Enumerating Chemical Graphs with Mono-block 2-Augmented Tree Structure from Given Upper and Lower Bounds on Path Frequencies

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Abstract. We consider a problem of enumerating chemical graphs from given constraints concerning their structures, which has an important application to a novel method for the inverse QSAR/QSPR recently proposed. In this paper, the structure of a chemical graph is specified by a feature vector each of whose entries represents the frequency of a prescribed path. We call a graph a 2-augmented tree if it is obtained from a tree (an acyclic graph) by adding edges between two pairs of non-adjacent vertices. Given a set of feature vectors as the interval between upper and lower bounds of feature vectors, we design an efficient algorithm for enumerating chemical 2-augmented trees that satisfy the path frequency specified by some feature vector in the set. We implemented the proposed algorithm and conducted some computational experiments.

1 Introduction

Development of novel drugs is one of the major goals in chemoinformatics and bioinformatics. To achieve this purpose, it is important not only to investigate common chemical properties over chemical compounds having certain structural pattern [8, 7, 16], but also
to enumerate all the chemical compounds having a specified structural pattern. The enumeration of chemical compounds has a long history, which can be traced back to Cayley [8], who addressed the enumeration of structural isomers of alkanes in the 19th century.

A multigraph is a graph that can have multiple edges between the same pair of vertices, where multiple edges represent double bonds or triple bonds in a chemical compound. Let us call a multigraph a \( k \)-augmented tree if it is connected and it becomes a tree possibly with multiple edges after removing edges between \( k \) pairs of adjacent vertices, where a 0-augmented tree and a 1-augmented tree are also called an acyclic graph and a monocyclic graph, respectively. In the 97,092,888 chemical compounds in the PubChem database, the ratio of the number of chemical compounds of a \( k \)-augmented tree structure to that of all registered chemical compounds is around 2.9%, 13.3%, 28.2%, 24.2% and 16.0% for \( k = 0, 1, 2, 3 \) and 4, respectively.

Quantitative Structure Activity/Property Relationships (QSAR/QSPR) analysis is a major approach for computer-aided drug design. In particular, inverse QSAR/QSPR plays an important role [17, 22], which is to infer chemical structures from given chemical activities/properties. As in many other fields, Artificial Neural Network (ANN) and deep learning technologies have recently been applied to inverse QSAR/QSPR. In these approaches, new chemical graphs are generated by solving a kind of inverse problems on neural networks, where neural networks are trained using known chemical compound/activity pairs. However, there was no mathematical guarantee for the existence of solutions in these approaches. In order to solve the inverse problem mathematically, a novel approach has been proposed by Akutsu and Nagamochi [2] for ANNs, using mixed integer linear programming (MILP).

Recently Chiewvanichakorn et al. [9], Azam et al. [3, 4] proposed a novel framework for the inverse QSAR/QSPR by combining the MILP-based formulation of the inverse problem on ANNs [2] and efficient enumeration of acyclic graphs and monocyclic graphs. This combined framework for inverse QSAR/QSPR mainly consists of two phases. The first phase defines a function \( f \) that converts each chemical graph \( G \) into a feature vector \( f(G) \) that consists of several descriptors on the structure of \( G \) and then solves (1) Prediction Problem, where a prediction function \( \psi_N \) on a chemical property \( \pi \) is constructed with an ANN \( N \) using a data set of chemical compounds \( G \) and their values \( a(G) \) of \( \pi \). The second phase solves (2) Inverse Problem, where (2-a) given a target value \( y^* \) of the chemical property \( \pi \), a feature vector \( x^* \) is inferred from the trained ANN \( N \) so that \( \psi_N(x^*) = y^* \) and (2-b) then a set of chemical structures \( G^* \) such that \( f(G^*) = x^* \) is enumerated. Methods applied to the case of inferring acyclic or monocyclic chemical graphs have been implemented as computer programs, through which chemical graphs \( G^* \) are inferred from given target values \( y^* \) of actual chemical properties such as heat of atomization, heat of formation, boiling point and octanol/water partition coefficient [3, 9, 4]. In this framework, an efficient algorithm for enumerating acyclic or monocyclic graphs that satisfy given descriptors is an important building block to solve (2-b). A natural next target to apply the framework for the inverse QSAR/QSPR is to construct a system of inferring chemical 2-augmented trees. To attain this, we design an
efficient algorithm for enumerating chemical 2-augmented trees.

Some useful tools such as MOLGEN [12], OMG [20], and similar, have been developed and are available for enumeration of chemical graphs. However, they are not always very efficient in enumerating chemical graphs that satisfy a given condition on structures such as frequency of certain types of subgraphs, because they treat general graph structures. In particular, it is known that the number of molecules (i.e., chemical graphs) with up to 30 atoms (vertices) C, N, O, and S, may exceed $10^{60}$ [5].

Fujiwara et al. [11] and Ishida et al. [14] studied the enumeration of acyclic chemical graphs that satisfy a given feature vector which specifies the frequency of all paths of up to a prescribed length in a chemical compound to be constructed. Their results have been recognized as establishing new methodologies in this track of research in chemoinformatics [25]. Instead of giving a single feature vector $f$ on the frequency of prescribed paths, Shimizu et al. [21] treated a set $F$ of feature vectors on path frequency given as the set of all vectors between a pair of upper and lower feature vectors, and designed a branch-and-bound algorithm of enumerating acyclic chemical graphs each of which satisfies some feature vector $f$ in the set $F$. Afterward Suzuki et al. [23] proposed an improved and more efficient algorithm. For monocyclic graphs, Suzuki et al. [24] proposed an efficient algorithm that constructs a monocyclic chemical graph by adding an edge to an acyclic chemical graph. Such acyclic chemical graphs in turn, can be obtained by an existing algorithm [11, 23]. The above-mentioned algorithms for enumerating acyclic and monocyclic chemical graph with given path frequencies now play a crucial role in the novel methods for inverse QSAR/QSPR [3, 9, 4].

For 2-augmented trees, we distinguish two types: (i) those with two edge-disjoint cycles and (ii) those with a single bi-connected component, where every two cycles share an edge. We call a 2-augmented tree in type (ii) a mono-block 2-augmented tree. In this paper, we design an algorithm for enumerating mono-block 2-augmented trees that satisfy given upper and lower bounds of path frequencies of graphs. We implemented the proposed algorithm and conducted some computational experiments.

2 Preliminaries on Graphs

This section reviews some basic definitions on graphs and introduces the notion of chemical graphs as used in this paper.

2.1 Multigraphs

Let $\mathbb{Z}_+$ denote the set of positive integers. For two integers $a$ and $b$, let $[a, b]$ denote the set of all integers $i$ with $a \leq i \leq b$.

A graph is defined to be an ordered pair $(V, E)$ of a finite set $V$ of vertices and a finite set $E$ of edges. In this paper, we do not consider self-loops, and an edge in $E$ joining two vertices $u, v \in V$ is denoted by $uv$.

Let $G$ be a graph. We denote the vertex set and the edge set of $G$ by $V(G)$ and $E(G)$, respectively. An ordered pair $(V', E')$ of subsets $V' \subseteq V(G)$ and $E' \subseteq E(G)$ is called a
Figure 1: (a) An example of a labeled graph, each vertex is labeled with a unique label $v_i$, $i = 1, 2, \ldots, 19$, and each edge with a unique label $e_j$, $j = 1, 2, \ldots, 25$. (b) A monocyclic graph $G$ with a unique cycle $C$, where the pendant tree $G(v)$ for a vertex $v \in V(C)$ is the subtree enclosed by a dashed line.

subgraph of $G$ if $(V', E')$ forms a graph, i.e., $\{u, v \in V \mid uv \in E'\} \subseteq V'$. We say that a subset $X \subseteq V(G)$ induces a subgraph $G'$ if $V(G') = X$ and $E(G')$ contains every edge in $E(G)$ between two vertices in $X$.

We call a graph where each vertex and edge has a unique name or an index a labeled graph. Throughout the paper, graphs are considered to be labeled, to distinguish or enumerate vertices, edges or some other structures in a graph. Figure 1(a) shows an example of a labeled graph. A rooted graph is a graph in which either a vertex or an edge is designated as a root.

A graph is called a multigraph when there can be more than one edge between the same pair of endvertices. Let $G$ be a multigraph. For a vertex $v \in V(G)$, we call the number of edges incident to $v$ the degree of $v$ and denote it by $\deg(v)$. Let $\{u, v\} \subseteq V(G)$. The multiplicity, i.e., the number of edges, between two vertices $u$ and $v$ is denoted by $\mul_G(u, v)$, where for two non-adjacent vertices $u$ and $v$, it holds $\mul_G(u, v) = 0$. We refer to the set of $\mul_G(u, v) \geq 0$ simple edges with endvertices $u$ and $v$ as a multiple edge with multiplicity $\mul_G(u, v)$. Let $E(G)$ denote the set of pairs $\{u, v\} \subseteq V(G)$ with $\mul_G(u, v) = 0$. For a pair of adjacent vertices $u$ and $v$, let $G - uv$ denote the graph $G'$ obtained by removing $\mul_G(u, v)$ simple edges between $u$ and $v$ from $G$. Conversely, let $G + q \cdot uv$ denote the graph $G'$ obtained by adding $q \in \mathbb{Z}_+$ simple edges between $u$ and $v$, i.e., $\mul_{G'}(u, v) = \mul_G(u, v) + q$. In particular, we denote $G + 1 \cdot uv$ by $G + uv$. If $G$ is clear from the context, then we denote $\mul_G(u, v)$ by $\mul(u, v)$.

