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Concomitant occurrence of anthropogenic air pollutants, mineral dust and fungal spores during long-distance transport of ragweed pollen☆

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ABSTRACT

Large-scale synoptic conditions are able to transport considerable amounts of airborne particles over entire continents by creating substantial air mass movement. This phenomenon is observed in Europe in relation to highly allergenic ragweed (*Ambrosia* L.) pollen grains that are transported from populations in Central Europe (mainly the Pannonian Plain and Balkans) to the North. The path taken by atmospheric ragweed pollen often passes through the highly industrialised mining region of Silesia in Southern Poland, considered to be one of the most polluted areas in the EU. It is hypothesized that chemical air pollutants released over Silesia could become mixed with biological material and be transported to less polluted regions further North. We analysed levels of air pollution during episodes of long-distance transport (LDT) of ragweed pollen to Poland. Results show that, concomitantly with pollen, the concentration of air pollutants with potential health-risk, i.e. SO_2, and PM_{10}, have also significantly increased (by 104% and 37%, respectively) in the receptor area (Western Poland). Chemical transport modelling (EMEP) and air mass back-trajectory analysis (HYSPLIT) showed that potential sources of PM_{10} include Silesia, as well as mineral dust from the Ukrainian steppe and the Sahara Desert. In addition, atmospheric concentrations of other allergenic biological particles, i.e. *Alternaria* Nees ex Fr. spores, also increased markedly (by 115%) during LDT episodes. We suggest that the LDT episodes of ragweed pollen over Europe are not a "one-component" phenomenon, but are often related to elevated levels of chemical air pollutants and other biotic and abiotic components (fungal spores and desert dust).

1. Introduction

Ragweed (*Ambrosia* L.) pollen is considered to be a highly potent aeroallergen worldwide (Oswalt and Marshall, 2008). It has been estimated that about 26% of the US population is sensitized to ragweed pollen (Arbes et al., 2005). In severely infested areas in Europe, such as Hungary, the clinically relevant sensitization rate among allergic patients exceeded 49% (Burbach et al., 2009). Recently, *Ambrosia* was appointed as one of the most important allergenic plants in China (Lou et al., 2017; Wang et al., 2017). It is projected that, due to climate change, the distribution range of *Ambrosia* will increase towards Northern and Eastern Europe (Rasmussen et al., 2017) resulting in substantial increase in problems associated with ragweed pollen allergy (Lake et al., 2017). In addition, the duration and intensity of ragweed pollen seasons as well as the allergenic potential of ragweed pollen may increase in the coming decades (Lake et al., 2017; Ziska et al., 2011; Hamaoui-Laguel et al., 2015; Choi et al., 2018).

The impact of *Ambrosia* is not only limited to heavily infested areas but due to the ability of ragweed pollen to be transported over

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long distances it can also affect sites located hundreds of kilometers from the source areas (Prank et al., 2013; Smith et al., 2013; Cecchi et al., 2006; Fernandez-Llamazares et al., 2012; Celenk and Malyer, 2017). Ragweed pollen transported from distant sources still possess immunoreactive activity, however its role in inducing new sensitizations is under debate (Cecchi et al., 2010; Grewling et al., 2016). Western Poland (Central Europe) is a region with well-documented examples of episodes of long-distance transport (LDT) of ragweed pollen, mainly from the Pannonian Plain (Smith et al., 2008; Stach et al., 2007), and to a lesser extent from Ukraine (Kasprzyk et al., 2011). One potential mechanism of LDT of ragweed pollen from the Pannonian Plain was described by Šikoparija, Skjøth (Šikoparija et al., 2013). The authors describe how a pressure gradient, created by high pressure around in the region of European Russia and the Black Sea and low pressure centred over Northwest Europe, result in surface winds that move west through a narrow gorge on the Danube River called the Iron Gates. This produces the gusty jet-effect wind over the Pannonian Plain termed the Kossava that, in association to sunny weather and orographic foehn winds, create the southeast-northwest movement of air forcing pollen northward to Poland and Scandinavia.

Previous studies (Grewling et al., 2016; Smith et al., 2008; Stach et al., 2007; Šikoparija et al., 2013) show that the atmospheric pathway of ragweed pollen from the Pannonian Plain to Northern Europe often passes through one of the most polluted areas in Europe, the Silesia province in Southern Poland (Leśniok et al., 2010; Kobza et al., 2018). Due to an extensive coal mining industry and combustion processes of coal in the Silesia province, elevated levels of sulphur dioxide (SO_2), nitrogen oxides (NO and NO_2), and particulate matter ($PM_{2.5}$ and PM_{10}) are recorded in the air (Bokwa, 2008). According to a report by the European Environmental Agency (EEA) (EEA, 2014), Poland is one of the largest contributors of $PM_{2.5}$ and PM_{10} emissions in the EU-28, and was the only country with increased trends in PM_{10} concentrations (2003–2012). Consequently, we hypothesize that air pollutants released over Silesia could be mixed with airborne pollen grains and simultaneously transported by air masses northwards to less polluted areas. This is important because air pollutants may interact with pollen grains in the air; agglomerating on their surface, affecting pollen vitality, altering physiologic and allergenic properties, and act as an adjuvant promoting allergic disease (Schiavoni et al., 2017; Behrendt et al., 1997; Konishi et al., 2014).

The proposed hypothesis was tested by analysing the concentration of selected primary air pollutants (PM_{10}, SO_2, CO and NO_2) in Poznań (Western Poland) recorded before, during, and after the LDT episodes of ragweed pollen (2005–2015). The transport pathways of air masses through Silesia were examined by back-trajectory analysis (HYSPLIT), while the transport of PM_{10} from Silesia has been modelled by chemical transport model (EMEP). In addition, we examined the potential impacts of elevated levels of other hazardous components that were recorded during LDT episodes of ragweed pollen (Fig. 1S), namely airborne concentrations of fungal spores from two of the most abundant allergenic species (*Alternaria* Nees ex Fr. and *Cladosporium* Link ex. Fr.) (Damialis et al., 2017; Twaroch et al., 2015) and mineral dust from the Sahara Desert (Karanasiou et al., 2012; Schuerger et al., 2018). This is the first time that the large-scale concomitant transport of airborne allergenic pollen, fungal spores, chemical air pollutants and mineral dust has been described.

2. Methods

2.1. Aerobiological data

The monitoring of airborne ragweed pollen grains and *Alternaria* and *Cladosporium* spores was conducted between 2005 and 2015 in Poznań, the biggest city in Western Poland (52°24′14″N, 16°53′20″E) (Fig. 1). This area is known to be free from permanent ragweed populations, and the nearest dense patches of ragweed are located 250 km away (Grewling et al., 2016). In addition, mean daily *Alternaria* spore levels from nine stations located in Hungary have been included to verify whether the Pannonian Plain could be a source area of *Alternaria* spores to Poland (Table 1S, Fig. 1). In both countries aerobiological sampling (sampler type and site selection) was conducted according to the recommendations of the European Aerobiology Society (Galán et al., 2014). In brief: airborne particles were collected by 7-day volumetric traps of the Hirst (1952) design located at roof level. Air containing pollen grains and fungal spores was sucked into the trap (10 l/min) and impacted on the adhesive tape that was later divided into segments corresponding to 24 h periods. Each segment was mounted on a microscope slide, stained with basic fuchsine, and examined by light microscopy (400x). The following counting methods were applied (Mandrioli et al., 1998): fungal spores - 1 longitudinal transect of the slide in Poznań and 12 vertical transects in Hungary; pollen grains - 4 longitudinal transects of the slide in Poznań. Vertical and horizontal counting methods are the most commonly applied methods for the identification of spores and pollen, and produce comparable results (Kapyla and Penttinen, 1981; Cariñanos et al., 2000; Ghabri et al., 2016; Sterling et al., 1999). Daily average (00:00–24:00) ragweed

