Localization of source regions of odd nitrogen through a receptor climatology of trajectories for the region of Western Siberia

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Air parcel trajectories arriving at Tall Mast (60°26' n.lat., 89°24'e.lon.) in the Western Siberia (see Fig.1) are calculated for a six month period from March to September 2007 using 24h isosigma trajectories in the lower troposphere basing on NCAR/NCEP final analysis data. Model output along with the observed surface concentrations of O3, NO and NO2 are analyzed to describe the regional transport climatology by examining the distribution of back trajectory end points over a latitude-longitude grid as a function of measured concentrations at the monitoring site. Statistically significant regions of the most important NOx sources in the West-Central Siberia have been identified along with seasonal variation of the primary transport paths of anthropogenic pollutants in the region. The results are found to be in a good agreement with the previous data on surface O3 and NOx concentrations measured during TROICA campaigns for the last decade along the Trans-Siberian Railroad. The seasonal variability of the measured species along with the sensitivity of the regional boundary layer photochemistry to the primary emission sources in the region are investigated basing on the developed source-receptor model to assess regional influence of climatologically significant regional sources of NOx.
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1. Tall Mast observations. The measurements of surface concentrations of NO, NO2 and O3 have been conducting by the Obukhov Institute of Atmospheric Physics at Tall Mast since 15 Mar 2007. The observational station is located in the plane boreal area approximately 300 m a.s.l. The surrounding terrain is almost desolate with the nearest large settlements located about 500 km south (the towns of the Southern Siberia such as Krasnoyarsk and Kansk). One of the primary goals of the measurements is to study the various effects of long-range and regional atmospheric transport. Our study employs measurements of NOx to test the validity and usefulness of conditional frequency analysis proposed earlier by Ashbaugh (1985) and further developed by L. Vasconcelos et al. (1996) to assess the influence of regional anthropogenic emission sources on the measured data and to study some characteristic peculiarities of the regional transport.

Fig.1. Tall mast in the Western Siberia (60°26' n.lat., 89°24'e.lon).

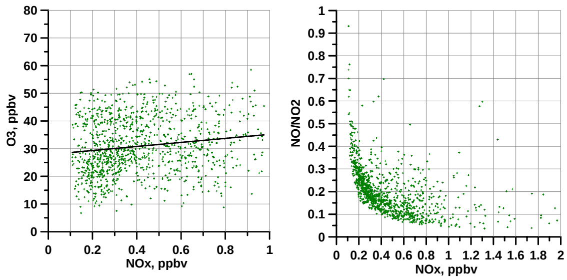

Fig.2. Daily ozone concentrations versus NOx (left) and ratio NO/NO2 versus NOx (right) for the measurement data shown on Fig.3.

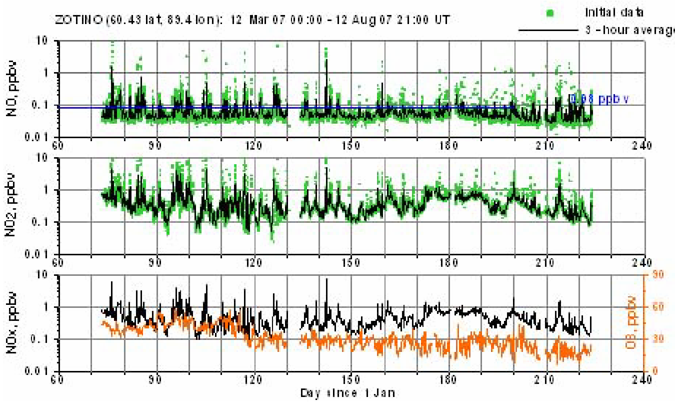

Fig.3. Surface (4m a.g.l.) concentrations of NO, NO2, and O3: 10 min averages (initial data, green) and 3 – hour averages (black). Orange – 3-hour average ozone concentrations.

As follows from the Fig.3, the higher values of surface O3 during the day time generally correspond to higher NOx (=NO+NO2) under the ratio NO/NO2 less compared to unity, which corresponds to the conditions of NOx-minimizing photochemical system, peculiar to the background as well as slightly contaminated surface air.