For a nonnegative integer $k$, a graph $P$ which consists of $k+1$ distinct vertices $v_0, v_1, \ldots, v_k$ and $k$ multiple edges $v_iv_{i+1}$, $i \in [1, k-1]$, is called a path (or a path of length $k$), and is denoted by $P = (v_0, v_1, \ldots, v_k)$. The length, i.e., the number of edges in a path $P$ is also denoted by $|P|$. A graph $C$ which consists of a path $(v_0, v_1, \ldots, v_\ell)$ of length $\ell \geq 2$ and a multiple edge between $v_\ell$ and $v_0$ is called a cycle and is denoted by $C = (v_0, v_1, \ldots, v_\ell, v_0)$. In this paper, a graph which consists of two vertices and two
edges between them is not considered as a cycle.

A block in $G$ is defined to be a maximal vertex subset $X \subseteq V(G)$ such that for any two vertices $u, v \in X$, there is a cycle of $G$ that passes through $u$ and $v$. We call $G$ a mono-block graph if $G$ has exactly one block $X$ with $|X| \geq 2$.

A connected multigraph with no cycles is called a multitree. Every multitree $T$ has either a vertex $v$ or an adjacent vertex pair $\{v, v'\}$ removal which leaves no connected component with more than $\lfloor |V(T)|/2 \rfloor$ vertices [15]. Such a vertex or an adjacent vertex pair is called a centroid, where a centroid $v$ is called a unicentroid and a centroid $\{v, v'\}$ is called a bicentroid.

Let $T$ be a rooted multitree and $v \in V(T)$. Let $u \in V(T)$ be a vertex such that $v$ is on the unique path between $u$ and the root. We call $u$ a descendant of $v$ and call $v$ an ancestor of $u$. In particular, if $u$ and $v$ are adjacent, we call $u$ a child of $v$ and call $v$ the parent of $u$. The parent of $v$ is denoted by $p(v)$. The set of children of a vertex $v$ is denoted by $\text{Ch}(v)$. The depth of $v$ represents the length of the unique path between $v$ and the root, and is denoted by $d(v)$. If $v$ is the root vertex or an end-vertex of the root edge, then $v$ has no parent, and $d(v) = 0$. We denote by $T_v$ the subtree of $T$ induced by $v$ and the set of descendants of $v$. For an edge $vw \in E(T)$ such that $w = p(v)$, we denote by $T_{vw}$ the subtree of $T$ induced by $w$, $v$, and the set of descendants of $v$. That is, $T_{vw}$ consists of the subtree $T_v$ and the vertex $w = p(v)$ joined by a multiple edge between $v$ and $w$ with multiplicity $\text{mul}_T(v, w)$. We regard $T_{vw}$ to be rooted at $w$.

For a connected multigraph $G$ with at least one cycle and a vertex $v \in V(G)$ such that $v$ is included in some of the cycles in $G$, the pendent tree $G(v)$ of vertex $v$ is defined to be the subgraph of $G$ induced by $v$ and the set of vertices reachable from $v$ without passing through any edge in a cycle of $G$, where $T$ becomes a tree, possibly only consisting of vertex $v$. We treat $G(v)$ as a tree rooted at $v$. For a vertex $u \in V(G(v))$ we define $\rho_G(u) = v$. For convenience, for a pendent tree $T = G(v)$ and a vertex $u \in V(G(v))$, we denote the subtree $T_u$ of $G(v)$ rooted at $u$ by $G(u)$. In addition, for the parent $w = p(u)$ of $u$ in $G(v)$, we denote by $G(w, u)$ the rooted tree $T_{wu}$.

An example of a pendent tree is illustrated in Fig. 1(b). For a multigraph $G$ with $n$ vertices, we say that a pendent tree of $G$ is exceeding if it has at least $n/3$ vertices.

### 2.2 $k$-Augmented Trees

A $k$-augmented tree with $n$ vertices is a connected multigraph such that the number of pairs of adjacent vertices is $(n - 1) + k$, i.e., it is constructed from a multiree with $n$ vertices by adding edges between $k$ pairs of non-adjacent vertices.

#### 2.2.1 1-Augmented Trees

Let $G$ be a monocyclic graph, which has a unique cycle $C$. Throughout this draft we will also call 1-augmented trees monocyclic graphs. For a vertex $u \in V(G)$, let $\rho_G(u)$ denote the vertex $v \in V(C)$ such that $u \in V(G(v))$. Note that in the case when $u \in V(C)$, it holds that $u = \rho_G(u)$. See Fig. 1(b) for an example of a monocyclic graph, where $\rho_G(u)$ is denoted for a vertex $u$. 

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For a monocyclic graph $G$ and two vertices $u, v \in V(G)$ such that $\rho_G(u) = \rho_G(v)$ (i.e., $u$ and $v$ are contained in the same pendent tree of $G$), let $P(u, v)$ denote the unique path in $G$ between $u$ and $v$.

### 2.2.2 Mono-block 2-Augmented Trees

Let $H$ be a mono-block 2-augmented tree, and $C_1, C_2,$ and $C_3$ be the three distinct cycles of $H$. We observe that there are exactly two vertices contained in all of the cycles of $H$. We call these vertices junctions. We call a pair of a junction $u$ and a neighbor of $u$ on some of the cycles of $H$ a junction pair. Note that the number of junction pairs in one mono-block 2-augmented tree is either six or five (see Fig. 2(a) and (b), respectively).

Let $u$ and $u'$ be the junctions of $H$. For $i \in [1, 3]$, we define $P(u, u'; C_i)$ to be the unique path from $u$ to $u'$ that is not in $C_i$. Note that either $P(u, u'; C_i)$ is itself a junction pair, or it contains exactly two junction pairs. See Fig. 2 for an example of mono-block 2-augmented trees and junctions.

![Figure 2: Mono-block 2-augmented trees $H_a$ and $H_b$ with junctions $u$ and $u'$. (a) The pair $\{u, x\}$ is one of the junction pairs of $H_a$. The path $P(u, u'; C_1)$ is drawn in bold. (b) The path $P(u, u'; C_1)$ drawn in bold consists of a single edge $uu'$.](image)

### 3 Problem Formulation

In this section, we formalize the problem to be addressed in this paper.

#### 3.1 Chemical Graphs

To treat chemical compounds as multigraphs, we introduce a color for every vertex in a graph. Colors represent chemical elements, such as $\text{O}$, $\text{N}$, or $\text{C}$. Let us denote the set of colors by $\Sigma$, and the color of a vertex $v$ by $\text{col}(v)$. The valence of a chemical element $c \in \Sigma$ is denoted by an integer function $\text{val}(c) \in \mathbb{Z}_+$. The size of a bond between two adjacent atoms is indicated by the edge multiplicity between the two vertices that correspond to those atoms. A multigraph $G$ is called a $\Sigma$-colored graph if each vertex $v \in V(G)$ is assigned a color $\text{col}(v) \in \Sigma$. A chemical compound can be viewed as a
Σ-colored multigraph without self-loops, where vertices and colors represent atoms and elements, respectively. Throughout this paper, we call Σ-colored multigraphs without self-loops chemical graphs.

Let \( G \) be a chemical graph. Chemical compounds, especially organic compounds, are rich with hydrogen atoms. Since the valence of hydrogen is 1, we can determine the structure of chemical graphs without considering hydrogen atoms. On this ground, we suppress hydrogen in order to enumerate chemical graphs more quickly. As a result, for some vertex \( v \) in a hydrogen-suppressed chemical graph, the degree \( \deg(v) \) may be smaller than the valence \( \val(col(v)) \). For a vertex \( v \in V(G) \), we define the residual degree of \( v \) to be \( \val(col(v)) - \deg(v) \) and denote it by \( \res(v) \). In a hydrogen-suppressed chemical graph, for a vertex \( v \), \( \res(v) \) represents the number of hydrogen atoms adjacent to \( v \) in the corresponding chemical compound.

### 3.2 Isomorphism on Chemical Multigraphs

In enumerating chemical graphs, we must avoid duplication of equivalent graphs. For example, two chemical graphs \( G \) and \( G' \) may have the same graph structure, and imply the same chemical compound even if they are different as labeled graphs. This case is formalized by the notion of isomorphism as follows. Let \( G = (V, E) \) and \( G' = (V', bE') \) be two chemical multigraphs. The following bijection \( \psi \) from \( V(G) \) to \( V(G') \) is called an isomorphism from \( G \) to \( G' \):

(i) for each vertex \( x \in V(G) \), it holds that \( \col(x) = \col(\psi(x)) \); and

(ii) for each pair \( \{x, y\} \subseteq V(G) \), it holds that \( \mul_G(x, y) = \mul_{G'}(\psi(x), \psi(y)) \).

If there exists an isomorphism from \( G \) to \( G' \), then we say that \( G \) and \( G' \) are isomorphic. We write \( G \approx G' \) if \( G \) and \( G' \) are isomorphic, and write \( G \not\approx G' \) otherwise. For two sets \( \mathcal{G} \) and \( \mathcal{G}' \) of chemical graphs, we say that \( \mathcal{G}' \) represents \( \mathcal{G} \) if

- for each chemical graph \( G \in \mathcal{G} \), there is a chemical graph \( G' \in \mathcal{G}' \) such that \( G \approx G' \); and

- for any two chemical graphs \( G_1', G_2' \in \mathcal{G}' \), it holds that \( G_1' \not\approx G_2' \).

An automorphism of a chemical graph \( G \) is defined to be an isomorphism \( \psi \) from \( V(G) \) to \( V(G) \) itself.

In addition, for two graphs \( G \) and \( G' \) and vertex subsets \( X \subseteq V(G) \) and \( Y \subseteq V(G') \) such that \( |X| = |Y| \), if there exists an isomorphism \( \psi \) from \( V(G) \) to \( V(G') \) such that for each vertex \( x \in X \) it holds that \( \psi(x) \in Y \), we say that \( G \) and \( G' \) are \( (X, Y) \)-isomorphic. In the case when the sets \( X \) and \( Y \) are singletons, i.e. \( X = \{x\} \) and \( Y = \{y\} \) and \( G \) and \( G' \) are \( (x, y) \)-isomorphic, we may write that they are \( (x, y) \)-isomorphic. Finally, if \( G \) is a graph rooted at vertex \( v_r \) and \( G' \) is rooted at \( v'_r \), and they are \( (v_r, v'_r) \)-isomorphic, then we say that they are rooted isomorphic and denote this by \( G \approx_r G' \).
3.3 Feature Vectors

In this paper, “feature vectors” represent occurrences of paths in a graph. To specify a feature vector space, we fix three parameters: a set $\Sigma$ of colors; the maximum multiplicity $d \geq 1$ among all pairs of vertices in multigraphs to be enumerated; and the maximum length $K \geq 0$ of the path structures to be specified.

