Summary of Professional Achievements

1. **Name:** Wioletta Rogula-Kozłowska

2. **Diplomas and scientific degrees:**

a)   Doctor of Technical Sciences (Doktor inżynier), 2009
       Silesian University of Technology in Gliwice,
       Faculty of Energy and Environmental Engineering
       Title of PhD Thesis: "Properties of Particulate Aerosol from Road Transport"
       Supervisor: Prof. Józef S. Pastuszka
       Reviewers: Prof. Jan Konieczyński and Prof. Jerzy Zwoździak

b)   Master of Sciences (Magister inżynier), 2003
       Silesian University of Technology in Gliwice
       Faculty of Energy and Environmental Engineering

3. **Information on previous employment:**

a) 2009 – present—adjunct, Department of Air protection of the Institute of Environmental Engineering, Polish Academy of Sciences, Zabrze

b) 2008 – 2010—assistant, Department of Air Protection of the Silesian University of Technology, Gliwice

c) 2005 – 2009—assistant, Department of Air Protection of the Institute of Environmental Engineering, Polish Academy of Sciences, Zabrze

d) 2003 – 2005—technical position, Department of Air Protection of the Institute of Environmental Engineering, Polish Academy of Sciences, Zabrze

4. My research achievement (publications), contributing significantly to environmental engineering, after receiving my PhD degree (according to the Art. 16(2) of the Act of 14 March 2003, Law on scientific degrees and titles and degrees and titles in the arts, Official Journal of Laws no. 65, item 595, as amended):

a) Title

Ambient particulate matter in selected urban and non-urban localities in Poland: Its origin oriented chemical characterization.

b) List of publications:

b1 **Rogula-Kozłowska W.**, Klejnowski K., Krasa A., Szopa S.: Concentration and elemental composition of atmospheric fine particles in Silesia Province, Poland. [In] Environmental Engineering III, Pawłowski L., Dudzińska M.R. & Pawłowski A. (eds.), Taylor & Francis Group, London, 2010, 75-81.

I designed a part of the research and measurements, performed a great part of the measurements (field works, gravimetric and chemical analyses of PM samples; analysis with the use of an X-ray fluorescence spectrometer I did together with S. Szopa), developed the results of the investigations, wrote and prepared the paper for publication and attended the whole review process (correcting the manuscript according to the reviewers' and editor's suggestions, responding to the reviewers' comments, holding correspondence with the editor, etc.). I estimate my contribution to the paper at 85%.

b2 **Rogula-Kozłowska W.**, Klejnowski K., Rogula-Kopiec P., Mathews B., Krasa A., Szopa S.: Composition of fine and coarse dust in Zabrze in winter and summer workdays and weekends. [In] Ochrona Powietrza w Teorii i Praktyce, Konieczyński J. (ed.), IPIŚ PAN, Zabrze 2010, Vol. 1, 307-319.

I planned the whole experiment (the idea was mine of such a selection of the chemical analyses, the sampling period, and the samples that all the main components of fine and coarse PM were possible to determine based on one continuous measurement series; K. Klejnowski supervised the field works), performed a great part of the investigations (field works, gravimetric analysis, chemical analysis of PM--analyses with the use of an X-ray fluorescence spectrometer I did together with S. Szopa), developed the results of the investigations, wrote and prepared the paper for publication and attended the whole review process (correcting the manuscript according to the reviewers' and editor's suggestions, responding to the reviewers' comments, holding correspondence with the editor, etc.). I estimate my contribution to the paper at 70%.

b3 **Rogula-Kozłowska W.**, Błaszczak B., Klejnowski K., Concentrations of $PM_{2.5}$, $PM_{2.5-10}$ and PM-related elements at two heights in an urban background area in Zabrze (Poland). Archives of Environmental Protection 37, 31-47, 2011; IF^{a}=0.444/0.619

I planned the whole experiment (selection of a proper and complete set of archive PM samples collected in the period 2009-2010 within the IEE PAS statutory researches and determination of their chemical composition with the use of an X-ray fluorescence spectrometer), elaborated a scheme for the result analysis, drew up a plan and determined the scope for computations. I wrote and prepared the paper for publication (except literature data which was collected by B. Błaszczak), attended the publishing process (correcting the manuscript according to the reviewers' and editor's suggestions, responding to the reviewers' comments, holding correspondence with the editor, etc.). I estimate my contribution to the paper at 80%.

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[a] IF value (valid in the publication year/ present 5-year IF)

b4   **Rogula-Kozłowska W.**, Klejnowski K., Rogula-Kopiec P., Mathews B., Szopa S.: A study on the seasonal mass closure of ambient fine and coarse dusts in Zabrze, Poland. Bulletin of Environmental Contamination and Toxicology 88, 722–729, 2012; IF=1.105/1.325

I designed the research (utilizing my own idea and performance of the PM chemical mass reconstruction and, based on this reconstruction, determination of the natural and anthropogenic components in fine and coarse PM), performed a great part of the investigations (field works, gravimetric and chemical analyses of PM samples; analysis with the use of an X-ray fluorescence spectrometer I did together with S. Szopa), developed the results of the investigations and relevant chemical mass reconstruction scheme, wrote and prepared the paper for publication and attended the whole publication process (correcting the manuscript according to the reviewers' and editor's suggestions, responding to the reviewers' comments, holding correspondence with the editor, etc.). I estimate my contribution to the paper at 80%.

b5   **Rogula-Kozłowska W.**, Błaszczak B., Szopa S., Klejnowski K., Sówka I., Zwoździak A., Jabłońska M., Mathews B.: $PM_{2.5}$ in the central part of Upper Silesia, Poland: concentrations, elemental composition, and mobility of components. Environmental Monitoring and Assessment 185, 581-601, 2013; IF=1.679/1.910.

I planned the whole experiment and drew up the investigation schedule, performed a part of the investigations (field works, gravimetric analysis, chemical analysis of PM with the use of an X-ray fluorescence spectrometry and sequential extraction of samples), applied my own approach in developing the investigation results (except for PCA and MLRA analyses done by I. Sówka and A. Zwoździak), wrote and prepared the paper for publication (with the help of B.Błaszczak in preparing the literature list and review, and of S. Szopa in writing the methodology part), attended the whole publication process (correcting the manuscript according to the reviewers' and editor's suggestions, responding to the reviewers' comments, holding correspondence with the editor, etc.). I estimate my contribution to the paper at 70%.

b6   **Rogula-Kozłowska W.**, Klejnowski K., Rogula-Kopiec P., Błaszczak B., Szopa S., Mathews B.: Mass size distribution of PM-bound elements at an urban background site: results of an eight-month study in Zabrze. Annual Set The Environment Protection (Rocznik Ochrony Środowiska) 15, 1022–1040, 2013; IF=0.806/0.735.

I planned the experiment (selection of an appropriate and complete series of PM samples from the set of samples collected in several-year research into the fractional PM composition planned by K. Klejnowski), performed a large part of the research (field works, gravimetric analysis and chemical analysis of the PM samples with an X-ray fluorescence spectrometer), applied my own approach in developing the measurement results, developed the results statistically, wrote and prepared the paper for publication, attended the whole publication process (correcting the manuscript according to the reviewers' and editor's suggestions, responding to the reviewers' comments, holding correspondence with the editor, etc.). I estimate my contribution at 75%.

b7   **Rogula-Kozłowska W.**, Sówka I., Mathews B., Klejnowski K., Zwoździak A., Kwiecińska K.: Size-resolved water-soluble ionic composition of ambient particles in an urban area in Southern Poland. Journal of Environmental Protection 4, 371-379, 2013.

I planned the experiment (selection of an appropriate and complete series of PM samples from the set of samples collected in several-year research into the fractional PM composition planned by K. Klejnowski), performed a part of the research (field works and gravimetric analysis), applied my own ideas in developing the data from the chemical analyses, including the analysis of the specific dependencies between the ions and estimated recognition of the secondary aerosol composition in 13 PM fractions (the latter jointly with B. Mathews), participated in preparing the paper for publication (the literature part and conclusions were prepared by I. Sówka and A. Zwoździak), and attended the whole publication process (correcting the manuscript according to the reviewers' and editor's suggestions, responding to the reviewers' comments, holding correspondence with the editor, etc.). I estimate my contribution at 65%.

b8   **Rogula-Kozłowska W.**, Klejnowski K.: Submicrometer aerosol in rural and urban backgrounds in Southern Poland – primary and secondary components of PM_1. Bulletin of Environmental Contamination and Toxicology 90, 103-109, 2013; IF=1.216/1.325.

I planned the research (the research consisted in implementing my own ideas, one was in fact a new method for PM chemical mass reconstruction, another one was determining the shares of natural and anthropogenic components and primary and secondary matter in submicron PM in two localities based on the PM chemical

mass reconstruction done using my method), did a great part of the work (I participated in the field works supervised by K.Klejnowski, gravimetric analysis, chemical analysis using X-ray fluorescence spectrometry, and in validation of a thermo-optical method for carbon determination in PM), developed the results from the analyses and the scheme for PM chemical mass reconstruction taking secondary organic and inorganic aerosol into account to develop these results, wrote and prepared the paper for publication, attended the whole publication process (correcting the manuscript according to the reviewers' and editor's suggestions, responding to the reviewers' comments, holding correspondence with the editor, etc.). I estimate my contribution at 85%.

b9 **Rogula-Kozłowska W.**, Klejnowski K., Rogula-Kopiec P., Błaszczyk J.: Concentrations and chemical mass closure of ambient dust at a traffic site in Southern Poland: A one-year study. [In] Environmental Engineering IV, Pawłowski A., Dudzińska M.R. and Pawłowski L. (eds.), Taylor & Francis Group, London, 2013, 293-306.

I planned the research (the research consisted in implementing my own ideas, one was a new method for PM chemical mass reconstruction, another one was determining the shares of natural and anthropogenic components in fine PM in a locality under strong effect of road traffic emissions based on the PM chemical mass reconstruction done using my method), did a part of the work (I participated in the field works, controlled, validated and numerically developed incoming results of the gravimetric and chemical analyses; the field and laboratory works were supervised by K.Klejnowski), developed the results from the analyses and the scheme for PM chemical mass reconstruction adequate to this data, wrote and prepared the paper for publication (except for the review of the carbon research results worldwide compiled by J. Błaszczyk from literature), attended the whole publication process (correcting the manuscript according to the reviewers' and editor's suggestions, responding to the reviewers' comments, holding correspondence with the editor, etc.). I estimate my contribution at 75%.

b10 **Rogula-Kozłowska W.**, Błaszczak B., Rogula-Kopiec P., Klejnowski K., Mathews B., Szopa S.: Physicochemical characteristics of fine ambient aerosol from quasi-rural area in Southern Poland. International Journal of Environmental Research 8, 751-764, 2014; IF=1.100/1.129.

I planned the research (the research consisted in implementing my own ideas, one was a new method for PM chemical mass reconstruction, another one was determining the shares of natural and anthropogenic components and primary and secondary matter in fine PM based on the PM chemical mass reconstruction done using my method), did a great part of the work (I participated in the field works supervised by K.Klejnowski, gravimetric analysis, chemical analysis using X-ray fluorescence spectrometry, and in validation of a thermo-optical method for carbon determination in PM), developed the results from the analyses and the scheme for PM chemical mass reconstruction taking secondary organic and inorganic aerosol into account to develop these results, wrote and prepared the paper for publication, attended the whole publication process (correcting the manuscript according to the reviewers' and editor's suggestions, responding to the reviewers' comments, holding correspondence with the editor, etc.). I estimate my contribution at 75%.

b11 **Rogula-Kozłowska W.**, Klejnowski K., Rogula-Kopiec P., Ośródka L., Krajny E., Błaszczak B., Mathews B.: Spatial and seasonal variability of the mass concentration and chemical composition of $PM_{2.5}$ in Poland. Air Quality, Atmosphere & Health 7, 41-58, 2014; IF=1.804/1.925.

