Study of transport of atmospheric admixtures and temperature anomalies using trajectory methods at the A.M. Obukhov Institute of Atmospheric Physics

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Abstract. Based on the trajectory model NOAA HYSPLIT_4 and reanalysis NCEP/NCAR a method for calculating extra large sets (millions of trajectories) of forward and backward trajectories has developed in A. M. Obukhov Institute of Atmospheric Physics (OIAP) of RAS. Using the arrays and trajectory methods, the fields of potential sources of aerosol and gas species are identified. Analyzed are distributions of potential regions of sources of ammonium nitrate, ammonium sulfate, and silicate contributing the surface aerosol, as well as of tropospheric formaldehyde and stratospheric nitrogen dioxide at the Zvenigorod Scientific Station of OIAP, of stratospheric ozone at the Kislovodsk High Mountain scientific Station of OIAP, of column aerosol contents at AERONET stations of Tomsk and Ussuriysk. The method is also applied to identification of remote regions associated with anomalies of winter surface air temperature in Moscow and anomalies of precipitation in the Lake Baikal basin.

1. Introduction

Trajectory models like HYSPLIT_4 [1] or FLEXPART [2] associated with global reanalysis data like NCEP/NCAR [3], ERA-Interim [4] or NCEP1.0/0.5 (ftp://arlftp.arlhq.noaa.gov/pub/archives/) make possible calculations of large arrays of trajectories used for studying transport of atmospheric admixtures on regional and global scales. Growing amount of observational data of atmospheric pollutants within ground-based networks and from satellites promises further increase in publications on this subject. The study of the long-range transport of atmospheric species in Russia is particularly important because of the long state border and vicinity of densely populated and rapidly developing regions.

A method of calculations and analysis of extra large sets (millions of trajectories) of long (up to 20 days) forward or backward air parcel (elementary air particle) trajectories has been developed at the A.M. Obukhov Institute of Atmospheric Physics of Russian Academy of Science (OIAP). The method is based on freeware freely available trajectory model and reanalysis data. Using the method, potential source regions of a number of aerosol [5-7] and gas atmospheric admixtures were identified and the probability of air mass transport from arid regions of southern Russia was estimated [8]. Applicability
of trajectory methods for climate studies was shown on the examples of identifying of remote regions that could be responsible for extreme air temperatures in Moscow [9, 10] and source region of precipitation in the Russian part of the Baikal basin [11]. The present work is a brief review of some studies of the transport of atmospheric species at OIAP.

2. Methods and data

The NOAA HYSPLIT_4 model and NCEP/NCAR reanalysis data (resolution 2.5°×2.5°) and NCEP GDAS1.0 (resolution 1.0°×1.0°) for the periods, respectively, from 1948 and 2004 to present time were used to calculate the forward or backward trajectories. To determine the average contribution of sources of an admixture in the cells of the space [ij] to the concentration of this admixture in the studied point A, the CWT (concentration weighted trajectory) method [12] is used, according to which the backward trajectory (BT) in [ij] is summed in [ij] with a weight equal to the admixture concentration at the moment of arrival of the air mass in A. The average over all BTs gives the average contribution [ij] to the admixture concentration in A. In the case of local measurements, the BT is calculated for the height of point A above the surface. The weight of the admixture in the air column above point A can also be the weight. In this case, the BT is calculated and analyzed together for a number of heights within the air column [13]. If the admixture has a distinct vertical profile, then BT at different heights with weights corresponding to this profile are taken into account (see, for example, [6]).