Let $c_0, c_1, \dots, c_K \in \Sigma$ be $K+1$ colors and $m_1, m_2, \ldots, m_K \in [1, d]$ be $K$ integers, where it may hold that $c_i = c_j$ or $m_i = m_j$ for some $i$ and $j$. We call an alternating sequence $t = (c_0, m_1, c_1, \ldots, m_K, c_K)$ a colored sequence of length $|t| = K$. We denote the set of all colored sequences with length $K$ by $\Sigma^{K,d}$, and the union of $\Sigma^{0,d}, \Sigma^{1,d}, \ldots, \Sigma^{K,d}$ by $\Sigma^{\leq K,d}$. For a colored sequence $t = (c_0, m_1, c_1, \ldots, m_K, c_K) \in \Sigma^{K,d}$, let $\text{rev}(t)$ denote its reverse sequence $(c_K, m_K, c_{K-1}, \ldots, m_1, c_0) \in \Sigma^{K,d}$.

Let a chemical graph $P$ be a path $P = (v_0, v_1, \ldots, v_K)$ of length $K$ with root $v_0$. We define the colored sequence $\gamma(P) \in \Sigma^{K,d}$ of $P$ to be

$$
\gamma(P) \triangleq (\text{col}(v_0), \text{mul}_P(v_0, v_1), \text{col}(v_1), \ldots, \text{mul}_P(v_{K-1}, v_K), \text{col}(v_K)).
$$

Let $G$ be a chemical graph. For a colored sequence $t \in \Sigma^{\leq K,d}$, the frequency $\text{frq}(t, G)$ of $t$ in $G$ is defined to be the number of vertex-rooted subgraphs $G'$ of $G$ such that $G' \approx P$ for a rooted path $P$ with $\gamma(P) = t$. We define the feature vector $f(G)$ of level $K$ of $G$ to be the $|\Sigma^{\leq K,d}|$-dimensional vector such that $f(G)[t] = \text{frq}(t, G)$ for each colored sequence $t \in \Sigma^{\leq K,d}$.

Given a color set $\Sigma$ and integers $d$ and $K$, the set of $|\Sigma^{\leq K,d}|$-dimensional vectors whose entries are nonnegative integers is called a feature vector space and is denoted by $f(\Sigma, K, d)$. Equivalently, each vector $g \in f(\Sigma, K, d)$ is a mapping $g : \Sigma^{\leq K,d} \to \mathbb{Z}_+$. For two vectors $g, g' \in f(\Sigma, K, d)$, we write $g \leq g'$ if for each entry $t \in \Sigma^{\leq K,d}$, it holds that $g[t] \leq g'[t]$. For two given vectors $g, g' \in f(\Sigma, K, d)$, a chemical graph $G$ is called feasible if $g \leq f(G) \leq g'$ and $\text{res}(v) \geq 0$ holds for all vertices $v \in V(G)$. Let $G(g, g')$ denote the set of all chemical graphs feasible to $(g, g')$.

4 Problem of Enumerating Mono-block 2-Augmented Trees

Let $G_1$ denote the set of $\Sigma$-colored labeled monocyclic graphs, and $G_2$ denote the set of $\Sigma$-colored labeled mono-block 2-augmented trees. Note that each of $G_1$ and $G_2$ conceptually contains infinitely many labeled graphs unless a way of expressing labeled graphs is restricted.

For two given vectors $g_e$ and $g_u$, the set of feasible graphs $H \in G_2$ is denoted by $G_2(g_e, g_u)$. Our goal in this paper is to construct a set $G'_2$ that represents the set $G_2(g_e, g_u)$. Since we can use an existing algorithm to obtain a set $G'_1$ that represents $G_1$ (for example, the algorithm due to Suzuki et al. [24]), we design an algorithm to our goal so that our target set $G'_2$ is constructed (1) by adding some number $p \in [1, \min\{\text{res}(x), \text{res}(y)\}]$ of edges between some non-adjacent vertices $x$ and $y$ in each $\Sigma$-colored labeled monocyclic graph $H$ in $G'_1$; and
(II) by discarding from the graphs $H$ those that are infeasible, i.e., $g_\ell \nleq f(H)$ or $f(H) > g_u$.

We design procedures for tasks (I) and (II) separately. In the rest of this subsection, we examine the task (I). Testing if a graph $H \in \mathcal{G}_2$ satisfies $g_\ell \leq f(H) \leq g_u$ can be handled in a relatively easy way. We remark that all non-isomorphic $\Sigma$-colored labeled monocyclic graphs might not be needed for us to generate our target set $\mathcal{G}_2'$ which is restricted by bounds $g_\ell$ and $g_u$. In other words, we may be able to introduce a pair $(g'_\ell, g'_u)$ of lower and upper feature vectors on $\Sigma$-colored labeled monocyclic graphs so that a smaller set of non-isomorphic $\Sigma$-colored labeled monocyclic graphs can produce all necessary graphs in our target set $\mathcal{G}_2'$.

For two vectors $g_1$ and $g_2$, the set of feasible graphs in $\mathcal{G}_1$ is denoted by $\mathcal{G}_1(g_1, g_2)$. Following the idea of Suzuki et al. [24], we modify a given lower vector $g_\ell$ into a vector $g'_\ell$ so that any graph in $\mathcal{G}_2(g'_\ell, g_u)$ can be constructed from some graph in $\mathcal{G}_1(g'_\ell, g_u)$. For a given lower vector $g_\ell \in f(\Sigma, K, d)$, we define $g'_\ell$ as follows:

(i) for each $t \in \Sigma^{0,d}$, let $(g'_\ell)[t] = g_\ell[t]$;

(ii) for each $t = (c, m, c') \in \Sigma^{1,d}$, let

$$
g'_\ell[t] = \begin{cases} 
\max\{0, g_\ell[t] - 1\}, & \text{if } c \neq c' \\
\max\{0, g_\ell[t] - 2\}, & \text{if } c = c', \text{ and}
\end{cases}
$$

(iii) for each $t \in \Sigma^{\leq K,d} \setminus \Sigma^{1,d}$, let $(g'_\ell)[t] = 0$.

**Lemma 1.** For a set $\Sigma$ of colors and integers $K \geq 0$ and $d \geq 1$, let the vectors $g_u, g_\ell \in f(\Sigma, K, d)$ be such that $g_\ell \leq g_u$, and $H \in \mathcal{G}_2(g_\ell, g_u)$. Then, for each junction pair $\{x, y\} \subseteq V(H)$, it holds that $H - xy \in \mathcal{G}_1(g'_\ell, g_u)$.

*Proof.* Let $\{x, y\}$ be a junction pair in $H$. Let $G$ be the graph $H - xy$. We show that $G \in \mathcal{G}_1(g'_\ell, g_u)$ i.e., $G$ is a graph in $\mathcal{G}_1$ with $g'_\ell \leq f(G) \leq g_u$ and $\text{res}(v) \geq 0$ for each vertex $v \in V(G)$. Obviously, $G = H - xy$ is a graph in $\mathcal{G}_1$ because removing from $H$ all edges between a junction pair leaves only one cycle in $G$.

Since $H \in \mathcal{G}_2(g_\ell, g_u)$, we have $g_\ell \leq f(H) \leq g_u$. First we check the upper bound of $f(G)$. Since removing all edges between $x$ and $y$ does not increase the frequency of any colored sequence, it holds that $\text{frq}(t, G) \leq \text{frq}(t, H)$ for any colored sequence $t$. Hence it holds that $f(G) \leq f(H) \leq g_u$. Next we check the lower bound of $f(G)$. For a colored sequence $t \in \Sigma^{0,d}$, we consider the entry $f(G)[t]$. We have $f(G)[t] = f(H)[t] = g_\ell[t] = g'_\ell[t]$ for all colored sequences $t \in \Sigma^{0,d}$, since removing edges does not change the frequency of colored sequences of length 0. A colored sequence $t \in \Sigma^{1,d}$ is given by $(c, m, c')$, and we have the following observations: If $t = \gamma(xy)$ or $t = \gamma(yx)$ for some path $xy$ in $H$, it holds that

$$
f(G)[t] = \begin{cases} 
\max\{0, f(H)[t] - 1\} & \text{if } t \neq \text{rev}(t) \\
\max\{0, f(H)[t] - 2\} & \text{if } t = \text{rev}(t);
\end{cases}
$$
and otherwise, we have \( f(G)[t] = f(H)[t] \). For all colored sequences \( t \in \Sigma^{1,d} \), since \( g_t \leq f(H)[t] \), it holds that

\[
f(G)[t] \geq \begin{cases} 
\max\{0, g_c[t] - 1\} & \text{if } c \neq c' \\
\max\{0, g_c[t] - 2\} & \text{if } c = c'. 
\end{cases}
\]

According to the definition of \( g^\dagger_t \), we have \( f(G)[t] \geq g^\dagger_t[t] \) for each colored sequence \( t \in \Sigma^{1,d} \). For a colored sequence \( t \in \Sigma^{L,K,d} \), it is clear from the definition of \( g^\dagger_t \) that \( f(G)[t] \geq g^\dagger_t[t] = 0 \). From above, we have \( g^\dagger_t \leq f(G) \leq g_u \).