I planned the research (the research consisted in implementing my own ideas for developing, performing, and comparing chemical mass reconstructions of PM, and based on the received results, determining the shares of natural/anthropogenic and primary/secondary components in fine PM in three localities in Poland), did a part of the work (I participated in the field works, controlled, validated and numerically developed incoming results of the gravimetric and chemical analyses; the field and laboratory works were supervised by K.Klejnowski), developed the results from the analyses and the scheme for PM chemical mass reconstruction adequate to this data, wrote and prepared the paper for publication (except for the literature review, partly prepared by B.Błaszczak, and the part of the result discussion concerning back trajectories, prepared by L. Ośródka i E. Krajny), attended the whole publication process (correcting the manuscript according to the reviewers' and editor's suggestions, responding to the reviewers' comments, holding correspondence with the editor, etc.). I estimate my contribution at 70%.

b12 **Rogula-Kozłowska W.** Traffic-generates changes in the elemental composition of size-segregated urban aerosol. [In] Ochrona Powietrza w Teorii i Praktyce, Konieczyński J. (ed.), IPIŚ PAN, Zabrze 2014, Vol. 1, 133-149.

I planned the whole experiment, all the field and the laboratory works. All the measurements and chemical analyses I did myself. I developed numerically, interpreted and described the results by myself. I prepared and wrote the paper 100% by myself.

b13 Rogula-Kozłowska W.: Traffic-generated changes in the chemical characteristics of size-segregated urban aerosols. Bulletin of Environmental Contamination and Toxicology 93, 493-502, 2014; IF=1.255/1.324.

I planned the whole experiment, all the field and the laboratory works. All the measurements and chemical analyses I did myself. I developed numerically, interpreted and described the results by myself. I prepared and wrote the paper 100% by myself.

b14 Rogula-Kozłowska W.: Chemical composition and mass closure of ambient particulate matter at a crossroads and a highway in Katowice, Poland. Environment Protection Engineering 41, 15-29, 2015; IF=0.652/0.557.

I planned the whole experiment, all the field and the laboratory works. All the measurements and chemical analyses I did myself. I developed numerically, interpreted and described the results by myself. I prepared and wrote the paper 100% by myself.

b15 Rogula-Kozłowska W., Majewski G., Czechowski P.O.: The size distribution and origin of elements bound to ambient particles: a case study of a Polish urban area. Environmental Monitoring and Assessment 187, Article ID:240, 2015; IF=1.679/1.918.

I planned the experiment (selection of an appropriate and complete series of data from the archive set of data on PM received within the frame of my own project), developed the scenario for the data development, and determined the scope of the data numerical development. I wrote and prepared the paper for publication, attended the whole publication process (correcting the manuscript according to the reviewers' and editor's suggestions, responding to the reviewers' comments, holding correspondence with the editor, etc.). I estimate my contribution at 70%.

b16 Rogula-Kozłowska W.: Size-segregated urban particulate matter: chemical composition, primary and secondary matter content and mass closure. Air Quality, Atmosphere & Health, 2015 (published on-line 15.06.2015; doi: 10.1007/s11869-015-0359-y); IF=1.804/1.925.

I planned the whole experiment, all the field and the laboratory works. All the measurements and chemical analyses I did myself. I developed numerically, interpreted and described the results by myself. I prepared and wrote the paper 100% by myself.

My publications I present below are written based on the collection and development of more than four thousand suspended dust samples. The sampling in the field, laboratory analyzes of the samples, and statistical development of the data sets required employing my co-workers; twelve out of all the seventeen publications in the series I co-authored with them. Nevertheless, all the papers I wrote myself based on my own ideas, the contributions from the co-authors have never gone beyond providing me with data I needed to materialize my ideas, and the creativity of the co-authors, often inspired by my suggestions, concerned mere acquiring of these data. The solutions I applied in achieving my scientific goals are mine own, and they have never been used nor described by anybody in any project or work before I published them in the presented papers.

Formally, the publications **b1-b16** can be divided into three groups:

1. The publications **b1, b2, b3, b5-b7**, and **b15**, based on the projects in which I took part as a project manager. The schedule and methodology for ambient particulate matter sampling, as well as the interpretation of the results of the laboratory and statistical analyses I performed myself according to the concept I assumed a priori. The participation of the co-authors consisted in making laboratory or statistical analyses within the scope I outlined myself, making their technical descriptions in view of their later publication, and preparing literature data.

2. The publications **b4** and **b8-b11**, based on the projects realized by a team. The projects were launched to monitor air quality within some areas. The co-authors participated in these projects as the leaders or the members of teams that performed the field or laboratory works; they provided me with descriptions of the methods they used. The interpretation of the data was mine in all the cases and all the conclusions concerning the ambient particulate matter concentrations within the investigated areas I drew by myself.

3. The publications **b12-b14, b16**, based on my own research projects which I realized totally by myself. I defined the scope and the goals of these projects, got funding, planned and carried out the research, and finally wrote the publications.

The statements on the shares in preparing the publications **b1, b2, b3, b4, b5-b7, b8-b11**, and **b15**, received from all the co-authors, I attach in Appendix 7.

c) Scientific goals and major results of presented works:

Background

Investigating the near-ground layer of the Earth atmosphere we investigate multicomponent polydisperse aerosol, whose disperse medium is air and the dispersed phase consists of solid particles and liquid droplets (of diameters from 0.001 to 100 μm). It very dynamically changes in time its chemical properties and mutual proportions of the ambient concentrations (mass, number or surface ones) of its size fractions between the limits changing from site to site on Earth as well as within a very small area such as an urban agglomeration. Specific particles of the dispersed phase (of the specific chemical composition, origin, etc.) make atmospheric aerosols that have their common names (e.g. bio-aerosol, mist, transportation aerosol, marine aerosol), or assume the names of these particles (e.g. soot, marine salt). But it is important to remember that they are aerosols, at least two-phased systems [1,2], because the atmospheric aerosols interact with the natural environment just as such systems [II.E.:E31[b]]: globally, affecting the climate [3,4,5,6] or visibility [7,8,9;10,11,12,13], and locally, affecting ecosystems and humans [14,15,16,17,18,19,20,21]. The division of the atmospheric aerosol into natural, predominant on the global scale [22,23], and anthropogenic, more harmful to humans, especially within urbanized areas [24,25,26,27,28,29,30,31,32,33; IIA.:A7.,A10.,A12.-A15.,IIE.:E44.;**b10,b11,b16**[c]] is most important and apparent. While the human influence on the properties of the former is impossible, the properties of the latter, as well physicals as chemical, can be controlled through the control of emissions. Most often, the scientific interest in atmospheric aerosol is due to the necessity of limiting its harmful effects on the natural environment and humans. However, the chemical composition of atmospheric aerosol and the distribution of its properties with respect to its particle size, as phenomena occurring in the Earth atmosphere, are interesting on their own.

The ambient particulate matter (PM) consists of solid and liquid particles together with all the substances adsorbed to or dissolved in them. The necessity of assuming and the adequateness of such a definition of PM arise from the common use of a very important class of methods for atmospheric aerosol investigation, i.e. those starting from PM sampling using

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[b] The in-text references in the square brackets are the numbers of the papers listed at the end of this document and/or the symbols of the parts of Appendix 6 (here IIE.) followed by the symbols of the referred to papers (here E31).

[c] The papers in the monothematic cycle of publications in 4b are referred to using the symbols **b1-b16**.

manual (inertial or filtration) techniques. A sample of PM on a filter collected at specific time and site is assumed to represent well the PM properties at these time and site. The comparison between these and other techniques (e.g. real time measurements) justifies this assumption, the artefacts arising during manual sampling do not affect drastically the sampling results and usually can be compensated for in theoretical reasoning [34,35,36]. Moreover, the manual techniques are so far irreplaceable in PM sampling as only these techniques allow for examining PM for many properties at the same time, because in the same PM sample (e.g. simultaneously for many element and chemical compound contents).

The related with each other toxicity to humans and origin of PM can be determined based on the chemical composition of PM and (most often mass) distribution of the PM components with respect to particle size (most often particle aerodynamic diameter). PM affects humans mainly through the respiratory system, and the smaller particle is, the deeper into lungs it transports the substances it consists of. In turn, a specific substance in PM can have many sources, and the PM origins are reflected in this substance distribution among the PM size fractions[d].

In Poland, so far, no investigations of the chemical composition of PM have given results that can be applied in the determinations of the PM origins. The works concerning the distribution of the PM components with respect to particle size in Poland are very few. Usually, PM is investigated for its chemical composition to determine its most toxic components [e.g.3738,39,40,41,42,43,44,45,46;II.A.:A1.,A4.,A6.,A7.,A10.,A13-A15.;II.E.: E2.,E8.,E14.,E16.,E22.,E23.,E30.,E40.], or the components accounting for its particular properties (reaction, hygroscopicity, mutagenicity, morphology, magnetic susceptibility, etc.) [e.g.47,48,49,50,51,52,53,54,55,56,57,58,59,60,61,62,63,64;II.E.:E3.-E7.,E10.,E19.,E25., E32.,E35.,E41.]. Till now, I have come across mere few papers by Polish authors presenting attempts to identify PM sources from the PM elemental composition, and based only on short-term measurements of elemental composition of only one PM size fraction [28,29,65]. While such data could be sufficient for determining the PM origins in Western Europe urban areas, these works are rather a strong evidence that in the Polish urban areas the PM origins cannot be credibly determined based only on the PM elemental composition. In Southern Poland, where the PM sources are more densely located and more diversified than they are in Western Europe cities [4,25,27,28,51,56,62], some additional information on PM is needed to determine the PM major sources, and such information can be provided by the elemental

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[d] Size fractions of PM, denoted PM_{c-d}, PM_c, or $PM_{>d}$, are suspended particles with the aerodynamic diameters between c and d μm, not greater than c μm, and not smaller than d μm, respectively [2,4].

composition of some number of PM size fractions. However, even this can be too little because the standard methods for determining the PM elemental composition allow for determining only metals and metalloids in PM with the atomic number greater than 13 (aluminum atomic number), which in an urban area usually make together less than 20% of the fine PM mass [e.g..**b1,b2,b4,b5,b15,b16**;18,28,29,54], therefore even if deriving the PM origins from the elemental composition of fractionated PM is considered a better method than the reasoning based on the elemental composition of one PM fraction ($PM_{2.5}$ or PM_{10}), determining of the PM chemical composition (also macro-components in PM, such as C, N, O, etc.) seems to be indispensable anyway. In the Polish circumstances of the almost total lack of data on the PM chemical composition, its recognition, and the development of the method for determining the PM origins based on it, are necessary at least for the southern part of Poland, from where PM is transported to other European regions [66,67,68,69,70].

PM is a mixture of elemental carbon, organic compounds, ammonium compounds, nitrates, sulfates, mineral particles, trace elements, and water[e]. Their mass size distributions and mutual proportions of their masses at different locations differ depending on the PM sources active in these locations[f] [71,72,73,74]. Each PM source, a physicochemical process (combustion, erosion, resuspension, nucleation, etc.) as well as physically existing emitter (tail pipe, power station chimney, coking battery, etc.) has its own emission profile, i.e. the PM emitted from this source has, more or less, constant chemical composition and mass size distributions of its properties [2,33,71,75,76,77]. In general, the chemical compounds that are emitted by a source occur also at a sampling site, what provides a link between the PM at a sampling site and the PM sources. Chemical (most often elemental) composition of PM has been used as an important clue to identifying the PM sources worldwide for a long time now. Various methods have been used, from the simplest determinations of the correlations between PM monitoring data and meteorological conditions (wind velocity and direction, precipitation, air temperature, etc.) [e.g.78,79;80,II.A.:A17;IIE.:XIV, XV,E15.,E36.,E42.], or comparison of the ambient concentrations of PM and its components at urban sampling sites (crossroads, urban background, street canyon, etc.) with concentrations in the background, e.g, regional [e.g.81,82,83,II.A.:A1.,A3.,A8.,A12.], to advanced statistical models (principal components analysis and its modifications [e.g.25,27,28,29,37,83,84,85,86;IIA.:A17., A18;**b5,b15**]), and receptor models [np.30,83,87,88,89,90]. Receptor models are most

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[e] Fine particles, $PM_{2.5}$, are atmospheric aerosol having acidic reaction; it contains majority of ambient sulfates, nitrates, hydrocarbons, elemental carbon, toxic metals, and water. Coarse particles, $PM_{2.5-10}$, are alkaline aerosol; it contains majority of ambient crustal matter such as silicon, iron, calcium, aluminum, and their oxides, big marine salt particles, and plant debris [2,4,71].

