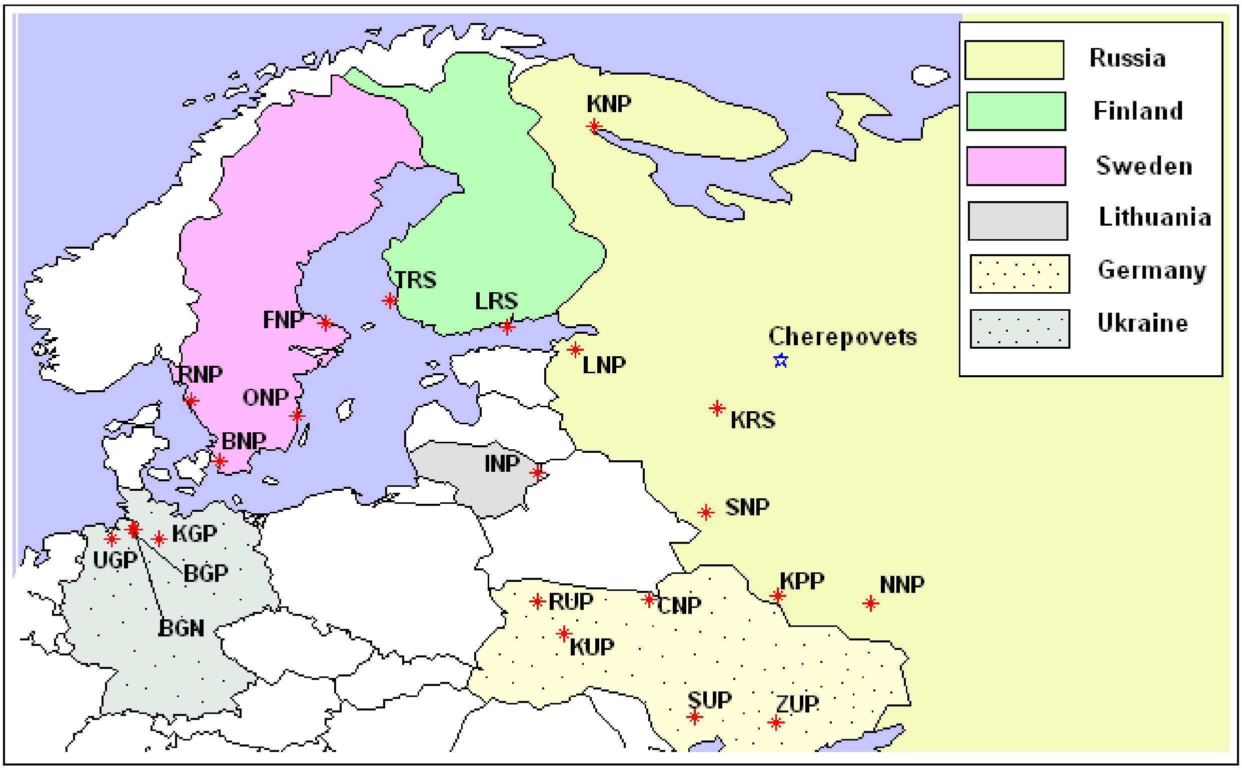


Figure 2.1*: Locations of selected nuclear power plants.*

***Table 2.1**: Nuclear power plants selected for the study.*

#	Site	Lat,°N	Lon,°E	Site Name of NPP	Country
1	KNP	67.75	32.75	Kola	Russia
2	LNP	59.90	29.00	Leningrad	Russia
3	KPP	51.67	35.60	Kursk	Russia
4	KRS	57.88	34.98	Kalinin	Russia
5	NNP	51.27	33.23	Novovoronezh	Russia
6	SNP	54.15	33.23	Smolensk	Russia
7	INP	55.50	26.00	Ignalina	Lithuania
8	LRS	60.50	26.50	Loviisa	Finland
9	TRS	61.50	21.50	Olkiluoto (TVO)	Finland
10	FNP	60.40	18.25	Forsmark	Sweden
11	ONP	57.25	16.50	Oskarshamn	Sweden
12	RNP	57.75	12.00	Ringhals	Sweden
13	BNP	55.75	13.00	Barsebaeck	Sweden
14	CNP	51.23	30.05	Chernobyl	Ukraine
15	RUP	51.19	25.53	Rovno	Ukraine
16	KUP	50.18	26.38	Khmelnitskiy	Ukraine
17	SUP	47.49	31.13	South Ukraine	Ukraine
18	ZUP	47.30	34.35	Zaporozhye	Ukraine
19	UGP	53.25	8.28	Unterweser	German
20	BGP	53.51	9.20	Brokdorf	German
21	BGN	53.53	9.12	Brunsbuettel	German
22	KGP	53.24	10.24	Kruemmel	German

2.2 Modeling of Atmospheric Transport Patterns

2.2.1 Trajectory modelling approach

The atmosphere represented by complicated physical and chemical processes and being in continuous motion is responsible for transport of air pollutants, tracking of which is important aspect for solution of different problems such as identification of sources, evaluation of contamination levels, elaboration of methods and models for emergency response, validation of models, etc.

A simple but rather effective method of tracking transport of air parcels in the atmosphere is the trajectory modelling technique. It is used to simulate transport pathways of pollutants in the atmosphere by making use of meteorological data and mathematical equations. Trajectory itself represents the time integrated advection of each particle and is calculated by making use of three-dimensional (more often) velocity field.

Trajectory modelling method has been widely used in atmospheric transport studies (*Stohl, 1998*) starting with the simple box-models (*Sportisse B., 2001*), and further using the Lagrangian approach for calculation of air parcels trajectories (*Stohl et al., 2002*).

In general, atmospheric trajectory modelling has found its applicability in different tasks of environmental studies (*Mahura et al., 2003*) including evaluation of source-receptor relationship (*Mahura & Baklanov, 2003, INTAS, 2003*). Tracking of substances pathways from the source of emissions and identification of receptors areas is realized by calculating a set of forward trajectories. Backward trajectories in turn can be used for identification of potential sources of substances arrived at receptor site.

As more advanced approach the atmospheric dispersion modelling can be used allowing following the variations of pollutants concentrations in time in the ambient air (*Koopman et al., 1989; Cheol-Hee Kim et al., 2008*). To simulate these variations different meteorological data is used for input

and numerical methods - for calculation. Depending on the system of equations as well as computational methods four basic types of air pollution dispersion models can be named: the simplest box models, Gaussian, Eulerian, and Lagrangian models. The last use moving frame of reference, i.e. mathematically follow pollution plume particles. Eulerian methods apply so-called fixed three-dimensional grid as a frame of reference and are typically used for solving more complex emission cases in opposite to Lagrangian methods (*Turner, 1994*).
Radioactive noble gases are good tracers for atmospheric studies with usage of trajectories due to their physical and chemical characteristics. These "convenient" properties of RNGs can be used for example in validation of air transport models (*Draxler, 1982ab*). The most relevant feature of noble gases is that they are always gaseous from production until radioactive decay. Particles attach to aerosols and can be removed from the atmosphere by precipitation. The volatility of noble gases results in transport properties which are significantly different from the transport properties of particles. Noble gases of xenon and krypton are generally inert and form very few chemical compounds that appears to be useful for their tracking in the atmosphere.
Evaluation of the atmospheric transport to the measurement site and identification of emission sources of ^{133}Xe and ^{85}Kr were the main objectives of this study. In addition, the same backward trajectory modelling approach applied had been used to evaluate atmospheric chemical transport and climatological patterns for elevated ozone episodes in Denmark (*Mahura et al., 2010*).

2.2.2 HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model

To calculate multiple individual backward trajectories arriving at the site at times corresponding to measurements the on-line HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT v.4.5) NOAA (National Oceanographic and Atmospheric Administration) model was applied (*http://www.arl.noaa.gov/HYSPLIT.php*). The Air Resources Laboratory's (ARL) HYSPLIT model is a complete system for computing both simple air parcel trajectories and complex dispersion and deposition simulations. The model calculation method is a hybrid between the Lagrangian approach, which uses a moving frame of reference as the air parcels move from their initial location, and the Eulerian approach, which uses a fixed three-dimensional grid as a frame of reference. In this model, advection and diffusion calculations are made in a Lagrangian framework following the transport of the air parcel *(Draxler and Rolph, 2003; Rolph, 2003).*
Meteorological data for trajectory calculations are obtained from meteorological dataset produced by models. HYSPLIT minimum requirements for calculations are horizontal wind components (U and V), temperature (T), height (Z) or pressure (P), and surface pressure P_o. To use the initial data some pre-reprocessing procedure is required, at first. According to with *Draxler and Hess, (1998)* in the beginning the meteorological data profiles at each horizontal grid point are linearly interpolated to a terrain-following σ-coordinate system:

$$\sigma = 1 - z/Z_{top}, \qquad (1)$$

where:

z_{top} – top of HYSPLIT's coordinate system,
z – heights expressed relative to terrain.

The advection itself is calculated from the average of the three-dimensional velocity vectors which are linearly interpolated in time and space. Final position will be determined by the formula:

$$P(t+\Delta t) = P(t) + 0.5\,[V(P,t) + V(P',t+\Delta t)]\,\Delta t, \qquad (2)$$

where:

P and P' – initial and first-guess positions,
Δt – integration time step,
V – velocity.

Finally, using the HYSPLIT modelling system and setting up on-line the date, arrival height, plotting options, meteorological parameters calculation, etc. atmospheric trajectories are simulated in a chosen domain.

2.2.3 Meteorological input dataset

In this study the National Center for Environmental Prediction (NCEP) and the National Center for Atmospheric Research (NCAR) Reanalysis dataset (global, from 1948 - present) for the HYSPLIT 4 model was used.
The NOAA's ARL in their air quality transport and dispersion modelling calculations routinely uses the NCEP data. Some of these datasets have been archived by ARL starting from 1989 for future research studies. They can be freely used for research and educational purposes.
The NCEP/NCAR Reanalysis dataset is a joint product of the NCEP and NCAR dating back to 1948 (*Kalnay et al., 1996*). It is continually updated and represents the state of the Earth's atmosphere, incorporating observations and global climate model output.
The reanalysis of more than 50-year data on global distribution fields of atmospheric parameters was done by USA National Centers. The measurements from surface stations, ships, and planes, rawinsonde sounding, weather-balloon, and satellite observations data (since 1970s) were used in order to reconstruct atmosphere state data in knots of a regular grid. All the data was carefully checked and assimilated by making use of an assimilation system saved without any changes during the reanalysis period.
The dataset available from 1 January 1948 to present contains data on geopotential height, wind, and temperature on 1 degree latitude-longitude global grid. It consists of 17 pressure levels from 1000 hPa to 10 hPa and 28 sigma levels.
The Reanalysis dataset advantages are a state-of-the-art data assimilation, increased number of observations, improving of quality control, remaining the model/ data assimilation procedure unchanged during the project and global adaptation.