Finally, we prove that \( \text{res}(u) \geq 0 \) for each vertex \( u \in V(H) \). Removing edges from \( H \) does not decrease the residual degree of any vertex. Hence it holds that \( \text{res}(v) \geq 0 \) for each vertex \( v \in V(G) \).

\[\square\]

Lemma 1 states that for each graph \( H \in G_2(g_t, g_u) \), there is at least one monocyclic graph \( G \in G_1(g^\dagger_t, g_u) \), a pair \( \{x, y\} \in E(G) \), and an integer \( p \) giving \( H \) as \( G + p \cdot xy \). For this, we first generate all monocyclic graphs in \( G_1(g^\dagger_t, g_u) \). Then we enumerate all mono-block 2-augmented trees in \( G_2(g_t, g_u) \) by adding edges between pairs of non-adjacent vertices in each of the given monocyclic graphs, where the pair of endvertices of the newly added edges becomes a junction pair of the newly created mono-block 2-augmented tree. The problem to deal with in this paper is formalized as follows.

**Enumerating mono-block 2-augmented trees from given monocyclic graphs with given Upper and Lower Path Frequency**

**Input:** A set \( \Sigma \) of colors, integers \( K \geq 0 \) and \( d \geq 1 \), two vectors \( g_u, g_t \in f(\Sigma, K, d) \) such that \( g_t \leq g_u \) and for all colored sequences \( t \in \Sigma^{0,d} \) it holds that \( g_t[t] = g_u[t] \), and a set \( G'_1 \) that represents the set \( G_1(g^\dagger_t, g_u) \).

**Output:** A set \( G'_2 \) that represents the set \( G_2(g_t, g_u) \).

In the rest of the paper, we focus on designing an enumerating procedure for the task (I).

### 4.1 Sketch of the Enumerating Procedure

Suppose that a set \( G'_1 = \{G_1, G_2, \ldots, G_4\} \) that represents the set \( G_1(g_t, g_u) \) is given. When \( \Sigma \)-colored labeled mono-block 2-augmented trees are generated by adding a multiple edge \( xy \) between a pair \( \{x, y\} \in E(G) \) to each graph \( G_i \in G'_1 \), we have to check whether the same graph has already been generated in the process or not, i.e., whether a newly generated graph \( H = G_i + p \cdot xy \in G_2 \) is isomorphic to another graph \( H' = G_j + q \cdot x'y' \in G_2 \) that has already been obtained from some graph \( G_j \in G'_1 \). We call the duplication arising when \( H = G_i + p \cdot xy \) and \( H' = G_j + q \cdot x'y' \) are isomorphic with \( i \neq j \) inter-duplication, and that when \( H = G_i + p \cdot xy \approx H' = G_i + q \cdot x'y' \) intra-duplication. This section provides some ideas on how to avoid such duplications without storing all generated graphs and explicitly comparing the new one with each of them.
In enumeration algorithms, the concept of a family tree is widely employed \[18, 19, 24\] in order to efficiently cope with inter-duplication. In order to define a family tree for graphs, we need to define a parent-child relationship between graph structures so that the parent structure of a given chemical graph \(H\) is uniquely determined from the topological structure of \(H\). A parent-child relationship over classes \(\mathcal{H}\) and \(\mathcal{G}\) of chemical graphs is defined by an injective mapping \(\pi : \mathcal{H} \rightarrow \mathcal{G}\) as follows. Given a chemical graph \(H \in \mathcal{H}\), we define a labeled graph \(G = \pi(H) \in \mathcal{G}\) so that for any two chemical graphs \(H_1\) and \(H_2\) with \(H_1 \approx H_2\), it holds \(G_1 \approx G_2\) with chemical graphs \(G_i = \pi(H_i), i = 1, 2\), implying that \(\pi(H)\) is determined only from the topological structure of \(H\). For two chemical graphs \(G \in \mathcal{G}\) and \(H \in \mathcal{H}\) with \(G = \pi(H)\), the graph \(G\) is called the parent of \(H\) (the parent of \(H\) as an unlabeled graph which is unique up to automorphism), and the graph \(H\) is called a child of \(G\). Let \(\text{Ch}_\pi(G)\) denote the set of chemical graphs \(H \in \mathcal{H}\) such that \(G = \pi(H)\). Therefore, we easily observe the next.

**Lemma 2.** Let \(\pi : \mathcal{H} \rightarrow \mathcal{G}\) be a parent-child relationship over classes \(\mathcal{H}\) and \(\mathcal{G}\) of chemical graphs. For two chemical graphs \(G, G' \in \mathcal{G}\), if \(G \not\approx G'\), then \(\text{Ch}_\pi(G) \cap \text{Ch}_\pi(G') = \emptyset\).

Then, Lemma \[2\] ensures that we do not need to explicitly check for inter-duplication, that is, if a chemical graph \(H \in \mathcal{G}_2\) that is a child of a chemical graph \(G_i \in \mathcal{G}_1\) is isomorphic to some chemical graph \(H' \in \mathcal{G}_2\) generated from a chemical graph \(G_j \in \mathcal{G}_1\) with \(i \neq j\).

This section closes by presenting a high level description of a procedure that, given a chemical graph \(G \in \mathcal{G}_1\) and a maximum multiplicity \(d\), constructs a set \(\mathcal{G}_2(G, d)\) that represents the set of chemical graphs in \(\text{Ch}_\pi(G) (\subseteq \mathcal{G}_2)\) with multiplicity at most \(d\). For this, we will introduce

(I-a) an appropriate choice of a parent-child relationship \(\pi : \mathcal{G}_2 \rightarrow \mathcal{G}_1\); and

(I-b) a way of avoiding intra-duplication from the same chemical graph \(G \in \mathcal{G}_1\).

The technical details on (I-a) and (I-b) will be discussed in Sections \[7\] and \[8\] respectively. Now the task (I) is divided into two subtasks, (I-a) and (I-b).

We here explain ideas on (I-a) and (I-b). For (I-a), we define a parent-child relationship \(\pi : \mathcal{G}_2 \rightarrow \mathcal{G}_1\) such that the parent \(\pi(H)\) of \(H\) is a \(\Sigma\)-colored labeled monocyclic graph obtained from \(H\) by removing all \(\text{mul}_H\{x, y\}\) edges between a junction pair \(\{x, y\}\) which meets a certain condition derived in Section \[7\]. The reason why such a junction pair \(\{x, y\}\) is carefully chosen is that the resulting function \(\pi\) must satisfy the definition of parent-child relationships over classes \(\mathcal{G}_2\) and \(\mathcal{G}_1\).

To handle (I-b) for a chemical graph \(G \in \mathcal{G}_1\), we call a subset \(F \subseteq \overline{E}(G)\) proper if

- for any \(H \in \text{Ch}_\pi(G)\), there is a pair \(\{u, v\} \in F\) and an integer \(p \in [1, \min\{\text{res}(u), \text{res}(v)\}]\), such that \(H \approx G + p \cdot uv\); and

- for two distinct pairs \(\{x, y\}, \{u, v\} \in F\), and integers \(p \in [1, \min\{\text{res}(x), \text{res}(y)\}]\) and \(q \in [1, \min\{\text{res}(u), \text{res}(v)\}]\) it holds that \(G + p \cdot xy \not\approx G + q \cdot uv\).

Section \[8\] provides a procedure for testing if a given pair \(\{x, y\} \in \overline{E}(G)\) belongs to a proper set \(F\) after executing a preprocessing to construct \(F\).

With the above ideas, a procedure for constructing a set \(\mathcal{G}_2(G, d)\) is given as follows.
Procedure 1

Input: An integer $d$ and a chemical graph $G \in \mathcal{G}_1$ with a unique cycle $C$ and multiplicity at most $d$.

Output: A set $\mathcal{G}_2(G, d)$ that represents the set of chemical graphs in $\text{CH}_\pi(G) (\subseteq \mathcal{G}_2)$ with multiplicity at most $d$.

1: Execute a preprocessing to construct a proper set $F \subseteq E(G)$; /* by Procedure 5 in Section 8 */
2: for each pair $\{x, y\} \in F$ do
3: for each integer $p \in [1, \min\{d, \text{res}(x), \text{res}(y)\}]$ do
4: if $G + p \cdot xy$ is a child of $G$ /* tested by Procedure 4 in Section 7 */ then
5: Generate the mono-block 2-augmented tree $G + p \cdot xy$ as part of the output
6: end if
7: end for
8: end for.

5 Signature and Code

In order to define our parent-child relationship $\pi : \mathcal{G}_2 \to \mathcal{G}_1$ for the task (I-a), we introduce the notion of “signature” of graphs.

For a class $\mathcal{G}$ of graphs, if we have a way of choosing a labeling of each graph $G \in \mathcal{G}$ which is unique up to the graph’s automorphism, then we can test the isomorphism of the two graphs directly by comparing their labels. Such a labeling for a graph $G$ is called a canonical form of $G$. Once such a canonical form for a class $\mathcal{G}$ of graphs is obtained, we can easily encode each graph $G \in \mathcal{G}$ into a sequence $\sigma(G)$, called the signature of $G$, such that two graphs $G, G' \in \mathcal{G}$ are isomorphic if and only if $\sigma(G) = \sigma(G')$.

5.1 Lexicographical Order

We fix a total order of the colors in $\Sigma$ arbitrarily, e.g., $\emptyset < \mathbb{N} < \mathbb{C}$. We introduce a lexicographical order among sequences with elements in $\Sigma \cup \mathbb{Z}_+$ as follows. A sequence $A = (a_1, a_2, \ldots, a_p)$ is lexicographically smaller than a sequence $B = (b_1, b_2, \ldots, b_q)$ if there is an index $k \in [1, \min\{p, q\}]$ such that

(i) $a_i = b_i$ for all $i \in [1, k]$; and

(ii) $k = p < q$ or $a_{k+1} < b_{k+1}$ with $k < \min\{p, q\}$.