2.1. Aerosol admixtures

To determine the average contribution of sources of ammonium nitrate, NH$_4$NO$_3$ [μg/m$^3$], and silicates [μg/m$^3$], data on the chemical composition of the aerosols with particle sizes of 1-2 μm obtained at the Zvenigorod Scientific Station (ZSS, 55.7 N, 36.8 E) of OIAP in 2004-2017 [7]. The sources of aerosols for the south of Siberia and the south of the Russian Far East were determined using aerosol volume concentrations [μm$^3$/μm$^3$] in the range of particle sizes of 0.1-5.0 μm in the atmospheric column, according to data of AERONET [14, 15] sites in Tomsk (56.5 N, 85.0 E) and Ussuriysk (43.8 N, 131.95 E). Trajectories were calculated simultaneously for seven heights in the range ≤ 5.0 km, in which the bulk of the aerosol mass is concentrated [16]. Trajectories of different heights were taken into account with a weight proportional to the aerosol backscattering coefficients at the appropriate height by the method described in [6].

2.2. Probability of air particle transport

The probability of transport of air particles from the near-surface layer (start height is 50 m) in the arid region of southern Russia (Kalmykia and Astrakhan Oblast) to spatial cells [ij] of size 1.0°×1.0°, $P_{ij}$, was estimated using 5-day forward trajectories (FTs) by the relationship $P_{ij} = M_{ij}/N×100\%$, where $M_{ij}$ is the number of FTs hit the [ij], N is the number of all FTs [8].

2.3. Gas admixtures

To determine the source regions of the anomalies of tropospheric formaldehyde (HCHO) content [molecules/cm$^2$ (mol/cm$^3$)], stratospheric ozone (O$_3$) content [DU], stratospheric nitrogen dioxide (NO$_2$) content [mol/cm$^3$], the datasets of these admixtures, obtained respectively at ZSS in 2009-2017 [17-19], see figure 1a, at the Kislovodsk High Mountain scientific Station (KHMS, 43.7 N, 42.7 E) of OIAP in 2002-2016 [20], see figure 1c, and at ZSS in 1990-2017 [21, 22], see figure 1e, are used. By subtracting annual cycles, anomalies of tropospheric formaldehyde, HCHO' (figure 1b), stratospheric nitrogen dioxide, NO$_2'$ (figure 1d), and stratospheric ozone, O$_3'$ (figure 1f) were obtained, which were used to calculate the source distributions of the admixtures’ anomalies.
2.4. Precipitation

In view of assumption of the leading role of water vapor in the formation of precipitation the contributions of different regions to daily precipitation [mm/day] in the Russian part of the Lake Baikal basin according to the datasets of daily precipitation at 24 meteorological stations (MSs) of Roshydromet [23], see figure 2, in 1948-2016 were determined in [11]. Further, the analysis was extended by daily precipitation at 19 MSs in the Mongolian part of the basin (figure 2) over the same period obtained from the NOAA meteorological data archive at https://www7.ncdc.noaa.gov/CDO/cdoselect.cmd. To account for the wind turn with altitude, the backward trajectories were calculated for heights of $1/3H_0$, $2/3H_0$ and $H_0$, where $H_0$ is the mixed layer depth above an MS in the time of arriving of air particle to the MS. Using the massifs of trajectories arrived to all MSs in Baikal basin, the contribution of the regions to the daily precipitation in the whole basin in 1948-2017 was calculated.
2.5. Air temperature

Under the assumption of the advective nature of temperature extrema in Moscow, the backward trajectories of air masses associated with cold and warm outbreaks in different seasons were studied in [9, 10]. In the case of winter (December-January-February) extreme cooling and warming in Moscow daily surface air temperatures, $T$ [$^\circ$C], were used at the VDNH meteorological station [23] of Roshydromet in 1948-2017, see figure 1g. By subtracting annual trends for each day of winter, temperature anomalies, $T'$ [$^\circ$C], were obtained (figure 1h) which were used as weights of the corresponding 5-day backward trajectories of air particles arriving in Moscow at a height of 50 m.