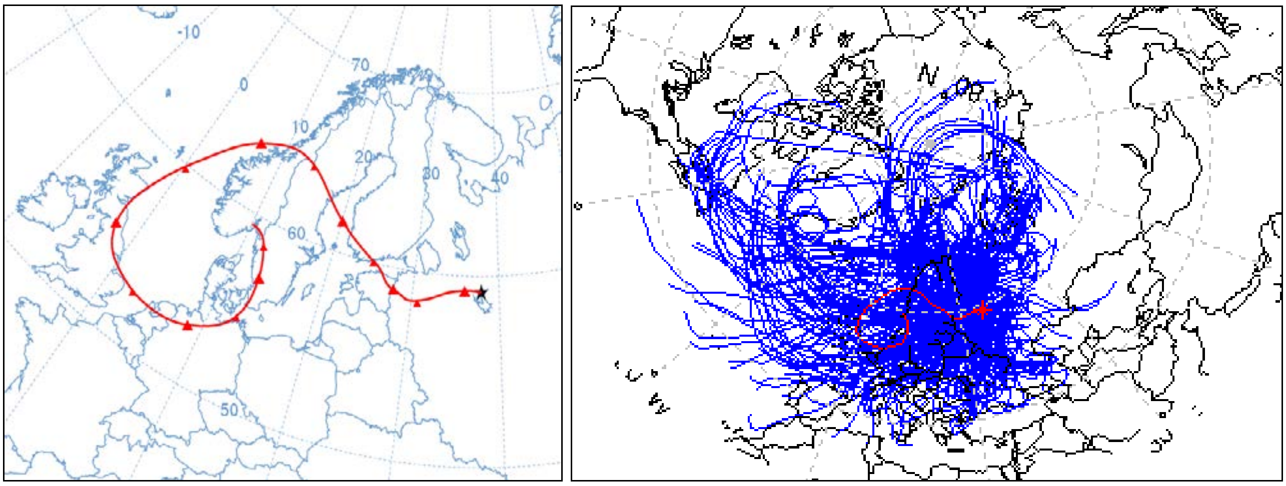
2.2.4 Calculation of backward trajectories

All backward trajectories were simulated in accordance with a sampling time corresponding to the measurements performed. The sampling time of 12 cases[1] for krypton was precisely specified. The sampling time for ^{85}Kr 33 other cases and for all 62 xenon cases was assumed to be equal to 36 hours on the average. Trajectories were calculated with a 6 hour interval for Kr and 12 hour interval for Xe for each separate sampling time interval. The duration of trajectories arrived at Cherepovets (measurement site is located at 59.13˚ N; 37.93˚ E) was chosen to be 168 hours (i.e. 7 days). Note, that trajectories of more than 5 days of duration might experience problems in accuracy of calculation, and hence, are less reliable. Although for specific case studies, especially, related to identification of the far remote source regions the trajectories of a longer duration can be a valuable choice (*Mahura & Baklanov, 2003*).
Calculations were performed for two levels in height. The first level of 100 meters above ground level (agl) was chosen as attributed to the surface layer of the atmosphere, and the second level at 500 meters agl – as a layer contained within the boundary layer of the atmosphere. In total 622 and 473 backward trajectories were simulated for ^{85}Kr and ^{133}Xe episodes, correspondingly. It should be noted that calculations are performed at the Universal Coordinated Time (UTC) and hence, times of measurements were converted into UTC terms. For example, measurements in Cherepovets were made at 10 a.m. of the local time which is equivalent to 07 UTC. In addition, for each trajectory at 1 hour interval an extra dataset was also recorded. It contained (backward in time) information

[1] *Under specific cases we will consider all chosen episodes with elevated or lowered krypton concentrations*

about spatial coordinates (latitude, longitude and altitude) of moving air parcel as well as values of pressure, air temperature and mixing height at each position. In general, calculation of the main atmospheric variables and parameters in the grids of the latitude vs. longitude domain appears to be rather useful approach.

Along with individual (Figure 2.2a) backward trajectories obtained the ensembles (Figure 2.2b) of trajectories were also simulated. By combining different episodes and visualizing them in the way of ensembles additional information about the general directions of contaminated air transport, spatial coverage of associated airflow patterns, distant regions from which air parcels can travel until reaching the measurement site, as well as temporal variation peculiarities can be obtained.



(a) *(b)*

***Figure 2.2**: Krypton case: (a) trajectory arrived at 100 m agl at the Cherepovets site on 20 December 2007 at 03 UTC, and (b) all 622 trajectories arrived at the Cherepovets site at 100 m agl during 27 Sep 2006 – 28 Jul 2008.*

The simulated individual backward trajectories can be used as a preliminary indicators of 1) geographical source regions with its approximate boundaries, 2) possible noble gas sources of emissions, 3) spatial extension characterizing an average speed of travelling air parcels and its' temporal variability, as well as 4) time spent by trajectories within the boundary layer vs. free troposphere.

2.3. Cluster Analysis of Trajectories

2.3.1 General approach

Although trajectory modelling approach is used successfully to study different problems connected with atmospheric transport processes: radioactive (*Stohl et al., 2002*) or anthropogenic pollutants studies (*Mahura et al., 2003*) for example, it still has a degree of uncertainty. In order to derive more representative single trajectories as well as trajectory ensembles and evaluate additional probability information a cluster analysis technique can be applied.

Cluster analysis, also called segmentation analysis or systematic analysis, is a way to create groups of objects, or clusters, in such a way that the profiles of objects in the same cluster are very similar and the profiles of objects in different clusters are quite distinct (*Wilks, 1995*).

Statistical method considered has found its application for different scientific fields including atmospheric sciences (*Saucy et al, 1987; Baker, 2009*). As applied to trajectory modelling the cluster analysis technique can be used for getting qualitative as well as quantitative estimation of tracers transport, identification of dominant atmospheric flows for the sites of interest, source regions of air pollutants, etc.

2.3.2 Realization in MATLAB

In this study the cluster analysis was performed by making use of MATLAB (MATrix LABoratory) (*http://www.mathworks.com*) programming language. The m-program has been written, tested and applied to calculate a set of trajectories. Among different available cluster calculation methods the Euclidian distance technique (*Romesburg C.H., 1984*) was selected. In clustering the pairs of latitude and longitude values of trajectories at each time step were chosen as criteria. The time step was equal to trajectory calculation time interval i.e. 6 (for Kr) or 12 (for Xe) hours. Hence, the Euclidean distances (*DE*) can be calculated by means of the formula:

$$DE(X,Y) = \sqrt{\sum_{t=1}^{NT} kw_t (X_t - Y_t)^2}, \quad (3)$$

where:

X , Y – longitude and latitude of trajectories at moment t (t = 0, NT),

NT – number of considered time intervals for trajectories (or number of pairs of values of latitude and longitude),

kw_t – weight coefficient (selected in order to regulate the number of clusters, in the first approximation kw_t =1).

At the first step the output, '*A*', was obtained as a vector of length, containing the distance information. The distances were arranged in the order *(1,2), (1,3), ..., (1,m), (2,3), ..., (2,m), ..., ..., (m-1,m).* '*A*' is also commonly known as a similarity or dissimilarity matrix. '*A*' is formatted as a vector in order to save space and computation time.

At the second step a hierarchical cluster tree was derived by using the matrix '*A*' as the input. It was performed by making use of a '*centroid*' method that identifies way to create the cluster hierarchy. The output, '*Z*', is an $(m-1)\times 3$ matrix containing cluster tree information.

Finally, clusters were constructed from the hierarchical tree and plotted in a form of histograms. The input values were matrix '*Z*' and threshold '*c*' for cutting '*Z*' into clusters. The output '*T*' was a vector of size '*m*' that contained the cluster number for each observation in the original data. Several runs were also realized by varying a maximum number of clusters (or mean trajectories corresponding to atmospheric transport pathways) between 5 and 9 in order to select the best number of clusters which depended on synoptical patterns in the geographical region as well as number of latitude/ longitude pairs within each cluster.

2.3.3 Visualization of clustering results

In order to visualize results of the cluster analysis and estimate mean atmospheric flow of backward trajectories arriving at the measurement site the programming on MATLAB was done. For that points corresponding to pairs of averaged latitude and longitude values for each cluster at each time step were derived and classified. Finally mean backward trajectories were calculated and plotted in accordance with the classification organized and criteria set. For plotting the built-in MATLAB program '*plotm*' was used that displays projected line objects (latitude and longitude coordinates) within the current map axes.

The averaged trajectories obtained from the cluster analysis allowed to estimate the prevailing directions of RNG atmospheric transport, in order to assess geographical source regions that could be the reasons for episodes with elevated as well as lowered values of volumetric activity of RNGs, to evaluate airflow climatology for the area of interest, and to give a quantitative estimation of pollution transport for each case studied.

3. Results and Discussions

3.1 Analysis of Times Series of Monitoring Results

3.1.1 Time-series of krypton measurements

In time series of ^{85}Kr measurements (see Figure 3.1) covering a two-year period all episodes with elevated and lowered concentrations above/ below mean values were identified. The mean value for ^{85}Kr was found to be equal to 1,54 Bq/m^3. The specific cases in total have accounted 28 episodes for elevated (i.e. more than 1,50 Bq/m^3) and 17 episodes for lowered (i.e. less than 1,50 Bq/m^3) concentrations, with the highest 1.79±0.27 Bq/m^3 during 25 - 27 May 2007 and the lowest 1,31±0,20 Bq/m^3 during 25 - 27 November 2006. One-two measurements were carried out every month. The monthly mean square deviation has been varying from 3% to 11%, with a mean – 6%. Measure of variability amounted 8% and the median value (1,54 Bq/m^3) was almost the same as the mean one. The minimum value of 1,31 ± 0,20 Bq/m^3 was considered as the minimum background value of krypton for the region. The maximum value exceeded this value in 1,4 times. According to *Dubasov (2009),* the results of observations carried out in Cherepovets showed that the volumetric activity of ^{85}Kr in the North-Western region of Russia for the last 15 years has increased by almost 50% and has reached to 1,55 Bq/m^3.
As seen in the Figure 3.1 the time-series of ^{85}Kr concentration can be divided into periods with elevated or lowered values observed. The analysis of ensembles of backward trajectories will be implemented for the most representative periods i.e. *'A'* (27 Sep – 27 Nov 2006) and *'B'* (30 Nov 2006 – 9 Feb 2007) as well as two maxima (1.79±0.27 Bq/m^3 and 1.79±0.18 Bq/m^3) of the gas concentration in order to define if some temporal correlations exist and identify sources (nuclear plants) responsible for these elevated values.

3.1.2 Time-series of xenon measurements

The two-year (Aug 2006 – Jul 2008) time series of ^{133}Xe measurements comprised of all episodes with concentration values above (elevated) and below (lowered) mean value are represented in the Figure 3.2. The xenon mean value was found to be equal to 0,79 ± 0,46 Bq/m^3. In total 26 elevated and 36 lowered cases were selected and analyzed. Periods of elevated/ lowered concentrations cannot be clearly distinguished for Xe compared with Kr. However, two maxima of 2,47 Bq/m^3 during 25 - 27 September 2006 and 2,00 Bq/m^3 during 14-16 January 2008, and one minimum of 0,09 Bq/m^3 during 11 - 13 February 2007 were identified in the time-series.
The median was found to be equal to 0,71 Bq/m^3. The minimum concentration of 0.09 Bq/m^3 was considered as the background value for the region (see Figure 3.2). Thus, it can be noticed that the highest concentration of 2,47 Bq/m^3 exceeded this value more than 27 times.
As krypton and xenon enter the atmosphere easily because of their chemical inertness, the monitoring of their levels is currently a high priority for the environmental radiation control. Thus, further estimation should take into account peculiarities of atmospheric transport of these sampled gases. It appears to be important to know how these inert gases behave in the atmosphere after being emitted and what happens with them under influence of complex atmospheric processes as well as weather conditions during the whole period of its travelling and residence in the atmosphere. To answer these questions 1) individual backward trajectories associated with elevated/ lowered concentration episodes and 2) trajectory ensembles were analyzed applying cluster analysis.

