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Airborne Platinum, Palladium, and Rhodium as Indicators of Traffic-Related Emissions: A Zagreb Case Study

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Abstract

Platinum group elements (PGEs), especially platinum (Pt), palladium (Pd), and rhodium (Rh), are analyzed as emerging airborne contaminants in urban environments. This study aimed to monitor the spatial and temporal distribution of PGEs in urban air and to evaluate their potential as indicators of traffic-related emissions. The paper presents a five-year monitoring of Pt, Pd, and Rh mass concentrations in airborne particulate matter collected from three urban locations (North, Center, and South) with different traffic loads in Zagreb, Croatia. Weekly samples were digested in acid under high temperature and high pressure, and analyzed using inductively coupled plasma mass spectrometry (ICP-MS). At the monitoring location South, mass concentrations of all PGEs were generally 20–40% higher than at other locations, consistent with its higher traffic density. The PGEs showed seasonal variability, with 40–60% higher mass concentrations in winter and autumn than in spring and summer. The spatial and temporal distribution of PGE mass concentrations across urban locations demonstrates their potential as indicators of traffic-related activity. Palladium mass concentrations were consistently the highest, as a result of its increased use in modern catalytic converters. These findings underscore the relevance of long-term PGE monitoring for understanding urban atmospheric pollution dynamics within changing environmental conditions.

Keywords: environmental monitoring; particulate matter; platinum group elements; traffic-related pollution

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1. Introduction

Urban air pollution is still one of the most pressing environmental challenges in densely populated areas, with transportation emissions strongly influencing the chemical composition of ambient air. While regulated pollutants such as nitrogen oxides (NO_x), particulate matter (PM), sulfur dioxide (SO_2), carbon monoxide (CO), and volatile organic compounds (VOCs) have long been monitored, increasing attention is directed toward emerging contaminants, including platinum group elements (PGEs) such as platinum (Pt), palladium (Pd), and rhodium (Rh). These elements are primarily emitted from automotive catalytic converters, which use PGEs to reduce harmful vehicle emissions. Although these technologies are designed to reduce gaseous pollutants, they inadvertently contribute to the release of trace metals into the atmosphere [1–10]. These metals are widely used in various industries, but the automotive industry is the dominant consumer of PGEs [11–13]. As a result of their widespread use in vehicle converters, elevated concentrations of these

metals have been detected in urban environments, particularly near high-traffic areas. PGEs have been quantified in urban soils, road dust, airborne particulate matter, sediments, aquatic systems, vegetation, and biota [14–30]. Although these findings demonstrate broad environmental dispersion, the magnitude and spatial patterns of PGE contamination vary considerably across cities, suggesting that local traffic density, fleet composition, and urban morphology strongly influence their distribution. Enclosed environments, such as tunnels show particularly high concentrations, highlighting the importance of dispersion conditions [31–33]. Historically considered inert, PGEs are now being recognized as potentially bioactive. Recent studies indicate that certain PGE soluble species and nanoparticles may induce oxidative stress, immunomodulatory effects, and cardiovascular or reproductive impacts [34–36]. Human exposure has been documented in occupational settings and, increasingly, in urban populations living near dense traffic [37–43]. These findings underscore the need to better understand long-term atmospheric levels, especially in regions where monitoring is limited. Unlike many regulated air pollutants, PGEs are not routinely monitored, and their long-term behavior in urban atmospheres remains insufficiently characterized, particularly in southeastern Europe. This gap limits the ability to evaluate temporal trends, assess the effectiveness of emission control technologies, and understand population exposure as well as environmental risks. Because PGE emissions are closely linked to traffic density, vehicle type, and driving conditions, their concentrations in ambient air may serve as potential indicators of traffic-related activity and urban mobility patterns. Zagreb, the capital and largest city of Croatia, displays many of these urban characteristics. With its dense traffic corridors, mixed land use, and seasonal heating patterns, the city experiences recurring air quality challenges, particularly during colder months. Its moderate continental climate, combined with topographical features such as the Medvednica mountain and the Sava river valley, further influences pollutant dispersion and accumulation by restricting horizontal airflow. These features also create conditions that support the development of temperature inversions and reduce vertical mixing within the lower atmosphere during stable meteorological conditions. As a growing urban center with over 770,000 inhabitants, Zagreb provides a relevant case study for investigating traffic-derived PGE emissions. This study focuses on their occurrence and distribution across selected urban zones, contributing to the broader understanding of emerging metal pollutants within the framework of environmental science and sustainable air quality management.

2. Materials and Methods

Weekly PM_{10} samples (particulate matter with an aerodynamic diameter <10 μm) were continuously collected (24 h/7 days) from 1 December 2015 to 31 December 2020 at three urban monitoring locations (North, Center, and South). Sampling was performed using 90 mm quartz fiber filters (Pall Life Sciences, Port Washington, NY, USA), with a weekly airflow of approximately 800 m^3. The selected locations enabled spatial coverage of different urban environments, while the weekly sampling period ensured representative accumulation of airborne particulate matter.

2.1. Monitoring Locations

Location North (45°50′07″ N, 15°58′38″ E) is an urban background station near the Institute for Medical Research and Occupational Health. It is adjacent to a two-lane road in each direction (north–south and south–north), and local air pollution is primarily influenced by residential heating and moderate traffic, with reduced impact from major roadways (Figure 1a). In contrast, location Center (45°48′38″ N, 15°58′04″ E) is an urban traffic station, situated within the dense city center, in proximity to three lanes running

west–east and east–west, two lanes heading south–north, and one lane in the opposite direction, as well as multistory urban buildings. It reflects the influence of substantial traffic density, central heating systems in older buildings, and limited air dispersion due to narrow streets and compact urban structures (Figure 1b). Location South (45°46′23″ N, 15°58′47″ E), positioned in Novi Zagreb, is also classified as an urban traffic station. It is strongly influenced by local traffic along broad avenues and a nearby intersection with three lanes in each cardinal direction (north–south, south–north, east–west, and west–east), domestic heating in large residential blocks, and additional contributions from nearby industrial activities and waste management facilities (Figure 1c). The positions of the sampling locations, together with the street view of the sampling locations, are presented in Figure 1.