If $A$ is lexicographically smaller than $B$, then we denote $A < B$. If $p = q$ and $a_i = b_i$ for all $i \in [1, p]$, then we denote $A = B$. Let $A \preceq B$ mean that $A < B$ or $A = B$.

We often rely on lexicographically sorting a collection $S = (s_1, s_2, \ldots, s_k)$ of $k$ sequences. We will represent a lexicographically ascending (resp., descending) order on collection $S$ by a permutation $\pi : [1, k] \to [1, k]$ such that for $1 \leq i < j \leq k$ it holds that $s_{\pi(i)} \preceq s_{\pi(j)}$ (resp., $s_{\pi(j)} \preceq s_{\pi(i)}$).
For a collection $S$ of sequences, let us denote by $||S|| = \sum_{s \in S} |s|$ the total length of the sequences in the collection $S$. A known algorithm due to Aho et al. [1] can be used to lexicographically sort a collection $S$ of sequences over an alphabet of size $n$ in $O(||S|| + n)$ computation time.

5.2 Canonical Form and Signature of Trees

In this subsection, we review the concept of a canonical form of rooted trees [18, 19].

5.2.1 Ordered Trees

An ordered tree is a rooted tree with a fixed total order among the children of each vertex. By convention, we assume that the order of children in an ordered tree is from left to right.

Let $T$ be a multitree with $n$ vertices, rooted at a vertex $r$. There may be many different ordered trees on $T$. A canonical form of $T$ is given by an adequately chosen ordered tree on $T$. When we conduct a depth-first-search, we assume that we visit children from left to right. We denote the vertices of $T$ by $v_1, v_2, \ldots, v_n$, indexed in the order visited by a depth-first-search starting from the root. Let $\tau$ be an ordered tree on $T$ and let $\delta(\tau)$ denote the alternating color-depth sequence $(c_1, d_1, \ldots, c_n, d_n)$ that consists of the color $c_i$ and the depth $d_i$ of the $i$-th vertex $v_i$ for $i \in [1, n]$. Let $M(\tau)$ denote the sequence $(m_2, m_3, \ldots, m_n)$ of the multiplicity $m_i = \text{mul}(v_i, p(v_i))$ between the $i$-th vertex $v_i$ and its parent $p(v_i)$ for $i \in [2, n]$.

5.2.2 Left-heavy Trees

A left-heavy tree of a rooted tree $T$ is an ordered tree $\tau$ that has the lexicographically maximum code $\delta(\tau)$ among all ordered trees of $T$. Note that a left-heavy tree has the following recursive structure: for every vertex $v \in V(T)$, the subtree $T_v$ is also a left-heavy tree, and $\delta(T_v)$ and $M(T_v)$ are continuous subsequences of $\delta(T)$ and $M(T)$, respectively. We define the canonical form of a rooted tree $T$ to be the left-heavy tree $\tau$ that has the lexicographically maximum sequence $M(\tau)$ among all left-heavy trees of $T$, and define the signature of $T$ to be $\sigma(T) \triangleq (\delta(\tau), M(\tau))$. We give a procedure to calculate the signature $\sigma$ of a rooted tree as Procedure 2 in Section 5.2.3.

5.2.3 Calculating the Signature of Rooted Multi-Trees

For any two sequences $S_1$ and $S_2$, let $S_1 \oplus S_2$ denote the concatenation of $S_1$ and $S_2$. Given an ordered multi-tree $T$ on $n$ vertices indexed $v_1, v_2, \ldots, v_n$ as visited in a depth-first traversal, let $\delta(T) = (c_1, d_1, c_2, d_2, \ldots, c_n, d_n)$ be its color-depth sequence as defined in Section 5.2.2. For an integer $k \geq 1$ we define the $k$-shift $\delta^k(T)$ of the sequence $\delta(T)$ to be the sequence $(c_1, d_1 + k, c_2, d_2 + k, \ldots, c_n, d_n + k)$ obtained by adding $k$ to each of the depth entries of $\delta(T)$.

Let $T$ be an ordered multi-tree rooted at a vertex $r$, and let $u_1, u_2, \ldots, u_{\text{deg}(r)}$ denote the children of $r$ indexed according to their left-to-right ordering. Let $M(u_i)$ denote
The signatures \(\sigma(T(u_i)) = (\delta(T(u_i)), M(T(u_i)))\) for all \(i \in [1, \deg(r)]\). Given the signatures \(\sigma(T(u_i)) = (\delta(T(u_i)), M(T(u_i)))\) for all \(i \in [1, \deg(r)]\), we devise a way to represent \(\sigma(T)\) by \(\sigma(T(u_i))\) via the following observation.

**Observation 3.** Let \(T\) be an ordered multi-tree rooted at a vertex \(r\), and let \(\text{CH}(r) = \{u_1, u_2, \ldots, u_{\deg(r)}\}\) denote the set of children of \(r\) indexed according to their left-to-right ordering. Given the sequences \(\delta(T_u)\) and \(M(T_u)\) for all \(u \in \text{CH}(r)\), for the sequences \(\delta(T)\) and \(M(T)\) it holds that:

\[
\begin{align*}
\delta(T) &= (\text{col}(r), 0) \oplus \delta^1(T(u_1)) \oplus \delta^1(T(u_2)) \oplus \cdots \oplus \delta^1(T(u_{\deg(r)})); \\
M(T) &= M(u_1) \oplus M(u_2) \oplus \cdots \oplus M(u_{\deg(r)}).
\end{align*}
\]

**Lemma 4.** Given a rooted multi-tree \(T\), let \(\tau\) denote the ordered multi-tree such that \(\sigma(T) = (\delta(\tau), M(\tau))\), and let \(\prec\) denote the left-to-right ordering among siblings in \(\tau\). For any two siblings \(u\) and \(v\) in \(\tau\), if \(u \prec v\), then it holds that \((\delta(\tau_v), \text{mul}(v, p(v)), M(\tau_v)) \prec (\delta(\tau_u), \text{mul}(u, p(u)), M(\tau_u))\).

**Proof.** To derive a contradiction, suppose that there exist siblings \(u\) and \(v\) in \(\tau\) such that \(u \prec v\) and \((\delta(\tau_u), \text{mul}(u, p(u)), M(\tau_u)) \prec (\delta(\tau_v), \text{mul}(v, p(v)), M(\tau_v))\) holds. Let \(\tau'\) denote the ordered multi-tree obtained by switching the places of \(u\) and \(v\) in \(\tau\). Clearly, \(\tau'\) is isomorphic to \(T\) and \((\delta(\tau), M(\tau)) \prec (\delta(\tau'), M(\tau'))\). This contradicts the assumption that \(\sigma(T) = (\delta(\tau), M(\tau))\), i.e., that \(\sigma(\tau)\) is lexicographically maximum among all of the ordered multi-trees isomorphic to \(T\).

By Observation 3 and Lemma 4, we show an algorithm to calculate the signature of a given rooted multi-tree in Procedure 2. As an added benefit, the procedure in fact calculates the signatures of all rooted subtrees of a given tree.

**Procedure 2** SUBTreeSignature

**Input:** A \(\Sigma\)-colored multi-tree \(T\) with multiplicity at most \(d\) rooted at a vertex \(r \in V(T)\).

**Output:** The signatures \(\sigma(T_v)\) of each rooted tree \(T_v, v \in V(T)\).

1. \(s := \emptyset;\)
2. **for each** \(v \in V(T)\) in DFS-post order **do**
3. \(\text{if } v \text{ is a leaf} \text{ then} \)
4. \(\delta[v] := (\text{col}(v), 0); M[v] := \emptyset\)
5. **else**
6. /* The signatures \(s[u]\) of all children of \(v\) are already obtained */
7. **for each** \(u \in \text{CH}(v)\) **do**
8. \(\delta' := 1\text{-shift of } \delta[u];\)
9. \(M' := \text{mul}(u, p(u)) \oplus M[u];\)
10. \(s'[u] := (\delta', M')\)
11. **end for;**
12. \(S := (s'[u] \mid u \in \text{CH}(v));\)
13. **Let** \(k := |\text{CH}(v)|;\)

14
/* Represent \( S \) as \((s_i = (\delta_i, M_i) \mid i \in [1, k])\) */

13: Sort \( S \) in lexicographically descending order \( \pi \);
14: \( \delta[v] := (\text{col}(v), 0) \oplus \delta_\pi(1) \oplus \delta_\pi(2) \oplus \cdots \oplus \delta_\pi(k) \);
15: \( M[v] := M_\pi(1) \oplus M_\pi(2) \oplus \cdots \oplus M_\pi(k) \)
16: end if
17: \( s[v] := (\delta[v], M[v]) \)
18: end for;
19: output \( s[v] \) as \( \sigma(T_v) \) for each \( v \in V(T) \).

Lemma 5. Given a \( \Sigma \)-colored rooted tree \( T \) on \( n \) vertices and multiplicity at most \( d \), Procedure 2 computes the signatures \( \sigma(T_v) \) of all rooted subtrees \( T_v, v \in V(T) \), of \( T \) in \( O(n \cdot (n + |\Sigma| + d)) \) time.

Proof. Let \( n_v \) denote the number of vertices in the subtree \( T_v \) rooted at vertex \( v \), and \( d_v \) the maximum depth of a leaf in the rooted tree \( T_v \), where \( v \) is taken to have depth 0, and it holds that \( d_v \leq n_v \).