3. Results

3.1. Aerosol admixtures

As it was shown in [7] the 20% of the highest concentrations of NH$_4$NO$_3$ in surface aerosols with particle sizes in the range 1-2 μm with a probability of about 80% are observed at ZSS when air masses are transported from the region of Western Europe extending from the south of Britain to the north of Italy. According to data on all concentrations (not only the highest ones), it is shown that the maximum average concentration of NH$_4$NO$_3$ at ZSS in 2004-2017 are contributed by the air masses from central France and northern Italy (figure 3a). Aerosols with an extreme concentration of silicates enter the ZSS from the Caspian and Aral regions of Kazakhstan [7], for which the maximum average contribution to the concentration of silicates at ZSS is also typical, see figure 3b. Note that the contributions of the regions to the east and west of ZSS are different for the admixtures at times (up to one order).

An aerosol transport study based on data from the AERONET station in Tomsk showed that the largest contribution to the volume concentration of aerosols in the range of particle sizes of 0.1-5.0 μm in the atmospheric column is provided by sources located on the border of Mongolia and China near the Gobi Desert, in the Middle East (Iran and Arabian peninsula), as well as on the border of Russia and eastern Kazakhstan (figure 3c). Less significant sources are in the western and central parts of Kazakhstan, as well as in northern Africa (figure 3c).

For the south of the Russian Far East (Ussuriysk), the Yellow Sea, as well as the eastern provinces of China near the Yellow Sea (figure 3d), are the most significant sources of aerosols in the range 0.1-5.0 μm.
μm, where according to satellite data [26, 27] a regional maximum of the aerosol optical depth is observed. The influence of this region is greatest in the warm season, which is probably due to the change in the dominant southeastern direction of the East-Asian monsoon winds in the warm season to the opposite direction in the cold season. Figure 3d also shows that the area of high contribution to the aerosol concentration in Ussuriysk is stretched in the west direction, which may be due to the transport of mineral aerosols from the Gobi Desert.

**Figure 3.** The average contribution of different regions to the content of ammonium nitrate (a) and silicates (b) in aerosols in the range 1-2 μm in the western Moscow Oblast. The cells' size is 1°×1°.

The average contribution of regions aerosol volume concentration (aerosol in the range of 0.1-5.0 μm) in the atmospheric column above Tomsk (c) and Ussuriysk (d). The cell's size is 5°×5°.

The probability of transport of air particles from the deserted region (marked by the black triangle) of Kalmykia and the Astrakhan Oblast for the whole atmosphere column above the 1°×1° (e) cell, the same for the mixed layer (f).

The average contribution of the potential source to the tropospheric formaldehyde content anomaly at Zvenigorod Scientific Station (ZSS) in October-March (g) and April-September (h). The cells' size is 0.13°×0.13°. Border of Moscow megacity is marked with white line.
3.2. Probability of polluted air mass transport

Air particles, starting at a height of 50 m above the deserted regions of the south of Russia in Kalmykia and the Astrakhan Oblast, are most likely to move in the northeast, west and south directions (figure 3e), and fall with the same probability (0.4-1.0%) into the atmospheric column with a 1°×1° base in the south of the West Siberia, the central part of the Black Sea, the southern Caspian and Moscow, see figure 3e. The probability of the transport to Saint Petersburg is 2.5 times less than to Moscow, 6 times less than to Saratov, Voronezh and Sochi and 16 times less than to Volgograd (figure 3e). The decrease in the probability of falling into the mixed layer in comparison with the entire atmosphere is strongly dependent on the direction of transport and the distance from the trajectory start site (figures 3e and 3f). For example, the probability of falling into a mixed layer, compared with the entire atmosphere, for Saint Petersburg is 6 times lower, for Saratov is 3 times lower, and for Sochi and the northern Caspian remains constant (figures 3e and 3f).