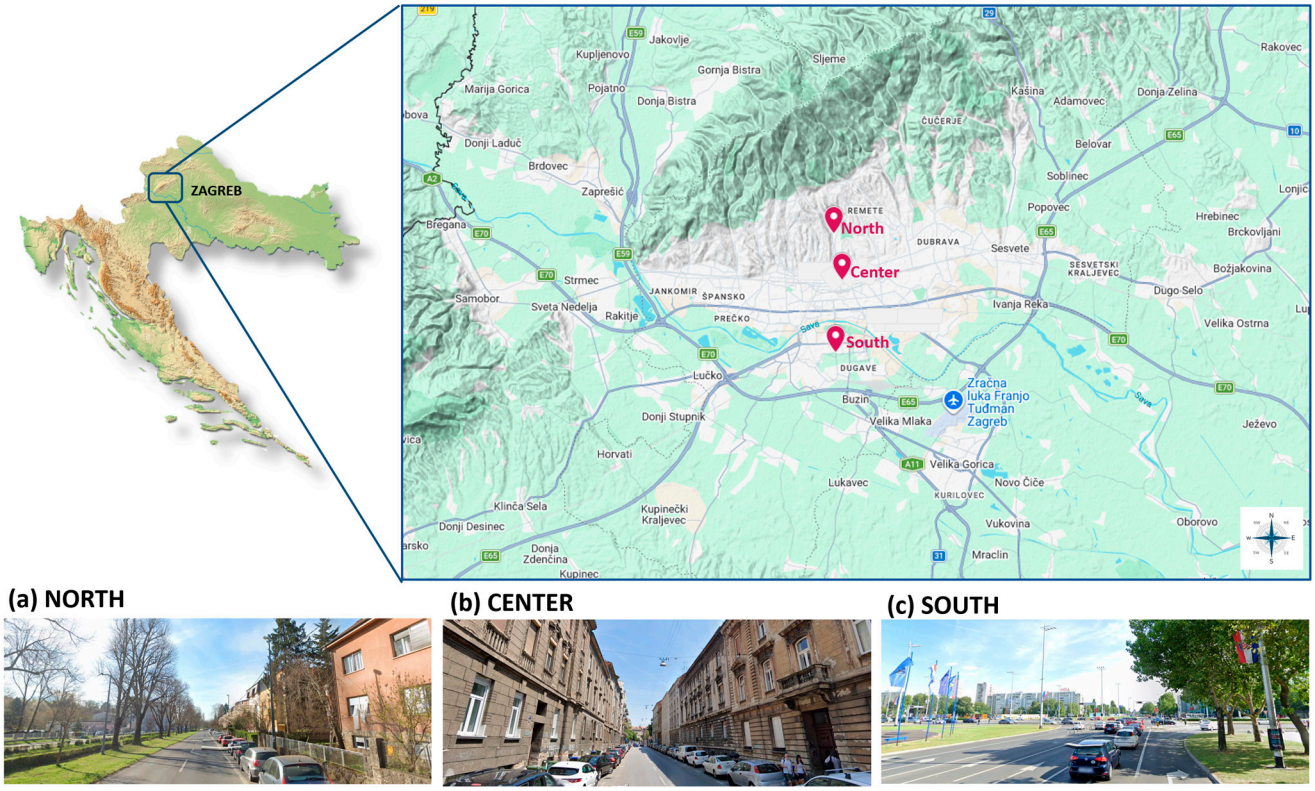

Figure 1. Geographic location of (**a**) Zagreb, Croatia, and street view of sampling locations (**a**) North (45°50′07″ N, 15°58′38″ E), (**b**) Center (45°48′38″ N, 15°58′04″ E), (**c**) South (45°46′23″ N, 15°58′47″ E). Source: Google Earth.

2.2. Determination of Platinum, Palladium, and Rhodium

The analytical method for the determination of Pt, Pd, and Rh has been developed and reported in our earlier publications [44,45]. Weekly samples of PM_{10} collected on quartz filters (Pall Life Sciences, Port Washington, NY, USA) were prepared in acid (25% HNO_3, *v*/*v*) by microwave digestion at high pressure and temperature using an Ultraclave system (Milestone, Sorisole, Italy). Following digestion, a small portion of hydrochloric acid was added to stabilize the analytes, after which the solution was diluted with distilled water and stored until analysis. An inductively coupled plasma mass spectrometer (ICP-MS, 7500cx, Agilent Technologies, Santa Clara, CA, USA) was used for analysis, with the selected isotopes ^{103}Rh, ^{105}Pd, ^{195}Pt, and ^{115}In as the internal standard. The ICP-MS was adjusted to maintain oxide and doubly charged ion ratios below 1%. The carrier and make-up flow were tuned to enhance sensitivity while reducing spectral interferences. Calibration standards (in 5% HNO_3/1% HCl, *v*/*v*) were prepared from certified stock solutions (Inorganic Ventures, Christiansburg, VA, USA; 1000 µg/mL) of Rh, Pd, and Pt,

covering nine concentration levels from 0.05 to 4.80 ng/mL. Calibration was performed before each sample analysis.

Quality Control

Analytical performance was evaluated through limits of detection (LOD), accuracy, and precision. The LOD were 0.177 pg/m^3 for Rh, 0.908 pg/m^3 for Pd, and 0.045 pg/m^3 for Pt. Accuracy and precision were evaluated by spiking known amounts of Pt, Pd, and Rh standards onto reference materials (NIST 1648 and ERM CZ120, PM10-like). Recoveries ranged from 86 to 104% for Pt, 91–115% for Pd, and 90–108% for Rh. Precision, expressed as the relative standard deviation (RSD) of 10 consecutive CRM measurements, was 5% for Pt, 6% for Pd, and 5% for Rh. These results confirm the robustness and reliability of the analytical procedure.

2.3. Data Analysis

The analysis was conducted using the Python programming language (www.python.org, v3.10, accessed on 1 February 2026). The dataset included weekly mass concentrations of Pt, Pd, and Rh measured at three urban locations in Zagreb (North, Center, and South) over five years (2016–2020). Mass concentrations were expressed in picograms per cubic meter (pg/m^3). Before statistical analysis, all values below the limit of detection (LOD) were set to one-half of the corresponding LOD for each element to ensure consistency across the dataset [46]. Sensitivity analysis using alternative LOD imputations (0, LOD, and LOD/$\sqrt{2}$) showed minimal impact on descriptive statistics (Table S1), ANOVA results, correlation structure (Table S2), and PCA loadings. Also, a seasonal comparison was conducted. Rather than the calendar year, the seasons were defined by meteorological conditions: winter (December–February), spring (March–May), summer (June–August), and autumn (September–November). This approach aligns with standard practices in urban air quality research, where meteorologically defined seasons better capture the dynamics of pollutant behavior. Seasonal differences among the three locations were evaluated separately for each metal using one-way analysis of variance (ANOVA). Homogeneity of variances was assessed using Levene's test. When variances were homogeneous, classical one-way ANOVA was applied, followed by Tukey's honestly significant difference (HSD) post hoc test for pairwise comparisons. When variances were unequal, Welch ANOVA was used, followed by Games–Howell post hoc comparisons. All tests were two-sided with $\alpha = 0.05$. As a sensitivity analysis, two-way ANOVA including Location, Year, and their interaction (Location $\times$ Year) was conducted by season for each metal to assess the temporal stability of spatial contrasts. Associations among Pt, Pd, and Rh across stations were evaluated using Spearman correlation. Principal component analysis (PCA) was performed on standardized (z-scored) weekly Pt, Pd, and Rh mass concentrations to explore shared variability and potential common sources. To assess station separation along the first principal component (PC1), differences in PC1 scores were tested using the Kruskal–Wallis test, followed by pairwise Mann–Whitney U tests with Holm adjustment. All tests were two-sided with $\alpha = 0.05$.