The for-loop of lines 2 to 18 is executed for each vertex \( v \) in \( T \). Since vertices are iterated in an DFS-post order, in each iteration, the signatures \( \sigma(T_v) \) are already computed for each child \( u \) of \( v \) in \( T \). Then, in the for-loop of lines 6 to 10, their signatures are gathered and the depth entries are offset by 1 in line 7. This obviously takes at most \( O(n_v) \) time. Then, in line 13 the gathered sequences are sorted lexicographically. The total length of the sequences is \( O(n_v) \), and they are over the alphabets \( \Sigma \) for the color of each vertex, \([1, d_v]\) for the depth, and \([1, d]\) for the multiplicity, thus the total alphabet size is \(|\Sigma| + d + d_v\). By the algorithm for lexicographical sorting due to Aho et al. [1], the lexicographical sorting in line 13 takes \( O(n_v + |\Sigma| + d + d_v) \) time. Finally, summing over all vertices \( v \) in \( T \), for the computational complexity we get

\[
\sum_{v \in V(T)} O(n_v + |\Sigma| + d + d_v) = O\left( \sum_{v \in V(T)} (n_v + d_v) + n \cdot (|\Sigma| + d) \right)
= O(n^2 + n \cdot (|\Sigma| + d)),
\]

as required.

5.2.4 Ranking of Rooted Trees

Let \( T \) be a finite set of rooted multi-trees, and let \( Z = \{\sigma(T) \mid T \in T\} \) denote the set of signatures of the trees in \( T \). We define a lexicographical order over \( Z \) in the usual sense, i.e., for \( \sigma_1 = (\delta_1, M_1), \sigma_2 = (\delta_2, M_2) \in Z \) we write \( \sigma_1 < \sigma_2 \) if “\( \delta_1 < \delta_2 \)” or “\( \delta_1 = \delta_2 \) and \( M_1 < M_2 \).” Then, we use the lexicographical order over the set \( Z \) to define a ranking \( \text{rank}_T : T \rightarrow [1, |Z|] \), such that for two trees \( T_1, T_2 \in T \), \( \text{rank}_T(T_1) < \text{rank}_T(T_2) \) if \( \sigma(T_1) < \sigma(T_2) \), and \( \text{rank}_T(T_1) = \text{rank}_T(T_2) \) means that \( \sigma(T_1) = \sigma(T_2) \), i.e. \( T_1 \) and \( T_2 \) are isomorphic. It follows that having a rank function over a set of multi-trees, we can check whether two trees in the set are isomorphic to each other by comparing their ranks.

There exist algorithms in the literature that can calculate the rank of each subtree of a given tree [10] and rooted subgraph of an outerplanar graph [13] in time linear in...
the number of vertices in the graph. In our implementation we use simpler algorithms for this purpose at the cost of a higher time complexity.

For a set \(T\) of rooted trees, let \(T^*\) denote the set of all rooted subtrees of trees in \(T\). We give a procedure to calculate a ranking of a given set \(T\) of rooted trees in Procedure 3. By Procedure 2, we in fact obtain a ranking in the set \(T^*\) at no additional cost.

**Procedure 3 TreeRanking**

**Input:** A set \(T\) of \(\Sigma\)-colored rooted multi-trees with multiplicity at most \(d\).

**Output:** A ranking function \(\text{rank}_{T^*}\) of \(T^*\).

1. \(R := \emptyset; h := |T^*|;\)
2. \(S := (\sigma(T_i) \mid i \in [1, h]); /* Calculate \(\sigma(T)\) by Procedure 2 in Sec. 5.2.3 */

   \(* /\) Treat \(S = (s_1, s_2, \ldots, s_h)\) as an ordered set */
3. Sort \(S\) in lexicographically ascending order \(\pi;\)
4. \(R[T_{\pi(1)}] := 1; r := 1;\)
5. for each \(i \in [2, h]\) do
6. \(\text{if } s_{\pi(i-1)} < s_{\pi(i)} \text{ then } r := r + 1 \text{ endif};\)
7. \(R[T_{\pi(i)}] := r;\)
8. end for;
9. output \(R\) as \(\text{rank}_{T^*}\).

**Lemma 6.** Let \(T\) be a given set of \(\Sigma\)-colored rooted multi-trees with multiplicity at most \(d\), and let \(n\) denote the total number of vertices over trees in \(T\). Then, the rank of each rooted subtree of all trees in \(T\) can be computed in \(O(n(n + |\Sigma| + d))\) time in total.

**Proof.** Let \(T = \{T_1, T_2, \ldots, T_k\}\), and let \(n_i, i \in [1, k]\) denote the number of vertices in tree \(T_i\), where \(n = \sum_{i=1}^k n_i\). The signature \(\sigma(T_i) = (\delta(T_i), M(T_i))\) of each tree \(T_i\) is a sequence with \(O(n_i)\) entries with \(|\Sigma| + n_i + d\) possible values, for the color of vertices and depth in a tree in \(\delta(T_i)\), and multiplicity with the parent in \(M(T_i)\), respectively.

By Lemma 5, computing the signatures of all rooted subtrees in line 2 takes \(O(n_i \cdot (n_i + |\Sigma| + d))\) time for each tree \(T_i\), and therefore \(O(n \cdot (n + |\Sigma| + d))\) time in total. Now, each tree \(T_i\) has \(n_i\) rooted subtrees, and the total number of vertices over these subtrees is \(O(n_i^2)\). Therefore, the collection of signatures for the rooted subtrees of tree \(T_i\) has in total \(O(n_i^2)\) elements taking at most \(|\Sigma| + n_i + d\) different values (alphabet size). Over all trees \(T_i\), the elements of the subtree signatures take at most \(|\Sigma| + \max_{i \in [1, k]} \{n_i\} + d \leq |\Sigma| + n + d\) different values. Summing over all trees \(T_i \in T\), we get that the total length of the signatures over all subtrees is \(\sum_{i \in [1, k]} O(n_i^2) = O(n^2)\). Then, all these signatures can be lexicographically sorted in \(O(n^2 + |\Sigma| + d)\) time [11], which is dominated by the time to calculate the signatures.

Finally, having the lexicographically sorted signatures, we assign rank to trees in a straightforward manner by iterating over the sorted signatures as in lines 5 to 8 in Procedure 3, and the claim follows. \(\square\)
5.3 Code on Substructures of Mono-block 2-Augmented Trees

We can use the signature defined for rooted trees in Section 5.2 to devise codes for substructures of $k$-augmented trees, $k = 1, 2$. Let $H$ be a mono-block $k$-augmented tree with a block $B$, and let $\mathcal{T}(H) = \{H(v) \mid v \in B\}$ denote the set of all pendant trees in $H$. Recall that for a multi-tree $T \in \mathcal{T}(H)$, $\text{rank}_{\mathcal{T}(H)}(T)$ as defined in Section 5.2.3 gives the rank of $T$ according to the lexicographical order of the signature $\sigma(T)$. To simplify our notation, when the graph $H$ and hence the set $\mathcal{T}(H)$ is clear, we write $\text{rank}(\cdot)$ for the rank function $\text{rank}_{\mathcal{T}(H)}(\cdot)$.

Let $H$ be a mono-block 2-augmented tree. Recall that there are exactly two vertices called junctions, which are contained in all of the cycles in $H$, as defined in Section 2.2.2. Also recall that for a junction $u$ in $H$ and a neighbor $u'$ of $u$ on a cycle of $H$, we call the pair $(u, u')$ a junction pair. Recall that there are at most six junction pairs in a mono-block 2-augmented tree. Let $u$ and $v$ denote the junctions of $H$. For a junction $u$, we define $\text{code}(u)$ to be the sequence $([V(H(u))], \text{col}(u), \text{deg}(u), \text{rank}(H(u)))$. For junctions $u$ and $v$ and a cycle $C_i$ of $H$, recall that $P(u, v; C_i)$ denotes the $uv$-path $(v_1, v_2, \ldots, v_p)$ such that $v_j \notin V(C_i)$ for $j \in [2, p - 1]$, let $n(P(u, v; C_i))$ denote the number of vertices $\sum_{j=2}^{p-1} |V(H(v_j))|$, and we define the code $\text{code}(P(u, v; C_i))$ of the path $P(u, v; C_i)$ to be the following sequence:

$$\text{code}(P(u, v; C_i)) \triangleq (n - n(P(u, v; C_i)), |P(u, v; C_i)|, \text{rank}(H(v_1)), \text{mul}(v_1, v_2), \text{rank}(H(v_2)), \ldots, \text{mul}(v_{p-1}, v_p), \text{rank}(H(v_p)))$$

For a junction $u$, we define $\text{code}^*(u)$ to be the sequence obtained by arranging the codes of $P(u, v; C_i)$, $i \in [1, 3]$, in lexicographically non-ascending order.

6 Parent of a Mono-block 2-Augmented Tree

Let $H$ be a mono-block 2-augmented tree. Let $C_i$, $i \in [1, 3]$, denote the cycles of $H$, and let $u$ and $v$ denote the junctions of $H$. Without loss of generality, we assume that $(\text{code}(u), \text{code}^*(u)) \preceq (\text{code}(v), \text{code}^*(v))$. For the cycle $C^*$ such that $\text{code}(P(u, v; C^*)) \preceq \text{code}(P(u, v; C_i))$, $i \in [1, 3]$, let $e^*$ denote the edge between $u$ and the neighbor of $u$ in $P(u, v; C^*)$. We define the parent of $H$ to be the graph $H - e^*$.

**Lemma 7.** Let $H$ be a mono-block 2-augmented tree with $n$ vertices. Let $C_i$, $i \in [1, 3]$ denote the cycles of $H$, and let $u$ and $v$ denote the junctions of $H$ such that $(\text{code}(u), \text{code}^*(u)) \prec (\text{code}(v), \text{code}^*(v))$. For the cycle $C^*$ such that $\text{code}(P(u, v; C^*)) \preceq \text{code}(P(u, v; C_i))$, $i \in [1, 3]$, the number of edges in $P(u, v; C^*)$ is at least 2.