[f] Inventories of data on chemical composition of various PM size fractions at various sites I present in, among others, publications **b3-b5, b7-b12** and **b16** from the monothematic cycle of publications in 4b.

credible because besides determining the origins of primary PM, what they do by comparing chemical composition of PM samples with source profiles, they can attribute the transformations of gases in the air to secondary PM as its origins. For instance, the amounts of sulfates and nitrates in the air cannot be accounted for by any emissions because these chemicals do not occur in the emissions, only their gaseous precursors are emitted [74,91,92]. Sulphur dioxide (SO_2), ammonia (NH_3), and nitric oxides (NO_x) are precursors to sulphur acid (H_2SO_4), ammonium hydrogen sulphate (NH_4HSO_4), ammonium sulphate (($NH_4)_2SO_4$), and ammonium nitrate (NH_4NO_3) [93,92]. Volatile organic compounds transform to secondary organic aerosol, generally in photochemical reactions; its particles can aggregate to make greater particles [74,94,95].

Statistical and receptor models need considerable amount of input data (from measurements or analyses). Moreover, if PM at a sampling site comes from a number of PM sources having similar chemical profiles (e.g. road transport and some industrial processes, or coal combustion and coal coking [25,33,71,77,96,97;IIA:A15.,A17.;IIE.:E14.,E44.;**b1,b3-b6, b15**]), these models, to perform properly, need data on some source-specific gaseous emission markers or/and relations between emitted PM components (for all significant PM sources in the neighborhood) [98,99]. Finding them is practically impossible, and the emission profiles are usually taken from the literature or/and from the measurements of emissions often done in circumstances entirely different from those in which they are later applied.

The simplest methods, such as analysis of enrichment factors for PM-bound elements, or chemical mass closure[g] (reconstruction) of PM using the masses of the chemical compounds and/or groups of chemical compounds which contain analytically determined PM components   [e.g.25,26,31,66,67,71,73,81,82,100,101,102,103,104,105,106,107,108,109], allow (at some loss of precision) for lowering the number of samples by relaxing the criteria for covering the investigated time period with the sampling time[h]. Usually, these methods are applied to determinations of natural and anthropogenic, or primary and secondary components of PM [83,102,103,104,110,111,112,113,114; **b4,b5,b8-b14,b16**]. Such methods are usually the first stage in the reasoning about the origins of PM, and most often they are, or they should be, introductory to time and cost consuming collecting of information (PM sampling) for further more credible apportionment of PM among sources using, for instance, receptor models [113,115,116]. However, in some areas, such as urban areas in Southern Poland, the

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[g] I use the term "chemical mass reconstruction" instead of commonly used in the literature "chemical mass closure" in the whole text [105].
[h]. For instance, in routine monitoring using manual measurements, it is required that the sample-taking is uniformly distributed over a year and that the sampling time covers the year in at least 14%.

simple methods of finding PM sources, such as inferring from (mass) contributions of the component groups to PM (chemical mass reconstruction), can be as credible as more advanced methods are. This may occur in heavily urbanized and industrialized regions, where there exists multiplicity of densely situated small PM sources, where the effects of some important PM sources (e.g. inflow of PM from other regions, a number of industrial plants, household ovens, etc.) are hard to be taken into account and must be omitted, or where many PM sources have similar emission profiles (emitting PM and the same PM precursors), and where the unambiguous attribution of PM amounts to sources is impossible.

Goals of the research

1. Determining of the chemical composition of several PM fractions in several various localities in Poland and assessment of this knowledge applicability in the determinations of the PM origins.

2. Determining of the PM chemical composition in several various localities in Poland and application of this knowledge to simplified determination of the origins of particular PM size fractions.

3. Applying of the mass distributions of the PM chemical components with respect to the particle aerodynamic diameter to determination of the PM origins.

My publications that I present here recapitulate my attempts to realize the three presented goals. Achieving the goals, I drew some conclusions concerning the investigated areas, presented in *Detailed description of the research*, and some general conclusions, of wider meaning, presented below, in *General description of the research*.

General description of the research

The general conclusions I drew in the course of realizing of the first goal (I have analyzed more than 700 PM samples of various PM size fractions [**b1,b3,b5,b6,b12**]) concerned the applicability of the information on the PM elemental composition in determinations of the PM origins. I have shown that:

1. Within the areas where PM and the majority of PM-bound elements are almost entirely municipal in a heating season, and are not in a non-heating one, the PM origins can be derived from the data on the element contents of PM only separately in these two seasons.

2. Within the areas where a multiplicity of various PM sources occur, and their effects on the PM concentrations differ, even a relatively small set of data on the elemental composition of several PM size fractions can be more useful in the PM origin determinations than a big set of such data concerning only one PM size fraction. I proved this, for instance, by trying to find the characteristic elements for some PM sources within the Upper Silesia Agglomeration, where the elemental analysis of PM split into at least several size fractions appeared to be necessary. Based on the elemental compositions of 13 PM size fractions, I found that in Zabrze Ni, V, Sc, and Sb are characteristic of road traffic, and Cl, S, Se, B, and As of power production from coal.

3. The sources of a single PM size fraction (e.g. $PM_{2.5}$ or PM_{10}) in areas of densely located PM sources of various type, as in Silesian cities, can be determined only based on the macro-component content of PM and/or, besides on their ambient concentrations, on other properties of the PM-bound elements. The mobility of a PM-bound element is a promising parameter in determinations of the PM sources. If the concentrations in PM and the mobilities of some number of elements are known then the amounts can be estimated of easily soluble salts (sulfates, chlorates, nitrates, some bicarbonates, etc.), non-soluble or sparingly soluble compounds (majority of oxides, sulfides, nitrides, carbonates), and complexes (e.g. aluminosilicates, minerals) that contain these elements. In my opinion, the methods for determinations of the PM origins based on mobility of PM-bound elements should be intensely developed in future and determinations of the mobility of the PM-bound elements for the purposes of PM source apportionment should become a standard.

Besides the above three conclusions from the research presented in **b1, b3, b5, b6,** and **b12**, I formulated some findings about the localities where I conducted the researches, and although as such they concern only these localities, they can be applied to the whole Silesia Region. I found that the ambient concentrations of the majority of the fine PM ($PM_{2.5}$) bound elements (except Na, S, Cl and some heavy metals, e.g. Cd, Cr, Zn, and Pb) in the Silesia Region resemble those in other urbanized regions in Europe. The ambient concentrations of the coarse PM ($PM_{2.5-10}$) bound elements in various localities are not comparable, they clearly depend on the level of soil contamination at the sampling site and on the measurement circumstances.

In the cities of the Silesia Region, the $PM_{2.5}$-bound elements come mainly from industrial power production (e.g. S, Ca, Se, Sb), combustion in local ovens of coal (e.g. S, Se, K, Cl), biomass (e.g. K, S), and waste (e.g. Cl, Br, Cu, Mn, Pb, Sb, Zn), and combustion of liquid fuels in car engines (e.g. Cu, Zn, Sb, Pb). Depending on a city, their summary

contribution to the mass concentration of $PM_{2.5}$, as the contribution of fuel combustion, is 46-82%. $PM_{2.5}$-bound Al, Si, Ca, Fe, and Ti come from soil. I found that in the cities of the Silesia Region no less than 2-3% of the $PM_{2.5}$ mass is mineral-soil matter composed of their oxides. Resuspended road dust and soil are the basic source of $PM_{2.5-10}$; they determine the $PM_{2.5-10}$ elemental composition.

In the Silesia Region cities, it is practically impossible to apportion PM-bound elements among particular PM sources only through analyzing one PM size fraction (e.g. $PM_{2.5}$ or PM_{10}). My attempts at doing this yielded ambiguous results; I present them in *Detailed description of the research (Ad 1)*. In *Ad 1*, I also give the particular conclusions from the investigations of PM for its elemental composition.

The majority of the results received during the first goal realization are qualitative and quantitative data on PM in some particular localities in Poland; however, as detailed data on the PM concentrations in some specific areas in the Upper Silesian Agglomeration, they make a unique update to the basis of datasets on PM in our part of Europe. Especially the ambient concentrations of the considerable number of 40 elements from 4 or 13 PM size fractions seem to be valuable. In the literature, the results on the elemental composition of very fine PM ($PM_{0.1}$, PM_1) are scarce; there is a need for such results from all kinds of regions, but especially for those concerning the PM within such industrial areas as the Upper Silesian Agglomeration [18,19,20,25, 32,33,68,69,70,96,99,109,117,118,119; IIE.:E44.; **b5,b12,b15**].

The conclusions from the second part of my work are derived from the results of the analyses for the chemical composition of more than 2500 samples of various fractions of PM [**b2,b4,b8,b9,b10,b11,b13,b14**]. PM was sampled at sites of various importance: national (Diabla Góra), regional (Racibórz), urban (Zabrze, Katowice, Gdańsk) sampling sites, and at sites of very local importance (crossroads and highway in Katowice). Based on the literature and the analytically determined amounts in PM of organic carbon, elemental carbon, main ions extracted from water-soluble compounds, and main and trace elements:

1. I developed a scheme for the PM chemical mass reconstruction that is applicable not only to data from measurements, but it can also utilize specific information on the area where the data was collected;

2. Applying my PM chemical mass reconstruction scheme to various datasets, I managed to identify the chemical composition of from 50 to 100% of the mass of such PM size fractions as $PM_{0.1}$, PM_1, $PM_{1-2.5}$, $PM_{2.5}$, $PM_{2.5-10}$, PM_{10}, and PM_{10-40};

3. I determined the mass shares of primary and secondary matter in various PM size fractions, and indicated the properties of PM (e.g. elemental carbon content of PM) and environmental

conditions (e.g. concentrations of ozone and gaseous precursors of secondary aerosol, air temperature) that are favorable to and those that inhibit the creation of secondary matter in the air;

4. I pointed out the applicability of a simple division of the PM components into groups to qualitative and quantitative determinations of contributions of a source/group of sources of PM and PM precursors to the PM concentrations in several localities in Poland.

The method I mention in 4 enabled me to formulate my most important findings about the PM origins in the areas I investigated. Namely, I substantiated the assumption, having been taken for granted for a long time, that the emissions from local sources, mainly obsolete and inefficient household ovens, are the most serious air pollution source in the Upper Silesian Agglomeration [27,120;II.A.:i,iii-v,A2.,A3.,A4.;II.B.:B3.,B4.,B7.,B8.;II.E.:X,XIII, XVII-XIX,E8.-E11.,E14.,E15.]. Presently, in Katowice, municipal or/and industrial emissions make from 29 to 82% of the $PM_{2.5}$ mass depending on the season of the year The municipal emissions, in fact uncontrollable, blur the differences between the properties of fine PM (concentrations, chemical composition) in urban (Zabrze) and non-urban (Racibórz) localities. I also found that in the southern cities (Katowice), from 1.5 to 5%, and in the northern ones (Gdańsk) from 4 to 7% of the $PM_{2.5}$ mass can come from road traffic; from industrial power production, it is from 8 to 12% and from 19 to 23%, respectively.

High elemental carbon content of $PM_{2.5-10}$ (12 and 20% of the mass in the non-heating and heating seasons, respectively) makes $PM_{2.5-10}$ in Zabrze different from $PM_{2.5-10}$ in other parts of Europe. It also proves clear effects of municipal emissions on $PM_{2.5-10}$ concentrations in the Upper Silesia Agglomeration, also visible in $PM_{2.5}$ concentrations.