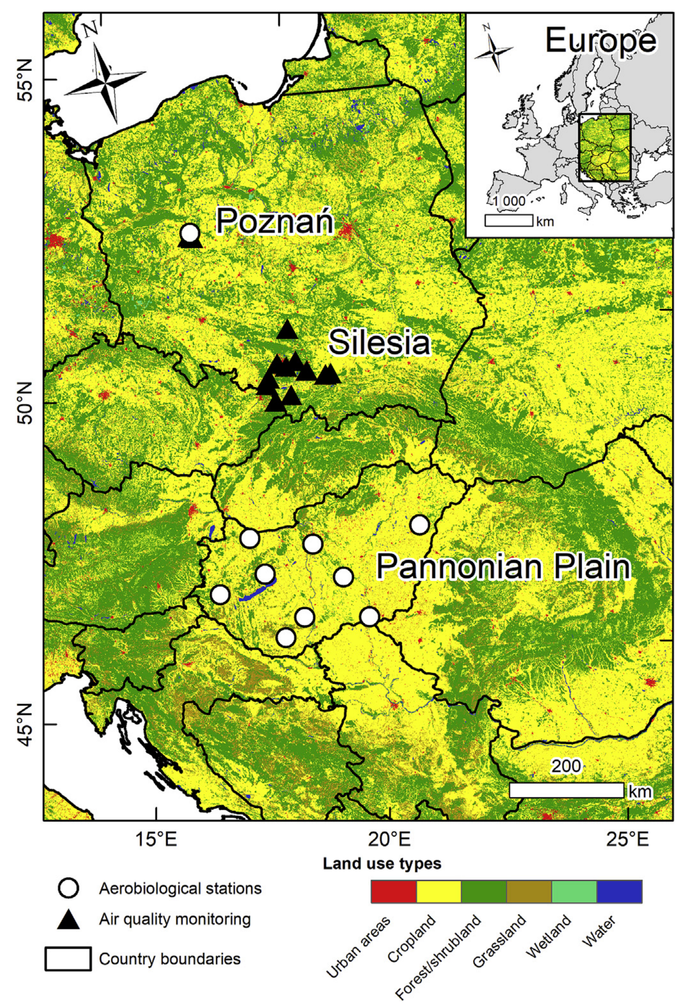

Fig. 1. Location of aerobiological and air quality monitoring stations.

pollen and fungal spore counts were converted into concentrations and expressed as pollen/m^3 and spores/m^3, respectively (Galán et al., 2017).

2.2. Air pollution data

The following mean daily air pollutant levels (2005–2015) have been extracted from the Chief Inspector of Environmental Protection database (www.gios.gov.pl/en/): carbon monoxide (CO), sulphur dioxide (SO_2), nitrogen dioxide (NO_2), and particulate matter (PM_{10}). The air pollution data were collected with hourly resolution (00:00–24:00) from two monitoring stations located in Poznań (receptor area), and nine stations located in Silesia (source area) according to the methods described in Kobus, Iwanek (Kobus et al., 2007) (Table 2S, Fig. 1). However, the data from Silesia were only available from 2014 and so the information is simply used to describe the "spatial background" of pollutant levels in the region. In order to include long-term air pollution data for Southern Poland in the study, we extracted daily air pollution levels from two stations in Kraków (data available from 2006) located around 50 km east from the Upper Silesian Industrial Region (Table 2S, Fig. 1): (1) Nowa Huta (SO_2, CO, NO_2); (2) Aleje Krasińskiego (PM_{10}). The mean monthly pollution levels in September (2006–2015) have been calculated for both Poznań and Kraków. September was chosen as the majority (70.5%) of the LDT episodes with air masses crossing Silesia were recorded in this month.

2.3. Air mass back trajectory analysis

The pathways of air masses containing ragweed pollen recorded in Poznań have been calculated using back trajectory analysis. Back trajectories were computed using a cluster approach employed for numerous airborne pollen studies (e.g. Skjøth, Sommer (Skjøth et al., 2012) and references therein). The Lagrangian Integrated Trajectory model (HYSPLIT) (Draxler et al., 2013; Stein et al., 2015) with the meteorological data originating from version 3.5 of the WRF model (Skamarock et al., 2008) have been used. More details on initial and boundary conditions, as well as physical options and nested domains for the model used in this study are listed in Bilińska et al. (2017). To produce the input files for the HYSPLIT model, the output WRF data at 12-km spatial and 1-h temporal resolution were transformed to ARL format. The approach of using WRF and a 12 km spatial resolution has previously been shown to produce much better results compared to the standard HYSPLIT setup that use the coarser global data set available through the online HYSPLIT webpages (Hernández-Ceballos et al., 2014). HYSPLIT trajectories were calculated 72 h back in time with receiving heights of 500 m, 1000 m and 2000m (de Weger et al., 2016) and for 29 days with mean ragweed pollen level >10 pollen/m^3. Analysis showed that air masses passed through Silesia on 17 days before reaching Poznań (Table 3S). Data from these episodes were entered into statistical analysis.

2.4. Transport of PM_{10} and mineral dust

To check whether PM_{10} originating from Silesia could reach Poznań, the atmospheric transport of PM_{10} during three LDT episodes with the highest mean daily ragweed pollen levels have been modelled; i.e. in 2005, 2006, and 2011 (Table 1). We ran a complex Eulerian chemical transport model EMEP MSC-W. The model has been used by the Meteorological Synthesizing Centre-West (MSC-W) of the European Monitoring and Evaluation Program (EMEP) in support of the Convention on Long Range Transboundary Air Pollution and is one of the key tools within European air pollution policy assessment (Simpson et al., 2012). The EMEP model has previously been used to analyse air pollution at a regional scale in Poland (Werner et al., 2018).

Full details of the EMEP model are given in Simpson, Benedikctow (Simpson et al., 2012). We used EMEP model version 4.10 (Simpson et al., 2015-). The model is coupled offline with meteorology and, in this study, was driven by meteorological parameters from the WRF meteorological model. The model domain was defined on the polar-stereographic projection at a 12 km $\times$ 12 km grid and covers Europe and northern Africa. Anthropogenic emissions of NOx, NH_3, SO_2, primary $PM_{2.5}$ and PM_{10}, CO and NMVOC were included from the TNO MACC III data base at 1/8° x 1/16° spatial resolution (Kuenen et al., 2014). Natural emissions include biogenic emissions calculated internally in the EMEP model as a function of underlying vegetation cover and meteorology, sea salt aerosol emissions, and the import of Saharan dust. Boundary conditions are responsible for the import of Saharan dust, and boundary conditions were based on monthly average dust concentrations for a single year from the global model CTM2 at the University of Oslo. Therefore the representation of the source strength of Saharan dust may vary for individual events. The atmospheric flow and deposition processes transporting the dust and controlling its distribution over Europe are fully represented within the model (Vieno et al., 2016).

For each analysed episode the simulation with EMEP was run twice. In the first simulation (BASE) we used all the emissions sources as described above, whereas in the second simulation (RE, reduced emission) we reduced the emissions of primary PM_{10} for southern Poland (Upper Silesia and Małoposka region) by 15% as in Clappier, Fagerli (Clappier et al., 2017). The BASE simulation was used to analyse spatial and temporal concentrations of air pollution over Europe, with particular focus on Saharan dust contributions to total PM_{10} concentrations over Poland. Both simulations were used for source-receptor analysis (Clappier et al., 2017). For this purpose we calculated the differences in mean daily primary PM_{10} concentrations between the BASE run and the simulations with reduced emission an plotted it on the map. The difference (D) was expressed in percentages as:

$$D = \frac{BASE - RE}{BASE} * 100$$

D is presented in maps and shows the relative contributions of the Upper Silesia and Małopolska region to emissions of primary PM_{10} in the EMEP model domain. Differences were calculated for selected days of high ragweed pollen concentrations observed in Poznań.

2.5. Statistical analysis

The mean daily concentrations of selected air pollutants have been calculated during days with mean daily ragweed pollen levels >10 pollen/m^3 (so called LDT days), and 1, 2, and 3 days before and after LDT days (air pollution levels recorded during corresponding before/after days were averaged). This threshold value (10 pollen/m^3) was based on atmospheric concentrations of ragweed pollen reported to evoke allergic symptoms (Bergmann et al., 2008). Differences between air pollutant levels were analysed by the Kruskal-Wallis H test and Dunn's procedure for multiple pairwise comparison. *P*-values have been adjusted using Benjamini-Hochberg correction. The same methods were applied to determine the difference between air pollution during LDT episodes in Poznań and Silesia. Previous studies investigating LDT episodes of ragweed pollen showed that the air masses need around 1–2 days to travel several hundred kilometres (Kasprzyk et al., 2011; Šikoparija et al., 2013; de Weger et al., 2016) therefore the mean daily

Table 1
The analysis of selected episodes of LDT of ragweed pollen to Poznań (with air masses passing through Silesia) with respect to the presence of particulate matter and desert dust (modelled levels), and *Alternaria* spores (actual levels).