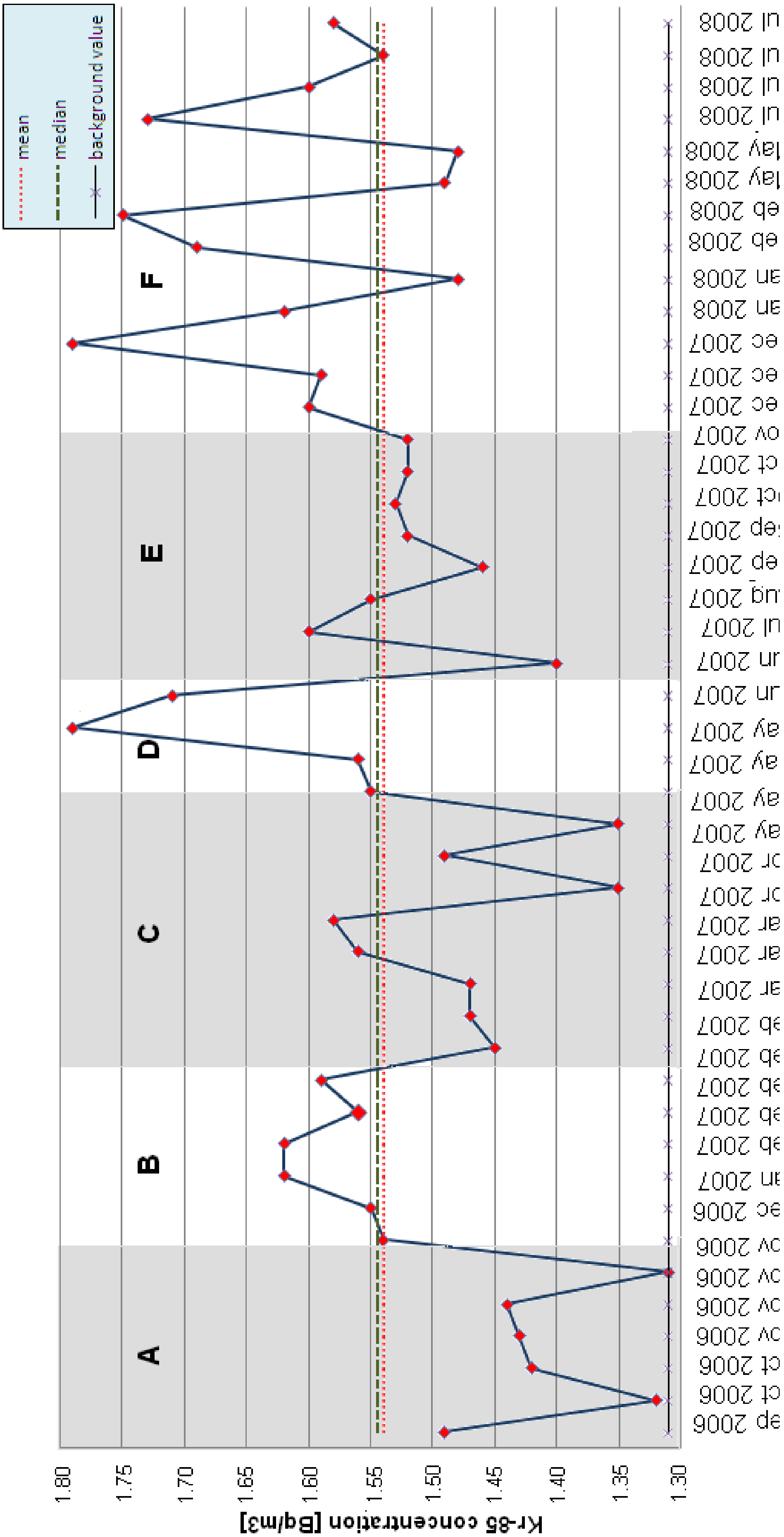


***Figure 3.1**: Time series of ^{85}Kr concentration measured at the Cherepovets measurement site during Sep 2006 – Jul 2008*

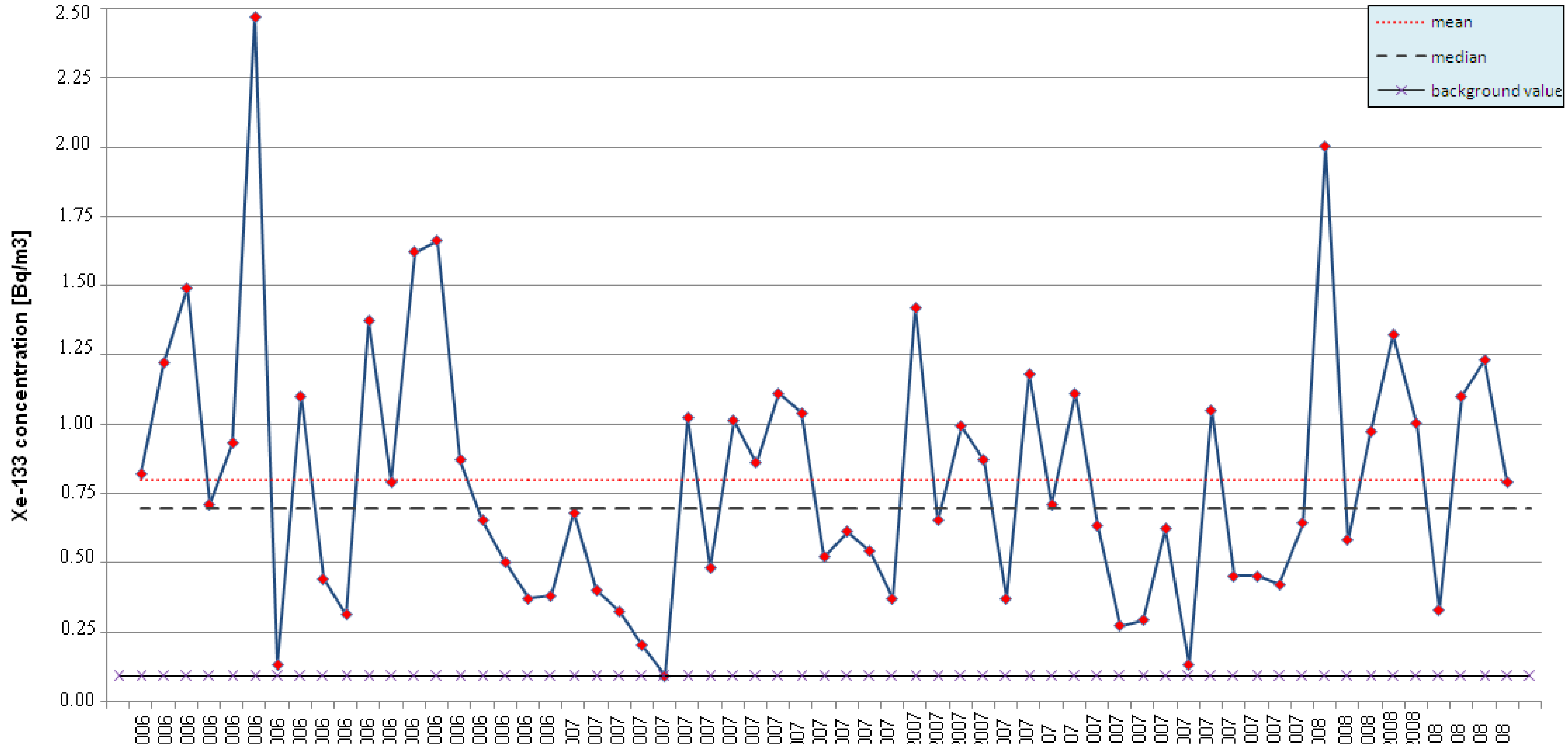


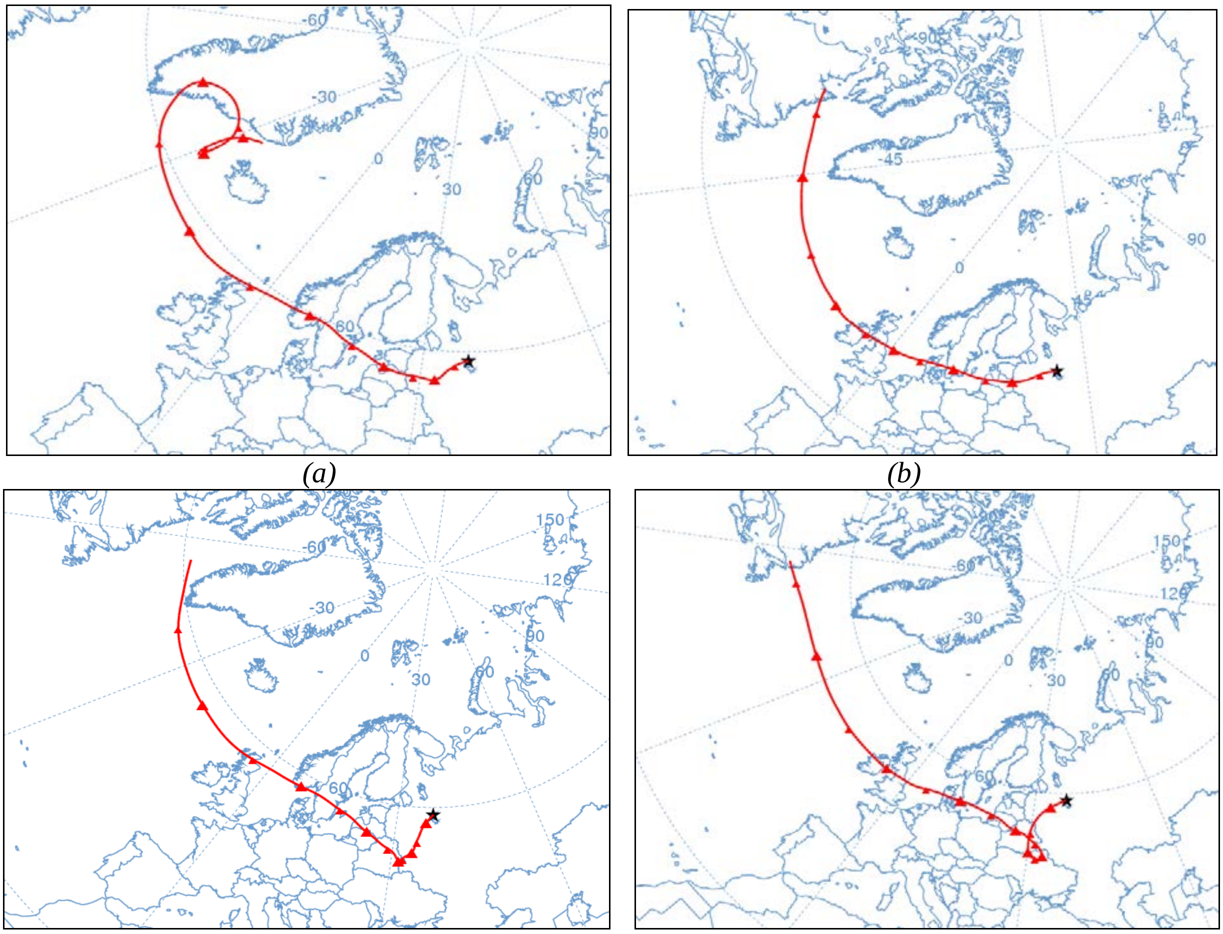
Figure 3.2: *Time series of* ^{133}*Xe concentration measured at the Cherepovets measuring site during Aug 2006 – Jul 2008.*

3.2 Analysis of Krypton Specific Episodes with Individual Trajectories

For ^{85}Kr, one of the main peculiarities and concerns at the same time is its ability to be accumulated in the ambient air without being deposited on the surface. This gas due to its long period of half-life (10.76 years) can be distributed to large distances and even took part in circulation process in the Earth's atmosphere. According to studies performed total equalizing of neutral admixture concentration occurs in two years after its emission into the atmosphere (*Krypton-85 in the atmosphere, 1975*). Thus, the process of ^{85}Kr tracking becomes more complicated due to large-scale circulation of already accumulated krypton in the atmosphere.
The simplest way to identify potential source regions of sampled noble gases for the specific dates is to calculate individual backward trajectories arriving at the site of measurements (receptor point). In total almost 1100 trajectories for both gases were analyzed. In the next sections (§3.2.1 and §3.2.2) two most representative cases with extreme (highest and lowest) values of ^{85}Kr volumetric activities measured at Cherepovets will be evaluated.