**Proof.** From the definition, a cycle has at least three vertices. Since $H$ has three distinct cycles, there exists an integer $i \in [1, 3]$ such that $|P(u, v; C_i)| \geq 2$. Suppose that $P(u, v; C^*) = uv$. We have $\text{code}(P(u, v; C_i)) \prec \text{code}(P(u, v; C^*))$ (resp., $\text{code}(P(u, v; C^*))$ is $n - n(P(u, v; C_i)) < n - 1$ (resp., $n - n(P(u, v; C^*)) = n - 1$). This, however, contradicts that $\text{code}(P(u, v; C^*))$ is the lexicographically minimum among $\text{code}(P(u, v; C_i))$, $i \in [1, 3]$. As a result, we see that $P(u, v; C^*) \neq uv$, and the number of edges in $P(u, v; C^*)$ is at least 2. \qed
7 Necessary and Sufficient Conditions for Generating Children

Let $G$ be a monocyclic graph with a cycle $C$. For a non-adjacent vertex pair $\{x,y\}$ in $G$, and an integer $p \in [1, \min\{\res(x), \res(y)\}]$, if $\rho_G(x) = \rho_G(y)$ holds, then the graph $G + p \cdot xy$ will have two blocks, and will not be a mono-block 2-augmented tree. Hence, in order to generate a mono-block 2-augmented tree, we must choose a vertex pair $\{x,y\}$ satisfying $\rho_G(x) \neq \rho_G(y)$. Observe that then $\rho_G(x)$ and $\rho_G(y)$ will be the junctions in the mono-block 2-augmented tree $G + p \cdot xy$. Now, we derive necessary and sufficient conditions to determine whether $G + p \cdot xy$ is a child of $G$ or not.

Lemma 8. Let $G$ be a monocyclic graph, and let $C$ denote the cycle of $G$. Let $x$ and $y$ be non-adjacent vertices in $V(G)$ with $\rho_G(x) \neq \rho_G(y)$, and let $p \in [1, \min\{\res(x), \res(y)\}]$. Let $H$ denote the graph $G + p \cdot xy$, where $\rho_G(x)$ and $\rho_G(y)$ become the junctions in $H$, and let $C_i$, $i \in [1,3]$, denote the three cycles in $H$. Then $H$ is a child of $G$ if and only if the following conditions are satisfied:

(i) $x = \rho_G(x)$ and $y \neq \rho_G(y)$ (i.e., $x \in V(C)$ and $y \notin V(C)$);
(ii) $(\code(\rho_G(x)), \code^*(\rho_G(x))) \preceq (\code(\rho_G(y)), \code^*(\rho_G(y)))$; and
(iii) $\code(P(\rho_G(x), \rho_G(y); C)) \preceq \code(P(\rho_G(x), \rho_G(y); C_i))$, for all $i \in [1, 3]$.

Proof. Necessity. Let $H$ denote the mono-block 2-augmented tree constructed as $G + p \cdot xy$. Suppose that both $x$ and $y$ are in $V(C)$, then the junctions of $H$ are $x$ and $y$. From Lemma 7, we see that $H$ is not a child of $G$. Suppose that neither of $x$ and $y$ is in $V(C)$, then the edge $xy$ is not incident to a junction of $H$, and we see that $H$ is not a child of $G$, and hence Condition (i) must hold.

Next, we consider the case when vertices $x$ and $y$ satisfy Condition (i) but do not satisfy Condition (ii). In this case, the parent of $H$ is obtained by deleting an edge incident to $\rho_G(y)$, and we see that $H$ is not a child of $G$.

Finally, we assume that $x$ and $y$ satisfy Conditions (i) and (ii) but do not satisfy Condition (iii). In this case, there exists a cycle $C^* \neq C$ such that $\code(P(\rho_G(x), \rho_G(y); C^*)) \prec \code(P(\rho_G(x), \rho_G(y); C))$. The parent of $H$ is obtained by deleting the edge between $x$ and the neighbor of $x$ in $P(x, \rho_G(y); C^*)$ but not the edge $xy$. Hence we see that $H$ is not a child of $G$.

Sufficiency. By choosing $x = \rho_G(x) \neq \rho_G(y) \neq y$ we see that $H = G + p \cdot xy$ is a mono-block 2-augmented tree with junctions $x = \rho_G(x)$ and $\rho_G(y)$ and that $xy$ is a junction pair in $H$. The requirements of (ii) and (iii) in the lemma follow the definition of a parent in Section 6.

Let $G$ be a monocyclic graph with a cycle $C$. Let $x$ and $y$ be non-adjacent vertices in $G$ such that $x = \rho_G(x) \neq \rho_G(y)$, and $y \notin V(C)$, and let $p \in [1, \min\{\res(x), \res(y)\}]$. Let $H$ be the mono-block 2-augmented tree $G + p \cdot xy$. The pendent tree $H\langle x \rangle$ is equivalent to $G\langle x \rangle$. Let $v_y$ be the child of $\rho_G(y)$ such that $G\langle v_y \rangle$ contains $y$. Then the pendent
tree $H\langle \rho_G(y) \rangle$ is equivalent to $G\langle \rho_G(y) \rangle - G(v_y)$. From Condition (ii) of Lemma X if $|V(G\langle \rho_G(y) \rangle)| - |V(G\langle v_y \rangle)| < |V(G\langle x \rangle)|$ holds, then we have

$$(\text{code}(\rho_G(y)), \text{code}^*(\rho_G(y))) < (\text{code}(\rho_G(x)), \text{code}^*(x)),$$

and we see that $H$ is not a child of $G$. Let $C_i$ denote the cycles of $H$ for $i \in [1, 3]$, where $C_1 = C$. We have $n(P(u, v; C)) = |V(G\langle v_y \rangle)|$. From Condition (iii) of Lemma X if $|V(G\langle v_y \rangle)| < \min\{n(P(u, v; C_2)), n(P(u, v; C_3))\}$ holds, then code($P(\rho_G(x), \rho_G(y); C))$ is not the lexicographically minimum among code($P(\rho_G(x), \rho_G(y); C_i)), i \in [1, 3]$, and $H$ is not a child of $G$. From these observations, we have the following lemma.

**Lemma 9.** Let $G$ be a monocyclic graph with $n$ vertices, and let $C$ denote the cycle of $G$. Let $x$ and $y$ be two non-adjacent vertices in $V(G)$ such that $x = \rho_G(x) \neq \rho_G(y)$ and $y \notin V(C)$, and let $p \in [1, \min\{\text{res}(x), \text{res}(y)\}]$. If $G + p \cdot xy$ is a child of $G$, then it holds that $|V(G\langle \rho_G(y) \rangle)| \geq n/3$.

**Proof.** Let $H$ be the mono-block 2-Augmented tree $G + p \cdot xy$, and let $C_i, i \in [1, 3]$, denote the cycles of $H$, where $C_1 = C$. The junctions in $H$ are $\rho_G(x)$ and $\rho_G(y)$. Let $n_y$ and $n_y'$ denote the numbers $n(P(\rho_G(x), \rho_G(y); C_2))$ and $n(P(\rho_G(x), \rho_G(y); C_3))$, respectively. Without loss of generality, we assume that $n_y \leq n_y'$. Since the number of vertices in $G$ is $n$, we have $n = |V(G\langle x \rangle)| + |V(G\langle y \rangle)| + n_y + n_y'$. Let $v_y$ be the child of $\rho_G(y)$ such that $G\langle v_y \rangle$ contains $y$. If $H$ is a child of $G$, then from Conditions (ii) and (iii) of Lemma X we have $|V(G\langle \rho_G(y) \rangle)| - |V(G\langle v_y \rangle)| \geq |V(G\langle x \rangle)|$ and $|V(G\langle v_y \rangle)| \geq n_y'$.

Therefore, we have

$$|V(G\langle \rho_G(y) \rangle)| \geq |V(G\langle x \rangle)| + |V(G\langle v_y \rangle)|$$

$$\geq |V(G\langle x \rangle)| + n_y'$$

$$\geq n - |V(G\langle \rho_G(y) \rangle)| - n_y - n_y' + n_y'$$

$$\geq (n - n_y)/2.$$  

Since we have $n_y \leq n_y' \leq |V(G\langle v_y \rangle)|$ and $n = |V(G\langle x \rangle)| + |V(G\langle \rho_G(y) \rangle)| + n_y + n_y'$, we obtain $n_y \leq n/3$ and $|V(G\langle \rho_G(y) \rangle)| \geq (n - n_y)/2 \geq n/3$.  

As a consequence of Lemma X for a monocyclic graph $G$ with $n$ vertices and cycle $C$, a pair $\{x, y\}$ of non-adjacent vertices in $G$ with $x = \rho_G(x) \neq \rho_G(y)$ and $y \notin V(C)$, and an integer $p \in [1, \min\{\text{res}(x), \text{res}(y)\}]$, if $V(G\langle \rho_G(y) \rangle)| < n/3$ holds, then it holds that $G + p \cdot xy$ is not a child of $G$.

**Lemma 10.** Let $G$ be a monocyclic graph with a cycle $C$ and let $r \in V(C)$. If $G$ has a pendent tree $G(r^*)$ such that $r^* \neq r$ and $|V(G\langle r^* \rangle)| \leq |V(G\langle r \rangle)|$, then for any pair $\{x, y\}$ of non-adjacent vertices such that $x \in V(C)$ and $y \in V(G\langle r \rangle)$ and an integer $p \in [1, \min\{\text{res}(x), \text{res}(y)\}]$ it holds that $G + p \cdot xy$ is not a child of $G$.

**Proof.** Let $r^*$ denote the root of a pendent tree such that $|V(G\langle r \rangle)| \leq |V(G\langle r^* \rangle)|$. Let $H$ be the graph $G + p \cdot xy$, where $\rho_G(x)$ and $\rho_G(y)$ are the junctions in $H$, and let
Let $C_i, i \in [1, 3]$, denote the cycles of $H$, where $C_1 = C$. Suppose that $x \neq r^*$. Then, vertex $r^*$ is included in some path $P(\rho_G(x), \rho_G(y); C_j), j \in \{1, 2, 3\}$, and it holds that $n(P(\rho_G(x), \rho_G(y); C_j)) \geq |V(G(r^*))|$. Let $v$ be the child of $r$ such that $V(G(v))$ contains $y$. For the path $P(\rho_G(x), \rho_G(y); C)$, we have $n(P(\rho_G(x), \rho_G(y); C)) = |V(G(v))| < |V(G(r))|$. Hence we have $\text{code}(P(\rho_G(x), \rho_G(y); C)) < \text{code}(P(\rho_G(x), \rho_G(y); C^*))$, which contradicts Condition (iii) of Lemma 8.