Date	Ragweed pollen level (max. value)	Potential origin of ragweed pollen	PM_{10} from Silesia (max. value)	Mineral dust (max. value)	Potential origin of mineral dust	*Alternaria* spores level (max. value)	Potential origin of *Alternaria* spores
07–10.09.2005	53 (pollen m^{-3})	Pannonian Plain	NO	YES (~5 $\mu g/m^3$)	Sahara desert	YES (246 spore m^{-3})	Pannonian Plain
13–18.09.2006 (I episode)	51 (pollen m^{-3})	Pannonian Plain	YES (~20 $\mu g/m^3$)	NO	–	YES (268 spore m^{-3})	Pannonian Plain
24–27.09.2006 (II episode)	22 (pollen m^{-3})	Pannonian Plain/ Ukraine	YES (~40 $\mu g/m^3$)	YES (~30 $\mu g/m^3$)	Ukrainian steppe	YES (179 spore m^{-3})	Pannonian Plain/ Ukraine
26–27.08.2011	91 (pollen m^{-3})	Pannonian Plain	YES (~50 $\mu g/m^3$)	YES (~20 $\mu g/m^3$)	Sahara desert	YES (311 spore m^{-3})	Pannonian Plain

concentrations on the LDT days and one day before the LDT days were analysed. The mean monthly September (2006–2015) air pollution levels (background values of air pollution) in Poznań and Kraków were compared by Mann-Whitney *U* test ($\alpha = 0.05$). The mean monthly September (2005–2015) *Alternaria* spore levels in Poznań and ten stations in Hungary were analysed by the Kruskal-Wallis H test and Dunn's procedure for multiple pairwise comparison. The statistical analysis has been performed using R statistical software version 3.5.1 (R_Core_Team, 2017).

3. Results

3.1. Co-occurrence of chemical air pollutants and ragweed pollen during LDT episodes

During the selected 17 LDT days, the mean ragweed pollen level in Poznań was significantly higher than 1–3 days before (Fig. 2). The mean daily levels of all investigated air pollutants during LDT days also increased from 3.8% to 104.2% for NO_2 and SO_2, respectively. Statistically significant increases were observed in relation to SO_2 (Chi square = 13.3, $p = 0.004$, df = 3) and PM_{10} (Chi square = 10.0, $p = 0.018$, df = 3). Daily mean temperature also significantly increased (on average by 3.0 °C). Three days after LDT days, air pollution levels returned to background levels (mean monthly September level).

The concentrations of all air pollutants (except of NO_2) were significantly higher in Kraków than Poznań during the 17 LDT days (Fig. 3). Similarly, the mean September pollutant levels (2006–2015) in Kraków were significantly higher than in Poznań for SO_2, PM_{10} and CO ($p < 0.0001$). The mean September (2014–2016) SO_2 and PM_{10} concentrations in most of the cities in Silesia (10/10 and 8/10 sites, respectively) were also higher than in Poznań (Fig. 2S). In contrast, the mean monthly CO and NO_2 concentrations in Poznań did not differ markedly from the pollutants concentrations in Silesian stations (Fig. 2S).

During the two episodes in 2006 and the episode in 2011 the source-receptor analysis shows that emission sources from Upper Silesia and Małopolska influenced PM_{10} concentrations over the Poznań area, (Fig. 4). For the first episode in 2006, it can be seen that the influence of the Upper Silesia and Małopolska region reaches as far as the Baltic Sea and Scandinavia. A fifteen percent reduction in primary PM_{10} emissions over Upper Silesia and Małopolska caused a decrease in primary PM_{10} concentrations of up to 5–10% over central Poland and 3–4% over the Poznań area. For the second episode in 2006 and the episode in 2011, the decrease in primary PM_{10} concentrations is in the range of 1–2% over Poznań. The situation is different for 2005, as influence of the Upper Silesia and Małopolska region is to the north and north-east and there is very little or no influence of emissions from this region on the Poznań area.

3.2. Co-occurrence of fungal spores and ragweed pollen during LDT episodes

The mean daily concentrations of *Cladosporium* and *Alternaria* spores during LDT days were higher than the 1–3 days before/after LDT days (Fig. 2). Significant increases were observed with respect to *Alternaria* spores (Chi square = 19.5, $p = 0.0002$, df = 3). Levels of *Cladosporium* and *Alternaria* spores returned to background levels within two days of the LDT episodes (background relative to mean monthly September concentrations). Daily variations in airborne concentrations of *Alternaria* spores and *Ambrosia* pollen showed similar patterns during the most intense LDT episodes (Fig. 5). Mean monthly September levels (2005–2015) of *Alternaria* spores were significantly higher in all of the selected Hungarian stations than in Poznań ($p < 0.05$) (Fig. 6).

3.3. Co-occurrence of mineral dust and ragweed pollen during LDT episodes

The increased levels of mineral dust were calculated by the EMEP model in Poznań during LDT episodes of ragweed pollen (Fig. 4, Table 1). The concentration of mineral dust particles exceeded 30 and 20 $\mu g/m^3$ in 2006 (24–27 September) and 2011 (26–27 August), respectively. In 2006 the total PM_{10} and mineral dust concentrations were similarly high, while in 2011 the amount of PM_{10} was twice the level of mineral dust. Lower concentrations of mineral dust particles (<5 $\mu g/m^3$) were recorded in 2005. The presence of desert dust was observed in air masses arriving from the South (2005, 2011) and Southeast (2006 II episode). Back trajectory analysis revealed the mineral particles may have originated from the Mediterranean Basin in 2005 and 2011, and from Eastern Ukraine in 2006 (Fig. 7).

4. Discussion

4.1. Co-occurrence of chemical air pollutants and ragweed pollen during LDT episodes

The northward progression of air masses transports ragweed pollen grains long distances through the Moravian Gate into Poland (Šikoparija et al., 2013; Stępalska et al., 2017). In this study, we have shown that the same conditions required for the LDT of ragweed pollen also result in elevated levels of air pollution in Poznań, Western Poland. The most striking increase was observed in relation to SO_2 and PM_{10} (their concentrations increased by 104% and 37%, respectively).

Before reaching Poznań the air masses passed through Southern Poland (Upper Silesia region), where air pollution levels are markedly higher than in Western Poland. This region may therefore be considered as a source of air pollutants transported to Poznań. Indeed, the Silesia province has previously been identified as a source area of air pollutants (including PM_{10} and SO_2) for