2.4. Traffic Data

Information on the number and types of vehicles was obtained from the Croatian Vehicle Centre (CVH), which provides the only publicly available long-term dataset describing the vehicle fleet for the City of Zagreb and Zagreb County. These data include annual counts of vehicles undergoing first registration, registration renewal, or regular technical inspection, categorized by fuel type (petrol, diesel, hybrid, electric). Site-specific or continuous traffic counts for the three monitoring locations are not available; therefore, these broader fleet-level data represent the most consistent indicator of vehicle compo-

sition relevant to the urban area. Previously published traffic measurements for these locations [45] are referenced in the Results and Discussion section, where they support the interpretation of spatial patterns.

3. Results and Discussion

3.1. Spatial and Temporal Distribution of Pt, Pd, and Rh

Using data from an ICP-MS analysis of weekly PM_{10} samples collected over five years at three locations with different traffic loads enabled a detailed assessment of the spatial and temporal distribution of PGEs in Zagreb's urban air.

The variations in Pt, Pd, and Rh mass concentrations measured at three locations in Zagreb (North, Center, and South) during the sampling period from January 2016 to December 2020 are shown in Figure 2. The sampling period on the x-axis represents the week, month, and year of sample collection. All three analyzed metals exhibit comparable temporal trends, characterized by fluctuations and recurrent winter peaks, suggesting the influence of seasonal factors, such as increased vehicular emissions during colder months as well as unfavorable meteorological conditions that reduce air circulation and cause pollutant accumulation near the ground.

At all three locations, Pd concentrations were higher than those of Pt and Rh, indicating its broader use in modern catalytic converters. The first catalytic converters used Pt and Rh in ratios of 5–20:1, but this composition gradually shifted mainly due to fluctuations in metal prices and availability. Later, in three-way converters, the ratio of platinum, palladium, and rhodium shifted to Pt:Pd:Rh = 0–1:8–16:1 [47], reflecting the increasing substitution of Pt with Pd because of their similar catalytic properties, greater availability, and lower cost. Given the oscillations in Pt and Pd prices shown in Figure S1 (source: LBMA Platinum and Palladium Price Data [48]), with Pt being more expensive until 2019, when Pd prices surpassed it, a future shift in elemental concentrations within converters may occur, potentially influencing urban emission profiles. The South location generally exhibited slightly elevated levels of all PGEs, as expected due to the higher traffic density in the surrounding area. A gradual decrease in Pt and Rh mass concentrations toward the end of the monitoring period may reflect changes in vehicle technology or fuel composition, whereas the increase in Pd concentrations suggests element-specific emission dynamics.

Descriptive statistics for Pt, Pd, and Rh mass concentrations by location and sampling year are given in Table S3. The highest mean mass concentrations for all three metals were generally observed in 2016, while the lowest values occurred in 2019. At all three locations, the highest values were obtained for palladium and the lowest for platinum. As shown in Figure 2, the most pronounced short-term peaks for all three elements occurred during winter and, to a lesser extent, autumn, reflecting reduced atmospheric dispersion and increased seasonal emissions. These seasonal and interannual patterns are consistent with the overall temporal trends observed across the full monitoring period.

In France, Omrani et al. (2021) [9] sampled PM_{10} along a boulevard in Nantes, a high-density traffic site with a speed limit of 30 km/h. They collected eight samples from November to May and reported mean values below the quantification limit (1.6 pg/m^3) for Pt, 1.29 pg/m^3 for Pd, and 0.10 pg/m^3 for Rh. In the United States, Das and Chellam (2020) [49] obtained average mass concentrations of 3.28 pg/m^3 for Pt, 10.64 pg/m^3 for Pd, and 1.48 pg/m^3 for Rh. These concentrations are higher than those determined in this study, which is expected given the sampling locations. Specifically, the Houston site was situated in an area characterized by exceptionally high traffic density, with multiple major roadways and highways in proximity (245 m), resulting in a strong and continuous influence of vehicular emissions. Similar comparisons have been made in previous studies, in which Pt, Pd, and Rh concentrations were compared with those reported in other cities

worldwide, yielding similar conclusions. Higher concentrations were consistently observed at high-density traffic locations. Across all studies, Pd tended to dominate, followed by Pt and Rh, and mass concentrations remained within the pg/m^3 range [43,45].