Unlike road traffic in Western Europe urban localities, road traffic does not alter visibly the chemical composition of particular PM size fractions in the Silesian cities. In Katowice, in localities under strong effect of traffic emissions, the only traits of this effects in PM are the lack of ammonium nitrate (NH_4NO_3) among the secondary inorganic PM components and the mass contribution of secondary organic matter to PM lower than in localities beyond the road traffic effect. Greater amounts of sodium chlorate (NaCl) in coarse PM and of elemental carbon in fine PM can be found in a road vicinity.

I described the results and conclusions drawn from the examinations of chemical composition and mass closure of various PM size fractions in various Polish localities in *Ad 2*. I also present there the results on quantitative shares of various categories of substances, including organic and inorganic secondary matter, in masses of various PM size fractions.

The most important general conclusions from the works on the chemical mass reconstruction method for fractionated PM and from the attempts at its applications to determine the qualitative and quantitative contributions of the sources of both PM and its gaseous precursors to the PM concentrations are as follows:

1. The soil (natural) and anthropogenic matter contents of PM should be estimated based on analyzing the factors of enrichment of PM in those elements whose chemical compounds form these two PM component categories. It is an essential element of the PM chemical mass reconstruction, especially of the fine PM mass, and it should replace the arbitral assumption that the PM-bound Si, K, Ca, Ti, and Fe always and as a whole are natural and that the rest of the elements is anthropogenic.

2. Within the areas where there are many sources or where the activity of PM sources depends on a season of the year, the investigation of PM for chemical composition should be at least as long as one whole year. The activity of anthropogenic sources, such as combustion of fuels for heating in the cities of Southern Poland, can be relatively easily noted in any period of the year. But, for example, an arbitral assumption that in a near-sea locality marine salt is a component of PM in any period in the year can lead to erroneous PM source determination. By analyzing the seasonal fluctuations of some ion ambient concentration and their mutual proportions in Gdańsk, I proved that the salt brought with marine water by winds contributes significantly to the $PM_{2.5}$ mass only in summer and autumn. Whole-year investigations can catch the effects of such periodically active natural sources, and also of such incidental but periodically active sources as open fires (grass burning), construction works, etc.

3. Within the areas where municipal emissions strongly affect PM concentrations, PM-bound elemental carbon is not a good tracer for road traffic. The proportion of the masses of the PM-bound organic to elemental carbon is a better indicator of the magnitude of the traffic contribution to the PM concentrations and composition.

So, the accomplishment of my second goal, like that of the first one, also led to the formulation of entirely new conclusions, this time based on applications of my schemata of the PM chemical mass reconstruction (e.g. using enrichment factors to divide the PM components into natural and anthropogenic) to PM analysis in several areas in Poland (the shares of the main PM components in the PM mass and classification of the PM-bound matter by its origin). These areas were so characteristic as to make these conclusions valid for other areas where the PM and its precursors come from as a complex system of sources as they do in Southern Poland. The new schemata, taking into account local conditions, applied in such areas instead of the used up to now schemata prepared for areas with much simpler systems of

PM sources (the USA, Western and Southwestern Europe), can yield better approximates of the shares of the PM component groups in PM.

Determination of the chemical composition of the PM fractions, including the finest ones, $PM_{0.1}$ and PM_1, was a subtask of the second goal. The chemical composition of $PM_{0.1}$ and PM_1 is a very rarely presented in the literature.

The accomplishment of my third scientific goal consisted in the application (one of the first in the world) of the (mass) distribution of the PM chemical components with respect to particle aerodynamic diameter in the determinations of the PM origins [b7,b13,b15,b16]. In each of b7, b13, b15, and b16, I present a number of specific conclusions concerning the characterization and origins of PM in the areas in which I conducted the investigations that I describe in *Ad 3* in detail.

However, the most original findings of this part of my research arose from putting together all the specific conclusions from b7, b13, b15, and b16. Analyzing the mass size distributions of the PM components in Katowice [b16] and the findings from b7 and b13, I proved that in this city, typical of the Upper Silesia Agglomeration, in summer, PM occurs in three particle populations differing in the chemical composition and origins. The first one makes about 13% of the PM mass. It is the population of the finest PM particles (mainly with the aerodynamic diameters up to 0.26 μm, i.e. containing $PM_{0.26}$), primary, coming from combustion of fossil fuels and biomass for power production and in car engines. In fact, it consists of primary organic compounds and of oxides and salts of various metals. The second population, mainly secondary PM (ammonium sulfate $(NH_4)_2SO_4$, ammonium nitrate NH_4NO_3), contains the fraction of PM particles with aerodynamic diameters between 0.26 and 1.6 μm ($PM_{0.26-1.6}$). In Katowice, its average mass contribution to PM is about 59%, its chemical composition and concentrations are shaped by municipal or/and industrial emissions; its gaseous precursors come also from road traffic. The third population, about 28% of the PM mass, is the population of coarse particles, containing the particles with the aerodynamic diameters from1.6 to 40 μm ($PM_{1.6-40}$). They are composed mainly of soil, road dust, organic matter, and elemental carbon. This population contains no more than 4.6% of secondary inorganic matter, and the sulfate and nitrate ions, differently than in the second population (in fact in $PM_{0.26-1.6}$), come mainl y from calcium sulfate ($CaSO_4$), potassium sulfate (K_2SO_4), and sodium nitrate ($NaNO_3$).

Analyzing the mass size distributions of 18 PM-bound elements, factors of PM enrichment in these elements, and applying principal component analysis (PCA) combined with cluster (CA) analysis, I proved that also in Zabrze in a heating season PM occurs in three

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particle populations similar to those in Katowice, also differing from each other in the chemical composition and origins [b15]. Each of b7, b13, b15, and b16, written in the course of realization of my third scientific goal, gives some scheme that can be used to determine the origins of PM divided into any number of size fractions, within any area, and in any season of the year. Properly used combinations of these schemes, depending on the case, allow for quite precise determination of secondary matter content in PM. The detailed analysis of the chemical composition of the sub-fractions of PM_1, based on one such a scheme and described in *Ad 3*, allowed me to prove that in Katowice, in the non-heating period of 2012, PM_1 consisted of secondary matter in 49% [b16]. Using another, simpler, approach described in *Ad 2*, I proved that PM_1 at an urban background sampling site in Zabrze, in both the non-heating and heating periods of 2009, consisted in more than 50% of secondary matter [b8]. It not only confirms the noticed earlier similarity of PM in the two cities of the Upper Silesian Agglomeration, Zabrze and Katowice, but also verifies my method for analyzing PM for its chemical composition in different areas.

All the schemata I applied to data on PM from various areas and various averaging periods, the ones based on the analysis of elemental composition of fractionated PM (*Ad 1*), and chemical mass reconstruction of the PM fractions with separating the PM components into primary and secondary matter (*Ad 2*), through those that in the determinations of the PM origins combine chemical mass reconstruction and modalities of mass size distributions of PM components (*Ad 3*), gave almost identical results, both qualitative and quantitative, concerning the PM origins within several investigated areas.

Detailed description of the research
Ad 1 (publications b1, b3, b5, b6, b12)

In the period 2007—2013, I investigated the elemental composition of various size fractions of PM in several cities of the Silesian Province [b1,b3,b5,b6, and b12]. I used energy dispersive X-ray fluorescence spectroscopy (EDXRF), the protocol for determinations of the PM elemental composition I developed myself [II.A.:vi;II.E:XVI,XXVII,XXVIII,E1., II.J:J.6.,J.11.,J.13.]. I analyzed 168 samples of $PM_{2.5}$ collected in 2002—2004 and 2007 for 40 elements, and from the descriptive statistics of the series of the 24h concentrations of these elements in these periods, from the linear correlations between these series, and from more advanced statistical analyses of these data (principal component analysis, or PCA; multidimensional linear regression analysis, or MLRA), I concluded that:

- The majority of the averages over a measuring period (one calendar year) of the ambient concentrations of $PM_{2.5}$-bound elements (except Na, S, Cl, and some heavy metals, e.g. Cd, Cr, Zn, and PB) in the Silesian Region resemble their concentrations in other urbanized European regions [b1 and b5];

- The lowest 24h concentrations of the investigated $PM_{2.5}$-bound elements, except Al, Si, Ca, Fe, and Ti that within the investigated areas are probably crustal (from soil erosion and resuspension), occur in a non-heating, and the highest in a heating period [b1];

- Except for Al, Si, Ca, Fe, and Ti, all the $PM_{2.5}$-bound elements come from combustion of coal (e.g. Se, K, Cl), biomass (e.g. K, S), waste (e.g. Cl, Br, Cu, Mn, Pb, Sb, Zn), and liquid fuels in car engines (e.g. Cu, Zn, Sb, Pb); however, the exact assignment of the PM-bound elements to the PM sources is impossible [b1 and b5]; in 2007, the contribution of combustion (the sum of the yearly contributions of the mentioned sources) to the $PM_{2.5}$ mass was 78% in Katowice and 36% in Zabrze [b5];

- In Katowice, $PM_{2.5}$ containing Cr, Mn, and Fe, therefore most probably coming from high-temperature industrial processes, is 14% of the total $PM_{2.5}$ mass; $PM_{2.5}$ from industrial power production, containing S, Ca, Se, and Sb, is 10% in Zabrze and 4% in Katowice [b5];

- In the areas where in a heating period municipal emissions strongly, and in a non-heating one hardly, affect the $PM_{2.5}$ concentrations, and where they contribute to the majority of chemical elements in $PM_{2.5}$, the $PM_{2.5}$ origins can be derived from these element contents in $PM_{2.5}$ only separately in a heating and in a non-heating period [b5];

- Assignment of some elements or groups of elements to particular emission sources may require determination of the elemental composition of some PM size fractions [b5];

- About 2—3% of the $PM_{2.5}$ mass is mineral and soil matter [b1]; determinations of the origins of the rest of the $PM_{2.5}$ mass would need to determine the concentrations of the $PM_{2.5}$ macro components or/and to use, besides the ambient concentrations, other properties of the $PM_{2.5}$-bound elements [b1 and b5].

The last three conclusions gave the directions to my further research. Namely:

- I determined the elemental composition of $PM_{2.5}$ and $PM_{2.5-10}$ sampled simultaneously in the same urban background locality in a non-heating period (sampling site in Zabrze, 26 April-3 July 2008) both at two heights (6 and 2 m above ground level, a.g.l.) [b3];

- I determined and analyzed (for origins) elemental composition of four PM size fractions, PM_1, $PM_{1-2.5}$, $PM_{2.5-10}$, and PM_{10-40}, at an urban background sampling site in a heating and a non-heating period (Zabrze, two four-month sampling periods in 2009) [b6];

- I determined the elemental composition of 13 PM size fractions at an urban background site and at a crossroads (Zabrze, five-month sampling period between the mid-June and mid-October 2013), and pointed to both the PM size fractions and PM-bound elements that were clearly linked to road traffic (exhaust or motion of vehicles) [b12];

- Using my own method, I determined the mobility of $PM_{2.5}$-bound elements (the percentage of the mass of an element from its PM-bound water or ammonium acetate soluble compounds in its total mass in PM) [b5].