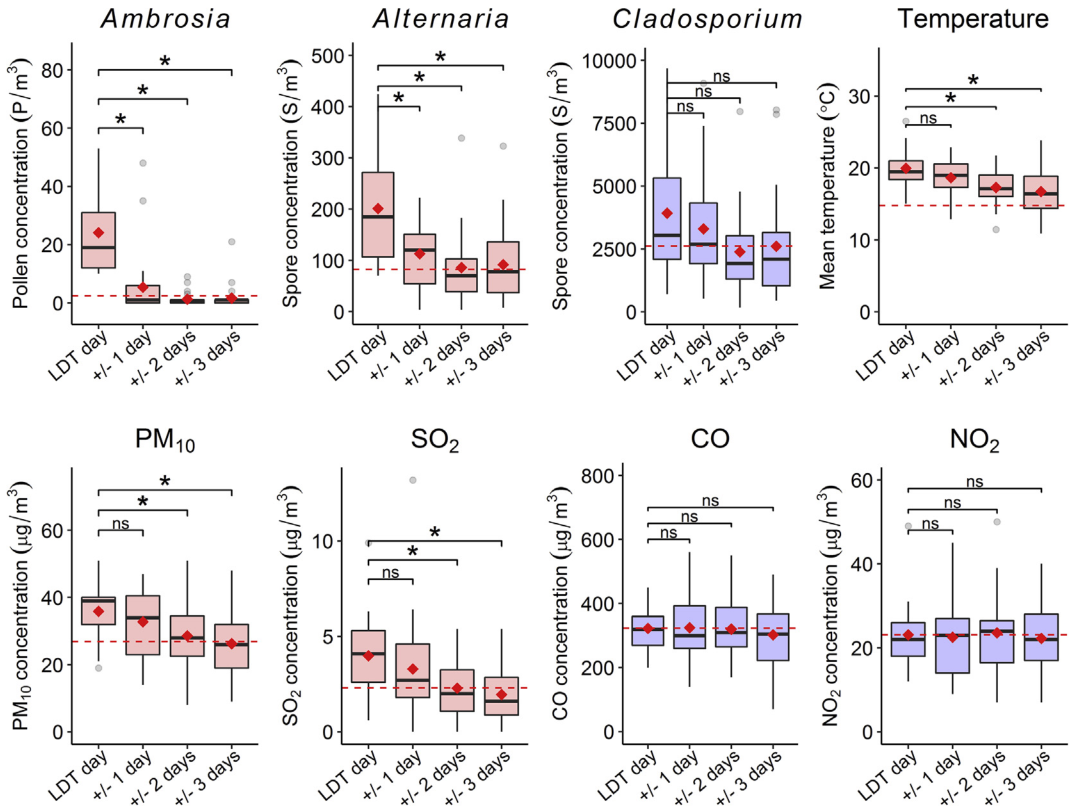

Fig. 2. Comparison of mean daily concentrations of biological and chemical air pollutants and temperature recorded during LDT day and 1,2, 3 days before/after LDT day in Poznań (Kruskal-Wallis and Dunn's post-hoc test). Red color – significantly higher particles level during LDT, blue – lack of significant differences in particle concentration, dotted line – mean monthly September level, "*ns*" – not significant, "*" – significant difference ($p < 0.05$). The grey dots represent outliers.

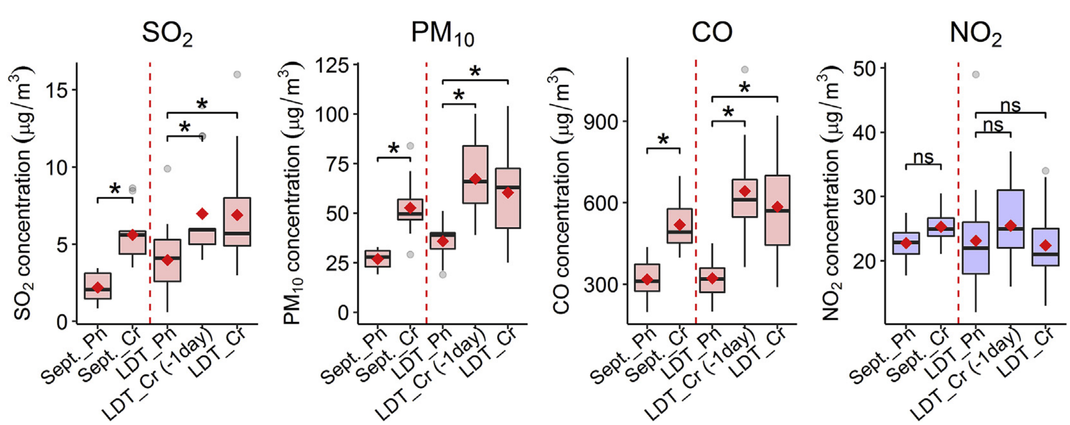

Fig. 3. Comparison of mean monthly September levels of air pollutants in Poznań (Sept._Pn) and Silesia (Sept._Cr) and mean daily air pollution level during LDT day (in Poznań) and LDT day and one day before (in Silesia). Red color – significantly higher particles level during LDT, blue – lack of significant differences in particle concentration, "*ns*" – not significant, "*" – significant difference ($p < 0.05$). The grey dots represent outliers.

neighbouring regions such as Northern Poland (~400 km away) (Reizer and Orza, 2018) and the Czech Republic to the south (~70 km) (Buzek et al., 2017; Kozakova et al., 2019; Černikovský et al., 2016). Our observations were further supported by the EMEP model, which showed that considerable levels of PM_{10} released over Silesia can travel several hundreds of kilometres to Northern Poland and Scandinavia. The exception is 2005, for which the EMEP model shows that the Upper Silesia region contributes very little to primary PM_{10} concentrations recorded in Poznań. Furthermore, decreases (~20–30%) seen in SO_2 and PM_{10} levels recorded in Kraków after LDT of ragweed pollen (Fig. S3) supports our theory that the northward movement of air masses takes polluted air away from the south of Poland and transports it to the north.

During the analysed LDT episodes of ragweed pollen to Poznań the average increase in PM_{10} was 9.6 $\mu g/m^3$, with the highest PM_{10} increases recorded in 2006 and 2009 (21.0 $\mu g/m^3$ and 19.0 $\mu g/m^3$, respectively). According to a WHO report on the health effects of particulate matter, an increase of PM_{10} by 10 $\mu g/m^3$ results in increases in 'all-cause daily mortality' by 0.2–0.6% (WHO, 2013). Similarly, several studies showed that even low SO_2 concentrations (5–20 $\mu g/m^3$) might negatively affect human health (Burnett et al.,