3.2.1 Elevated concentration episode: May 2007

The first case is associated with the highest (1.79 ± 0.27 Bq/m^3) concentration of ^{85}Kr among all measured. The sampling time lasted for 36 hours. For this case the 7 - day backward trajectories were calculated with 6 hour interval for almost three days of sampling period starting from 25 May 2007 at 19 UTC to 27 May 2007 at 07 UTC (Figure 3.3). The simulation was carried out at two levels: 100 m and 500 m agl.



(a) *(b)*

(c) *(d)*

***Figure 3.3**: Backward trajectories arrived at the Cherepovets site on 25 May 2007, 19 UTC at (a) 100 m agl and (b) 500 m agl; and on 27 May 2007, 07 UTC at (c) 100 m agl and (d) 500 m agl.*

In total 14 backward trajectories were obtained for this episode. The general atmospheric transport for these days had a very similar pattern and hence, only first and last days are shown in Figure 3.3, as examples.
As seen in Figure 3.3, the general transport of air parcels was carried out from the western sector. The well-distinguished cyclonic influence with its strong upwelling flows over the south-eastern part of Greenland was observed during the first two days of transport, i.e. on 19 – 20 May (Figure 3.3a). The air parcels passed over the Atlantic Ocean at a relatively high speed and after crossing the British Islands the speed gradually decreased. The noticeable slowing-down and clockwise vorticity of the flow was observed over territories of Belorussia during the last days, i.e. on 24 - 27 May (Figure 3.3cd). The descending motion occurred from the free troposphere and took place almost during the whole atmospheric transport with an exception of strong upwelling flows due to the cyclonic influence. The air parcels were travelling above the boundary layer for almost half of its way. Because almost all of the airborne pollutants emitted into the ambient atmosphere are transported and dispersed within the mixing layer the impact of sources will be more significant within this layer.
The ^{85}Kr is emitted during the nuclear fuel reprocessing directly into the lower layers of the troposphere and being mixed it travels in accordance with the general atmospheric flows as well as under influence of advection and dynamic factors determining cyclonic and anticyclonic activity.
By comparing the trajectory modelling results with the geographical location of the nuclear power plants on the transport pathways the most probable sources of krypton emissions were identified. Taking into account that the total number of backward trajectories calculated for this specific case was equal to 14, in the 7 cases the trajectories were travelling not far from the Swedish (Oskarshamn, Ringhals and Barsebaeck) NPPs and Lithuanian (Ignalina) NPP. Five times trajectories passed nearby the Smolensk nuclear plant (Russia). The Kalinin NPP was situated on a way of 13 trajectories. As the result backward trajectories, arrived at the Cherepovets site and resulted in elevated volumetric activity, travelled in a vicinity of 6 mentioned nuclear power plants.

3.2.2 Lowered concentration episode: November 2006

Trajectories for the second case of the extreme (lowest, 1.31 ± 0.20 Bq/m^3) concentration of ^{85}Kr measured at the Cherepovets site are shown in the Figure 3.4. The simulation was carried out starting from the 25 Nov 2006, 19 UTC till 27 Nov 2006, 07 UTC. The main peculiarity of this case is a significant divergence of the flow at two heights. The trajectories arrived at the level of 100 m showed the southern – south-eastern transport (Figure 3.4ac) while those at 500 m arrived from the west - south-western sector (Figure 3.4bd). This can be related to a strong horizontal inhomogeneity due to a large horizontal temperature gradient presence in latitudinal direction that causes the additional rotation of the wind. The vertical profiles of trajectories for the 100 m did not exceed the level height and did not experience a significant vertical motion transport. The air parcels arrived at 500 m (as opposite to the 100 m level) experienced upwelling and descending motions but the general transport occurred within the mixing layer.
The last day (27 Nov 2006, 07 UTC) (Figure 3.4d) represents the south-western direction of air particles transport at 500 meters and the south-eastern direction gradually becoming the southern at 100 meters (Figure 3.4c). For trajectories arrived at 500 m agl the speed of atmospheric transport on the whole was rather slow although at the beginning it was higher over the Atlantic Ocean (Figure 3.4b).
Based on the analyses of all 14 trajectories the southern transport occurred in a vicinity of the following nuclear plants: Kalinin, Kursk, Smolensk, and Novovoronezh. The trajectories arrived from the south-western - western sector travelled not far from the Swedish (Barsebaeck), Lithuanian (Ignalina) and a number of German and French nuclear plants.
As seen, although the atmospheric transport pathways brining the lower value of ^{85}Kr can be explained by the meteorological situation the number of NPPs on the paths of parcels in the lowest

concentration cases is comparable with the highest ones. The general analysis of all calculated trajectories showed that different meteorological situations as well as arrival directions had some common distinctive features corresponding to the case (highest/lowest) but nevertheless, the number of complex cases was also rather high.

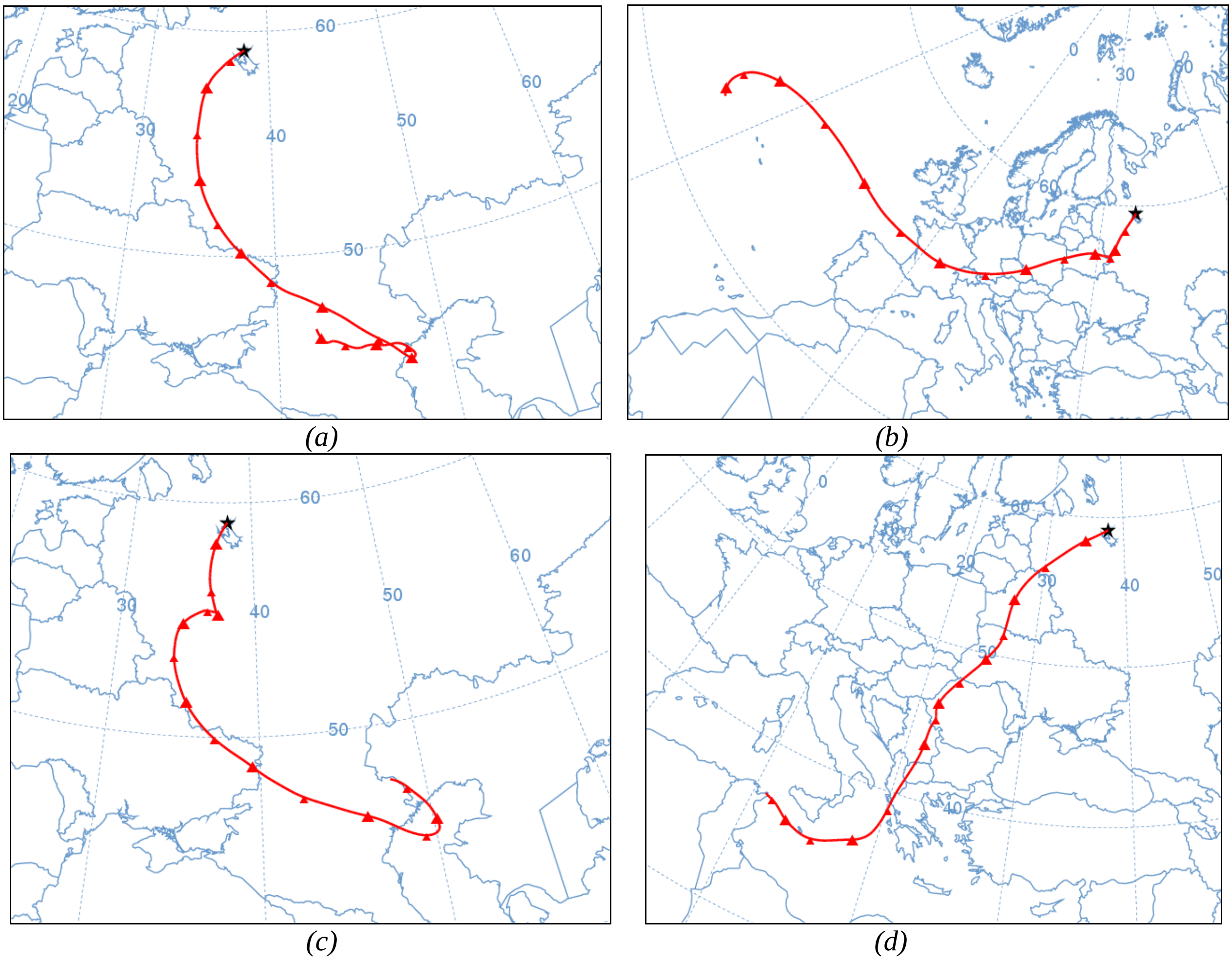


***Figure 3.4**: Backward trajectories arrived at the Cherepovets site on 25 Nov 2006, 19 UTC at (a) 100 m agl and (b) 500 m agl; and on 27 Nov 2006, 07 UTC at (c) 100 m agl. and (d) 500 m agl.*

In order to better understand the krypton atmospheric transport variations linked with concentration values (high/low) and obtain more representative averaged trajectories to derive dominating arrival directions the trajectory ensembles were derived as well as cluster analysis technique was applied.