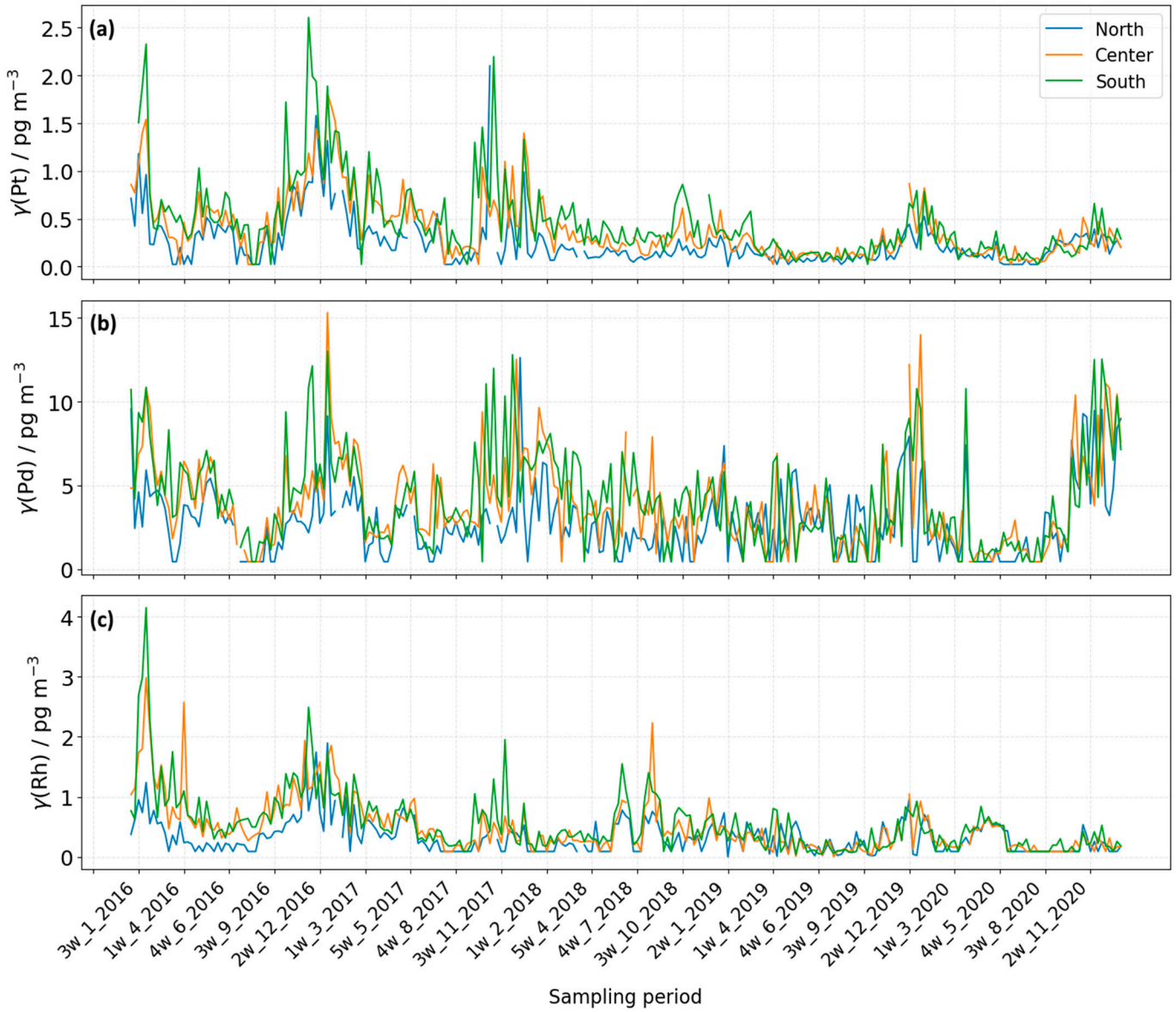

Figure 2. Temporal variation in Pt (**a**), Pd (**b**), and Rh (**c**) mass concentrations (pg/m^3) at three locations in Zagreb (2016–2020). The x-axis shows the sampling period expressed as week, month, and year.

According to the annual climate data provided by the Croatian Meteorological and Hydrological Service (Table S4), both 2016 and 2019 were warmer than the 1991–2020 climatological average in the Zagreb area, with thermal conditions classified as warm in 2016 and extremely warm in 2019. At the same time, total annual precipitation was categorized as normal in 2016 and wet in 2019. The elevated mass concentrations of PGEs observed in 2016 may be explained by unfavorable meteorological conditions, including reduced precipitation and frequent temperature inversions, which limit atmospheric dispersion and enhance pollutant accumulation. This combination likely contributed to atmospheric stability and reduced vertical mixing, resulting in the highest recorded PGE mass concentrations in 2016 compared to other sampling years. In contrast, although 2019 was classified as an extremely warm year, the above-average precipitation likely facilitated wet deposition

and atmospheric cleansing, leading to lower PGE mass concentrations than in the previous sampling years.

These findings highlight the importance of using meteorological and urban traffic data when assessing pollution dynamics. Such insights are valuable for developing targeted mitigation strategies and promoting sustainable urban planning in cities with similar environmental burdens. The elevated palladium mass concentration measured at location North in 2020 may be partially attributed to local construction activities associated with the project of the reconstruction and expansion of the Institute for Medical Research and Occupational Health (ReC-IMI), which spanned from early 2020 to late 2023 [50]. These activities included the demolition of one of the Institute's buildings, located approximately 100 m from the sampling site, as well as increased traffic in the immediate vicinity. Both of these factors may have contributed to temporary increases in airborne particulate matter and, consequently, Pd mass concentrations. According to the annual air-quality assessment reports for the city of Zagreb [51], the monitoring station Ksaverska cesta (45°50′07″ N, 15°58′38″ E) recorded higher PM_{10} mass concentrations in 2020 (23 μg/m^3) compared to 2019 (21 μg/m^3). In addition, the maximum recorded PM_{10} concentration in 2020 (185 μg/m^3) was substantially higher than in 2019 (64 μg/m^3), and the 98th percentile also increased from 55 μg/m^3 in 2019 to 78 μg/m^3 in 2020, suggesting more frequent or more intense short-term pollution episodes.

Seasonal differences in PGE mass concentrations are already evident in Figure 2. These seasonal patterns are significant in urban environments such as Zagreb, where the combination of traffic density, built-up infrastructure, and meteorological conditions contributes to the accumulation of pollutants. These seasonal peaks were statistically confirmed by one-way ANOVA and post hoc tests (Table S5), indicating that the observed temporal differences are not only descriptive but reflect significant underlying variability.

Seasonal differences in PGE mass concentrations were evident throughout the whole monitoring period (Figure S2), with higher concentrations observed during the colder part of the year. With this in mind, yearly seasonal differences were further examined. Figure 3 shows average seasonal mass concentrations of Pt, Pd, and Rh at all three locations, North, Center, and South, for each year of monitoring. Yearly seasonal distribution of PGE mass concentrations confirmed the previously mentioned observation of higher concentrations during the colder part of the year. It revealed an overall decreasing trend from 2016 to 2019, followed by a slight increase in 2020.

Understanding these temporal shifts is necessary for designing effective air quality strategies in urban areas, especially in relation to seasonal traffic patterns and heating-related emissions.

Regarding the slight increase in seasonal average mass concentrations in 2020, several overlapping factors may have influenced urban air pollution dynamics in Zagreb. The COVID-19 pandemic introduced substantial changes in mobility pattern, including travel restrictions between counties [52], and a marked decline in public transport usage [53,54]. Additionally, the March 2020 earthquake of magnitude 5.3 Mw (Richter scale) [55] disrupted daily routines and urban infrastructure, potentially affecting traffic flow and contributing to localized congestion due to reconstruction work. Although national vehicle mileage decreased by 2.6% [56] in 2020, these concurrent disruptions suggest that mobility patterns were atypical and spatially heterogeneous. In the absence of detailed traffic count data, these influences should be interpreted cautiously and cannot be confirmed as contributing factors to the observed 2020 anomaly. The highest mass concentrations of Pt, Pd, and Rh were usually determined during the colder seasons, particularly in winter and autumn, while the lower values were, as expected, determined for spring and summer. This pattern suggests a strong seasonal influence of traffic-related emissions, reduced atmospheric

dispersion, and meteorological conditions like temperature inversions and lower mixing heights during colder months.