The ambient concentrations of majority $PM_{2.5}$-bound elements in Zabrze, including crustal Ca, Fe, and Ti, are higher at 6 than at 2 m a.g.l., although the $PM_{2.5}$ concentrations are almost equal at both altitudes [b3]. PM_1- and $PM_{1-2.5}$-bound Al, Ca, Mg, Ti, and Rb, also considered crustal, have higher concentrations in winter than in summer [b6]. The ambient concentrations of the PM-bound Al, Ca, Fe, Mg, Ti, and Rb in Zabrze could have been affected by other processes than erosion or re-suspension of mineral matter. However, the $PM_{2.5-10}$-bound elements, including anthropogenic S, Cl, Zn, As, and Pb [b3 and b5], have much greater concentrations at 2 than at 6 m a.g.l., and the $PM_{1-2.5}$ concentrations at 2 m is almost three times higher than at 6 m [b3]. But the average ambient concentrations of each of these elements bound to $PM_{2.5-10}$ and to PM_{10-40} are almost the same in a heating and a non-heating period [b6]. Therefore, the core parts of the masses of these elements in coarse PM are anthropogenic, coming from road dust, construction works, etc., even if their ambient concentrations are due to re-suspension. On the other hand, some elements, e.g. Cu, Zn, Cd, and Pb, occur in soil in greater than the average concentrations within the investigated area [b5], therefore some elements in coarse PM might have come from polluted soil.

The described methods did not allow for unambiguous distinguishing between the elements related to road traffic (tires, brakes, road surface, chassis, etc.) and those coming from mineral matter (Earth crust, soil, etc.) in coarse PM [b3 and b6]. Neither they allowed to divide the PM-bound elements into those coming from power production and those from combustion of liquid fuels in car engines [b1,b3,b5, and b6]. It could be partly done by comparing the elemental compositions of 13 PM size fractions (defined by the impactors used: two the same thirteen-stage cascade DEKATIs) at a crossroads and at an urban background site in non-heating period [b12]. A non-heating period was selected to avoid overshadowing of the effects of road traffic emissions by the municipal emissions in a heating period, the latter negligible in the majority of the Western Europe cities and very intense in Zabrze. Each of Ni, Zn, Sn, and Sb from at least three out of all four sub-fractions of $PM_{0.03-0.26}$, and Mg, Cu, Sc, V, Fe, Rb, Sr, Mo, Pd, Te, I, Cd, Cs, and Ba from at least one, had much

higher ambient concentrations at the crossroads than at the urban background site, $PM_{0.06-0.108}$-bound Ni and Zn even 20 times. Thus, these elements are characteristic of the emissions from fuel combustion in car engines and additives to fuel and lubricants [b12]. This way, only the rest of the $PM_{0.03-0.26}$-bound elements, i.e. Na, Cl, Ca, K, S, Cr, Mn, Fe, As, Se, Br, and Pb, having at the two sites similar ambient concentrations, can indicate power production (from coal).

In $PM_{0.26-2.5}$, the elements characteristic of road traffic are V, Ni, Sb, and Sc, and in some its sub-fractions also Al, Ti, Co, Rb, Rh, Sb, Sc and Au [b12]. For coarse PM, $PM_{2.5-6.8}$-bound Na, Mg, Al, Si, K, Ca, Ti, Mn, Fe, V, Ni, Zn, and Sr, had ambient concentrations higher at the crossroads than at the urban background site. Majority of the $PM_{6.8-40}$-bound elements had the concentrations higher at the crossroads, the greatest differences between these two sites were in the concentrations of crustal Si, Al, Ti, Fe, Ca [b12].

Concluding, to recognize the road traffic effects on the PM concentrations in Zabrze, Ni, V, Sc and Sb in fine PM (combustion of fuels in car engines) and Na, Ni, V, Zn, and Sb in coarse PM (tires, brakes, road surface, chassis, etc.) should be investigated. The fine PM-bound Cl, S, Se, Br and As can reflect the effects of power production (from coal).

Ad 2 (publications b2, b4, b8, b9, b10, b11, b13, b14)

The available analytical methods allow to determine the masses of some number of the categories of chemical substances in PM. Such a reconstruction of the PM mass at a sampling site can be so inaccurate as to yield the sum of the masses of the determined in PM components greater than the whole PM mass (when determined masses are relatively high and the methods overestimate the results), and this despite leaving out many components of PM. However, still the distribution of the PM mass among these categories (substances) points to the PM origins.

The basic categories of the chemical PM components, most commonly used in PM chemical mass reconstructions, are [e.g. 25,26,31,66,67,71,73,81,82,100,101,102,103,104, 105,106, 107,108, 109;**b2,b4,b8,b9,b10,b11,b13,b14**]:
- Two carbon groups: elemental carbon (EC) and organic carbon (OC, from organic compounds);
- Main ions from water soluble compounds (SO_4^{2-}, NO_3^-, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, Na^+, Cl^-);
- Chemical elements, such as Al, Si, Cr, Mn, Fe, Ni, Cu, Zn, As, Cd, and Pb.

I developed a method for analyzing a set of daily twin-samples of two complementary PM fractions, $PM_{2.5}$ and $PM_{2.5-10}$. Each such a PM sample consisted of two PTFE or quartz fiber filters (2×132 samples), the kind of the filters changing every second day, exposed in parallel, one to $PM_{2.5}$, the second--to $PM_{2.5-10}$, at an urban background sampling site in Zabrze in 2009. The results of the analyses I present in **b2**; they were used in the first in Poland chemical mass reconstruction of $PM_{2.5}$ and $PM_{2.5-10}$, which I present and discuss in **b4**. Carbon (EC+OC), SO_4^{2-} and NO_3 prevailed in $PM_{2.5}$ and $PM_{2.5-10}$ in Zabrze [**b2**]. Na^+, Cl^-, NO_3^-, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Al, Si, Fe, Zn, and Pb had lower shares in the $PM_{2.5}$ mass, and those of the trace elements Cr, Mn, Ni, Cu, Ga, Ge, As, Se, Br, Rb, Sr, Y, Mo, Rh, Pd, Ag, Cd, Sn, Sb, Te, I, Cs, Ba, Au, Co, Sc, V, and La were the lowest. $PM_{2.5-10}$ contained considerable amounts of crustal elements, mainly Si [**b2**].

I divided the PM components determined in **b2** into the commonly accepted categories taking into account the PM origins, methods for the mass assessments, and minimum number of components needed for the chemical mass reconstruction [113]. There are six such categories: organic matter (OM), elemental carbon (EC), secondary inorganic aerosol (SIA), Na^+ and Cl^- (marine components, MC), crustal matter (CM, compounds of elements from natural sources), and other elements (OE, anthropogenic elements). I replaced the elsewhere used name "sea salt" (SS) with the "marine components" (MC) because I decided that sea salt does not occur in the Zabrze air [**b4**]. To divide the elements, for which I analyzed PM, into natural and anthropogenic, and then to define the CM and OE categories, I applied my original method based on computing enrichment factors (EF) for these elements [**b4**]. Up till now, Si, K, Ca, Ti, and Fe were arbitrarily assumed to be wholly natural and the whole rest of the elements—anthropogenic. However, the high EFs of $PM_{2.5}$-bound Si, K, Ca, Ti, and Fe in the heating period of 2009 suggest their anthropogenic origins. It was clear to me that not whole masses of these elements in $PM_{2.5}$ were in CM. I included into the $PM_{2.5}$-bound CM mass only these parts of the $PM_{2.5}$-bound Si, K, Ca, Ti, and Fe that could be attributed to their EFs in the non-heating period. The rest of the $PM_{2.5}$-bound Si, K, Ca, Ti, and Fe masses I considered anthropogenic and included into the OE mass.

The low EFs for $PM_{2.5}$-bound Rb, Sr, and Ba in the non-heating period, and for $PM_{2.5-10}$-bound Cr, Mn, Ni, Rb, Sr, and Ba in both heating and non-heating periods, meaning lack of the anthropogenic influence, made me add their masses to the CM mass for $PM_{2.5}$ in the non-heating period and for $PM_{2.5-10}$ in both periods [**b4**].

This categorization of the PM mass proved natural sources (erosion, re-suspension of soil matter) to account for 56% of the $PM_{2.5-10}$ mass in the non-heating and 41% in the heating

period in Zabrze. The rest of $PM_{2.5-10}$ consisted mainly of road dust and elemental carbon agglomerates with various adsorbed to them compounds (hydrocarbons for example); lesser contribution to $PM_{2.5-10}$ is that of biological matter (fungi, plant debris, pollen, etc.) [b4]. Even 23% of the $PM_{2.5}$ mass in the non-heating, and 16% in the heating one, could be in SIA, the matter that arises from gaseous precursors of PM (SO_x, NO_x, NH_3); it was 15 and 18% for $PM_{2.5-10}$, respectively.

The amounts of PM precursors in the air within such an area as Zabrze are practically determined by fuel combustion, so I suppose that even 80% of the $PM_{2.5}$ mass may come from combustion in Zabrze, directly or through precursors [b4]. Based on the conclusions from the elemental analysis of PM (*Ad 1*) and the most common chemical profiles of sources (in receptor models, for example) [b4], I showed that it was the combustion of:

- Liquid fuels in car engines, because the indicators of the road traffic influence, trace elements Ni, V, Cu, Sc, Sb [b12], and EC, in the non-heating period (June-October) of 2013, had their greatest mass shares in $PM_{2.5}$ on working days, and their daily ambient concentrations were higher on working days than on weekends [b2]; the chemical profiles of $PM_{2.5}$ in the heating period (January-March) of 2009 matched the profile of $PM_{2.5}$ coming from fuel combustion in car engines [b4];

- Solid fuels and biomass in various power producing utilities, because besides the similarities between the elemental compositions of $PM_{2.5}$ in Zabrze and $PM_{2.5}$ emitted from such utilities [b4] I noticed significantly higher ambient concentrations [b4] and shares in the $PM_{2.5}$ mass of $PM_{2.5}$-bound EC, SO_4^{2-}, Cl⁻, Br, Se, and As in the heating than non-heating period of 2009 [b2].

High ambient concentrations and shares in the $PM_{2.5-10}$ mass of $PM_{2.5-10}$-bound EC, Cl⁻, and K^+ in the heating period of 2009 (January-March) [b4], especially on weekends, accompanied by limited effects of road traffic emissions [b2], and similarity of the chemical compositions (excluding CM) of $PM_{2.5}$ and $PM_{2.5-10}$ in the heating period of 2009 prove that the combustion of coal, biomass, and probably waste in household ovens is a significant source of PM in Zabrze.

In **b11**, I show the differences in the urban $PM_{2.5}$ chemical composition between Southern and Northern Poland, compare the $PM_{2.5}$ compositions at two urban sites with the $PM_{2.5}$ chemical composition within a non-urban (clean) area in Diabla Góra (reference area for Poland, national background), and show the differences in the intensity of the influence of various PM sources on $PM_{2.5}$ concentrations and chemical composition based on three series of $PM_{2.5}$ samples collected in parallel during 2010 at three localities, Katowice in Southern

Poland and Gdansk and Diabla Góra both in Northern Poland (358, 356, and 365 samples, respectively) [b11]. For all the three localities, I defined the same categories of components of $PM_{2.5}$ as in b4 (I replaced the symbol "MC" with "Na_Cl" following the reviewer's suggestion). I analyzed the relations between the $PM_{2.5}$ components and concluded that:

- The shares of the main components in the $PM_{2.5}$ mass in Katowice and Zabrze were similar [b4 and b11]. The average share of carbonaceous matter (OM+EC) in $PM_{2.5}$ was from 42 to 49%, depending on an averaging period, except for the autumn of 2010 when it was about 20%; this is the highest share among all the category shares. The SIA mass share in $PM_{2.5}$ in Katowice reached 33% [b11].

- In Gdańsk, in Northern Poland, the share of OM+EC in the $PM_{2.5}$ mass was lower than in Katowice and Zabrze in Southern Poland, in 2010 it was in average from 12 (spring) to 42% (winter); as in Southern Poland, the yearly mas share of OM+OC in $PM_{2.5}$, equal to 32%, was greater than the yearly share of SIA, equal to 25.4% [b4 and b11].

- The SIA mass prevailed in $PM_{2.5}$ in Diabla Góra, its share in $PM_{2.5}$ in 2010 was from 19 (spring) to 44% (winter) [b11].

- In the winter of 2010, in each locality, SIA in $PM_{2.5}$ consisted as well of ammonium sulfate ((NH_4)$_2SO_4$) as of ammonium nitrate (NH_4NO_3). In the rest of the year NH_4NO_3 was less abundant in $PM_{2.5}$, in the summer there was no NH_4NO_3 in $PM_{2.5}$ at all. I found some evidences of $PM_{2.5}$ having acid reaction in the winter in all the three localities, and in Diabla Góra also in the autumn [b11].

