Green Metric Approaches in Gas Chromatography

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Introduction

Since introducing the first principles of green chemistry 25 years ago, ideas about sustainability and chemistry have progressed with solid and permanent steps, with companies adding sustainability officers and universities adding curriculum and chemistry-related processes. In this chapter, we will investigate the influence of green chemistry, which is not much talked about, on gas chromatography (GC). What is green chemistry by defining it? What is not? We will seek answers to your questions. Then is gas chromatography green? How can it be greener? We will put forward the latest developments in the literature by bringing these questions to the agenda.

The 12 Principles of Green Chemistry were explained in the late 1990s, and awareness was raised on this issue like many others. It was difficult and painful for humanity to enter the age of enlightenment after the dark age. However, scientific innovations and developments and the technology they bring are quite rapid. Especially the change after the Industrial Revolution is dizzying. Although these conveniences and changes make people happy, they have created another problem: environmental pollution. Although awareness of this situation and searches for a solution were expressed in the 1950s, the real attention-grabbing implementation was in the 1990s. In this regard, analytical chemistry and analysis methods have also played their part and interacted. In our age, analysis is everything; measuring, determining the existing amount, or seeking answers to what exists is a constant need in every field such as food, cosmetics, industry, medicine, etc. Therefore, it is important and necessary not to underestimate the effectiveness of analysis methods, to look for greener, easier methods, and to be ready for changes (Malissa & Roth, 1987; Pimentel & Coonrod, 1985).

More recently, the United Nations has established the 17 Sustainable Development Goals (UN SDGs) that guide policymaking across government and institutional platforms, including the 12 Principles of Green Chemistry and the UN Sustainable Development Goals, which focus on viable methods in chemical sciences and analytical chemistry research and development guides. This environmental movement is one of the most important points of development and sustainability. The understanding of quality also requires this. Because quality means sustainability, applicability, and convenience. Green chemistry touches upon sustainable points such as ease of application, less reagent use, analysis time, and analyzer health, and measures them with various metrics. These metrics appeal to visual perception as well as on a numerical scale. While it offers a general perspective on the developed or applied method, it also has detailed applications, examining each step. But no matter what view they present, their underlying measurement purpose is to protect human health, reduce environmental pollution, and consume less energy (Anastas, 1999; Keith et al., 2007).

Green analytical chemistry

Green analytical chemistry supports the reduction of toxic chemicals and waste and saves energy in the processes besides using minimum waste. In achieving these goals, the 12 principles of green chemistry serve as an important guide (Sajid & Płotka-Wasylka, 2022).

These principles of green analytical chemistry;

1. Waste management
2. Atomic economy
3. Less hazardous chemical synthesis
4. Scheming safer chemicals
5. More reliable solvents and excipients
6. Plan for energy capability
7. Greener syntheses with renewable raw materials
8. Reduction of by-products (derivatives)
9. Catalysis
10. Device of twist
11. Elimination of pollution for all time
12. More credible chemistry to prevent accidents (Sajid & Płotka-Wasylka, 2022; Sogut & Çelebi, 2020)

Over time, various measurements have been improved to evaluate the greenness of analytical procedures. These are specific or general and feasible for analytical methods. (Płotka et al., 2013; Sajid & Płotka-Wasylka, 2022).

Green analytical chemistry methods

Recent research indicates that to make analytical methodologies more environmentally friendly there needs to be a combination of small changes to current methods and big innovations that completely change how analysis is done. Some strategies include substituting reagents and solvents using automation and advanced flow techniques to reduce chemical usage miniaturization and measuring analytes directly in the field rather than through sampling (Płotka et al., 2013).

Analytical Method Volume Intensity (AMVI)

The analytical method of volume density is applied to evaluate liquid chromatographic methods. Determining the solvent used in this application and the total volume of waste resulting from the experiment with a specific procedure allows this method to be carried out (Imam & Abdelrahman, 2023).