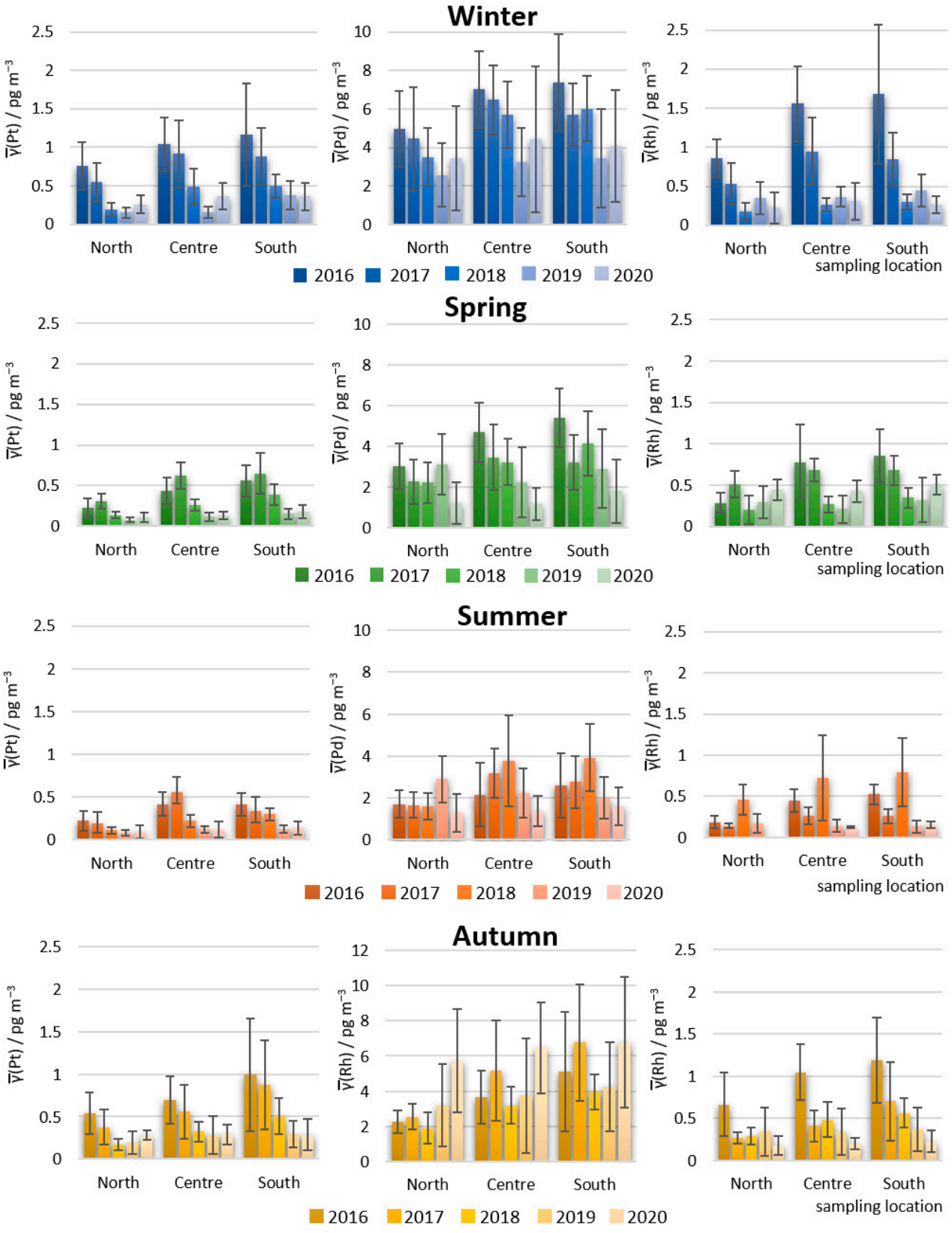

Figure 3. Average seasonal mass concentrations of Pt, Pd, and Rh at all locations (North, Center, South) throughout the whole monitoring period (2016–2020). Error bars represent the standard deviation (SD) of seasonal mass concentrations.

Seasonal differences among the three sampling locations were statistically significant for all three metals (Pd, Pt, and Rh), as confirmed by one-way ANOVA or Welch ANOVA ($p < 0.05$; Table S5). Post hoc comparisons (Tukey HSD or Games–Howell) showed that these differences were mainly driven by the urban background location North, which consistently exhibited lower concentrations relative to the more traffic-influenced Center and South locations. In contrast, differences between the two urban traffic locations were not consistently significant across seasons (Table S6), indicating that the main spatial pattern reflected the distinction between the urban-background site and the two traffic-influenced sites, rather than differences between the two urban traffic locations themselves.

A sensitivity analysis using two-way ANOVA (Location, Year, Location × Year) confirmed significant Location and Year effects across all seasons for Pt, Pd, and Rh (Table S7). The Location × Year interaction was significant for several metal-season combinations, suggesting that these spatial differences did not remain constant across years. Overall, PGE mass concentrations tended to increase from the urban-background location North toward the traffic-influenced Center and South locations, but the spatial contrast varied by season and year. Year-by-year seasonal analyses (results indicated in Table S8) showed that significant differences were most often observed between the location North and either the location Center or South, while differences between Center and South were generally rare. For Rh and Pd, significant differences between stations appeared mainly in the earlier years of the study (especially 2016–2018). Pt, on the other hand, showed the most consistent differences between stations in spring throughout the entire period, with occasional differences between Center and South (only in 2018). Taken together, these findings outline a clear and consistent spatial pattern across the dataset.

These differences are also visually evident in Figure 4, which illustrates spatial differences in PGE mass concentrations measured at the North, Center, and South locations across all of the sampling years. Median concentrations of all three elements followed a similar trend, with the location South generally exhibiting the highest concentrations, and the North the lowest. This gradient reflected the influence of local emission sources, particularly vehicular traffic and urban activity, which are more pronounced in the southern and central parts of the city. The spatial gradient was supported by significant differences in median concentrations (Figure 4) and by the Location effect in the two-way ANOVA (Table S7), confirming that traffic intensity is a consistent determinant of PGE levels across years. The higher variability and presence of outliers at location South further indicated episodic pollution events or stronger local contributions. This aligns with the urban layout and traffic density, particularly at the locations Center and South, which experienced greater vehicular flow compared to location North. Our findings highlighted how the spatial distribution of pollutants reflects a city's functional structure, underscoring the value of environmental data for understanding and managing urban systems.

The correlation matrix shown in Figure 5 reveals the relationships among Pt, Pd, and Rh mass concentrations measured at the North, Center, and South locations. Strong positive correlations were observed for the same metals across different locations; for example, the correlation coefficient for Pt_Center and Pt_South is 0.81, as is the case for Rh_Center and Rh_South, indicating coherent spatial trends driven by common urban emission sources, such as traffic.

Within each location, PGEs also showed moderate-to-strong correlations, particularly between Pt and Rh, supporting their shared origin from vehicular catalytic converters. Slightly weaker correlations for Pd suggest greater chemical variability in the atmosphere. Most correlations were statistically significant ($p < 0.05$), as indicated in Figure 5, confirming that these relationships likely reflect true associations rather than random variations. Over-

all, the consistent positive relationships across locations confirm traffic-related emissions as the dominant regional source of PGEs in Zagreb's urban air.