- The masses of sodium and chlorine (Na_Cl, MC in b4) made together 8, 13.5 and 9.2% in average of the $PM_{2.5}$ mass in 2010 in Katowice, Diabla Góra, and Gdańsk; in the winter, the Na_Cl mass shares in $PM_{2.5}$ in Zabrze and Katowice were equal [b4 and b11]. The Na_Cl mass share in $PM_{2.5}$ was very high in Katowice in the spring, in Gdańsk in the autumn, and in Diabla Góra in the spring, summer, and autumn (15-16%) [b11].

- The yearly (2010) mass shares of CM in $PM_{2.5}$ were about 8% in Katowice, Gdańsk, and Diabla Góra; in Zabrze the share of CM in $PM_{2.5}$ was 5% [b4]. In each locality it was highest in the summer (14-15%) and lowest in the winter (3.5-5.5%) [b11].

From this PM chemical mass reconstruction I inferred that fuel combustion influence on $PM_{2.5}$ concentrations is strongest in Southern Poland. In Katowice and Zabrze, more than 70% of the $PM_{2.5}$ mass can be accounted for by combustion (OM+OC, Na_Cl, and SIA). In Gdańsk, mainly in summer and autumn, effects of the sea (the Baltic Sea) on the $PM_{2.5}$ concentrations is observed (wind-blown sea water droplets containing sea salt), and stoichiometric calculations [113; b11] gave the sea salt share in the $PM_{2.5}$ mass at the level of

respectively 10 and 14% in these periods. The presence of sea salt in $PM_{2.5}$ cannot be excluded in Diabla Góra. However, it was not to derive directly from the values of the proportions of proper ion concentrations [b11], and because the southwestern winds prevail in Poland, therefore even if it occurs then only in small amounts.

The simultaneity of the measurements in Katowice, Gdańsk, and Diabla Góra in 2010, and Diabla Góra being the air quality background site for whole Poland [121] allow for using the results from b11 in the assessment of contributions of the most influential $PM_{2.5}$ sources to the $PM_{2.5}$ concentrations in all these three localities. The ambient concentrations of the $PM_{2.5}$-bound EC and SO_4^{2-} coming from power production in Katowice and Gdańsk were impossible to be measured directly because of very high emissions from other sources. I assumed that air pollutants in Diabla Góra are not of the local origin, being brought from other regions [b11], that $PM_{2.5}$-bound EC and SO_4^{2-} are spread uniformly over the whole Poland by high stacks of power stations, and that in Diabla Góra they come exclusively from power production. Consequently, the ambient concentrations of the $PM_{2.5}$-bound EC and SO_4^{2-} from power production in the three localities were the same. From the analysis of the proportions of the sum of the average concentrations of $PM_{2.5}$-bound EC and SO_4^{2-} in Diabla Góra to the concentrations of $PM_{2.5}$ in Diabla Góra, Gdańsk, and Katowice in various seasons, it results that from 8 to 12% of the $PM_{2.5}$ mass in Katowice, from 19 to 23% in Gdańsk, and from 29 to 39 % in Diabla Góra, depending on the season, can come from power production.

In Gdańsk, municipal emissions are negligible in summer because of well-developed heat distribution network, therefore $PM_{2.5}$-bound EC comes only from power industry and road traffic, and the EC concentrations are the same as in Diabla Góra in summer. I assumed that $PM_{2.5}$-bound NO_3^- in Gdańsk comes exclusively from road traffic. There were about 230,000 vehicles registered in each Gdańsk and Katowice in 2010, so the ambient concentrations of $PM_{2.5}$-bound EC and NO_3^- were the same in both cities, and the concentrations of EC without the power industry share and of NO_3^- in Gdańsk in the summer were equal to the concentrations from traffic in Gdańsk and Katowice in each season of 2010. From the proportions of the sum of the average concentrations of $PM_{2.5}$-bound EC and NO_3^- due to traffic to $PM_{2.5}$ concentrations in the two cities in various seasons I estimated the road traffic contribution to $PM_{2.5}$ at 1.5-5% in Katowice and 4-7% in Gdańsk, depending on a season.

For any of the three localities, the subtraction of the lowest in 2010 seasonal contributions of Na_Cl, power production and road traffic emissions, and crustal matter (for Gdańsk also marine salt) from 100% gives the approximate upper limit of the contribution of

the municipal emissions (local household ovens, fireplaces) and/or industrial emissions to $PM_{2.5}$ at this locality. Analogously, considering the highest contributions, and additionally undetermined matter, one can receive the approximate lower limit. This way, the mass contribution of municipal and/or industrial emissions to $PM_{2.5}$ in Katowice in 2010 are estimated to be between 29 and 82%, depending on the season of the year.

Although very simplified, this approach seems to yield quite correct results. The yearly contributions of road traffic and municipal emissions together, i.e. of combustion, to the yearly concentration of $PM_{2.5}$ in Katowice in 2010, computed using this method, is 68.5%, while the yearly contribution of OM+EC, Na_Cl and SIA to $PM_{2.5}$, computed in **b11** for Katowice, where they make together the whole combustion emissions, is 70%.

Some conclusions from the applications of the above scheme to estimate the contribution of several sources to $PM_{2.5}$ concentrations based on the results from **b11**, both those on the quantitative effects of road traffic on PM and those about domination of municipal emissions in the cities of the Upper Silesian Agglomeration [**b11**], I confirm in **b9** and **b14**. By analyzing a series of 358 diurnal PM_{10} samples collected in Katowice in 2010 at a sampling site affected strongly by emissions from the A4 highway, I proved that strong road traffic sources can elevate PM_{10} concentrations by about 14% on average in a year relative to urban background [**b9**]. By analyzing the monthly series of these samples, I proved that this difference is due to salt (NaCl) and sand that are sprinkled on the roads in winter, and, re-suspended, become components of PM_{10} [**b9**]. Analyzing 18 samples of 13 PM size fractions (from 13 stage impactor) collected at this site, I proved that only coarse particles, $PM_{2.5-10}$, account for this difference, and that the effect of combustion of solid fuels and waste in household ovens (plastic bottles, bags, and wrappings) on Na^+ and Cl^- content of fine particles, PM_1 and $PM_{1-2.5}$, cannot be excluded in any period in the year [**b14**].

In summer (especially in its middle: June-August), at the site near A4 in Katowice, SIA probably does not contain NH_4NO_3 [**b9**]. I associate this with low ambient concentrations of ammonia (NH_3) (much lower in the summer of 2010 than in the heating period [**a9**]; ambient concentrations of NO_x were high), too low for ammonia to neutralize sulphur and nitric acids in the air [**b9**]. In the middle of the summer of 2010, in the holiday season, I observed the same phenomenon in the Katowice center [**b14**]. In a road vicinity, when ambient concentrations of ammonia are low, PM has probably acidic reaction; it is possible that $Ca(NO_3)_2$ and $NaNO_3$ make the majority of PM-bound nitrates [**b9** and **b14**]. It is probably the most characteristic chemical property of the PM affected by road traffic.

The proportion OC/EC of the OC to the EC contents of PM, usually much lower in areas under the road traffic influence than elsewhere (urban background, rural area) [82; II.A.:A8., II.E.:XX], is a good indicator of the magnitude of the road traffic contribution to the concentrations and chemical composition of PM_{10}. However, nondispersive infrared spectrometry applied to pyrolysed samples, a standard method in such determinations, used in its determination in Katowice, distorted it [122,123,124,125,126,127] and gave it very low [b9]. I show in b9 and b11 that not being capable of separation of OC from EC in PM samples, this method can be used only to determine the summary EC+OC content of PM. Probably the method overestimates the EC content of PM because of great amounts of organic compounds in the samples (pyrolytic carbon arises, burnt in oxygen together with EC from samples). I suppose that this limitation of the thermal method concerns only its applications to PM from the area that I investigated or from the areas in which the OC and EC contents of PM are similar to those in Katowice. Elsewhere the thermal method performs well in the determinations of PM-bound OC and EC [66,67,128,129].

OC/EC for PM in Katowice, computed from the OC and EC contents in PM that were determined using a Sunset Laboratory Inc. thermo-optical analyzer [b14] [II.F:F17.,F21; II.J: J6.,J19], was lower at the A4 highway in March-June 2010 than in the Katowice center in the holiday period when traffic is usually limited. The difference was greatest for PM_1 and $PM_{1-2.5}$, proving that fine particles of EC from car engines account for lowering OC/EC for PM_1 from 9 at the crossroads to 6 at the highway [b14]. I ultimately proved the usability of OC/EC in reasoning about the effects of road traffic on PM by parallel measurements of 13 PM size fractions at the urban background site and at the A4 highway in Katowice [b13]. Depending on the fraction, OC/EC was from 1.15 ($PM_{0.06-0.108}$) to 5.8 ($PM_{4.4-6.8}$) times greater at the background site. Such great differences in OC/EC for coarse PM between traffic and background sites, specific of Katowice, distinguish this region from other regions [b13].

I also applied the proportion OC/EC in determinations of primary and secondary matter content of PM in several localities [b8,b10 and b14]. I analyzed the chemical composition of 464 PM_1 and $PM_{2.5}$ samples from Racibórz [b8 and b10] and of 149 PM_1 samples from Zabrze [b8]. In Katowice, in two localities strongly affected by road traffic, I examined PM_1, $PM_{1-2.5}$, $PM_{2.5-10}$, and PM_{10-40} for primary and secondary matter contents [b14].

The conclusion were as follows:

- Secondary organic compounds (secondary organic carbon, OC_{sec}) made in average 56% of the mass of organic compounds in PM_1 in Zabrze and 43% in Racibórz (average in August – December 2009 and 2010 respectively) [b8];

- In both Zabrze and Racibórz, the share of OC_{sec} in PM-bound OC is higher in a non-heating than in a heating period; in Zabrze it was from 40% in December to 66% in September 2009, the highest daily OC_{sec}/OC, 84%, in Racibórz I observed in August 2010; monthly averages of OC_{sec}/OC were between 29 and 38% [b8];

- The ambient PM_1-bound C_{sec} concentrations and C_{sec} mass shares in PM_1 are higher in Zabrze than in Racibórz because the ambient concentrations of the precursors of OC_{sec} in Zabrze (volatile organic compounds, VOC, from many sources, mainly from road traffic) are higher [b8];

- The above conclusion agrees with the observations of OC_{sec}/OC in the localities directly affected by road traffic emissions: at the A4 highway in Katowice the OC_{sec} shares in PM_1- and $PM_{1-2.5}$-bound OC were both equal to about 80%, and at the crossroads 65%; the OC_{sec} share in $PM_{2.5-10}$-bound OC at the highway was over 70% and at the crossroads 40%; the share of OC_{sec} in OC is lower for these PM fractions whose EC content is high (EC has the highest mass shares in $PM_{2.5-10}$ and in PM_{10-40}) [b14];

- The conditions for the OC_{sec} formation in the air are less favorable in Racibórz (lower VOC ambient concentrations) than in Zabrze, therefore the mass shares of secondary inorganic aerosol (SIA) and secondary organic aerosol (SOA) in PM_1 are lower in Racibórz than in Zabrze, even by10%; even 60% of the PM_1 in Zabrze and 50% of PM_1 and $PM_{2.5}$ in Racibórz is secondary matter (SIA+SOA) [b8 and b10];

- Small EC shares in PM_1 and in $PM_{2.5}$ and high ambient concentrations of ozone and of the gaseous precursors of organic aerosol in a non-heating period (especially in summer) are favorable to OC_{sec} creation [b8 and b10];

- In Racibórz, the shares of secondary matter (SOA+SIA) in PM_1 and $PM_{2.5}$ are similar, these two fractions have similar chemical compositions, the almost only differences between them are greater CM share in $PM_{2.5}$, and the secondary matter content: in $PM_{2.5}$ $(NH_4)_2SO_4$ and NH_4NO_3 occur, in PM_1 only $(NH_4)_2SO_4$ occurs; in a heating period, the NH_4NO_3 content of $PM_{2.5}$ can be even two times greater than the $(NH_4)_2SO_4$ content [b10].