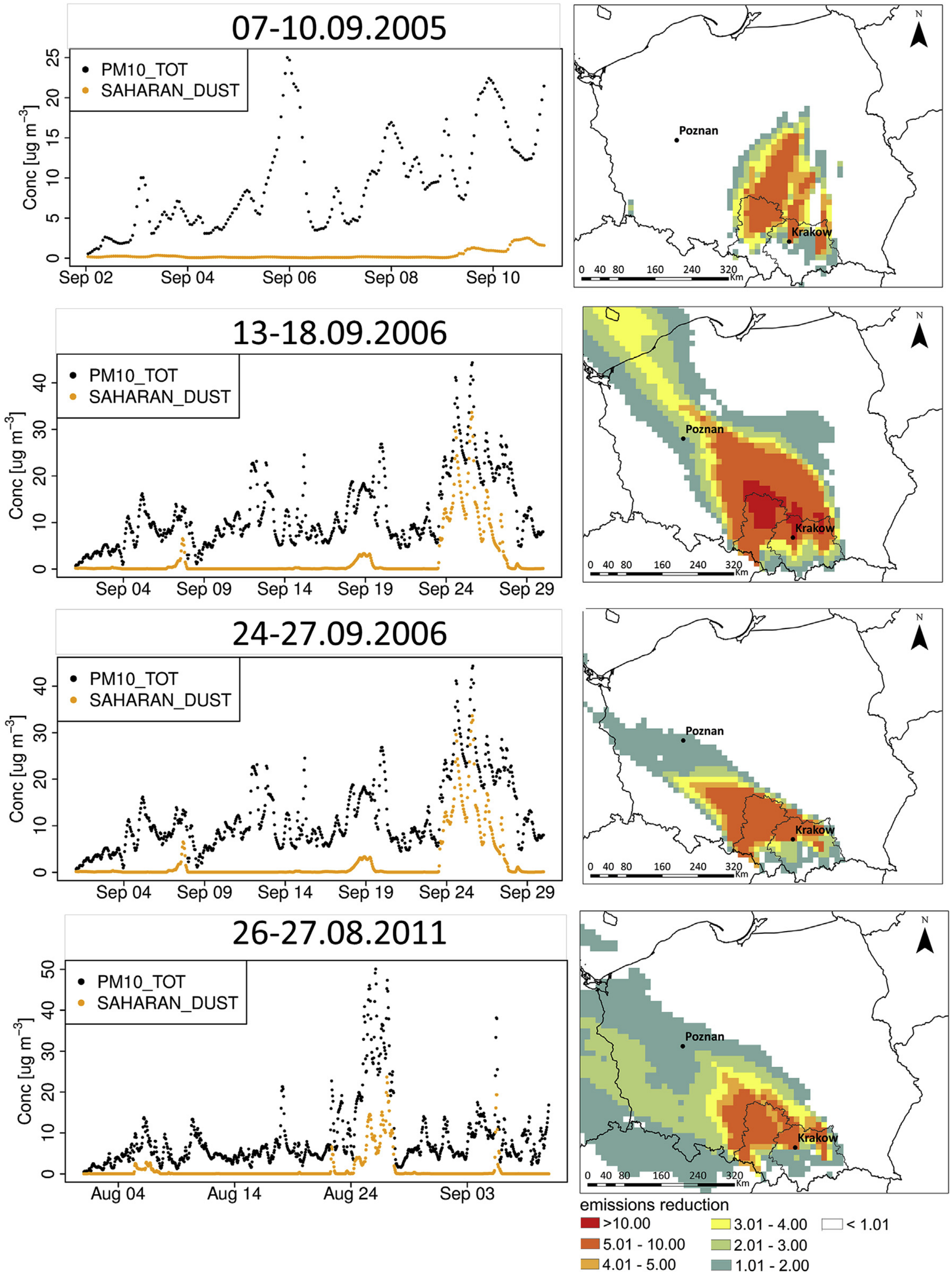

Fig. 4. Daily concentrations of total PM_{10} and desert dust in Poznań (left) and the atmospheric movement of PM_{10} originating from Silesia calculated by chemical transport model EMEP MSC-W (right) during the most intensive LDT episodes of ragweed pollen.

2004; Pope et al., 2002; Chen et al., 2012). The mean SO_2 level during investigated LDT episodes was rather low in Poznań (less than 5 μg/m^3) but, due to the synergistic interactions between air pollutants and pollen grains (Schiavoni et al., 2017), the adverse effects of "multi-pollutant mixture" on human health should be considered. For instance, the sales of antihistamines was higher when concomitantly high birch pollen and high air pollution was recorded than situations with high birch pollen alone (Grundström et al., 2017).

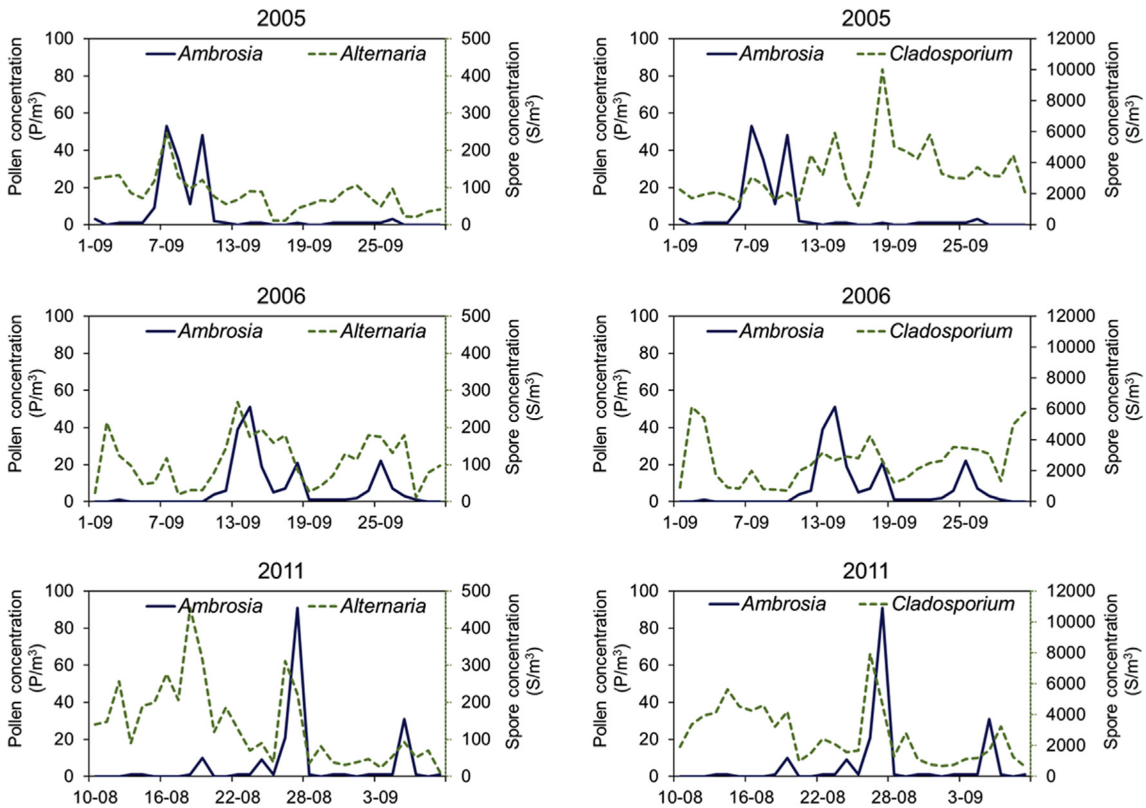

Fig. 5. Daily concentrations of ragweed pollen and *Alternaria* and *Cladosporium* spores in Poznań during the most intensive LDT episodes of ragweed pollen (see Methods 2.4).

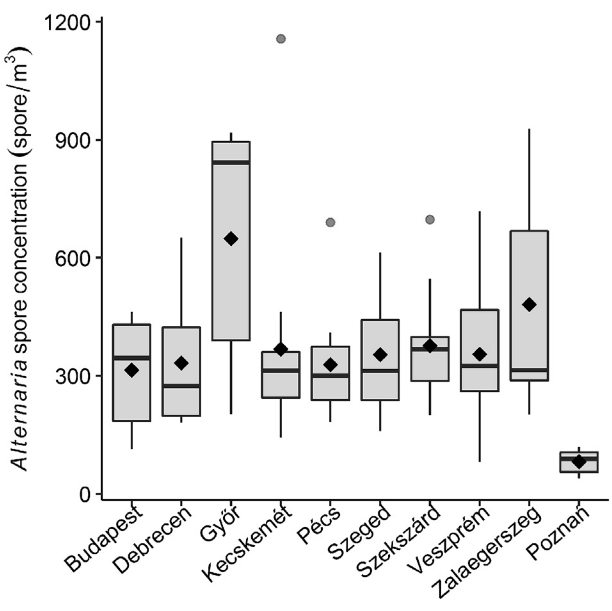

Fig. 6. Comparison of mean monthly September level of *Alternaria* spores concentration between Poznań and nine Hungarian cities (potential source region of spores). In every Hungarian city the *Alternaria* concentration was significantly higher than in Poznań ($p < 0.05$). The grey dots represent outliers.

4.2. Co-occurrence of fungal spores and ragweed pollen during LDT episodes

Among the two investigated fungal species, the atmospheric behaviour of *Alternaria* spores showed very similar patterns to LDT ragweed pollen. The mean daily airborne concentration of *Alternaria* spores during LDT days were significantly higher (~115%, $p < 0.05$) than during the 2–3 days before/after ragweed pollen peak days. Levels of *Cladosporium* spores did not show such a strong increase, although mean daily atmospheric *Cladosporium* spore concentrations were also higher (up to 43%) when ragweed pollen arrived over Poznań. *Alternaria* spores are well adapted for transport in large numbers over long distances. For instance, the sources of *Alternaria* spores recorded in Worcester, UK (Sadyś et al., 2015) and Badajoz, Spain (Fernandez-Rodriguez et al., 2015) were found to come from 10s or 100s of kilometers from the traps. Furthermore, high numbers of *Alternaria* spores have been found in samples collected at elevations over 1000 m a.s.l. (Heise and Heise, 1948), and the tropospheric transport of *Alternaria* spores from Eastern Asia to North America has also been reported (Smith et al., 2012).