3.3 Analysis of Xenon Specific Episodes with Individual Trajectories

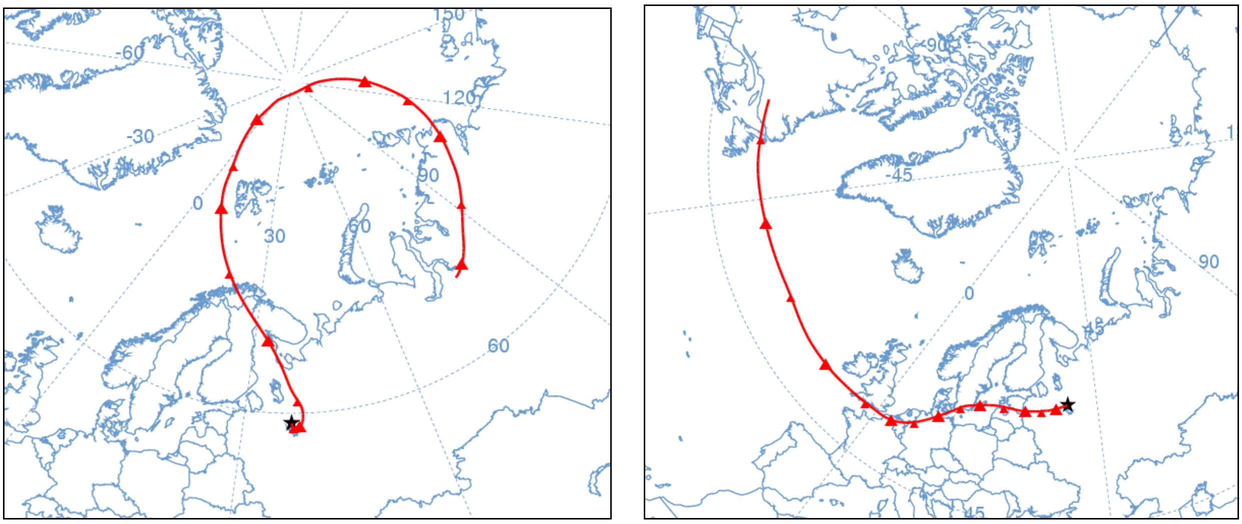
The radionuclide of ^{133}Xe with the half–life time of 5.2 days in opposite to krypton is not accumulated in the atmosphere and is associated with emissions and accidental releases from nuclear plants and pharmaceutical factories.
The analysis of all trajectories calculated for xenon showed that the cases related to elevated concentrations were connected with the northern and north-western atmospheric transport and associated mostly with trajectories passing near by the Finnish, Swedish NPPs as well as northern Russian Kola NPP by trajectories. The southern and south-western transport is also responsible for the elevated episodes and is associated with passing near the southern Russian NPPs, Lithuanian (Ignalina) as well as Ukrainian and German NPPs. The trajectories corresponded to lowered concentration episodes in majority cases had been associated with the northern and north-eastern directions. Note that some of these trajectories were travelling in a vicinity of NPPs when none of

RNG emissions occurred.

The most representative cases of trajectories associated with the highest and lowest concentrations of ^{133}Xe are described in the sections 3.3.1 and 3.3.2.

3.3.1 Elevated concentration episode: September 2006

The maximum of ^{133}Xe concentration (2,47 ± 0,26 Bq/m^3) was recorded during 25 - 27 Sep 2006 episode. In accordance with the 36 hour sampling time 8 backward trajectories in total for two levels in height were calculated for three day period starting from the 25 Sep at 9 UTC. The calculation interval was chosen to be 12 hours. In the beginning of sampling the atmospheric transport was by westerlies. The trajectories originated over the eastern coast of Canada, and after passing the Atlantic Ocean travelled over territories of the United Kingdom, Belgium, Germany, the Baltic Sea, and Estonia before reaching the measurement site. The air motion was descending during the whole transport time and four times slower over the land than over the water surface. The trajectory path was in a vicinity of the Swedish (Barsebaeck, Oskarshamn and Ringhals) as well as German and British NPPs.



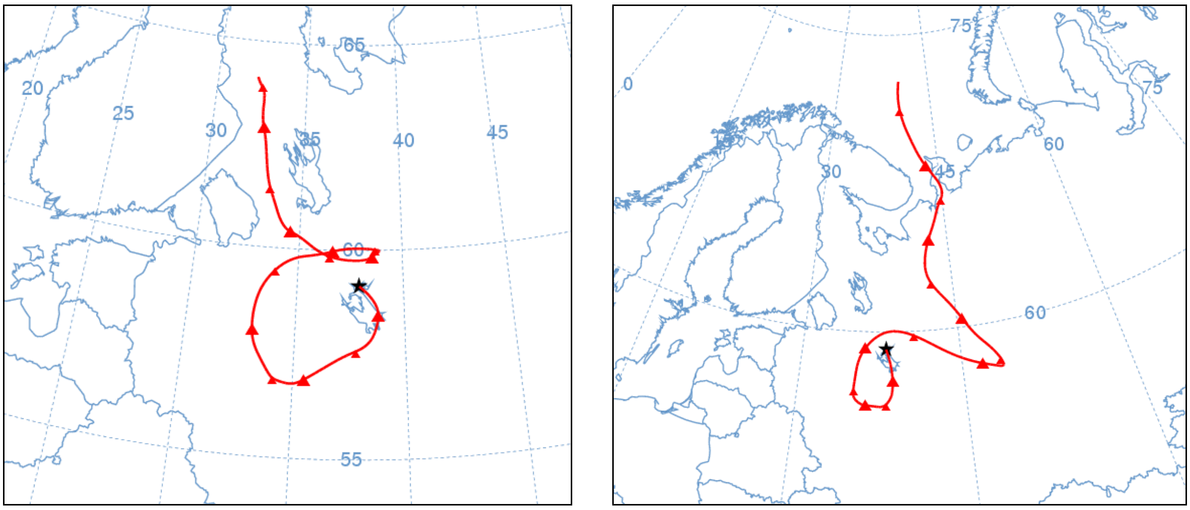
(a) (b)

***Figure 3.5**: Backward trajectories arrived at the Cherepovets measurement site on (a) 26 September 2006, 19 UTC at 100 m agl and (b) 24 September 2006, 07 UTC at 100 m agl.*

As shown in the Figure 3.5 starting from the 25 Sep at 19 UTC the atmospheric transport direction over Cherepovets region has changed to northern. The atmospheric transport of the first 4-5 days of transport was determined by the cyclone, originated over Taymyr Peninsula and passed through the Arctic regions. The Kola NPP was on the way of the three northern trajectories.

3.3.2 Lowered concentration episode: February 2007

The minimum ^{133}Xe concentration value of 0,09 ± 0,03 Bq/m^3 was registered from sample taken during 11 - 13 Feb 2007. In total 8 backward trajectories were calculated starting from the 11th Feb at 19 UTC with the interval of 12 hours. The general arrival direction for all trajectories was northern with well distinguished cyclonic circulation during the last four days before arrival at the site (Figure 3.6). The atmospheric transport took place over the North-Western Russian territories and the Barents Sea, and its speed was significantly higher at 500 m altitude especially on 10th and 11th Feb. For 10 trajectories there were no NPPs on the way of air parcels arrived at the measurement site. Nevertheless, 4 trajectories passed in the vicinity of Kalinin NPP. Note that a moment of passing near by the NPP is not necessarily connected with occurrence of noble gases emissions.



(a) (b)

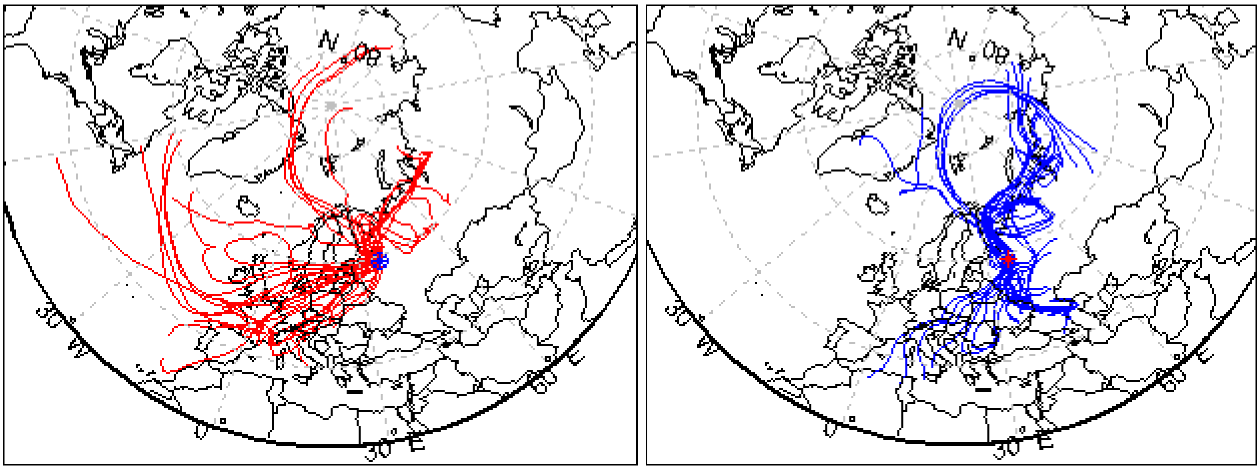
***Figure 3.6**: Backward trajectories arrived at the Cherepovets measurement site on 13 Feb 2007, 07 UTC at (a) 100 m agl and (b) 500 m agl.*

3.4 Analysis of Trajectory Ensembles

Additional information about potential source regions as well as emission sources, and estimation of prevailing directions of both ^{85}Kr and ^{133}Xe atmospheric transport can be realized by analyzing ensembles of backward trajectories. Depending on the method applied the peculiarities of seasonal and temporal variations of the general atmospheric flow as well as variations of height, etc. can be studied.

3.4.1 Trajectory ensembles for krypton episodes

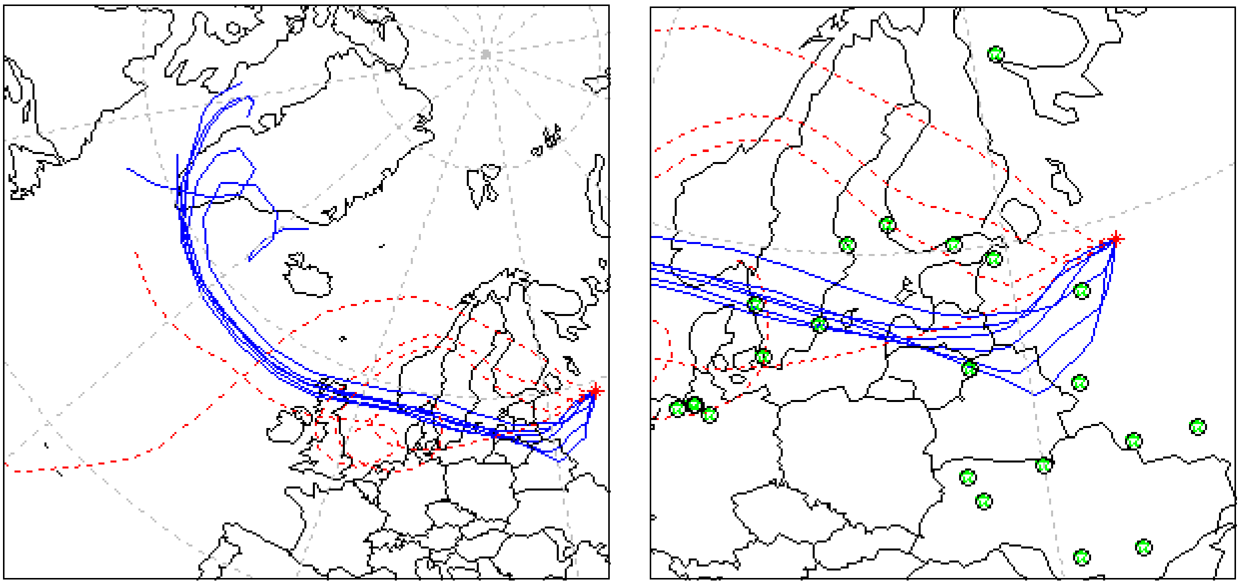
The time-series of ^{85}Kr measurements had been divided into several periods as was shown in the Figure 3.1. All trajectories corresponded to these periods were plotted and analyzed. The Figure 3.7 represents all trajectories corresponded to the periods *'A'* and *'B'* (see Figure 3.1) and arrived at the measurement site during 30 Nov 2006 – 9 Feb 2007 ('*B'* with elevated values) and of 27 Sep 2006 – 27 Nov 2006 ('*A'* with lowered values).
The majority of trajectories associated with the elevated concentrations travelled to the measurement site through the European countries with a "large" density of NPPs as well as over Nordic countries (Figure 3.7a).
The following nuclear plants are situated in these regions and could be related to the elevated concentrations within the period *'B'*: Russian (Kola, Kalinin, Leningrad, Smolensk), Swedish (Forsmark, Oskarshamn, Barsebaeck, Ringhals) and Finnish (Olkiluoto, Loviisa), Lithuanian (Ignalina) as well as German, French and British NPPs.
On the contrary, many trajectories of episode *'A'* (Figure 3.7b) passed over the northern territories where there are no NPPs. Some trajectories arrived from the southern sector. The following plants were situated on the way of these trajectories: Russian (Kola, Kalinin, Smolensk, Kursk, Novovoronezh), as well as Ukrainian (Chernobyl, Rovno, and Khmelnitskiy) NPPs.
The ensemble plots allow estimating general features of transport for the episodes and allow identifying regions from where the trajectories have arrived, but the emission sources itself cannot be clearly identified.