Chemical hazard assessment for management strategies (CHEMS-1)

It is a strategic evaluation method that aims to put chemical release data into a systematic mechanism to evaluate the toxic effects of the chemicals used within the procedure in the experiment and the toxicity effects that may occur in case of exposure to these chemicals (Swanson et al., 1997).

Chromatography environmental assessment tools

This approach offers an extremely effective yet simple application method used to profile gas chromatography (GC) methods in terms of their suitability for green chemistry. Its goal is to reduce the damage caused by organic reagents to the environment and the public health used in GC methods. The environmental assessment tool (EAT) carefully examines the health, environmental, and safety topics for all chemicals included in the chromatographic methods. Also, it calculates a total score by rating the suitability of the different methods used with an understanding of green chemistry. It allows the comparison of calculation methods about their suitability for green chemistry (Gaber et al., 2011; Shi et al., 2023).

Life cycle assessment (LCA)

This method is applied to products to measure environmental impact factors (Jacquemin et al., 2012).

PROMETHEE: Preference ranking organization method for enrichment evaluations

The purpose of PROMETHEE, which is used as a multi-metric priority-setting method, is to be used as a decision support tool in problems such as selection and clustering. Here, the advantages and disadvantages of the options available for decision are compared, and the aim is to reach the most appropriate solution (Karasakal et al., 2019).

Modified eco-scale with green certificate

Aiming at the quantitative method of green parameters, the modified eco scale with green certification evaluates the process of hazards, reagents, energy, and waste. The application can be applied to sample preparation and analytical measurement procedures, while values are analyzed semi-quantitatively. The result of this analysis interprets the inherent toxicity and risks related to the use of reagents and assists in correct classifications. The disadvantages are that only numerical data is presented and no evaluation is made for sampling (Shi et al., 2023).

- ***HEXAGON***

This method, which aims to evaluate the optimum conditions or tests for analytical methods, can be applied to sample preparation and analytical measurement procedures and allows qualitative and quantitative evaluation to be carried out (Shi et al., 2023).

TOPSIS: Order of preference technique based on similarity to the ideal solution

Multi-criteria decision analysis is a group of tools used to score and rank alternatives according to the evaluation criteria studied, and TOPSIS is one of the multi-criteria decision analysis tools applied in the preference of the best alternative among others (Al-Hazmi et al., 2016; Nowak et al., 2020).

- ***RGB***

In the RGB metric, a global evaluation of analytical methods or procedures is achieved by utilizing the main colors red, green, and blue. The red (R) color represents the achievement of these methods, typically evaluated through classical validation techniques. The green (G) color means reliable and eco-friendly, such as hazards related to reagents or waste, energy consumption, and vocational dangers. The blue (B) color represents productivity and practical effectiveness. Each attribute is assigned a color score (CS) ranging from 0% to 100%, providing a comprehensive and transparent measurement, qualitatively or quantitatively. This approach enables the equation of different analytical procedures and predicts all possible applications for newly developed methods. However, the evaluation procedure can be tiring and difficult, and there is no clear method to detect the weights of the different criteria used in the evaluation (Shi et al., 2023).

NEMI: National Environmental Method Index

It is one of the oldest methods used to measure the greenness of analytical procedures, and this method creates a circle divided into four areas. Each area explains an alternative requirement of this method and is colored red or green depending on the requirements (Sajid & Płotka-Wasylka, 2022).

The requirement for green labeling of the initial field is that the chemicals used in the process must be toxic, bio-accumulative, and have no lasting effects. The second area requires no chemicals to be the harmful waste D, F, P, or U lists. If the pH range of the sample is between 2 and 12, the third area is marked green. Finally, the fourth area is marked green when the amount of waste generated in the process is 50 grams. NEMI pictogram is given for example in Figure 1(Sajid & Płotka-Wasylka, 2022).

Figure 1
An Example of NEMI Pictogram

In addition to the advantage of being easily understandable, NEMI has two important disadvantages: it takes a long time to collect information, and it is very general.

