

## Using background air pollutants levels correlation analysis to identify periods of long-range transport of anthropogenic pollution

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Increasing trends of airborne lead and cadmium at background station within the central region of European Russia have been identified previously (e.g., Gromov & Konkova, 2016). In order to reveal the possible drivers of these trends, a deeper investigation of correlation among these heavy metals (HM) and other co-measured pollutants is done in this study. Based on the data for the 2001-2012 period, calculations have been carried out for the period from 2006 onwards, when the growth of HM concentrations is observed. Pairwise correlations of individual species abundances were derived for the entire time series and subsets for each calendar year, including warm (April to September) and cold seasons (October to March). The calculated values for the seasons and the whole years vary substantially, suggesting that that variable ratios of atmospheric HM emission sources could affect the final air concentrations at measurement site in these periods.

To distinguish the events of predominant influence of natural and anthropogenic sources, we assume that correlation between lead and cadmium levels must be greater in the case of natural sources being in effect. High values of the correlation coefficient are expected in cases when HM air abundances are induced by the long-range transport from the regions of anthropogenic sources (co-emission of these metals results from a number of same sources, and both of them are also present on same matrix aerosols). The results demonstrate a substantial correlation between Pb and Cd, with higher values for individual seasons (70% of 0.5 and higher) than for whole years.

Higher mass concentrations of airborne dust (TSP) in remote areas are to large extent promoted by large particles blown away from the surface at local surroundings. Captured better by filters, such events could be a particular indicator of local (mostly natural) sources. Low or insignificant correlation with HM indicates prevalence of long-range transport of them and could help identifying the periods of HM of likely anthropogenic origin as well as more contribution of fine matrix aerosol in TSP. Furthermore, correlation between HM and content of sulfur (SO<sub>2</sub> and SO<sub>4</sub>) or nitrogen (NO<sub>2</sub>) oxides is applicable to the analysis of distant HM sources. In particular, these compounds co-occur in background air due to atmospheric transport of gaseous by-product from industrial fossil fuel burning sources, mostly similar to that for HM. The calculated pairwise correlations of lead with other pollutants in each season are presented in Table.

Table. Pairwise correlation ( $R^2$ ) between lead and selected pollutants load in the background air of studied area.

year	Warm season					Cold season				
	Cd	TSP	SO <sub>2</sub>	SO <sub>4</sub>	NO <sub>2</sub>	Cd	TSP	SO <sub>2</sub>	SO <sub>4</sub>	NO <sub>2</sub>
2006	0.5775*	-0.6114	-0.6246	-0.2818	0.0249	0.6858*	0.3692**	0.4823*	0.2755	0.4700*
2007	0.5952*	0.7941**	0.5895	0.1872	0.7841**	0.4863*	0.4343*	0.4680*	0.2542	0.4947*
2008	0.4636*	0.1419	0.2717	0.3945*	-0.13	0.7975*	-0.0020	0.5507*	0.2393	0.6560*
2009	0.6412*	-0.0682	0.2381**	0.2937*	0.5031	0.4718*	0.1758	0.5144*	0.1798	0.4172*
2010	0.5559*	0.3442*	0.1469	0.4507*	-0.0274	0.3466*	-0.0286	0.6014*	0.2356	0.0088
2011	0.0669	0.1765	0.0268	-0.0182	0.0567	0.6552*	-0.1147	0.4820*	-0.0859	0.1821
2012	0.4299*	-0.0912	0.0113	0.0207	0.0982	0.5550*	0.2971*	0.2619**	0.4717*	0.3171*
2013	0.2766*	0.3435	0.1483	0.26*	-0.1347	0.5492*	0.3229*	0.5088*	0.3379*	-0.0158
2014	0.2293	-0.0598	0.1868	0.1464	0.126					

\*)  $p$ -value < 5%, \*\*)  $p$ -value of 5-10%, clear – not significant

We find no significant correlation between HM and SO<sub>2</sub>, NO<sub>2</sub> or TSP in most of warm seasons. It might indicate the influence of long-range transport of HM and mostly anthropogenic nature of fine aerosol fraction. Of several exceptional cases of significant correlation, one coincides with the event of heat wave and intense wildfires (mostly

in regions of Central European Russia). We conclude that main atmospheric pollution sources over rural areas in warm seasons was also natural biomass burning (including peat smoking). Events of significant correlation between HM and gases observed in many cold seasons could be attributed to effects of additional small local sources of heat fuel combustion widely distributed within surrounding areas.

### **References**

*Gromov S.A., Konkova E.S., 2016. Trends in atmospheric heavy metals abundances over the Russian part of EMEP region over 1990-2012 period. Geophysical Research Abstracts, 18: EGU2016-17241.*