(a) (b)

Figure 3.7: *All backward trajectories arrived at the Cherepovets site (colour star) at the height of 100 m agl during the sampling period of (a) 30 Nov 2006 – 9 Feb 2007 with elevated concentration (period 'B') and (b) 27 Sep 2006 – 27 Nov 2006 with lowered concentration (period 'A').*

Let us consider atmospheric transport of air parcels associated with two maxima of ^{85}Kr concentration which were recorded during 25-27 May 2007 and during 19-20 Dec 2007 (Figure 3.8). The Figure 3.8a represents general view of 10 backward trajectories that brought elevated concentration values of krypton at the Cherepovets site. The blue trajectories originated over the Baffin Bay and territory of Greenland, passed the Atlantic Ocean having a cyclonic vorticity, and travelled over territories of UK, southern parts of Norway and Sweden, as well as the Baltic States, and Belorussia. The red dashed trajectories passed over the territories of the following countries: UK, Germany, Scandinavian countries, Baltic States, and Belorussia. Hence, the possible source territories can be those of Sweden, Finland, Lithuania, Germany and North - West of Russia.
On the next step the air parcel paths were compared with NPPs locations (Figure 3.8b). As seen the trajectories passed not far from the Swedish (Oskarshamn), Finnish (Olkiluoto) and Lithuanian (Ignalina) nuclear plants and travelled near Russian (Kalinin), Swedish (Ringhals and Barsebaek), and German NPPs.



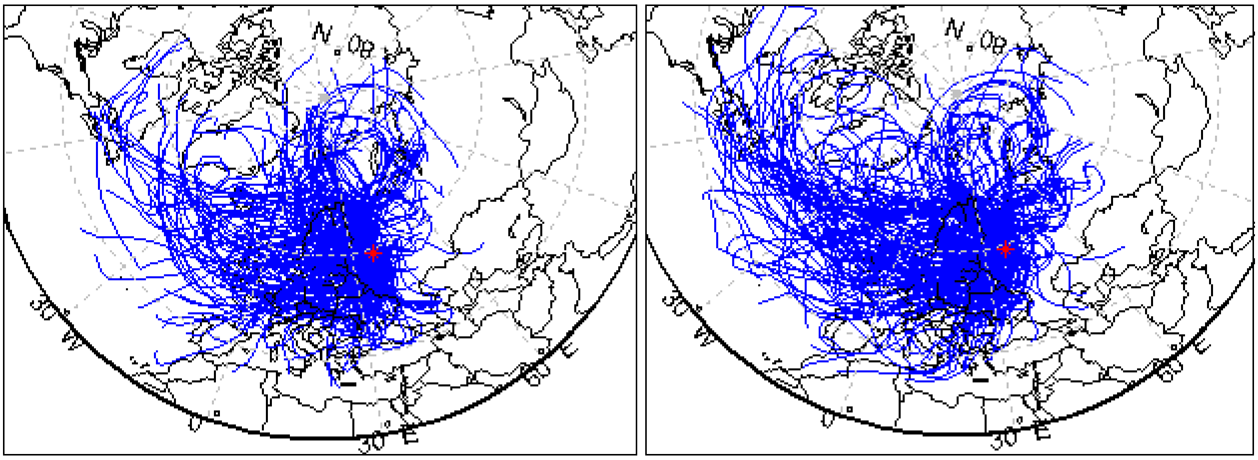
(a) (b)

Figure 3.8*: Trajectories arrived at the Cherepovets measurement site during the sampling intervals of 25-27 May 2007 and 19-20 Dec 2007 which are corresponded to two maxima of* ^{85}*Kr concentration.*

Finally, possible sources responsible for the elevated concentrations of ^{85}Kr recorded at the Cherepovets site were identified. This information can be used for further analysis of RNGs yield from the NPPs as well as identification of exact sources by analyzing the gases ratios.
Tracking of air parcels paths for elevated episodes of ^{85}Kr concentrations is rather complex task due to circulation of accumulated gas in the air. Krypton under mesoscale meteorological winds circulation can pass one site twice making the concentration to be increased (*Cheol-Hee Kim, 2008*). Hence, the more detailed results can be obtained by applying similar analysis to xenon measurements.

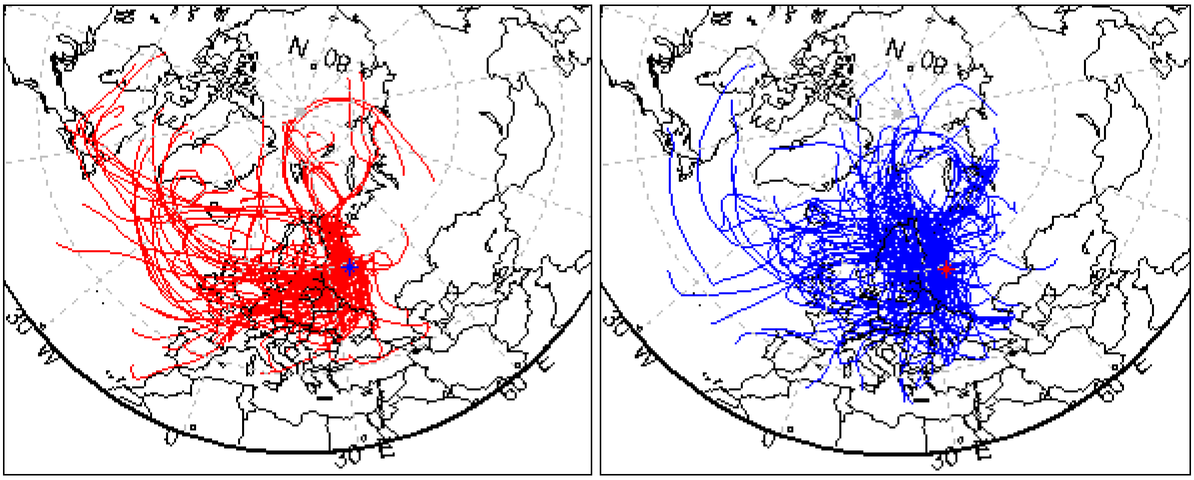
3.4.2 Trajectory ensembles for xenon episodes

In the case of xenon there are no distinguishable long-term time periods with elevated or lowered values only. However, it is important to get a general knowledge of the total territory covered by all trajectories arrived at the Cherepovets site. All (473) 7-day backward trajectories arrived at the site during 2 Sep 2006 – 28 Jul 2008 have been plotted for two levels of 100 and 500 m agl. (Figure 3.9). Note that both altitudes represent transport within the atmospheric boundary layer of the atmosphere. As seen, the spatial coverage by trajectories and consequently passage over potential source regions at 500 m level is larger than that at 100 m. It can be related to weaker winds at lower altitudes and more uniform temperature distribution.
In addition trajectories corresponded to elevated and lowered ^{133}Xe concentrations were plotted separately (Figure 3.10). As seen for the case with elevated values (Figure 3.10a) the more dense area (i.e. more trajectories passing one place) on the plot is situated in the south-western sector from the measurement site. In opposite to the above mentioned case an area with larger density of trajectories corresponded to the case with lowered concentrations (Figure 3.10b) is situated in the northern sector from the site. In this sector there are no so many NPPs compared with the south-western.



(a) (b)

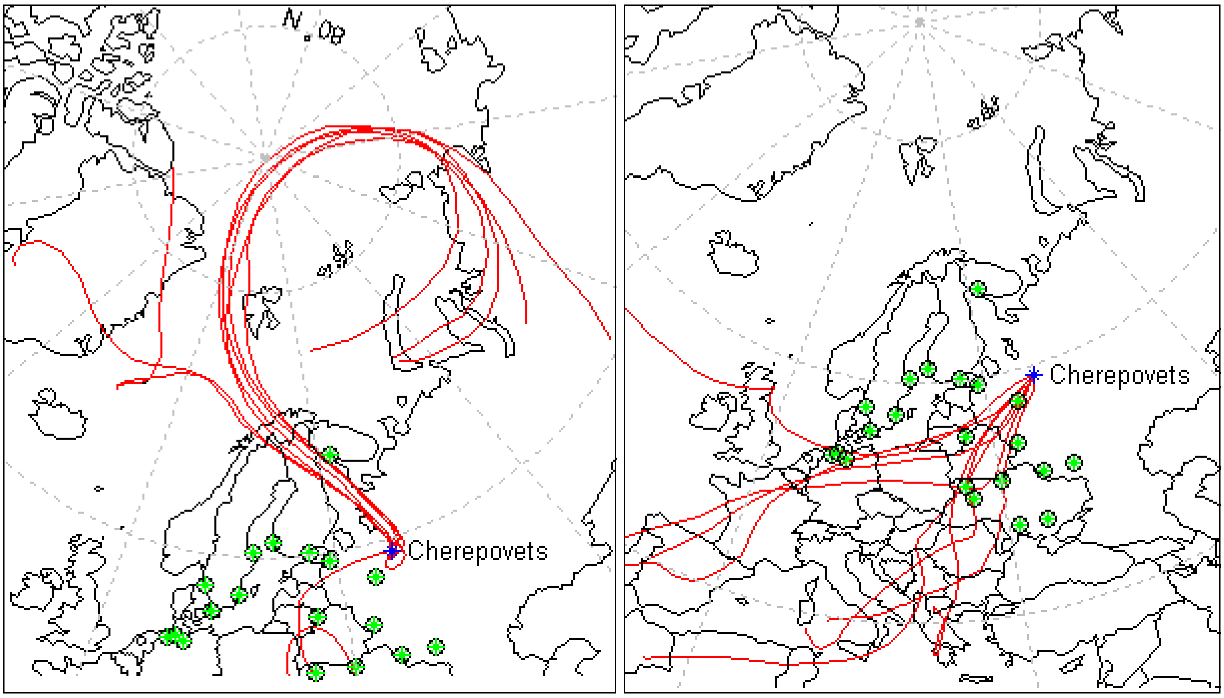
***Figure 3.9**: All backward trajectories arrived at the Cherepovets site (red point) during the sampling period of 2 Sep 2006 – 28 Jul 2008 at the heights of (a) 100 and (b) 500 m agl.*



(a) (b)

***Figure 3.10**: Backward trajectories corresponded to all (a) elevated and (b) lowered ^{133}Xe concentrations recorded at the Cherepovets measurement site during 2 Sep 2006 – 28 Jul 2008.*

At the next step in order to identify potential sources associated with elevated concentrations for xenon backward trajectories for two cases with maximum values were plotted and analyzed. The Figure 3.11 shows two sets of trajectories corresponded to two sampling periods of 25-27 Sep 2006 and 14-16 Jan 2008 when maximum concentration of ^{133}Xe was registered at the measurement site.