Analytical eco-scale

Analytical eco-scale, one of the most common green analytical chemistry methods, is measured by subtracting some penalty points from 100 points. Here, 100 points are defined as the ideal green analysis. After subtracting penalty points from this value, the closer the remaining value is to 100, the greener the analysis is. Penalty points vary depending on the quantity and quality of the chemicals to be used, the energy consumption in the process, occupational hazards in the conditions, the amount of waste to be generated, and the methods of processing the waste (Sajid & Płotka-Wasylka, 2022).

Penalty scoring begins with assigning penalty points to the solvent and reagent. Here, the pictograms of the chemical are considered. Each hazard pictogram on chemical bottles constitutes a penalty point; if the pictogram is classified as dangerous, this number is multiplied by two, and if the pictogram is classified as a warning, the number is multiplied by one. The resulting hazard score is multiplied by one if the chemical amount is less than 10 mL or 10 g, by two if it is between 10 and 100 mL (or grams), and by three if it is greater than 100 mL (or grams) (Sajid & Płotka-Wasylka, 2022; Soyseven et al., 2023).

When assigning penalty points to energy consumption, energy consumption per sample is considered (Sajid & Płotka-Wasylka, 2022).

- Energy consumption per sample ≤0.1 kWh = 0.
- Energy consumption per sample: ≤1.5 kWh = 1
- Energy consumption per sample: ≤1.5 kWh = 2

While assigning occupational hazard penalty points, three points are assigned for any gas or vapor released into the air. If the process is in seclusion, no points are assigned. Penalty points are given depending on the amount of waste and the treatment status of the waste (Sajid & Płotka-Wasylka, 2022; Tobiszewski et al., 2015).

Table 1

Scoring According To The Amount Of Waste And The Treatment Status Of The Waste

Amount of waste	**Penalty points**
<1 mL or 1 g	1
1–10 mL or 1–10 g	3
>10 mL or 10 g	5
Waste Treatment Method	**Penalty points**
Recycle	0
degradation	1
Passivation	2
No Purification	3

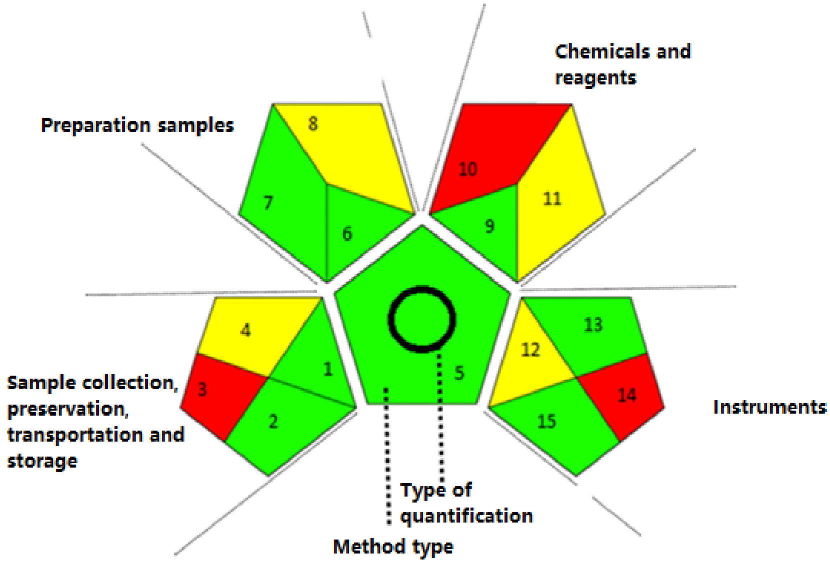
The final score is obtained by adding all the points and subtracting them from 100. A process with 75 or above is defined as optimum green analysis. A process with a scoring range of 50–75 is considered acceptable green analysis, while a process below 50 points is defined as insufficient green analysis (Sajid & Płotka-Wasylka, 2022).

Analytical eco scale gains an advantage by providing quantitative evaluation, as it is carried out with numerical evaluation out of 100. However, disadvantages arise due to the danger of pictograms and the effects of waste and chemicals on the environment not being fully taken into account (Sajid & Płotka-Wasylka, 2022; Tobiszewski et al., 2015).