3.3. Tropospheric formaldehyde

The lifetime of HCHO in the atmosphere is several hours [28]; therefore, 10-hour backward trajectories were calculated to determine the regions of sources of HCHO transported to ZSS. Assuming the dominant contribution of terrestrial sources to the HCHO content and hence the maximum of HCHO in atmospheric boundary layer, the backward trajectories were calculated for heights of 1/3H0, 2/3H0 and H0, where H0 is the mixed layer depth, and only the trajectory portions that entered the mixed layer participated in the calculation of the source field. Apparently, Moscow is the main source of high HCHO anomalies in the western part of the Moscow Oblast in the cold season (figure 3g). The areas highly contributed to the anomaly are allocated in the east and southeast of Moscow, where the Moscow oil refinery plant is located, a number of chemical industries, the most powerful Moscow thermal power plant, TPP-22, is located and where the densest motor traffic, especially in the cold season, is observed [29]. In other words, this is an area where the emissions of anthropogenic HCHO and volatile organic compounds, which are precursors of HCHO, are potentially high. In the warm season, the positive HCHO anomalies at ZSS are also caused by air transport from the east (figure 3h), but Moscow's contribution is significantly lower, which may be due to the shorter lifetime of the HCHO molecule with increased insolation (and an increased probability of reaction with the OH radical) in a warm season. Note that to the southeast of ZSS are areas (west and south-west of Moscow Oblast), which are also associated with increased HCHO anomalies, the origin of which has not yet been clarified.

3.4. Stratospheric ozone and nitrogen dioxide

The maximum contents of O3 and NO2 in the stratosphere is observed in the layers of 20-25 km and 25-30 km, respectively. For these altitude ranges, with an interval of 1 km, 5-day backward trajectories of air particles were calculated, arriving, respectively, at KHMS (O3 measurements) and at ZSS (NO2 measurements). Figures 4a and 4b show the average contribution of potential source regions into the O3 anomaly at KHMS (figure 4a) and NO2 at ZSS (figure 4b) during the cold period of the year from October to March. In general, the distribution of the source regions of the stratospheric O3 anomalies in October-March (figure 4a) reflects its latitudinal distribution in the cold season [30, 31]. The formation of stratospheric O3 is greatest in the tropics, but the most favorable conditions for its accumulation arise in the cold season in high and temperate latitudes [30, 31], where O3 moves to due to meridional circulation of the atmosphere.
Figure 4. Average long-term contribution the regions to the anomaly of stratospheric ozone (O₃) at Kislovodsk High Mountain Station (KHMS) (a) and stratospheric nitrogen dioxide (NO₂) at Zvenigorod Scientific Station (ZSS) (b) in October-March. The cells' size is 5°×5°.

The average contribution of regions to precipitation in the Lake Baikal basin (c) and the average specific humidity of air in the area of transport (d). Area of Baikal basin is marked with white polygon. The cells' size is 5°×5°.

The density of the backward trajectories associated with extreme warming (e) and extreme cold outbreaks (f) in Moscow in 1948-2017. The cells' size is 1°×1°.

The average contribution of the region to the anomaly of the winter surface air temperature in Moscow (g), the average air temperature in the area of air mass transport (h). The cells' size is 1°×1°.

For stratospheric NO₂ anomalies in the cold season another feature is obtained: the minimum anomalies of the stratospheric NO₂ content at ZSS are related to the air masses from the Arctic, and the maximum ones with an extended region in the latitudinal zone of 30-45 N over the Atlantic Ocean, northern Africa, the Mediterranean Sea and southern Europe (figure 4b). This distribution of NO₂ sources also reflects latitudinal variation of nitrogen dioxide in the cold season [32, 33].
3.5. Precipitation

The typical lifetime of the water vapor molecule in the atmosphere is about 10 days [34, 35], so 240-hour backward trajectories were calculated for heights of $1/3H_0$, $2/3H_0$ and $H_0$, where $H_0$ is the depth of mixed layer in which precipitation mainly occurs [34, 36]. Primary sources of water vapor are on the surface, so most of water vapor mass is transported in the atmospheric boundary layer. At the same time, it is obvious that a significant number of water vapor is transported above the boundary layer (clouds of upper levels). Thus, any layer of the troposphere can be a secondary source of water vapor, so in this paper, in contrast to [11], all sections of backward trajectories were taken into account, not just those that entered the mixed layer.