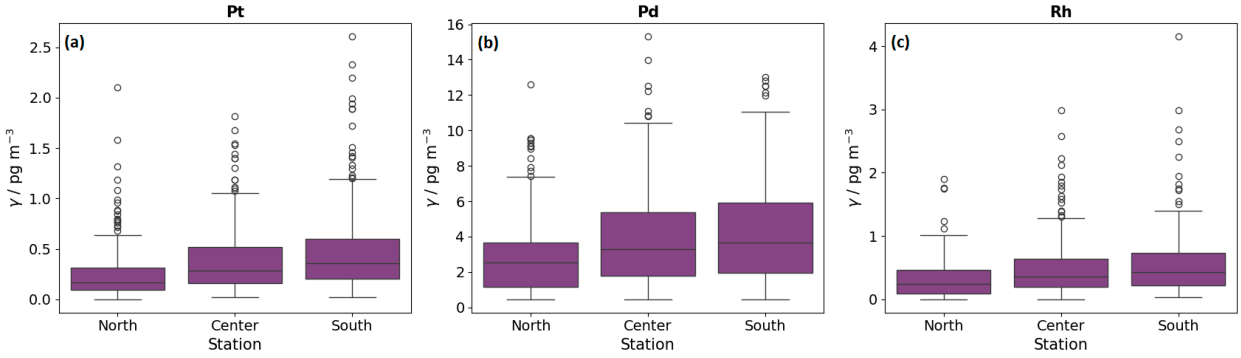

Figure 4. Spatial distribution of (**a**) Pt, (**b**) Pd, and (**c**) Rh mass concentrations at all of the monitoring locations (North, Center, South) for all of the sampling years (2016–2020). The top and bottom of the box represent the 75th and 25th percentiles, respectively. The line inside the box shows the median. The whiskers show the minimum and maximum non-outlier values, and points outside the whiskers indicate outliers.

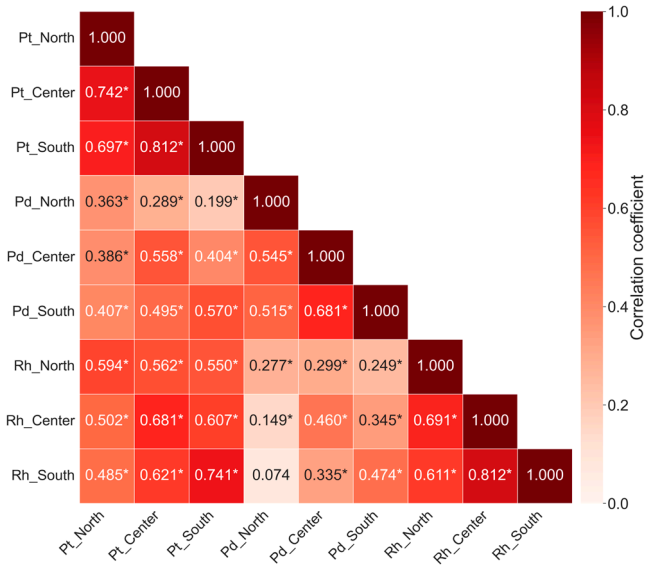

Figure 5. Spearman correlation matrix of Pt, Pd, and Rh for all monitoring locations (North, Center, South) for the period 2016–2020. * Statistically significant correlations ($p < 0.05$).

Principal Component Analysis (PCA) was applied to explore associations among Pt, Pd, and Rh mass concentrations and to confirm a potential common source influencing their variability. Before the analysis, weekly concentrations from all three monitoring stations were standardized (zero mean and unit variance) to eliminate the influence of different concentration scales between elements. PCA was performed on the standardized dataset using the covariance matrix, and the first two components (PC1 and PC2) were retained for interpretation. Principal Component 1 (PC1) represents the shared variance among Pt, Pd, and Rh and reflects their dominant common emission source, consistent with vehicular catalytic converters. Principal Component 2 (PC2) captures the residual variability not explained by PC1 and is associated with local influences such as site-specific traffic patterns, resuspension processes, and short-term meteorological effects. This multivariate approach enabled dimensionality reduction while explaining the dominant variance structure in the dataset, allowing identification of shared emission sources and subtle spatial or seasonal differences among stations.

PCA scores of weekly Pt, Pd, and Rh mass concentrations measured at three monitoring locations are shown in Figure 6. The first principal component (PC1), which explains 71.7% of the total variance, shows strong positive loadings for all three metals (Pt = 0.901, Pd = 0.773, Rh = 0.863), and reflects the overall concentration level of PGEs. In contrast, PC2, which accounts for 19.0% of the total variance, is dominated by Pd (loading = 0.629; 69.4% contribution) and places Pd in opposition to Rh (−0.379; 25.2%) and Pt (−0.177; 5.5%). This component, therefore, captures variability in the relative enrichment or depletion of Pd compared with Pt and Rh. Such a Pd versus (Pt, Rh) contrast may reflect temporal changes in catalyst formulation and fleet composition and/or differences in atmospheric behavior and resuspension of Pd-containing particles. Although the score clusters overlap, PC1 differed systematically among stations (Kruskal–Wallis H = 70.963, $p = 3.9 \times 10^{-16}$; $\varepsilon^2 = 0.089$), with higher median PC1 values at the location South and lower at the North. This indicates a modest but consistent spatial gradient in the overall PGE levels, reflecting stronger traffic influences at location South, while the overlap among clusters suggested that city-wide temporal drivers (e.g., seasonality and meteorology) affected all of the stations simultaneously. Pairwise comparisons (Mann–Whitney U with Holm correction, Table S9) indicated significant differences between locations North and Center, North and South, and a smaller difference between locations Center and South.

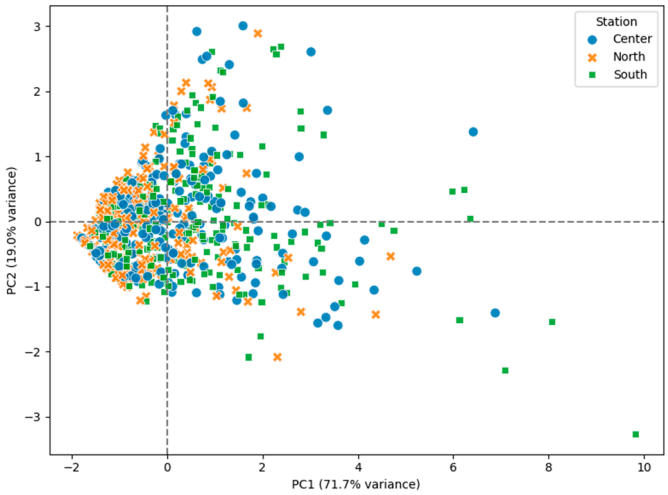

Figure 6. Principal component analysis (PCA) scores of weekly Pt, Pd, and Rh mass concentrations measured at locations North, Center, and South. PC1 and PC2 represent the first and second principal components, explaining 71.7% and 19% of the total variance, respectively.

The PCA results for individual monitoring locations (Figure S3) showed that the first component (PC1) explained the majority of the variance: 61.3%, 71.3%, and 73.2% for the locations North, Center, and South, respectively, reflecting a single dominant source influencing Pt, Pd, and Rh mass concentrations. The second component (PC2) accounted for less than 26% of the variance and showed no apparent seasonal clustering, indicating that the relative proportions of PGEs remained consistent throughout the year. The strong positive loadings of all three metals on PC1 further support a shared emission source, consistent with vehicular catalytic converters, while the PC2 contrast highlights temporal shifts in catalyst composition rather than independent sources.