Our study revealed that the concentration of *Alternaria* spores was significantly higher (up to 8-times) in Hungary than in Poznań, suggesting that the Pannonian Plain might be an important source of airborne *Alternaria* spores to Poland. In a European wide study it has been shown that the highest mean levels of *Alternaria* spores were recorded in the Pannonian Plain, reflecting its agricultural nature (Skjøth et al., 2016). It is worth mentioning that ragweed and *Alternaria* are associated with the same type of habitats (one as a crop weed, the other as a crop pathogen) and they both have similar release mechanisms and phenology. For instance, there are nine known *Alternaria* species reported to be associated with sunflower leaf blight worldwide (Wang et al., 2014), and the infestation of sunflower fields with ragweed is considered a serious weed problem interfering with the sustainability of sunflower production (Ozaslan et al., 2016). In addition, *Alternaria* spores are “dry-air spores” and release occurs during conditions of high

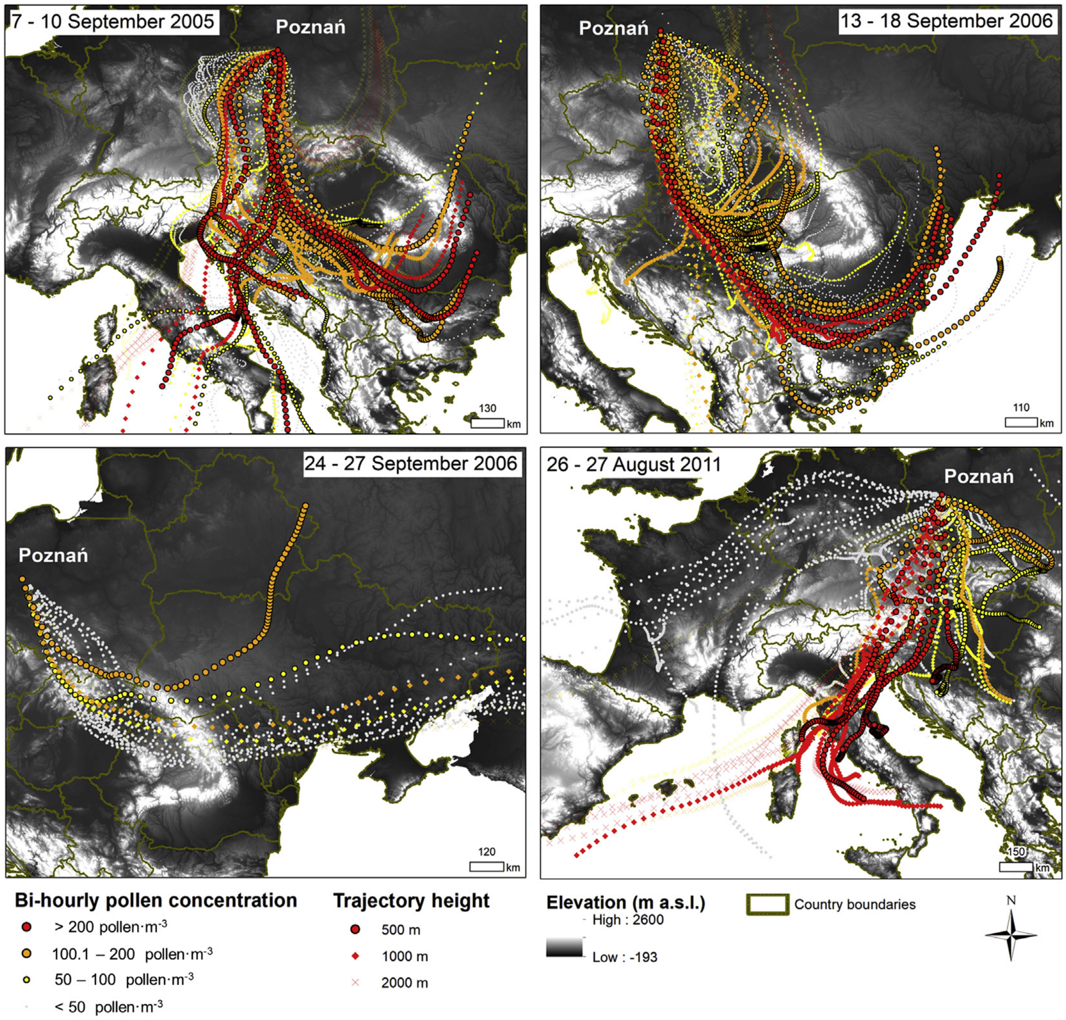

Fig. 7. Back trajectory analysis of air masses arriving to Poznań during the most intensive LDT episodes of ragweed pollen.

temperature, low humidity, and high wind speeds (Troutt and Levetin, 2001). These factors also promote the release of ragweed pollen from anthers (Bianchi et al., 1959). Gusty winds and high temperatures were shown to be crucial factors in LDT mechanism of ragweed pollen from the Pannonian Plain to Northern Europe (Šikoparija et al., 2013), and it is possible that ragweed pollen and fungal spores could be released simultaneously as one common plume.

In 2006, the concomitant increase in pollen and spores were recorded in Poznań when air masses were arriving from a south-eastern direction. As the airborne concentrations of *Alternaria* spores in September can be two to three times higher in Ukraine than in Poznań we suspect that also this region might be an additional source of *Alternaria* spores to Poland (Kasprzyk et al., 2015). However, we cannot exclude the possibility that the increase in spores was due to release from sources closer to Poznań because the LDT episodes were associated with increased air temperatures in Central Poland (up to 3–4 °C higher).

4.3. Co-occurrence of desert dust and ragweed pollen during LDT episodes

The EMEP model simulation showed a sudden increase in the atmospheric concentrations of mineral dust particles in Poznań between 26–27 August 2011, i.e. during one of the most intensive LDT episodes of ragweed pollen. Air mass trajectory analysis revealed that the dust originating from southern Europe, presumably caused by an intrusion of Saharan dust from North Africa. The Sahara Desert has traditionally been viewed as the largest source region of remotely transported mineral dust in Europe (Krasnov et al., 2016; Birmili et al., 2008; Athanasopoulou et al., 2016; Middleton, 2017). The advection of Saharan dust to Northern and Central Europe can occur several times a year with concentrations reaching 280μg/m^3 (Birmili et al., 2008; Ansmann et al., 2003; Mattsson and Martensson, 1994; Barkan et al., 2005). Saharan dust generally enters Europe via stable lofted aerosol layers (Birmili et al., 2008). The thickness of the dust layer varies from a few hundred to several thousand meters, and the main layer is located above the Planetary Boundary Layer (PBL) up to an altitude of 3–5 km (Ansmann et al., 2003; Papyannis et al., 2008). When the PBL depth attains its highest value (during hot sunny days) there are cases of dust intrusion inside the PBL leading to abrupt increases of aerosol concentration especially for southern Europe (Papyannis et al., 2008). Hot and dry weather on the Pannonian Plain aids the release of ragweed pollen during the flowering season and results in the PBL realising depths of several thousand meters during the day (Smith et al., 2008; Šikoparija et al., 2013). Released ragweed pollen grains are then transported up into the atmosphere reaching high concentrations at altitudes greater than 1000 m (Smith et al., 2008). During such conditions, desert dust mixed with ragweed pollen may intrude deep inside the PBL over the Pannonian Plain before being transported by air masses northwards.

However, another potential source area of desert dust should also be considered. The EMEP simulation for the second LDT episode recorded on 24–27 September 2006 showed a distinct increase in desert dust in Poznań (almost as high as the total level of PM_{10}). Interestingly, back trajectory analysis revealed that the air masses arrived from a south-eastern direction, particularly from Ukraine. The transport mechanism of desert dust originated from Ukraine has been comprehensively described by Birmili and Schepanski (Birmili et al., 2008) (based on an episode in May 2007). It was shown that the dust was emitted from vast areas of agricultural soil over the eastern and southern parts of Ukraine (over an area of 220 000 km^2) when surface wind speeds were high (20 m/s). It was suggested that due to the intensive agricultural development, the soil has become prone to wind erosion. As a result, the hourly concentrations of PM_{10} transported over Poland and Slovakia reached 1000 $\mu g/m^3$ (Athanasopoulou et al., 2016). The ragweed plants cover dense areas of Ukraine (Prank et al., 2013; Afonin et al., 2018) and the south-eastern part of the country is known to be a major source or airborne ragweed pollen for Western Europe (Kasprzyk et al., 2011; de Weger et al., 2016). Hot weather and strong winds favour both the release of ragweed pollen and the erosion of land creating mixed composition of mineral and biological materials and so it is presumed that both the airborne ragweed pollen and mineral dust recorded in Poznań in 2006 originated from the Ukraine.