(a) (b)

***Figure 3.11**: Backward trajectories for 100 and 500 m agl corresponded to two maxima of ^{133}Xe concentration recorded on (a) 25-27 Sep 2006 and (b) 14-16 Jan 2008.*

As seen the general atmospheric flows were carried out from the Arctic region having a cyclonic circulation for the highest case (2.47 Bq/m^3) among the maxima and from the European territory for another case (2.00 Bq/m^3). Trajectories brought the highest concentration at the site (Figure 3.11a) passed not far from the Russian Kola nuclear power plant. Trajectories corresponded to the second concentration maximum (Figure 3.11b) travelled near Russian (Kalinin and Smolensk) NPPs, Lithuanian (Ignalina), Ukrainian (Rovno, Chernobyl and Khmelnitskiy) and German (Brokdorf and Brunsbuettel) NPPs.

Analysis of ensembles of backward trajectories allows to identify potential sources of RNG emissions and to estimate atmospheric transport for particular cases. To get additional probability information for arrival directions of air parcels for multiple cases as well as to obtain mean backward trajectories (or atmospheric transport pathways) the cluster analysis technique was applied.

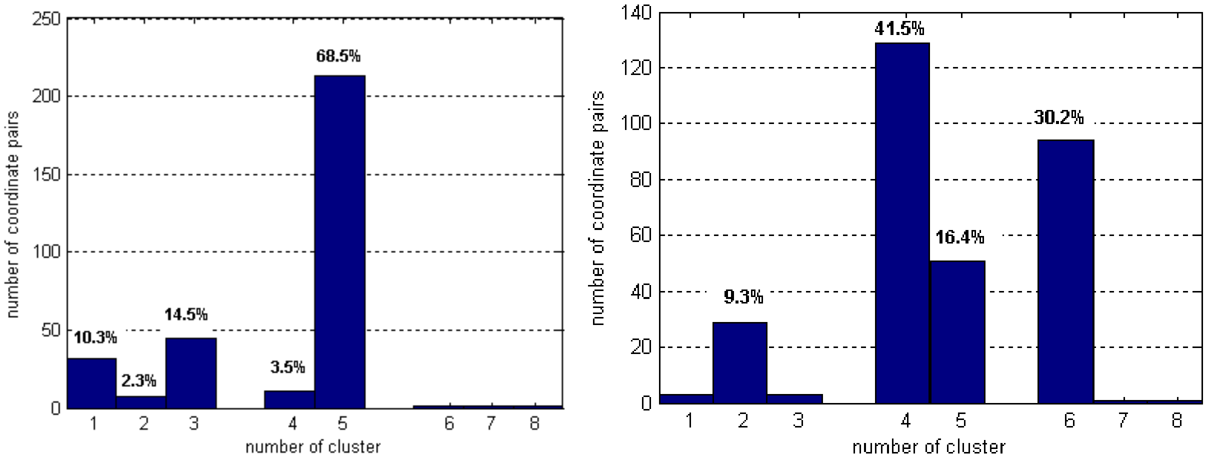
3.5 Cluster Analysis and Atmospheric Transport Pathways

3.5.1. Analysis of cluster statistics for krypton

At the first step the distribution of the cluster number versus the number of latitude/ longitude pairs (or 6 hour trajectories) was plotted as histograms. The analysis was carried out separately for two heights of 100 and 500 m agl. The numbers of clusters (i.e. further the number of mean trajectories corresponding to dominating atmospheric transport pathways) varied from 5 to 9; and the clusters numbers (identificators) are arbitrary. As an example, the number and percentage of trajectories evaluated for both heights at the first 6 hour interval is represented in the Table 3.1. The Figure 3.12 shows a case when all latitude/longitude pairs corresponded to the first time interval of 6 h (backward in time) were clustered.

Table 3.1*: Percentage and number of latitude/ longitude pairs per cluster for the first time interval of 6 hours (the levels of 100 / 500 m).*

^{85}Kr	**Percentage value [%] / number of coordinate pairs**							
# cluster	***1***	***2***	***3***	***4***	***5***	***6***	***7***	***8***
100 m	10.3 / 32	2.3 / 7	14.5 / 45	3.5 / 11	68.5 / 213	0.3 / 1	0.3 / 1	0.3 / 1
500 m	1.0 / 3	9.3 / 29	1.0/ 3	41.5 / 129	16.4 / 51	30.2 / 94	0.3 / 1	0.3 / 1



(a) *(b)*

Figure 3.12*: Number of latitude/ longitude pairs per cluster obtained for the first time interval of 6 h corresponding to the height of (a) 100 and (b) 500 m agl.*

By setting originally the number of clusters to be equal to 8 three most representative clusters were obtained for 100 m level (Figure 3.12a). These are clusters #1, #3, and #5 with percentage values of 10.3%, 14.5%, and 68.5%, correspondingly. In total, number of trajectories belonging to representative clusters accounted 93.3 % or in more than 93 % of the cases it was possible to assign

trajectory to a dominant transport pathway. The clusters having the percentage value of less than a few percent were linked with so-called "outliers" and non-representative clusters, and hence, these were not considered further as representing insignificant transport pattern.
As seen (from the Figure 3.12a and the Table 3.1) 68.5% (or 213 trajectories) of cluster #5 correspond to dominating arrival direction which was found to be western. The northern direction accounted for 14.5% (45 trajectories); and 10.3% (32) trajectories arrived from the south to the measurement site, for example, as shown in the Figure 3.13.

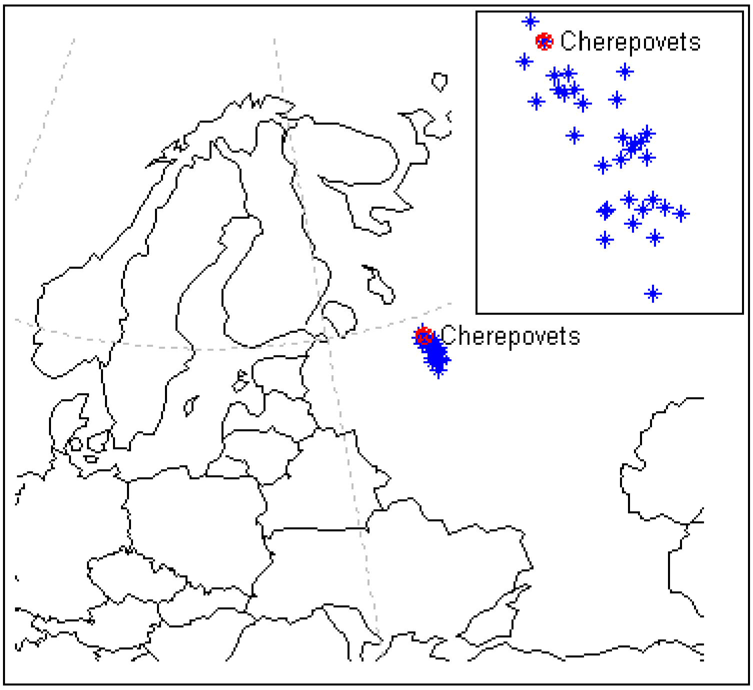


***Figure 3.13**: Trajectory coordinate pairs (time interval of 6 hours; level of 100 m) for the cluster #1 associated with the atmospheric transport from the southern sector.*

As the results of clustering analysis for 500 m level 4 representative clusters were obtained: #2, #4, #5, and #6 (Figure 3.12b, Table 3.1). These are accounted in total 97.4% of all calculated trajectories for this time interval. Dominating direction of arrival (cluster #4), which was found to be north-western, was represented by 41.5% (129) trajectories. One more prevailing direction (cluster #6) appeared to be northern with 30.2% (94) trajectories. Cluster # 5 represents southern direction with 16.4% (59) trajectories and cluster # 2 with the smallest number of trajectories - 9.3% (29) - corresponds to north-eastern direction.
The change of dominating direction for two levels can be related to the rotation of the wind with a height. Nevertheless, the general atmospheric flow associated with the western transport can cause an arrival of trajectories with elevated concentrations of krypton to the Cherepovets measurement site. The second dominating i.e. northern direction is better appeared at 500 m level and larger by 15.7% compared with 100 m level. The northern atmospheric transport can be associated with arrival of trajectories with lowered values.

3.5.2 Analysis of cluster statistics for xenon

Similar clustering approach was applied to ^{133}Xe trajectories. The Table 3.2 with number and percentage of trajectories per cluster for both heights at the first 6 hour interval is represented below. Similar to the above considered analysis for krypton the number of clusters originally selected for the procedure was equal to eight. As the result, 3 representative clusters were obtained for both levels in height (Figure 3.14).

Table 3.2: *Percentage and number of latitude/ longitude pairs per cluster for the first time interval of 6 hours (the levels of 100 / 500 m).*

^{133}Xe	Percentage value [%] / number of coordinate pairs							
# cluster	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**
100 m	0.4 / 1	30.7 / 73	44.5 / 106	0.4 / 1	18.5 / 44	0.4 / 1	1.3 / 3	3.8 / 9
500 m	1.7 / 4	7.6 / 18	0.4 / 1	3.8 / 9	0.8 / 2	39.7 / 94	41.4 / 98	4.6 / 11

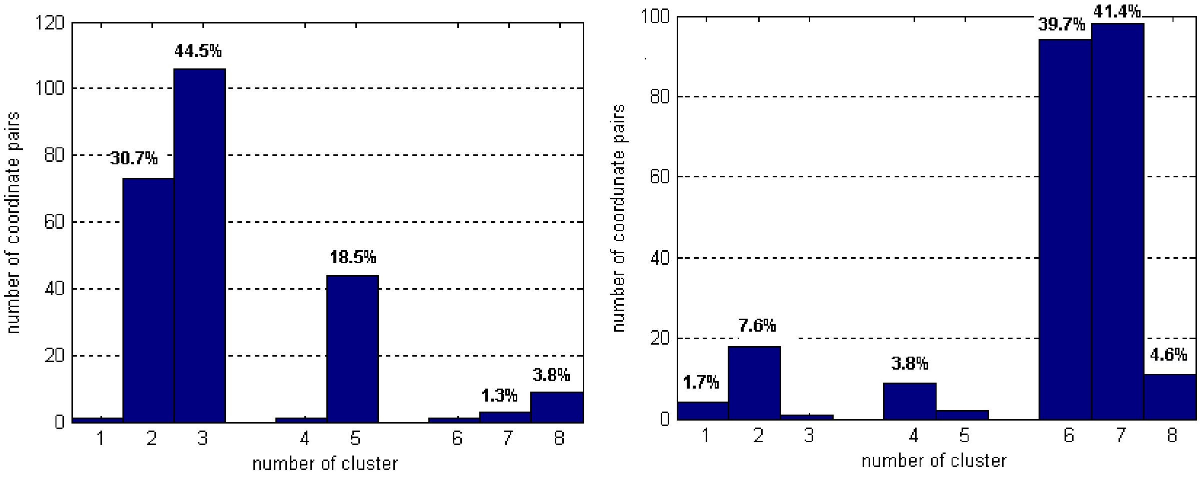


Figure 3.14: *Number of latitude/ longitude pairs per cluster obtained for the first time interval of 6 h corresponding to the height of (a) 100 and (b) 500 m agl.*