- ***GAPI: Green analytical procedure index***

GAPI is a pictogram consisting of five pentagrams, as shown in Figure 1, and each pentagram here describes the environmental impacts of the analytical method.

Figure 2
GAPI Pentagram (Sajid & Płotka-Wasylka, 2022)



The first pentagram is divided into four parts. The first of these parts is related to the collecting sample method. If the sample is collected online, it is marked green; if it is collected online or offline, it is marked yellow; and if it is collected offline, it is marked red. The second field provides information about the preservation method of the sample. The third area concerns the transportation of samples. When special conditions are required for storage, the fourth field is marked in red (Sajid & Płotka-Wasylka, 2022).

The second pentagram gives information about the type of method, and this field is indicated by the number 5. If the method does not require sample preparation, the field is green-marked. If simple methods are used such as filtering during the sample preparation, the area is labeled yellow, and if complex methods such as extraction are required, the area is labeled red. The circle within this area indicates whether the method is used for quality or quantity. This method is used only for quality reasons, a circle is not included (Sajid & Płotka-Wasylka, 2022).

The third pentagram consists of three areas numbered 6, 7, and 8, which contain the various measures of this preparation. The area numbered 6 contains the extraction scale. It is marked green if the extraction is at the nanoscale and yellow if the extraction is done. The sixth area is labeled in red for practicing micro or macro scale. The area numbered 7 is related to the solvents used in the extraction stage. If the method is solvent-free, the field is labeled green, yellow when green reagents are used, and red when solvents are used except green. The area numbered 8 relates to the operations required by a method in addition to extraction. If the method requires simple steps such as solvent cleanup or removal, the area is marked yellow, while the area is marked green if it does not require additional processing. When more advanced processing is required, the area is labeled in red (Sajid & Płotka-Wasylka, 2022).

The fourth pentagram shows the amounts of chemicals and solvents used and the effects of these amounts on health. The pentagram consists of three areas, numbered 9, 10, and 11. Field number 9 relates to the volume or quantity of substances used. If this amount is less than 10 mL or 10 grams, the field is marked green. If this ratio is between 10 and 100 mL (g), the area is marked in yellow, while when it exceeds 100 mL, the area is shown in red. The area numbered 10 shows the effects of the substances used on health. If the health hazard score of all chemicals or solvents is 0 or 1, the field is shown in green; if this score is 2 or 3, the field is marked yellow; and if it is 4, the field is marked red. Field 11 indicates the health hazards of chemicals. The area is colored according to its flammability or instability score. If this score is 0 or 1, the field is marked green; if it is 2 or 3, the field is colored yellow; and if it is 4, the field is marked red (Sajid & Płotka-Wasylka, 2022).

The fifth pentagram includes areas 12-15 and information about the amount of power consumed by the devices, occupational hazards, wastes that will be generated, and the procedures these wastes are processed. Field 12 shows the energy consumption per sample. The field is green-marked if the energy consumption is less than 0.1 kWh per sample, yellow if it is less than or equal to 1.5 kWh, and red if it exceeds 1.5 kWh. Field number 13 indicates occupational hazards. It is shown in green or red, depending on the danger situation. If the method does not leak gas or vapor, the area will be marked green; if there is vapor or gas release, the area will be marked red. Field 14 is associated with the amount of waste generated throughout the process. If the amount of waste is less than 10 mL (g), a green color is placed; if it is between 1 and 10 mL (g), the yellow color is placed; and if it is greater than 10 mL (g), a red color is placed. Field 15 gives information about the increase in waste to be generated. If the waste will be recycled, the area is shown in green; if the waste will be processed by degradation or passivation, the area is shown in yellow; if the waste will not be treated at all, the area is shown in red (Sajid & Płotka-Wasylka, 2022).

GAPI has many advantages over the analytical eco scale as it considers the method and process from many aspects. However, it is a disadvantage that the substances used in the synthesis and pre-extraction processes are not taken into account, and, for example, 10.1 mL and 400 mL are shown with the same label in waste generation (Sajid & Płotka-Wasylka, 2022).