5. Conclusions

Episodes of long-distance transported ragweed pollen to Northern Europe are often associated with elevated levels of anthropogenic and natural air pollutants, which may increase atmospheric concentrations by 100% within days. The action of high temperature and gusty winds favour the release of ragweed pollen, fungal spores and mineral dust (e.g. from the Ukrainian steppe) facilitating their concomitant occurrence and transport in the air. Furthermore, air masses with desert dust originating from the Sahara may intrude deep inside the PBL over the Pannonian Plain and mix with released ragweed pollen. Anthropogenic air pollutants, particularly SO_2 and PM_{10}, are gathered when air masses arrived over the highly polluted Silesia region. We suggest that the LDT of ragweed pollen from the Pannonian Plain to the North is not a simple “one-component” phenomenon but is often related to the simultaneous occurrence of various air pollutants, including chemical air pollutants and other biotic and abiotic components (fungal spores and desert dust). Synergistic interactions between aeroallergens and man-made air pollutants could change their physiologic and allergenic properties and act as an adjuvant promoting allergic disease. The impact of “multi-pollutant mixture” on human health should therefore be investigated further.

Conflicts of interest

The authors report no conflicts of interest. The authors alone are responsible for the content and the writing of the paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2019.07.116.

References

Afonin, A.N., et al., 2018. History of introduction and distribution of common ragweed (Ambrosia artemisiifolia L.) in the European part of the Russian Federation and in the Ukraine. EPPO Bull. 48 (2), 266–273.
EEA, 2014. In: Agency, E.E. (Ed.), Air Quality in Europe - 2014 Report. Publications Office of the European Union, Luxembourg.
Ansmann, A., et al., 2003. Long-range transport of Saharan dust to northern Europe: the 11–16 October 2001 outbreak observed with EARLINET. J. Geophys. Res. Atmos. 108 (D24).
Arbes, S.J.J., et al., 2005. Prevalences of positive skin test responses to 10 common allergens in the US population: results from the third National Health and Nutrition Examination Survey. J. Allergy Clin. Immunol. 116 (2), 377–383.
Athanasopoulou, E., et al., 2016. Long-range transport of Saharan dust and chemical transformations over the Eastern Mediterranean. Atmos. Environ. 140, 592–604.
Barkan, J., et al., 2005. Synoptics of dust transportation days from Africa towards Italy and central Europe. J. Geophys. Res. 110 (D07208).
Behrendt, H., et al., 1997. Air pollution and allergy: experimental studies on modulation of allergen release from pollen by air pollutants. Int. Arch. Allergy Immunol. 113 (1–3), 69–74.
Bergmann, K.C., et al., 2008. The threshold value for number of ambrosia pollen induced acute nasal reactions is very low. Allergo J. Int. 17, 375–376.
Bianchi, D.E., Schwemmin, D.J., Wagner Jr., W.H., 1959. Pollen release in the common ragweed (Ambrosia artemisiifolia). Int. J. Plant Sci. 120 (4), 235–243.
Bilińska, D., et al., 2017. Source regions of ragweed pollen arriving in south-western Poland and the influence of meteorological data on the HYSPLIT model results. Aerobiologia 33 (3), 315–326.
Birmili, W., et al., 2008. A case of extreme particulate matter concentrations over Central Europe caused by dust emitted over the southern Ukraine. Atmos. Chem. Phys. 8, 997–1016.
Bokwa, A., 2008. Environmental impacts of long-term air pollution changes in Kraków, Poland. Pol. J. Environ. Stud. 17 (5), 673–686.
Burbach, G.J., et al., 2009. GA2LEN skin test study II: clinical relevance of inhalant allergen sensitizations in Europe. Allergy 64, 1507–1515.
Burnett, R.T., et al., 2004. Associations between short-term changes in nitrogen dioxide and mortality in Canadian cities. Arch. Environ. Health 59 (5), 228–236.
Buzek, F., et al., 2017. Isotope composition of NH3, NOx and SO2 air pollution in the Moravia-Silesian region, Czech Republic. Atmos. Pollut. Res. 8, 221–232.
Cariñanos, P., et al., 2000. Comparison of two counting methods of slides from a hirst type volumetric trap. Aerobiologia 16, 339–346.
Cecchi, L., et al., 2006. Long distance transport of ragweed pollen as a potential cause of allergy in central Italy. Ann. Allergy Asthma Immunol. 96 (1), 86–91.
Cecchi, L., et al., 2010. Long-distance transport of ragweed pollen does not induce new sensitizations in the short term. Aerobiologia 26 (4), 351–352.
Celenk, S., Malyer, H., 2017. The occurrence of Ambrosia pollen in the atmosphere of Northwest Turkey: investigation of possible source regions. Int. J. Biometeorol. 61 (8), 1499–1510.
Černikovský, L., et al., 2016. Transboundary air-pollution transport in the Czech-polish border region between the cities of Ostrava and Katowice. Cent. Eur. J. Publ. Health 24 (Suppl. S45-S50).
Chen, R., et al., 2012. Short-term exposure to sulfur dioxide and daily mortality in 17 Chinese cities: the China air pollution and health effects study (CAPES). Environ. Res. 118, 101–106.
Choi, Y.-J., et al., 2018. Chamber and field studies demonstrate differential Amb a 1 contents in common ragweed depending on CO2 levels. Allergy Asthma Immun. 10 (3), 278–282.
Clappier, A., Fagerli, H., Thunis, P., 2017. Screening of the EMEP source receptor relationships: application to five European countries. Air Qual Atmos Health 10, 497–507.
Damialis, A., et al., 2017. Estimating the abundance of airborne pollen and fungal spores at variable elevations using an aircraft: how high can they fly? Sci. Rep. UK 7 (44535).
de Weger, L.A., et al., 2016. The long distance transport of airborne Ambrosia pollen to the UK and The Netherlands from Central and south Europe. Int. J. Biometeorol. 60 (12), 1829–1839.
Draxler, R., et al.. Hysplit4 Users Guide. http://www.arl.noaa.gov/documents/reports/hysplit_user_guide.pdf.209pp.2013.
Fernandez-Llamazares, A., et al., 2012. Ambrosia L. in Catalonia (NE Spain): expansion and aerobiology of a new bioinvader. Aerobiologia 28, 435–451.
Fernandez-Rodriguez, S., et al., 2015. Potential sources of airborne Alternaria spp. spores in South-west Spain. Sci. Total Environ. 15 (533), 165–176.
Galán, C., et al., 2014. Pollen monitoring: minimum requirements and reproducibility of analysis. Aerobiologia 30, 385–395.
Galán, C., et al., 2017. Recommended terminology for aerobiological studies.