2. Trajectory model. Calculations of air parcel Lagrangian trajectories have been performed basing on developed isosigma model in which it is assumed that air motions take place on surfaces of constant values of $\sigma=(p-p_t)/(p_s-p_t)$, ranging from σ=0.95 to σ= 0.8 in the present calculations, where $p_s(\varphi,\lambda,t)$ is the surface pressure and p_t is the top of the model (100 mbar). A trajectory path $\mathbf{r}(t)$ is related to the wind velocity field $\mathbf{V}(\mathbf{r},t)$ through the first order differential equation

$$\frac{d\,\mathbf{r}(t)}{d\,t}=\mathbf{V}(\mathbf{r}(t),t), \qquad (1)$$

whose solution for a arbitrary moment $t_N > t_0$ is

$$\mathbf{r}(t_N) = \mathbf{r}(t_0)+\int_{t_0}^{t_N}\mathbf{V}(\mathbf{r}(t),t)\,dt. \qquad (2)$$

Equation (2) at each subinterval (t_k,t_{k+1}) has been solved by second order in time Runge-Kutta method

$$\mathbf{r}(t_h) = \mathbf{r}(t_k)+\frac{\tau}{2}\cdot\mathbf{V}(\mathbf{r}(t_k),t_k), \quad t_h = t_k+\tau/2, \qquad (3)$$

$$\mathbf{r}(t_{k+1}) = \mathbf{r}(t_k)+\tau\cdot\mathbf{V}(\mathbf{r}(t_h),t_h)\;, \qquad (4)$$

where $\mathbf{V}(\mathbf{r}(t),t)$ is calculated by linear interpolation in space and quadratic interpolation in time with respect to the adjacent nodes of regular grid of the final analysis data whose original resolution is (1deg. Lat. x 1 deg.Lon.x 12h).

3. Statistical model Following a now-standard approach, the distribution of back trajectory segment endpoints aver a regular latitude-longitude grid 2.5x2.5 deg. is examined as a function of measured concentrations at the station. Grid cells in which segment endpoins are preferentially associated with high concentrations are then identified as the regions of climatologically significant sources basing on the calculated spatial probability distributions for individual air parcels.

4. Calculations. According to the calculations (see Fig.4) the 24 hour influence area (from the climatologic viewpoint) comprises a vast regions from the Ural Mountains on the west to the East-Siberian plateau on the east, and from tundra regions on the north to deserts of the Kasakhstan on the south. The directions of air mass arrival are distributed almost uniformly from north-west to south octants.

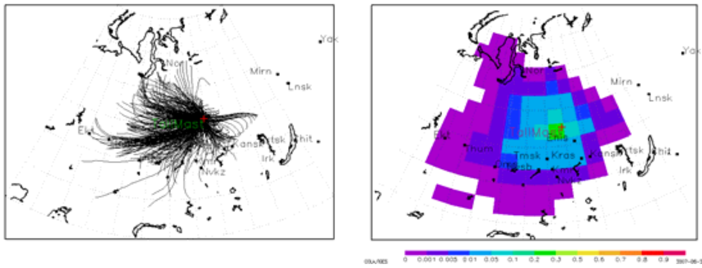

Fig.4. Ensemble of 24-hour 900 mbar isobaric trajectories arriving at Tall Mast for the period of 15 Mar – 15 Aug 2007 (left) and the derived influence area given in terms of a residence time in the given cell normalized by the total time of observations (right).

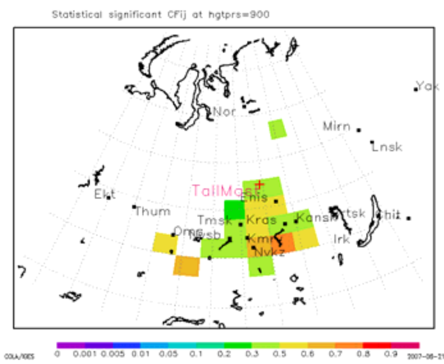

Рис.5. Statistically significant conditional frequency of the arrival of air with enhanced NOx from the given cell. Cells with CF>0.5 can be considered as areas of climatologically significant anthropogenic sources of emissions.