Calculations showed that the average precipitation in Lake Baikal basin with an average intensity of over 9 mm/day fall out when air masses from the Asia-Pacific region arrive to the basin (figure 4c). Precipitation with an average intensity of more than 18 mm/day is associated with the water area to the south and southeast of the coast of China, and precipitation of maximum intensity, more than 27 mm/day, with the South China and Philippine Seas (figure 4c). Average in the basin, low daily precipitation, 3-6 mm, is associated with air masses from Eurasia and China to the southwest of the basin, as well as from the Northern, Barents, Greenland, Norwegian Seas and the Bay of Biscay (figure 4c). With the Atlantic north of 40 N, as with the Middle East, the minimum precipitation is associated, less than 3 mm/day. The most unexpected was the relatively weak contribution of the Indian Ocean to precipitation in Baikal basin, which is probably due to the predominance of eastern air mass transport in the tropics and the influence of mountain systems (the Himalayas, Tibet) on the transport. The contribution of this or that region to the precipitations in the basin (figure 4c) is in good agreement with the average specific humidity of the air along the trajectory in the area of air mass transport (figure 4d). The Pacific Ocean is closest to the rest of the oceans to Lake Baikal basin, and the specific humidity of the air in the area of transport above it is greatest, which apparently determines that the maximum daily precipitation in Baikal basin is connected with Pacific Ocean.

3.6. Air temperature

As shown in [10], the remote regions with which extremely low and extremely high air temperature anomalies are associated in winter in Moscow are different (figures 4e and 4f), which confirms the assumption of the advective nature of winter temperature extremes in Moscow. Extreme cold outbreaks (anomalies < $-9.1^\circ$C, figure 3b) are associated with the air mass transported from a vast area from Moscow to western Siberia and from the Barents Sea to Kazakhstan with a center near 60 N, 50 E (figure 4f). Extreme warming (anomalies > 7.7 $^\circ$C) occur in winter in Moscow when air masses arrive from the latitudinal belt region of 40-50 N, which extends from the eastern Atlantic to the Caspian Sea (figure 4e). Assuming that even smaller (in modulus) anomalies can also be associated with the air mass transport, but from regions further away from sources of advective heat and "cold", the CWT method can determine the average contribution of regions to winter surface air temperature anomalies in Moscow in the entire range of anomalies, not just for extreme ones.

As can be seen from figures 4g and 4h, the distribution of the average contribution of the region to the surface air temperature anomaly in winter in Moscow does not contradict the fields of density of backward trajectories (figures 4e and 4f). Moscow itself turned out to be exactly on the line of zero anomalies. This line almost everywhere corresponds to a region with an average air temperature, as in Moscow (figure 4h). The coldest winter days in Moscow (anomalies < $-14^\circ$C) are associated with the region south of the Kara Sea (the north of the Yamal-Nenets Autonomous District and the Taimyr Autonomous District). The average temperature of air masses moving to Moscow in this region is very low: $-25...-30^\circ$C (figure 4h). Interestingly, in the north of the Greenland and Barents Seas, air masses are on average as cold (figure 4h), but the air mass transport there from is associated with much weaker temperature anomalies in Moscow in the range 0...6 $^\circ$C (figure 4g), which, apparently due to the "screening effect" of the warmer Norwegian and Barents seas. It can be seen from figure 4h that the main winter sources of advective heat for Moscow are at latitudes 40-50 N in the eastern Atlantic and in the northern part of the Mediterranean Sea. The average air temperature in the area of transport...
in these regions is > 10 °C, i.e. more than 20 °C higher than in Moscow (figure 4h). Thus, although air transport from these regions is associated with high temperature anomalies in Moscow, 6…8 °C (figure 4g), these anomalies could be higher by at least 12 °C. In other words, the air from these regions in the Atlantic and the Mediterranean Sea during winter to Moscow is cooled by not less than 12 °C.

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