Aerobiologia 33, 293–295.
Ghabri, D., et al., 2016. Comparison between the counting methods used by two aerobiology networks in southern Europe (Spain and Italy). Aerobiologia 33 (1), 87–92.
Grewling, Ł., et al., 2016. Mesoscale atmospheric transport of ragweed pollen allergens from infected to uninfected areas. Int. J. Biometeorol. 60 (10), 1493–1500.
Grundström, M., et al., 2017. The relationship between birch pollen, air pollution and weather types and their effect on antihistamine purchase in two Swedish cities. Aerobiologia 33 (4), 457–471.
Hamaoui-Laguel, L., et al., 2015. Effects of climate change and seed dispersal on airborne ragweed pollen loads in Europe. Nat. Clim. Chang. 5, 766–771.
Heise, H.A., Heise, E.R., 1948. The distribution of ragweed pollen and Alternaria spores in the upper atmosphere. J. Allergy 19 (6), 403–407.
Hernández-Ceballos, M.A., et al., 2014. Improvement in the accuracy of back trajectories using WRF to identify pollen sources in southern Iberian Peninsula. Int. J. Biometeorol. 58, 2031–2043.
Hirst, J.M., 1952. An automatic volumetric spore trap. Ann. Appl. Biol. 39, 257–265.
Kapyla, M., Penttinen, A., 1981. An evaluation of the microscopical counting methods of the tape in Hirst-Burkard pollen and spore trap. Grana 20, 131–141.
Karanasiou, A., et al., 2012. Health effects from Sahara dust episodes in Europe: literature review and research gaps. Environ. Int. 47, 107–114.
Kasprzyk, I., et al., 2011. The occurrence of Ambrosia pollen in Rzeszów, Kraków and Poznań, Poland: investigation of trends and possible transport of Ambrosia pollen from Ukraine. Int. J. Biometeorol. 55 (4), 633–644.
Kasprzyk, I., et al., 2015. Air pollution by allergenic spores of the genus Alternaria in the air of central and eastern Europe. Environ. Sci. Pollut. Res. 22, 9260–9274.
Kobus, D., et al., 2007. Organization of Air Quality Monitoring Data Collection in Poland. Review of Available Air Quality Data. Institute of Environmental Protection, Warsaw.
Kobza, J., Geremek, M., Dul, L., 2018. Characteristics of air quality and sources affecting high levels of PM10 and PM2.5 in Poland, Upper Silesia urban area. Environ. Monit. Assess. 190, 515.
Konishi, S., et al., 2014. Particulate matter modifies the association between airborne pollen and daily medical consultations for pollinosis in Tokyo. Sci. Total Environ. 499, 125–132.
Kozakova, J., et al., 2019. The influence of local emissions and regional air pollution transport on a European air pollution hot spot. Environ. Sci. Pollut. Res. Int. 26 (2), 1675–1692.
Krasnov, H., Katra, I., Friger, M., 2016. Increase in dust storm related PM10 concentrations: a time series analysis of 2001-2015. Environ. Pollut. 213, 36–42.
Kuenen, J.J.P., et al., 2014. TNO-MACC_II emission inventory: a multi-year (2003–2009) consistent high-resolution European emission inventory for air quality modelling. Atmos. Chem. Phys. 14, 5837–5869.
Lake, I.R., et al., 2017. Climate change and future pollen allergy in Europe. Environ. Health Perspect. 125 (3), 385–391.
Leśniok, M., Małarzewski, Ł., Niedźwiedź, T., 2010. Classification of circulation types for Southern Poland with an application to air pollution concentration in Upper Silesia. Phys. Chem. Earth 35, 516–522.
Lou, H., et al., 2017. Sensitization patterns and minimum screening panels for aeroallergens in self-reported allergic rhinitis in China. Sci. Rep. UK 7, 9286.
Mandrioli, P., Comtois, P., Levizzani, V., 1998. Methods in Aerobiology. Pitagora Editrice, Bologna.
Mattsson, J.O., Martensson, U., 1994. Yellow snow over the Alps and subarctic from dust storm in Africa. March 1991. AMBIO 23 (3), 233–235.
Middleton, N.J., 2017. Desert dust hazards: a global review. Aeolian Res. 24, 53–63.
WHO, Health effects of particulate matters, 2013. In: Organisation, W.H. (Ed.), Policy Implications for Countries in Eastern Europe, Caucasus and Central Asia. WHO Regional Office for Europe, Copenhagen, Denmark.
Oswalt, M.L., Marshall, G.D., 2008. Ragweed as an example of worldwide allergen expansion. Allergy Asthma Clin. Immunol. 4, 130–135.
Ozaslan, C., et al., 2016. Common ragweed: an emerging threat for sunflower production and human health in Turkey. Weed Biol. Manag. 16 (1), 42–55.
Papyannis, A., et al., 2008. Systematic lidar observations of Saharan dust over Europe in the frame of EARLINET (2000–2002). J. Geophys. Res. Atmos. (D10204), 113.
Pope III, C.A., et al., 2002. Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. JAMA 287 (9), 1132–1141.
Prank, M., et al., 2013. An operational model for forecasting ragweed pollen release and dispersion in Europe. Agric. For. Meteorol. 182–183, 43–53.
R_Core_Team, R., 2017. A Language and Environment for Statistical Computing. *R Foundation for Statistical Computing*, Vienna, Austria.
Rasmussen, K., et al., 2017. Climate-change-induced range shifts of three allergenic ragweeds (Ambrosia L.) in Europe and their potential impact on human health. PeerJ 5, e3104.
Reizer, M., Orza, J.A.G., 2018. Identification of PM10 air pollution origins at a rural background site. In: ES3 Web of Conferences. Air Protection in Theory and Practice, vol. 28, 01031.
Sadyś, M., Skjøth, C.A., Kennedy, R., 2015. Determination of Alternaria spp. habitats using 7-day volumetric spore trap, Hybrid Single Particle Lagrangian Integrated Trajectory model and geographic information system. Urban Clim. 14 (3), 429–440.
Schiavoni, G., D'Amato, G., Afferni, C., 2017. The dangerous liaison between pollens and pollution in respiratory allergy. Ann. Allergy Asthma Immunol. 118 (3), 269–275.
Schuerger, A.C., et al., 2018. Science questions and knowledge gaps to study microbial transport and survival in Asian and African dust plumes reaching North America. Aerobiologia 34 (4), 425–435.
Šikoparija, B., et al., 2013. A mechanism for long distance transport of Ambrosia pollen from the Pannonian Plain. Agric. For. Meteorol. 180, 112–117.
Simpson, D., et al., 2012. The EMEP MSC-W chemical transport model – technical description. Atmos. Chem. Phys. 12, 7825–7865.
Simpson, D., et al., 2015–2016. Transboundary particulate matter, photo-oxidants, acidifying and eutrophying components. In: Updates to the EMEP/MSC-W Model *EMEP Status Report 1/2015*2016. The Norwegian Meteorological Institute, Oslo, Norway, pp. 133–139.
Skamarock, W.C., et al., 2008. A Description of the Advanced Research WRF Version 3, Technical Report TN-475+STR. NCAR.
Skjøth, C.A., et al., 2012. Crop harvest in Denmark and Central Europe contributes to the local load of airborne Alternaria spore concentrations in Copenhagen. Atmos. Chem. Phys. 12, p. 11107e11123.
Skjøth, C.A., et al., 2016. Alternaria spores in the air across Europe: abundance, seasonality and relationships with climate, meteorology and local environment. Aerobiologia 32, 3–22.
Smith, M., et al., 2008. Long-range transport of Ambrosia pollen to Poland. Agric. For. Meteorol. 148 (10), 1402–1411.
Smith, D.J., et al., 2012. Free tropospheric transport of microorganisms from Asia to North America. Microb. Ecol. 64 (4), 973–985.
Smith, M., et al., 2013. Common ragweed: a threat to environmental health in Europe. Environ. Int. 61 (0), 115–126.
Stach, A., et al., 2007. Examining Ambrosia pollen episodes at Poznan (Poland) using back-trajectory analysis. Int. J. Biometeorol. 51, 275–286.
Stein, A.F., et al., 2015. NOAA'S HYSPLIT atmospheric transport and dispersion modeling system. Bull. Am. Meteorol. Soc. 96 (12), 2059–2078.
Stępalska, D., et al., 2017. Co-occurrence of Artemisia and Ambrosia pollen seasons against the background of the synoptic situations in Poland. Int. J. Biometeorol. 61 (4), 747–760.
Sterling, M., Rogers, C., Levetin, E., 1999. An evaluation of two methods used for microscopic analysis of airborne fungal spore concentrations from the Burkard Spore Trap. Aerobiologia 15, 9–18.
Troutt, C., Levetin, E., 2001. Correlation of spring spore concentrations and meteorological conditions in Tulsa, Oklahoma. Int. J. Biometeorol. 45, 64–74.
Twaroch, T., et al., 2015. Mold allergens in respiratory allergy: from structure to therapy. Allergy Asthma Immun. 7 (7), 205–220.
Vieno, M., et al., 2016. The UK particulate matter air pollution episode of March–April 2014: more than Saharan dust. Environ. Res. Lett. 11, 044004.
Wang, T., et al., 2014. Characterization of Alternaria species associated with leaf blight of sunflower in China. Eur. J. Plant Pathol. 140 (2), 301–315.
Wang, X., et al., 2017. Allergic sensitization feature under high pollen exposure in grassland of China. Biomed. Res. 28 (20), 9070–9074.
Werner, M., Kryza, M., Wind, P., 2018. High resolution application of the EMEP MSC-W model over Eastern Europe – analysis of the EMEP4PL results. Atmos. Res. 212, 6–22.
Ziska, L.H., et al., 2011. Recent warming by latitude associated with increased length of ragweed pollen season in central North America. P Natl. Acad. Sci. USA 108 (10), 4248–4251.