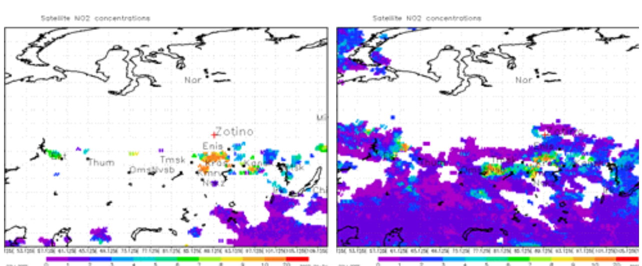

Fig.6. Monthly averaged column tropospheric NOx, (? 10^{16} mol. /cm2), derived from SCIAMACHY measurements (right – Jan 2006, left – Apr 2006). The both plates along with fig. 5, point to the towns of the Southern Siberia as the primary regions of high NOx emissions.

5. Comparison with TROICA-11 in-situ measurements. During 25 Jul – 5 Aug 2007 the measurements of surface concentrations of primary minor species along the Trans-Siberian railroad from Moscow to Vladivostok have been conducted with use of specially equipped carriage—observatory in the course of 11-th TROICA expedition. On the return pass the carriage-observatory have transected the same air mass which has arrived to the Tall Mast 1 day later, so the combined data from the both stationary point and moving observational system can be used to test the developed trajectory model as well as to study a characteristic episode of high NOx observed at Tall Mast associated to regional advection.

Fig.7. Carriage – observatory. The complex equipped with measuring instruments manufactured by the leading world manufacturers provides round-the-clock all-the-year-round continuous measurements of all basic gaseous species: ozone, nitrogen monoxide and dioxide, carbon monoxide and dioxide, methane, sum of non-methane hydrocarbons, sulfur dioxide and ammonia.

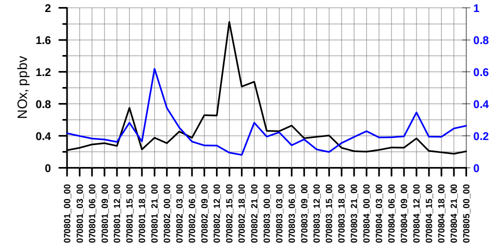

Fig.8. Observations of NOx surface concentrations (black) and NO/NO2 ratio (blue) at Zotino station for 1 Aug 07 00:00 – 5 Aug 07 00:00 GMT.

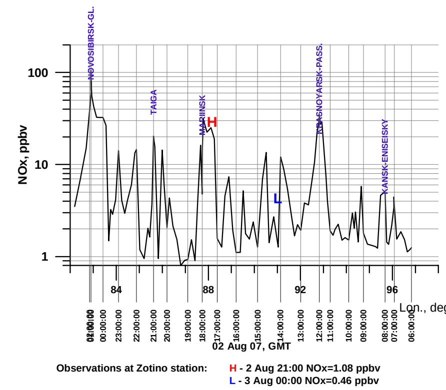

Fig.9. Observations of NOx surface concentrations along Trans-Siberian railroad on 2 Aug 07 06:00 – 3 Aug 07 00:00 GMT. Symbols H and L mark the times of observations of air parcels which were also observed at Zotiono station 2 day later during episodes of high and low NOx concentrations, correspondingly.

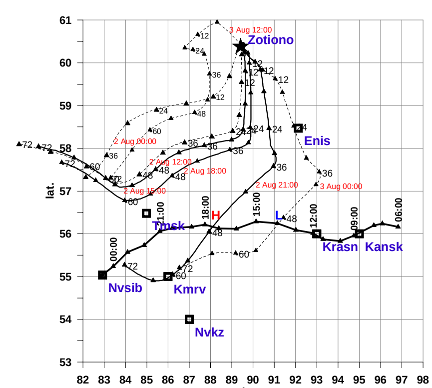

Fig.10. Air parcel trajectories at 950 mbar arriving at Zotino station for 2 Aug 07 00:00 – 3 Aug 07 12:00 GMT during episodes of high (solid) and low (dashed) NOx surface concentrations at the station. Red dates mark the time of arrival of the trajectory at the station. Bold solid line marks the Trans-Siberian Railroad. Symbols H and L are the same as for fig.7. Large towns in the region are also plotted.

6. Basic conclusions. The climatologically important sources of minor nitrogen species in the Western Siberia are primary associated with the industrial areas in the South, including those along the Trans-Siberian Railroad. The regional transport in the Western Siberia can influence significantly the spatial distribution of minor species affecting the lower tropospheric photochemistry in the distant regions of boreal forests. The background observational station Zotino can provide a valuable data to study many peculiarities of the atmospheric transport over the region, as well as over the Northern Eurasia as a whole.