Atmospheric aerosol of particular origins has characteristic and, more or less, constant both chemical composition and distributions of its properties (e.g. ambient number and mass concentrations, component contents, etc.) with respect to particle size (most often with respect to particle aerodynamic diameter; [2,33,71,75,75,76,77]).

At a sampling site, PM occurs in three main populations (modes) of particles: the nucleation mode (the finest particles, mainly from nucleation and condensation), accumulation mode (primary particles from combustion and aggregates of nucleation particles), and coarse mode (particles mainly from mechanical processes and big particles of salts) [130,131].The presence of each one at a sampling site can manifest itself as a maximum of the density function of the distribution of some property of these particles (most often mass or number concentration) with respect to the particle size (most often particle aerodynamic diameter) depending on the activity of the sources it comes from. The mass size distribution of PM at a site can have all the three maxima (modes) corresponding to these main three populations (modes), but usually only two occur, the mass of the nucleation population being much lower than of the two other [2,132,133]. Similarly, the mass size distribution of any PM component can be multimodal, and in fact many are (mainly those of trace elements) [II.E.:E16.,E23.,E30.,E51.;**b7,b13,b15,b16**].

I analyzed the distributions of the masses of the PM components among PM size fractions as well as the maxima of the density functions (or rather the intervals of particle aerodynamic diameters in which the distribution modes occur) of the mass distributions of various PM components with respect to the particle aerodynamic diameter to determine the PM origins, [**b7,b13,b15,b16**].

In **b15**, I present the results of the examinations for the chemical compositions of 13 PM size fractions, sampled with a 13 stage cascade impactor (12 sampling sessions, $12 \times 13 = 156$ samples), analysis of the distributions of masses of 18 PM-bound elements and of their sum among these fractions (distributions of the masses of elements with respect to PM particle aerodynamic diameter), and the maxima of their density functions (modality) and the particle diameter intervals of these maxima occurrence, and based on all this information, I find probable sources of these 18 elements and 13 fractions. The PM samples were taken at an urban sampling site in Zabrze, in winter (2009), when PM sources are more active than in summer.

The conclusions are as follows:

- The PM mass size distribution in Zabrze in the winter of 2009 is bimodal. One density function maximum (mode) occurs between 0.65 and 1 µm (accumulation mode), another between 6.8 and 10 µm (coarse particle mode) [b15]. Two particle populations, the accumulation mode and coarse particle mode, are dominant in the PM mass in Zabrze.

- Some elements (Cl, K, Cr, Ni, Cu, Zn, Sr, Cd, Sb, Pb) have bimodal mass size distributions, like PM, some trimodal (Mn, Ba), or even four-modal (As) [b15]. The modes of each of these distributions reveal PM particle populations that can have different origins.

- The analysis of the enrichment factors computed for all the elements in every PM fraction and of the density functions maxima (modes) supported the determination of the origins of PM-bound elements in Zabrze in the winter (2009). Almost all PM-bound elements (except Ca and Fe, which came from natural sources in all fractions) were anthropogenic [b15]. Their majority came from combustion either of solid fuels (S, Cl, K, Ni, Cu, Zn, Ge, As, Br, Cd, Sb, Ba, Pb; the absolute, sometime the only, density function maximum, between 0.17 and 1 µm) or liquid fuels in car engines and from high-temperature industrial processes (As, Ba, with the absolute maximum between 0.06 and 0.17 µm, and Sr, Mn with the absolute maximum between 0.06 and 0.17 µm and without the maximum between 0.17 and 1 µm). Road dust is also one of the great sources of the majority of the PM-bound elements in Zabrze, but lesser than combustion of fuels; it accounted for Cl, Cu, Zn, As, Br, Cd, Sb, and Pb in $PM_{>1}$. S, K, Ca, Cr, Mn, Fe, Ni, Sr, and Ba in coarse particles were more the effect of resuspension of soil and sand than of anthropogenic processes [b15].

- The combination of principal component analysis (PCA) and cluster analysis (CA) applied to 13 sets of the series of these element concentrations (for each fraction one set) also supported the determinations of the PM origins in Zabrze [b15]. Cluster analysis, using Ward's method, applied to the 13 first principal components from PCA applied before to the 13 sets of the series of ambient concentrations of 18 PM-bound elements, grouped the 13 PM size fractions in Zabrze in the winter into four groups: [$PM_{1.6-2.5}$, $PM_{2.5-4.4}$,], [$PM_{0.03-0.06}$, $PM_{0.108-0.17}$], [$PM_{0.06-0.108}$, $PM_{0.17-0.26}$, $PM_{0.26-0.4}$, $PM_{0.4-0.65}$, $PM_{0.65-1}$, $PM_{1-1.6}$], [$PM_{4.4-6.8}$, $PM_{6.8-10}$, $PM_{>10}$]. Each of these four groups is contained by one of four particle populations defined by mass size distributions of PM-bound elements (nucleation mode, accumulation mode, and two modes of coarse particles, related to different sources) [b15]. Probably flue gases from vehicles are the basic source of $PM_{0.03-0.06}$ and $PM_{0.108-0.17}$ within this area. $PM_{0.06-0.108}$, $PM_{0.17-0.26}$, $PM_{0.26-0.4}$, $PM_{0.4-0.65}$, $PM_{0.65-1}$, and $PM_{1-1.6}$ come also from burning of fuels, but in other combustion process; it is probably solid fuel combustion in local ovens and power industry.

Greater ambient particles in Zabrze come from resuspension of soil and sand ($PM_{2.5-4.4}$) and of road dust ($PM_{>4.4}$) [b15].

In **b13** and **b16**, I similarly characterize the origins of PM at an urban background site in Katowice in the non-heating period (March-June) of 2012. Along with the data on ambient concentrations of 26 elements in 13 PM size fractions, I used the concentrations of EC, OC, ions of water soluble compounds, and of 16 polycyclic aromatic hydrocarbons bound to these 13 PM fractions. I subdivided the mass of PM into parts corresponding to the groups of its chemical components (chemical mass reconstruction of fractionated PM) and examined the mass distributions of the components with respect to PM particle size. In **b13**, comparing these results from the urban background site with the results from parallel measurements carried out at the highway in Katowice exactly in the same way and at the same time (March-June 2012), similarly interpreted, I determined the road traffic effects on the chemical characteristics of PM in this typical of the Upper Silesia Agglomeration city. In **b16**, I present quantitative contributions of some sources/processes to the PM concentrations in Katowice that I determined from the distributions of masses of some chemical compounds from PM and their categories (groups) among the PM size fractions. I also present there my own scheme for chemical mass reconstruction of the 13 PM fractions.

In **b16** I show that although the mass size distribution of PM at an urban background site in Katowice in winter is monomodal, with the mode between 0.4-0.65 μm, the modalities of mass size distributions of the PM-bound chemical compound categories, and also of the PM-bound polycyclic aromatic hydrocarbons, divide PM into three size fractions very distinctively. Namely, about 13% of the PM mass in Katowice belongs to $PM_{0.03-0.26}$, 59% to $PM_{0.26-1.6}$, and 28% to $PM_{1.6-40}$. The fractions $PM_{0.03-0.26}$ and $PM_{1.6-40}$ consist mainly of primary components (mass of each in about 65%) while $PM_{0.26-1.6}$ of secondary ones (about 57% of the mass). In average, 21% of the $PM_{0.03-0.26}$ mass is anthropogenic matter (mainly oxides of anthropogenic elements), 37.6% primary chemical compounds, 3.1% elemental carbon, and 3.2% mineral matter (mainly crustal element oxides). About 25% of the $PM_{0.03-0.26}$ mass is secondary compounds both inorganic (ammonium sulfate and nitrate) and organic [b16]. At the highway in Katowice, $PM_{0.03-0.26}$ has a different chemical composition [b13]. Although at the highway, as at the urban background site, $PM_{0.03-0.26}$ consists mainly of primary matter, the share in PM and ambient concentrations of this primary matter (especially of elemental carbon) are higher, sometimes several times, than at the urban background site (mode of the PM-bound EC between 0.03 and 0.26 μm, it does not exist at the urban background site). Instead, the mass shares and ambient concentrations of the $PM_{0.03-0.26}$-bound

secondary inorganic matter, most probably consisting of sulfur and nitric acids, are much lower [b9,b13, and b14]. At the highway, the mass size distributions of some elements related with road traffic (tail pipe emissions) have modes between 0.03 and 0.26 µm; they have no maxima in this interval at the background site [b13 and b16]. I conclude from this that the concentrations of $PM_{0.03-0.26}$ in Katowice are mainly shaped by road traffic. Of course, the traffic most significant effects are observed at a source (the highway), and PM undergoes a variety of transformations changing its characteristics on the way from such a source to other city areas (the urban background site), however, still the traits of the traffic emissions, depending on the emission intensity, can be spotted in PM within these areas. It is also obvious that at the urban background site in Katowice the chemical composition of $PM_{0.03-0.26}$ is affected more by the combustion of fuels and biomass in power production facilities (very fine particles) than at the crossroads, where traffic emissions dominate. From this and b11 it can be seen that the estimation in b11 of the contribution of road traffic and power industry to the $PM_{2.5}$ concentrations in Katowice at from 9.5 to17%, depending on a season, well agrees with conclusions drawn in b16, and that road traffic and power industry are the main sources of $PM_{0.03-0.26}$ in Katowice.

PM$_{0.26-1.6}$ in Katowice consists mainly of secondary matter, organic and inorganic. Primary organic matter, elemental carbon, anthropogenic and mineral matter make no more than 35% of the $PM_{0.26-1.6}$ mass [b16]. In general, at both the urban background site and the highway, the $PM_{0.26-1.6}$ chemical composition is the same; only bound to some sub-fractions of $PM_{0.26-1.6}$ EC and some usually traffic related metals have the ambient concentrations and the mass shares in PM slightly higher at the highway [b13]. I consider the $PM_{0.26-1.6}$ chemical composition and its similarity at the two localities a sufficient proof of this PM size fraction being shaped by municipal emissions (household ovens, biomass combustion in gardens, grilling, local boiler installations, etc.). It is also confirmed by the estimation of the quantitative contributions of municipal emissions to the $PM_{2.5}$ concentrations based on the results from b11. So, municipal emissions in a typical city in the Upper Silesia Agglomeration can account for about 55% of the PM mass and contribute to PM mainly particles having aerodynamic diameters between 0.26 and 1.6 µm. Majority of these particles come from transformations of PM gaseous precursors coming from various sources [b16]. The concentrations of this dominant in the Katowice air PM size fraction are due to both (gaseous) emissions, mainly from combustion of fuels in local installations, and atmospheric conditions favoring or limiting the secondary PM formation [b9 and b14]. In b7, a more elaborated method for computing ambient concentrations of secondary inorganic chemical compounds

from various PM size fractions is presented. I used the data on the ambient concentrations of the ions of water soluble compounds bound to 13 PM size fractions sampled at the urban background sampling site in Zabrze in August-December 2008. As at the urban background site in Katowice, the two main secondary inorganic components of PM, ammonium sulfate and ammonium nitrate, I found in $PM_{>1.6}$, and those from $PM_{0.26-1.6}$ had the highest ambient concentrations. Therefore, the origins of particular PM size fractions and quantitative contributions of various emission sources to the PM concentrations at the urban background site in Katowice are typical of those in other cities of the Upper Silesia Agglomeration. In **b7**, I also show that the estimations of the ambient concentrations of ammonium sulfate $((NH_4)_2SO_4)$ and ammonium nitrate (NH_4NO_3) based on simple stoichiometric computations can lead to overestimating of these concentrations.