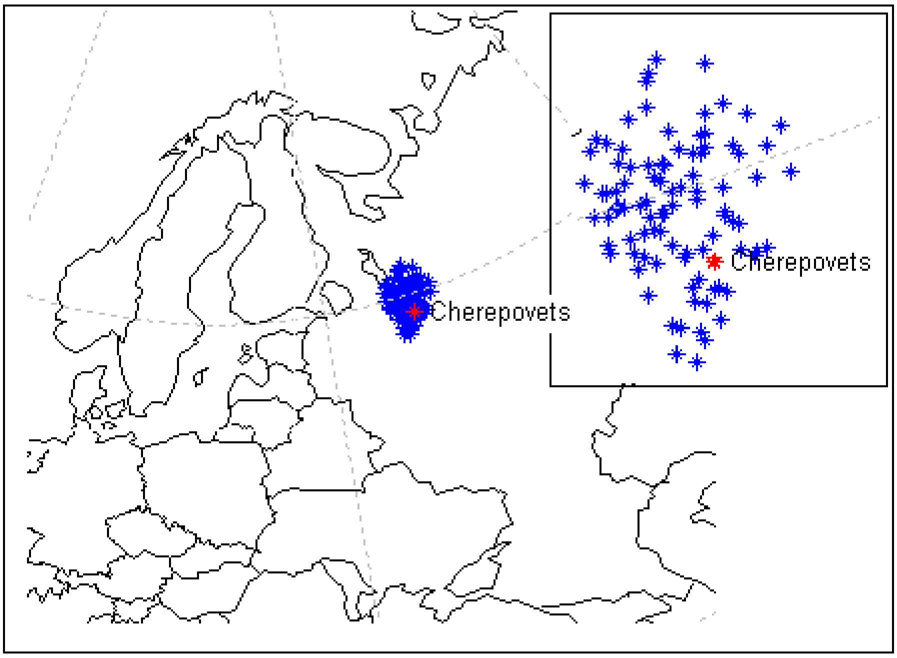


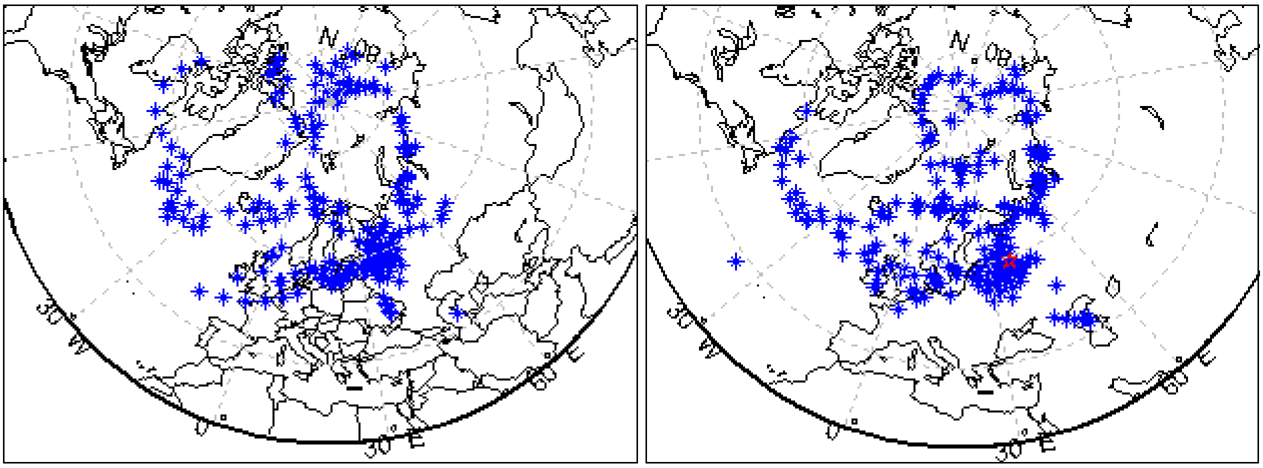
Figure 3.15: *Trajectory coordinate pairs (time interval of 6 hours; level of 500 m) for the cluster #6 associated with atmospheric transport from the northern sector.*

The northern direction (cluster #3) was dominating for the level of 100 m agl and comprised 44.5% (Figure 3.14a). The number of trajectories associated with this type of transport was equal to 106. Trajectories arrived from the Southern direction (cluster #2) amounted 30.7% (73 trajectories). The

western direction (cluster #5) was observed for 44 cases (trajectories) with percentage number of 18.5%. In total number of trajectories for representative clusters accounted 93.7%.
The prevailing direction for the level of 500 m agl (Figure 3.14b) was found to be western – south-western (cluster #7), and percentage value for the case was equal to 41.4% (98 trajectories). The northern direction (cluster #6) which was dominating at the lower level at 500 m level comprised 39.7% (94 trajectories), i.e. almost 4.8% less (Figure 3.15).
The southern direction (cluster #2) was 23.1% less for the 500 m level compared with 100 m level and accounted 7.6% (18 trajectories).
Thus, the prevailing western or south-western direction can be associated with arriving of contaminated air masses to the measurement site, while lowered concentrations of xenon can be transported to the site from the north.

3.5.3 Mean air trajectory - flow pattern

From the cluster analysis data obtained latitude/longitude averaged pairs were extracted from each cluster in order to estimate general (mean) atmospheric transport pattern of radioactive gases to the measurement site. According to the number of representative clusters the obtained 3 – 4 averaged coordinate pairs at each time-interval were calculated from all available at the exact time interval, plotted and then analysed for both radioactive gases.



(a) *(b)*

***Figure 3.16**: General atmospheric transport patterns represented by averaged coordinate pairs at all time intervals for (a) ^{85}Kr and (b) ^{133}Xe for the level of 100 m agl.*

Cluster analysis showed that in general, there are 3 dominating atmospheric transport pathways, i.e. from the western, northern, and "local" southern sectors and these can be distinguished.

- For ^{85}Kr, it was evaluated that 42.5% of trajectories were associated with the western transport (Figure 3.16a). Mean atmospheric flow from the northern sector accounted 34.9%, and 12.2% of trajectories arrived from the southern sector. Trajectories having no clear identification accounted 10.4% in total.
- For ^{133}Xe, 36.2% of trajectories were associated with atmospheric transport from the western sector and 31.6% of atmospheric transport was related to the northern (Figure 3.16b). Southern direction accounted 21.7%. Trajectories having no clear identification comprised 10.5% in total.

Therefore, for both noble gases the main dominating transport pathways was associated with the western sector and related to elevated concentrations measured at the Cherepovets site. The transport form the northern sector was associated with the lowered concentrations, and it occurred over territories with no presence of the nuclear power plants, as sources of noble gases emissions.

4. Conclusions

Following the main goal of this study the estimation of atmospheric transport for elevated and lowered (the above/ below a range of accepted standards and mean values) concentrations of radioactive noble gases (RNG) ^{85}Kr and ^{133}Xe was realized. The RNG monitoring was performed by the V. G. Khlopin Radium Institute (St. Petersburg, Russia) at the Cherepovets measurement site (Vologda region, Russia) considered as a background station during the period of two years (Aug 2006 – Jul 2008) within the framework of the international project «*Development of methodical bases and mobile equipment for monitoring of Xe and Kr radionuclides in the Northwest region of Russia*» (2005-2009).

In order to estimate geographical regions as well as identify potential sources of emissions responsible for elevated and lowered concentration episodes for both gases several approaches were applied in this study. The time-series of ^{85}Kr and ^{133}Xe measurements were analyzed at the first step, and episodes with elevated and lowered concentration values for both gases were identified. These are accounted 28 and 26 elevated and 17 and 36 lowered episodes for krypton and xenon, correspondingly. Maximum registered concentration of ^{133}Xe (2.47±0.26 Bq/m^3) exceeded its background value (0.09±0.03 Bq/m^3) in more than 27 times while for ^{85}Kr the highest value (1.79±0.27 Bq/m^3) was 1,4 times larger the minimum (1.31±0.20 Bq/m^3).

At the second step the trajectory modelling approach was applied. In total 622 and 473 individual backward trajectories were simulated for ^{85}Kr and ^{133}Xe selected episodes, correspondingly. The trajectory modelling took into account only variability of the atmospheric transport patterns without consideration of possible losses of pollutant during transport (since noble gases are inert gases). At third, ensembles of trajectories related to extreme episodes for both gases as well as periods of elevated and lowered concentrations of krypton were simulated. This allowed obtaining additional information about the general directions of contaminated air transport and identification potential sources of emissions associated with these episodes as well as estimate spatial coverage of airflow patterns. The identification of source regions by means of trajectory analysis showed that the general atmospheric transport related to high concentration episodes for both noble gases occurs from the western sector where a large number of nuclear power plants (NPPs) is situated. The lowered concentration episodes in turn are associated with the transport from the northern sector where there are no nuclear plants - with an exception of the Russian Kola nuclear plant.
Finally, to derive mean atmospheric pathways and flow patterns and obtain additional probability information the cluster analysis technique was applied. Clustered backward trajectories represented general atmospheric transport pathways to the Cherepovets measurement site averaged over a period of noble gases measurements. Approximate geographical boundaries of potential source regions as well as an average transport time to the site, and percentage characteristics of the transport are estimated. The number of representative clusters for all time intervals accounted for more than 90%. The maximal percentages for each time interval were associated with an arrival of atmospheric trajectories from the western sector, first of all, as well as from the northern. The common feature of atmospheric transport for ^{85}Kr and ^{133}Xe was identified. Nevertheless, it is important to note that a number of complex cases (mainly for krypton) also exists. This can be explained by using of a single trajectory representing the transport. The second reason can be an uncertainty caused by sampling technique for noble gases. Moreover, measurements are carried out at the ground level while the calculated air trajectories have arrived at higher levels, i.e. there might be influences due to local terrain features as well as local meteorological conditions.

The significance of studies connected with spatial and temporal variations of krypton and xenon atmospheric transport is rather large. Continuous accumulation of ^{85}Kr in the atmosphere and identification of not announced nuclear accidents as well as safety and quality of nuclear power plants operation by tracking of ^{133}Xe atmospheric transport are challenging problems that should be studied. The probability of further growth of krypton content in the atmosphere is rather high nowadays due to existing problems of its trapping from the gaseous emissions of reprocessing nuclear factories. Further growth of its content in the atmosphere could enhance its affect on atmospheric electro conductivity as well as cause a development of the global geophysical effects influencing climate. Furthermore, the continuous development of nuclear industry and building of nuclear power generating units needs expansion of Xe radionuclides monitoring sites and studying peculiarities of its transport and circulation in the atmosphere.

The results of this study were presented at the International Conference on Environmental Observations, Modeling and Information Systems (5-11 Jul 2010, Tomsk, Russia) as well as will be finalized in a form of manuscript for submission to “Atomic Energy” journal.

The future research plans include analysis of long-term time-series of ^{133}Xe and ^{85}Kr at the Saint-Petersburg measurement site. This city comparing with Cherepovets is a large populated agglomeration (more than 5 million people) that is situated in a proximity to the Leningradskaya NPP as well as there are many European nuclear plants to the west from the measurement site. Additionally, a probability field analysis will be applied in order to identify more detailed geographical boundaries of potential sources regions. Furthermore, the dispersion modelling approach will be applied using a modified Enviro-HIRLAM (Environment – High Resolution Limited Area Model) for radioactivity applications. Afterwards, results of trajectory modelling (cluster analysis over trajectories) will be compared with dispersion modelling results as well as dominating annual and monthly spatial-temporal patterns for the potential sources will be investigated.

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