Analytical Greenness Calculator Metric (AGREE)

The analytical greenness calculator consists of 12 principles of green analytical chemistry. Each principle is evaluated in the range of 0–1 point; the color chart changes from 1 to 0, from green to red; and the final score consists of 12 principles. As a result, it can be easily evaluated whether the method is suitable for 12 green parameters or not. It has been given in Figure 3 (Sajid & Płotka-Wasylka, 2022).

Figure 3

An Example of AGREE Pictogram And Its Color Scale (Pena-Pereira et al., 2020)

Analytical Method Greenness Score (AMGS)

Analytical method greenness score applied to analytical measurement procedures; It aims to determine the impact of method design and device selection by providing the evaluation in the categories of device energy consumption, the cumulative energy demand of solvents, environmental, health, and safety-related waste production. Although the greenness of analytical methods can be evaluated quantitatively and comprehensively by AMGS, their calculations are complex and have some limitations. Therefore, the analytical method requires much more information to calculate the greenness score. This process of providing too much information delays the time it takes to calculate the greenness score (Shi et al., 2023).

Greenery Index with Spider Diagram

It aims to evaluate a comprehensive measure of greenness by examining reagents under the headings of general properties, odor, fire safety, and stability. Evaluations are shown as -5 and +5 numerical values. -5 is the least green result, and +5 is the greenest result (Shi et al., 2023).

Green Aspects of Gas Chromatography

GC is often seen as more environmentally friendly than LC because it separates analytes in the gas phase without needing solvents. GC uses eco-friendly carrier gases like helium or hydrogen. GAC principles can be applied in GC by choosing the right carrier gases using short columns with small diameters and heating directly (Shaaban et al., 2017).

Selection of Appropriate Carrier Gas

Selection of Suitable Carrier Gas for GC Analysis Choosing the right carrier gas is crucial for environmentally friendly GC. Helium is commonly used because of its inertness and safety besides high linear velocity. However, helium is a non-renewable resource. Nitrogen is also used but has a lower linear velocity than helium resulting in longer analysis times and making it less desirable. Hydrogen on the other hand allows for high flow rates without efficiency loss making it the best option for green GC (Aly & Górecki, 2019; Shaaban et al., 2017).

- ***Speeding up GC Analysis***

GC analysis can be time-consuming and takes 10-30 minutes per run. Speeding up the process can reduce energy consumption and make it more environmentally friendly. Using shorter columns with small diameters is one way to shorten analysis times without compromising resolution and efficiency although small columns have limited loading capacity (Sciarrone et al., 2015).

Two dimensional GC

Conventional gas chromatography (1D-GC) is used for analyzing a broad range of volatile and semi-volatile substances, not ideal for complex samples due to limited peak capacity and selectivity. Comprehensive two-dimensional gas chromatography (GC×GC) is a powerful method for separating volatile and semi-volatile compounds in complex samples especially when paired with mass spectrometry (MS). GC×GC has been around for over 25 years and involves two columns with different retention mechanisms connected through a modulator interface. The modulator periodically collects fractions from the first column and reinjects them into the second column for further separation. With modulation periods ranging from approximately 2 to 8 seconds, GC×GC offers enhanced resolution sensitivity and peak capacity compared to traditional 1D-GC requiring a similar or slightly longer separation time with the same sample volume. The high resolving power of GC×GC particularly when combined with time-of-flight mass spectrometry (TOF-MS) allows for analyzing complex samples with minimal to no sample preparation thereby reducing solvent usage significantly(Dallüge et al., 2003; Tranchida et al., 2015).

Other Aspects

Portable gas chromatography-mass spectrometry (GC-MS) equipment for self-use was introduced in the late 1990s. Subsequently, advancements in heating systems, column construction, and detectors have persisted. To effectively utilize these devices, specific criteria must be fulfilled, including compact size and weight, battery-powered operation, rapid separation speed, and obtaining satisfactory chromatographic resolution for analytes with a broad range of volatility.