$PM_{1.6-40}$ in Katowice contains no more than 22% of secondary matter--4.6% is inorganic and 17.4% is organic [**b16**]. $PM_{1.6-40}$-bound sulfate and nitrate ions come mainly from $CaSO_4$, $NaNO_3$, K_2SO_4, etc. [**b7**]. Therefore, $PM_{1.6-40}$ consists mainly of primary matter, i.e. elemental carbon (5%), mineral matter (16%), and trace anthropogenic matter (mainly metal oxides, 19%) [**b16**]. The differences in the $PM_{1.6-40}$ chemical composition between the urban background site and the site at the highway occur mainly for $PM_{>4.4}$; $PM_{1.6-4.4}$ at both sites has similar chemical composition [**b13**]. So, some part of $PM_{1.6-40}$ in Katowice, mainly greater particles, is primary, and comes from soil and other material erosion (roads, construction sites, cars, etc.) and re-suspension. However some $PM_{1.6-40}$ part consists of great elemental carbon agglomerates and salt particles that come from solid fuel combustion in inefficient household ovens or from open fires (grass burning in allotment gardens, grilling, etc.)

5. My other scientific activities

Besides the research that I conducted during the past 6 years and whose results I present in the publication cycle in 4, there were three other strands in my scientific activity that I also consider important. The research in the first one, the properties of aerosol from vehicular sources and effects of the emissions from these sources on the physicochemical parameters of atmospheric aerosol (chemical composition of the surface layer of PM, PAH and selected metal content of PM, optical properties of PM) [II.A.:ii,vii; II.E.:V,VI,VIII,X,XIII,XV,XXII,XIV], I started in 2004, when I started to work on my PhD dissertation [II.J:3c,3e,3h, J4.]. In 2009-2014, after receiving my PhD degree, I continued to

work on the extended version of the subject in co-operation with the Department of Air Protection of the Silesian University of Technology in Gliwice within the frame of two my own projects [II.J.:J4.,J19.] and as a participant of the EUREKA project [II.J.:J3.] launched in the Department. Within the latter, I developed and applied a method for analyzing the elemental composition of PM emitted from car engines of various types, in various technical conditions, and under various driving cycles [II.E.:E34.,E38.]. The results from my own projects I applied in the works described in the papers in 4 [b12,b13,b14,b16] and in determinations of the effects and properties of vehicular aerosol [II.A.:A8.; II.E.:E28.,E44.,E49.,E50.,E54.]. The most noticeable features of both my projects were the scope of the PM chemical analyses (PM was analyzed for EC, OC, 10 main ions, 40 elements including Hg, 16 PAHs) and applying all these analyses to each of 13 PM size fractions (aerodynamic diameter range from 0.03 to 40 μm). The projects included also counting the PM particles of particular size (number concentrations, aerodynamic diameter range from 0.005 to 20 μm) [II.F.:F17.,F21.].

An outcome of these investigations was the publications II.A.:A1.,A3.,A8.,A12.; I.E.:E1.,E3.,E28.,E34.E38.,E44.,E45.,E46.,E49.,E50.,E54., monograph II.E.:E13., reports documenting the researches II.F.:F17.,F21., and two papers being now prepared for publication.

The second subject, toxic component content of PM in Poland, I have explored since 2008 within the frames of the design works that the Institute was commissioned to perform by the state administration [II.8.:B2.,B3.,B4.], research projects [II.J.:J4.,5.,J6.], research programs [III.A.:A1.5.], and as a subject for BSc and MSc theses I was advisor for at the Silesian University of Technology [III.J.:J1.1.]. The researches consist mainly of examining PM size fractions for PAH content in various localities in Poland. The researches resulted with papers that, besides data on air pollution with PAHs and comparisons of various areas for ambient PAH concentrations, propose simple numerical indicators of the hazard from PM-bound PAHs [II.A.:A4.,A6.,A7.,A10.,A13.,A14.,A15.; II.E.:E16.,E22.,E23.,E29.,E45.,E52.]. At present, our paper on the subject is submitted for publication in the Environmental Monitoring and Assessment (Springer). We are proceeding with developing the collected data on PM-bound PAHs.

Intending further and wider exploration of these two topics, I initiated joining of the Institute and the BOSMAL Automotive Research and Development Center Ltd. in Bielsko-Biała in a consortium to investigate PM-bound durable organic compounds released from car

engines [III.E.:E1.6.]. In the nearest future I will try to receive funds from the National Centre for Research and Development for the realization of this project.

In 2012, I joined the investigations on mass size distribution of PM-bound Hg, one of the first in the world [II.A.:A18.:II.E.:E30.,E40.,E53.]. We plan to carry on these just launched investigations in the future and extend them to include determinations of the origins of Hg in various PM size fractions.

The third direction of my research is the applicability and applications of selected statistical methods and models in the development of data on the chemical composition of PM and in the analyses of the relations between air pollution (also with PM) and the shaping it factors. I became interested in the applications of statistical methods in the air quality assessment in 2003, when I designed and applied neural networks to predict air pollutant concentrations for the purposes of my MSc thesis [II.E.:I,II,III,IV,IX], and since 2010, in cooperation with the scientists from the Gdynia Maritime University, Warsaw University of Life Sciences, Wrocław University of Technology, and Silesian University of Technology in Gliwice, I have continued the research. The results of these joined efforts are presented in **b5** and **b15**. Other papers on the subject that I co-authored are II.A.:A9.,A16.,A17., A19.,A20.;II.E.:E6.,E14.,E36.,E39.,E41.,E42., listed in Appendix 6. However, the most important outcome of my interest in statistical methods is a synthesis of the results of my investigations in this field and the results presented in the publications listed in 4, and using it to create a detailed scenario for exact determinations of the quantitative contribution of particular emission sources to the concentrations of PM_1 in two localities in Poland. I have been utilizing this scenario in my own research project (SONATA) since the end of 2013 [II.J.:J7.]. Another, also important and quantifiable, effect of my interest in these methods is their application in the analysis of data on the chemical forms of elements that occur in PM (4, *Ad 1*). The latter idea is to be exploited in the large project I plan to launch in the future [III.E.:E1.5.].

Besides my own research, I perform investigations as a member of the Immision Department of the IEE, taking part in all the projects realized in the Department. The works I take part in as a member of the Department are as follows:

- Design works commissioned by the state administration and large business entities [II.B.:1b.,2b.,3b.,4b.,5b.,6b.,7b.,8b.,9b.,10b.;B1.,B2.,B3.,B4.,B5.,B6.,B7.,B8.,B9., B10.];

- Projects financed from the funds for the statutory activity of the IEE PAS [II.J.:1j.,2j.,4j.,6j., 7j., 9j.,11j., 12j.; J9.,J10., J12.,J13.,J15.,J16.,J17.];

- Documenting the researches conducted within the statutory activity of the IEE PAS, final reports on the projects and investigations commissioned by the state administration and large business entities [II.F.:1f.,2f.,3f.,4f.,5f.,6f.,7f.,8f.,9f.,10f.,11f.,12f.,13f.,14f.,15f.,16f.;F2., F3.,F4.,F5.,F6.,F7.,F8.,F.9.,F10.,F11.,F12.,.F13.,F14.,F15.,F16.,F18.,F19.,F20.,F22.,F23.];
- Expert opinions and other commissioned reports (action in favor of practice) [III.M.:1m.,2m.,3m.,4m.,5m.,6m.,7m.,8m.,9m.,10m.;M1.1.,M1.2.,M1.3.,M1.4.,M1.5.,M1.6., M1.7.,M1.8.,M1.9., M1.10.,M1.11.,M1.12.,M1.13.],
- Developing the results from all the above mentioned projects and preparing them for publication [II.A.:i,iii,iv,v;A2.,A5.,A6.,A11.A20.;II.E.:E4.,E5.,E7.,E8.,E9.,E10.,E11.,E12., E15.,E21.,E25., E26.,E27.,E33.,E37.].

After I had received my PhD degree, since June 2009, I participated in three international projects (as a grant researcher, as a grant manager and researcher in one project), five research projects financed from the Ministry of Science and Higher Education and Polish National Science Centre (as a chief grant researcher in three, as a chief grant researcher and manager in two), ten projects realized as statutory tasks of the IEE PAS (as a chief grant researcher and manager in two, in the remaining—as a chief grant researcher), and I conducted one my own project which I financed by winning the IEE PAS internal competition for funds for an own subsidiary research (as a chief grant researcher and manager).

In 2013, I received funds from the Eko-Staż (Eco-Internship) project co-financed by the European Union within European Social Fund and the Regional Chamber of Commerce in Katowice [III.L.:L1.3.].

Since I received my PhD degree in 2009, till now, I have participated in ten-odd Polish and several international conferences (the titles of papers, the times and places of the conferences I give in II.L. and III.B.). Since 2004, I have been a member of the Organizing Committee of the Scientific Conference *Ochrona Powietrza w Teorii i Praktyce*; after 2009, as the Secretary of the Committee, I co-organized three conferences of this cycle [III.C.]. In December 2014, I was assigned an auxiliary advisor in a PhD degree conferment procedure at the Faculty of Environmental Engineering of the Wrocław University of Technology. In April of 2015, I was appointed a member of the Scientific Committee of the Scientific Conference *Jakość powietrza a zdrowie* [III.B.]. I actively participate in domestic and international research programs, research networks, and consortia [III.A. and III.E.]. Since 2008, I have been a member of the Scientific Board of the IEE PAS [III.Q.].

The list of my scientific publications contains 169[i] papers, of which:

- 38 are on the JCR[j] list (31 I published after receiving my PhD degree in 2009), 11 are in the publication cycle in 4;

- 70 are monographs, chapters in monographs in Polish and English, publications in international and domestic journals other than those in the JCR basis (50 I published after receiving my PhD degree); 5 of them are in the publication cycle in 4;

- 9 are international conference papers (6 I published after receiving my PhD degree);

- 12 are Polish conference communications (7 I published after receiving my PhD degree);

- 40 (23 after receiving my PhD degree) are unpublished (jointly developed reports, catalogues of air pollution data, reports on researches).

In the years 2013-2015, I have reviewed 42 scientific papers for Polish and foreign journals. I reviewed papers for: *Environmental Protection Engineering* (3 reviews), *Atmospheric Measurement Techniques* (2 reviews), *Architecture Civil Engineering Environment – ACEE* (3 reviews), *Water, Air & Soil Pollution* (4 reviews), *International Journal of Environmental Analytical Chemistry* (1 review), *Atmospheric Research* (2 reviews), *Annals of Warsaw University of Life Sciences – SGGW- Land Reclamation* (2 reviews); *Archives of Environmental Protection* (5 reviews), *Environmental Earth Science* (3 reviews), *Atmospheric Pollution Research* (4 reviews), *Chemosphere* (2 reviews), *Environmental Science & Technology* (4 reviews), *Przegląd Naukowy Kształtowanie i Inżynieria Środowiska* (2 reviews); *Environmental Science and Pollution Research* (1 review), *Polish Journal of Environmental Studies* (1 review); *Aerosol and Air Quality Research* (2 reviews), *Inżynieria i Ochrona Środowiska* (1 review).
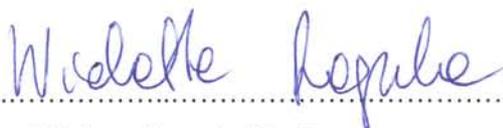
Wioletta Rogula-Kozłowska

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[i] 117 papers were published in 2009—2015, after I had received the PhD degree; 79 papers out of these 117 are scored and can be attributed 758 Ministry of Science and Higher Education points in total according to the criteria valid in the years of their publishing (papers **b1-b16** contributed 226 points).

[j] Presently, 35 of them are in the Web of Science base; they have 173 citations in total and 86 by other authors (15.09.2015; searching by author, time interval 2007—2015, the word combination: "Rogula-Kozlowska OR Rogula W"), what gives the h index equal to 8 (according to Google Scholar it is 328 citations and h=12).

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