3.2. Relationship Between Personal Vehicle Type and Airborne PGEs

To assess the impact of personal vehicle composition on urban air quality, temporal trends in vehicle types were compared with ambient average yearly mass concentrations of PGEs measured at the three monitoring locations in Zagreb over the period 2016–2020 (Figure 7). The vehicle categories included petrol, diesel, hybrid, and electric.

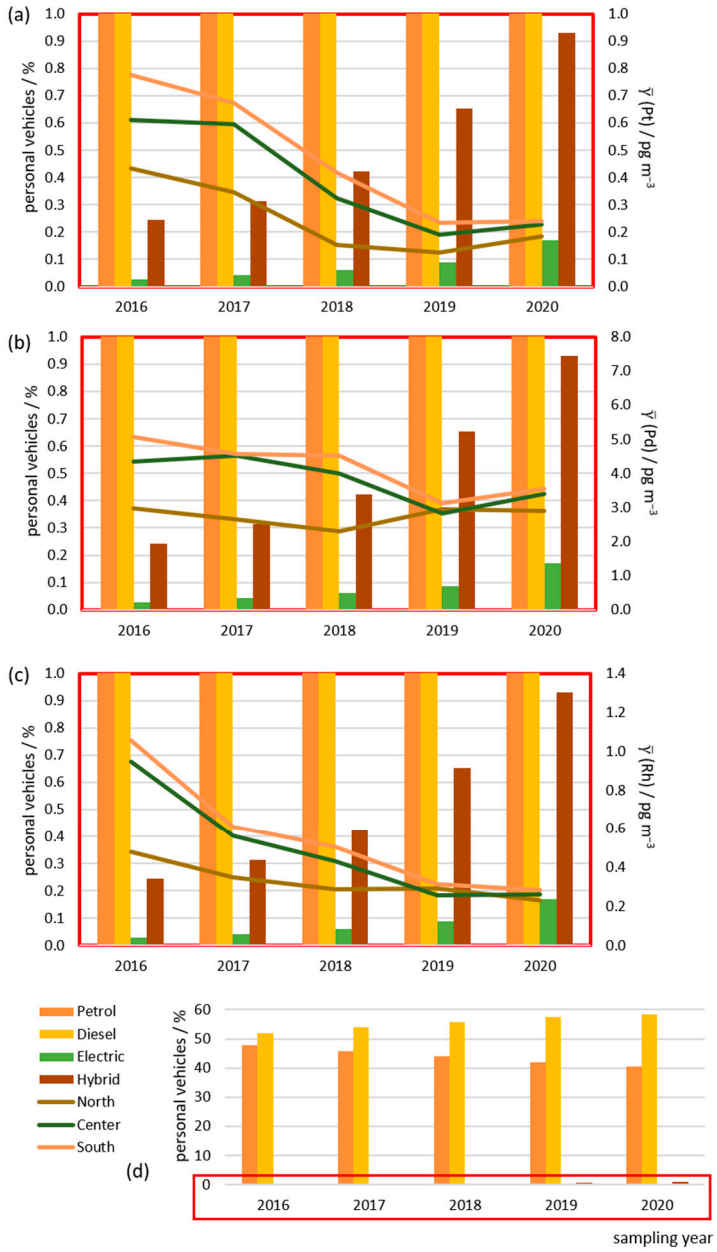

Figure 7. Percentage distribution of personal vehicle types (petrol, diesel, hybrid, and electric) compared with average yearly mass concentrations of Pt (**a**), Pd (**b**), and Rh (**c**) during the 2016–2020 sampling period. Graph (**d**) shows trends between petrol and diesel vehicles, while graphs (**a**–**c**) are expanded to highlight the increasing shares of hybrid and electric vehicles. Source: Center for Vehicles of Croatia (CVH).

Petrol vehicles are equipped with three-way catalytic converters that typically contain Pt, Pd, and Rh, with Rh playing a key role in reducing nitrogen oxides. Diesel vehicles, by contrast, rely mainly on oxidation catalysts and particulate filters, which predominantly use Pt and Pd but generally do not include Rh. Hybrid vehicles, depending on whether they are petrol- or diesel-based, emit PGEs in proportion to their conventional engine component. However, overall emissions are reduced due to lower fuel consumption [57]. Electric vehicles, which do not use combustion engines, are not associated with direct PGE emissions.

Figure 7 compares the distribution of vehicle types (petrol, diesel, hybrid, and electric) during the five-year sampling period (d) with the average yearly mass concentrations of Pt (a), Pd (b), and Rh (c).

Long-term, site-specific traffic density data for the sampling locations are not publicly available. The only detailed traffic information for these areas originates from measurements conducted within the project 'Master Plan of the Traffic System of the City of Zagreb, Zagreb County and Krapina-Zagorje County', which were provided for our previous study [45]. These measurements showed a clear spatial pattern: traffic density increased in the direction North < Center < South, corresponding to the observed gradient in PGE mass concentrations in Zagreb [45]. Since no continuous or historical traffic counts exist for the full sampling period of this study, the annual vehicle statistics from the Croatian Vehicle Centre (CVH) were used as the only consistent publicly available dataset describing the vehicle fleet relevant to the wider urban area. Moreover, because no substantial changes in traffic regulations occurred at the sampling locations during these years, the CVH data provide a stable and representative indicator of the traffic conditions affecting the monitored sites. Croatia is administratively divided into 20 counties and the City of Zagreb, which holds county-level status. For this study, data from the city of Zagreb and Zagreb County were included, as these areas are functionally integrated and daily commuting patterns result in shared traffic flows. Over the five years, diesel vehicles maintained a dominant share, while hybrid and electric vehicles showed a steady increase, particularly after 2018. The number of petrol vehicles gradually declined, from 48% in 2016 to 40% in 2020, as shown in Figure 7d. These shifts in the distribution of vehicle types were reflected in the measured PGE mass concentrations, though with varying sensitivity. Figure S4 provides additional insight into these associations, revealing distinct correlation patterns between specific vehicle categories and measured PGE levels across urban zones. Platinum levels showed yearly variability, with elevated concentrations coinciding with a higher share of petrol vehicles. Also, strong positive correlations were observed with the share of petrol vehicles across all locations (0.815, 0.924, 0.963 for the locations North, Center, and South, respectively), while diesel share showed strong negative correlations (−0.897, −0.949, −0.980 for the locations North, Center, and South, respectively). This pattern reflected the dominant contribution of petrol three-way catalytic converters to Pt emissions. Electric and hybrid vehicle shares also showed negative correlations with Pt, although these trends likely reflected their parallel increase as petrol share declined rather than a direct emission effect. In addition, the price of Pt and Pd may have contributed to the decrease in Pt levels and the relatively stable Pd levels across locations and years. Pd concentrations were also negatively correlated with diesel share and positively correlated with petrol share at the locations Center (0.887, −0.904) and South (0.840, −0.836). At the location North, Pd concentrations showed a weak positive correlation with diesel share, potentially reflecting local traffic patterns and the influence of nearby construction activities in 2020. Correlations with electric and hybrid vehicles were generally weak to moderate and likely reflect broader temporal trends rather than causal relationships. Rh mass concentrations displayed a consistent downward trend. Petrol share exhibited strong positive correlations with Rh mass concentrations (0.756, 0918, 0.907 for the locations North, Center, and South, respectively), whereas diesel share showed strong negative correlations at all sites (−0.808, −0.957, −0.949 for the locations North, Center, and South, respectively). Given that Rh is primarily used in petrol vehicle catalytic converters and is generally absent from diesel emission systems, its decline follows the decreasing share of petrol vehicles and the rise in number of hybrid and electric alternatives. This suggests that vehicle electrification could yield measurable reductions in Rh emissions in the future. Although a direct causal link cannot be established due to the absence of site-specific traffic counts, the strong and consistent correlations between the share of petrol vehicles and the corresponding Pt and Rh concentrations, together with the opposite pattern observed for diesel vehicles, provide evidence that changes in fleet composition meaningfully influence the observed PGE trends.