The literature also reports on the application of miniaturization as a further method for making GC more environmentally friendly. The analytical chemistry community is increasingly focusing on the diminution of capillary GC, MS analyzers, and other system components including vacuum pumps and electronics. The ion trap is the preferred choice for miniaturization among other mass analyzers because of its simplicity, ability to operate at high pressure, and the potential for tandem MS operation in a portable form. Nevertheless, the reduction in ion storage capacity imposes constraints on the downsizing of ion traps. One way to partially solve this difficulty is by confining the ions using a toroidal shape (Fanali et al., 2015; Ishii & Takeuchi, 1990).

Results

GAC objects to mitigate detrimental impacts on human health and the environment by using environmentally friendly practices in all analytical processes. The utilization of GAC principles in analytical methods is well regarded within the analytical chemistry community. Many researchers are striving to make chromatographic procedures more environmentally friendly by reducing the consumption of organic solvents and minimizing waste output. Several solutions have been suggested in this context to enhance the environmental sustainability of GC analyses.

Nevertheless, there are situations where it is not feasible to eliminate organic solvents. When faced with such situations, the optimum choice is to minimize the quantity of solvent. To decrease solvent usage, one can achieve this by faster analysis without increasing the consumption of the mobile phase, by reducing the usage of solvent and energy.

This technique can be executed by utilizing higher temperatures for the mobile phase and employing innovative column technologies such as porous sub-2-micron particles or superficially porous column packing. Additional methods, such as direct analysis, miniaturization, and online positioning of the chromatograph, serve as valuable tools for real-time monitoring (Aly & Górecki, 2019; Shaaban et al., 2017).

Choosing the appropriate carrier gas is crucial to making gas chromatography more environmentally friendly. Hydrogen is often regarded as the most effective carrier gas for gas chromatography (GC) because of its characteristic van Deemter curve, which remains consistent throughout a range of flow rates. This allows the separation process to be conducted at flow rates higher than the optimum level, resulting in reduced analysis time. In contrast to helium a finite resource, hydrogen may be generated sustainably. Another compelling approach to making GC more environmentally friendly is accelerating the analysis. Using short, narrow bore columns, nanotechnology can expedite the analysis process (Shaaban et al., 2017).

When developing a new analytical method, it should be advised to assess the environmental impact of the analytical methods at every stage. Soon, it is anticipated that there will be advancements in the development of small-scale analytical devices for on-site examination. These devices are likely to incorporate novel materials, particularly nanomaterials. When we compared it with GC, LC is less receptive to environmental sustainability efforts because it unavoidably requires using organic solvents. Greening LC substitutes conventional solvents with environmentally friendly alternatives such as ethanol, filtered water, or carbon dioxide (Byrne et al., 2016; Kerton & Marriott, 2015).

Conclusion

Implementing GAC technologies has been demonstrated to be a prudent approach for achieving both ecological and financial advantages. The advancement of novel sample preparation techniques that effectively minimize the usage of reagents and organic solvents also enhances the capabilities of other approaches, such as electrochemical and chromatographic procedures, which are not directly applicable to samples. The progress of streaming techniques has played a role in the development of GAC, but their full potential has not yet been completely utilized.

Chromatographic systems have demonstrated their cost-effectiveness and value as a viable substitute for fully automated analytical procedures in several fields such as pharmaceuticals, food, environmental studies, agree industries, and regular clinical analysis on a wide scale. Furthermore, they exhibit high speed, precision, and accuracy, while demanding less workload and maintenance.

The widespread adoption of GAC technologies for environmental and industrial applications will be facilitated by commercialization and acceptance by quality control laboratories and industrial facilities. This shift is needed within the realm of chemical education. By substituting obsolete experimental methods with appealing techniques that utilize current instrumentation, we may promptly decrease waste and foster essential environmental consciousness among our students for the future. This is crucial for the amount of work, expenses, and the well-being of analysts. Irrespective of the approach employed, it is important to remember that adopting environmentally friendly practices will consistently yield advantages for the practitioners and their laboratories.

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