It should also be considered that PGEs are bound to particulate matter and, once emitted into the environment, can be deposited and later subsequently released into the air through the resuspension of road dust. Their retention or removal may depend on local meteorology and topography, as well as on each metal's specific ability to enter surrounding vegetation or migrate into the soil. Consequently, changes in traffic emissions may not always be immediately reflected in the ambient air concentrations, but they will eventually become evident in long-term trends.

3.3. Study Interpretation Limitations

When interpreting these findings, several limitations should be acknowledged. The absence of site-specific traffic counts restricted the ability to directly quantify emission intensity at each monitoring location, requiring reliance on fleet-level vehicle statistics as the only consistent long-term dataset available for Zagreb. Although these data provided valuable insight into broader temporal trends, they cannot fully capture short-term or location-specific traffic dynamics, which may have contributed to some of the observed variability. The lack of information on PGE chemical speciation represents an additional constraint, as total mass concentrations do not reflect the toxicological relevance or environmental behavior of individual species. Furthermore, atypical events during the monitoring period, such as the 2020 earthquake, pandemic-related mobility changes, and local construction activities near the northern site introduced additional variability that could not be fully resolved with the available data. Despite these limitations, the consistency of the spatial and seasonal patterns, supported by statistical analyses and multivariate results, indicates that the main conclusions remain robust, while also highlighting clear directions for future research.

4. Conclusions

This study provides a comprehensive assessment of platinum group elements (PGEs) in PM_{10} across the city of Zagreb, revealing how urban structure, traffic density, and seasonal meteorology shape the spatial and temporal distribution of these emerging pollutants. Over a five-year monitoring period, all three of the analyzed elements, Pt, Pd, and Rh, exhibited comparable temporal trends, with recurrent winter peaks and a general decline in mass concentrations from 2016 to 2019. These patterns reflected the influence of colder season emissions, atmospheric stability, and gradual changes in vehicle technologies or fuel compositions. The consistently higher palladium levels aligned with its increasing use in modern catalytic converters while the overall decline in PGE mass concentrations suggests a slow shift in urban emission profiles.

Seasonal analysis confirmed that PGE mass concentrations were highest in winter and autumn, driven by increased vehicular activity, domestic heating, and meteorological factors that limit dispersion. Although a slight increase was observed in 2020, the available data do not allow attribution to a single cause, given the overlapping effects of pandemic-related mobility changes and earthquake-related disruptions. Spatial differences in PGE mass concentrations were most pronounced between the northern residential station and the more traffic-exposed central and southern locations which highlight the importance of local urban characteristics when interpreting air quality data.

Correlation analysis and principal component analysis (PCA) confirmed a shared dominant emission source, consistent with vehicular catalytic converters, with minor local variability. The observed associations between vehicle fleet composition and Pt and Rh mass concentrations, together with the opposite pattern for diesel vehicles, suggest that changes in fleet structure meaningfully influence long-term PGE trends. While PGEs can therefore provide useful insight into traffic-related emissions, their interpretation should

account for the broader urban context, including meteorology, resuspension processes, and non-traffic activities. In addition, ongoing urban mobility and air-quality measures in Zagreb, such as the expansion of cycling infrastructure, the introduction of a public bike sharing system, and actions outlined in the city's Air Quality improvement plan, may further influence emission patterns. These elements highlight the need to interpret PGE trends within dynamic and evolving urban environment rather than attributing changes to a single factor. Even with these considerations, our findings indicate the PGEs remain useful indicators of traffic related emissions and that their spatial patterns are broadly consistent with the city's functional and morphological structure. However, these interpretations should be viewed in light of the complex and evolving urban context described above. It is important to note that no regulatory limit values currently exist for PGEs in ambient air, and that their toxicological relevance depends strongly on chemical speciation. The mass concentrations measured in this study fall within the range typically reported for urban environments and do not indicate acute health risks. However, given the limited understanding of long-term exposure effects and the potential bioaccessibility of certain PGE species, continued monitoring and further toxicological research remain essential for a more complete assessment of their environmental and public health significance.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/environments13050254/s1, Table S1. Mean concentrations (pg/m^3) by substitution method and % deviation from LOD/2; Table S2. Spearman correlation coefficients between locations by substitution method; Table S3. Descriptive statistics for Pt, Pd and Rh mass concentrations (pg/m^3) at three locations (North, Center, South) by sampling year (2016–2020); Table S4. Annual climate indicators for Zagreb (2016–2020): temperature anomalies (°C), mean temperatures (°C), precipitation amount (%) and totals (mm), and percentile-based classifications provided by the Croatian Meteorological and Hydrological Service; Table S5. Summary of ANOVA-based statistical test assessing seasonal patterns in Pd, Pt, and Rh mass concentrations across monitoring locations in 2016–2020 period ($p < 0.05$); Table S6. Post hoc comparisons (Tukey HSD and Games–Howell) for seasonal differences in Pd, Pt, and Rh mass concentrations across the three monitoring locations in 2016–2020 period ($p < 0.05$); Table S7. Summary of two-way ANOVA tests evaluating spatial, temporal, and interaction effects on seasonal Pd, Pt, and Rh mass concentrations in 2016–2020 period ($p < 0.05$); Table S8. Seasonal average of Pt, Pd, and Rh mass concentrations (pg/m^3) and corresponding spatial differences at three locations (North, Center, South) by sampling year (2016–2020); Table S9. Pairwise location comparisons of PC1 scores (Mann–Whitney U with Holm-adjusted *p*-values) from PCA of standardized weekly Pt, Pd and Rh concentrations; Figure S1. Daily prices of platinum and palladium in US dollars from 4 January 2000 to 31 December 2022; Figure S2. Seasonal variability of Pt, Pd, and Rh mass concentrations at all monitoring locations (North, Center, South) throughout the whole monitoring period (2016–2020); Figure S3. PCA bi-plot graph for different monitoring locations (North, Center, South) throughout the whole monitoring period (2016–2020); Figure S4. Pearson correlation coefficients between vehicle types and PGE mass concentrations for all monitoring locations (North, Center, South). *Statistically significant correlation ($p < 0.05$).

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