Next, suppose that $x = r^*$. The junctions of $H$ are $r = \rho_G(y)$ and $r^* = x$. Let $v$ be the child of $r$ such that $V(G(v))$ contains $y$, and we have $|V(H(r))| = |V(G(r))| - |V(G(v))|$ and $|V(H(r^*))| = |V(G(r^*))|$. Therefore, since we have $|V(H(r^*))| > |V(H(r))|$ it holds that $(\text{code}(\rho_G(x))), \text{code}^*(x)) > (\text{code}(\rho_G(y)), \text{code}^*(\rho_G(y)))$, and therefore $G + p \cdot xy$ is not a child of $G$, as required.

Finally, from Lemmas 8, 9 and 10 we have the following necessary and sufficient conditions that a pair of non-adjacent vertices in a monocyclic graph $G$ must satisfy in order to obtain a child mono-block 2-augmented tree by adding multiple edges between them.

**Lemma 11.** Let $G$ be a monocyclic graph with $n$ vertices and a cycle $C$. Let $x$ and $y$ be non-adjacent vertices in $V(G)$ with $\rho_G(x) \neq \rho_G(y)$, and $p \in [1, \min\{\text{res}(x), \text{res}(y)\}]$. Let $H$ denote the graph $G + p \cdot xy$, where $\rho_G(x)$ and $\rho_G(y)$ are the junctions in $H$, and let $C_i, i \in [1, 3]$ denote the three cycles in $H$. Then $H$ is a child of $G$ if and only if the following conditions are satisfied:

(i) $|V(G(\rho_G(y))))| \geq n/3$;

(ii) $|V(G(\rho_G(y))))| > |V(G(r))|$ for each $r \in V(C) \setminus \{\rho_G(y)\}$;

(iii) $x = \rho_G(x)$ and $y \neq \rho_G(y)$, (i.e., $x \in V(C)$ and $y \not\in V(C)$);

(iv) $(\text{code}(\rho_G(x)), \text{code}^*(\rho_G(x))) \preceq (\text{code}(\rho_G(y)), \text{code}^*(\rho_G(y)))$; and

(v) $\text{code}(P(\rho_G(x), \rho_G(y); C)) \preceq \text{code}(P(\rho_G(x), \rho_G(y); C_i)), i \in [1, 3]$.

### 7.1 Preprocessing for Efficient Computation

Notice that in Lemma 11(iv) and (v), in order to check whether a mono-block 2-augmented tree $H$ obtained by adding an edge to a pair of nonadjacent vertices in a monocyclic graph $G$ is indeed a child of $G$ or not, requires our knowledge of the rank of pendent trees of $H$ in the set $T(H)$ of all pendent trees in $H$. This computation might seem wasteful, as a single monocyclic graph $G$ may have many candidates for children mono-block 2-augmented trees. We here give an observation that there exists a set $T$ of selected pendent trees of $G$ and their subtrees, such that this set will contain as a subset the set of pendent trees of any graph $H$ obtained by adding an edge between a pair of non-adjacent vertices in $G$. Then, to save on computation effort, we calculate the rank of rooted trees in this set $T$ only once per monocyclic graph $G$.  

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Let $G$ be a monocyclic graph with a cycle $C = (v_0, v_1, \ldots, v_{n-1}, v_0)$. Then, in addition to the set $\mathcal{T}(G) = \{G(v_i) \mid i \in [0, n-1]\}$ of pendent trees, we define the following sets of rooted trees.

- $\tilde{T}(G) = \{G(u) \mid u \in V(G(v_0)) \setminus \{v_0\}\}$
- $\hat{T}(G) = \{G(p(u)) - G(u) \mid u \in V(G(v_0)) \setminus \{v_0\}\}$.

Finally, we define the union $\mathcal{T}(G) = T(G) \cup \hat{T}(G) \cup \tilde{T}(G)$.

**Lemma 12.** Given a monocyclic graph $G$ with a cycle $C = (v_0, v_1, \ldots, v_{n-1}, v_0)$, let $\{x, y\} \in \mathcal{E}(G)$ be a pair of non-adjacent vertices such that $x \in V(C) \setminus \{v_0\}$ and $y \in V(G(v_0)) \setminus \{v_0\}$, and let $H = G + xy$ denote the graph obtained from $G$ by adding an edge $xy$. Then it holds that $\mathcal{T}(H) \subseteq \mathcal{T}(G) \cup \hat{T}(G)$.

**Proof.** By the choice of $x$ and $y$ the graph $H$ is a mono-block 2-augmented tree and the junctions of $H$ are the vertices $x$ and $v_0$. Let $P_i$, $i = 1, 2, 3$, denote the three $x, v_0$-paths in $H$, such that $xy \in E(P_3)$, i.e., $P_3 = P(x, y; C)$. The pendent trees $G(v_i)$, $i \in [1, n-1]$ are preserved between $G$ and $H$. Therefore, we focus on the pendent tree $G(v_0)$. Denote by $Q = (v_0 = v_0, u_2, \ldots, u_{k-1} = y)$ the $v_0, y$-path in the tree $G(v_0)$. Then, adding the edge $xy$ to path $Q$ we obtain $P_3$ in $H$, and for each rooted tree $G(u_i)$, $i \in [0, k-2]$, the tree $G(u_i - G(u_{i+1})$ becomes the rooted tree $H(u_i)$ in the mono-block 2-augmented tree $H$ (see Fig. 3). Equivalently, for $i \in [1, k-1]$ the tree $G(u_{i-1}) - G(u_i)$ becomes the pendent tree $H(u_i)$ in $H$. Since for $i \in [1, k-1]$ it holds that $p(u_i) = u_{i-1}$ in $G(v_0)$, the claim follows.

![Diagram](image)

(a) A monocyclic graph $G$ with $\{x, y\} \in \mathcal{E}(G)$.

(b) The mono-block 2-augmented tree $H = G + xy$.

Figure 3: (a) A monocyclic graph $G$ where the unique cycle is denoted by $C = (v_0, v_1, \ldots, v_{n-1}, v_0)$, with a pair $\{x, y\}$ of non-adjacent vertices such that $x \in V(C)$ and $y \in V(G(v_0)) \setminus \{v_0\}$. (b) The mono-block 2-augmented tree $H = G + xy$ obtained by adding an edge $xy$ to the monocyclic graph $G$ in (a). The subtrees denoted by dark gray are preserved from $G$ in $H$. 

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Lemma 13. Given an \( n \)-vertex \( \Sigma \)-colored monocyclic graph \( G \) with multiplicity at most \( d \) and a unique cycle \( C = (v_0, v_1, \ldots, v_{m-1}, v_0) \), the rank of all trees in \( T(G) \) can be computed in \( O(n \cdot (n + |\Sigma| + d)) \) time in total.

Proof. The set \( T(G) \) is composed of the set \( T(G) \) of pendent trees of the graph \( G \), the set \( \hat{T}(G) \) of rooted subtrees of pendent trees of \( G \) and trees in the set \( \tilde{T}(G) \) that are obtained as a difference between rooted subtrees of \( G \). Then, by the observation made in Section 5.2.2 that for a left-heavy tree \( T \) and any rooted subtree \( T' \) of \( T \) the sequences \( \delta(T') \) and \( M(T') \) are continuous subsequences of \( \delta(T) \) and \( M(T) \), it is not difficult to obtain the signatures of all trees in the set \( \tilde{T}(G) \). Finally, since the total number of vertices of trees in the set \( \tilde{T}(G) \) is not more than that of the set \( \tilde{T}(G) \), and Lemma 6, the claim follows. \( \square \)

7.2 A Procedure to Verify Child Conditions

We show an algorithm that for a given monocyclic graph \( G \), a pair \( \{x, y\} \) of non-adjacent vertices in \( G \), and an integer \( p \in [1, \min\{\text{res}(x), \text{res}(y)\}] \), based on Lemma 11 determines whether \( G + p \cdot xy \) is a child of \( G \) or not in Procedure 4 \( \text{CHILDCHECK} \).

Procedure 4 \( \text{CHILDCHECK}(G, \{x, y\}, p) \)

Input: A monocyclic graph \( G \) with \( n \) vertices and a cycle \( C \), a pair \( \{x, y\} \in \overline{E}(G) \) of non-adjacent vertices, and an integer \( p \in [1, \min\{\text{res}(x), \text{res}(y)\}] \).

Output: True if \( G + p \cdot xy \) is a child of \( G \), and False otherwise.

1: Answer := False;
2: if \(|V(G(\rho_G(y)))| \geq n/3\) then
3:   if \(|V(G(\rho_G(y)))| > |V(G(r))|\) for each \( r \in V(C) \setminus \{\rho_G(y)\} \) then
4:     if \( x = \rho_G(x) \neq \rho_G(y) \neq y \) then
5:       Construct \( H := G + p \cdot xy \); /* \( \rho_G(x) \) and \( \rho_G(y) \) are the junctions in \( H \) */
6:       Let \( C_i, i \in [1, 3] \) denote the cycles of \( H \);
7:       if \( \text{code}(\rho_G(x)) < \text{code}(\rho_G(y)) \) then
8:         if \( \text{code}(P(\rho_G(x), \rho_G(y); C)) \leq \text{code}(P(\rho_G(x), \rho_G(y); C_i), i \in [1, 3] \) then
9:           Answer := True
10:       end if
11:     end if
12:   end if
13: else if \( \text{code}(\rho_G(x)) = \text{code}(\rho_G(y)) \) then
14:     if \( \text{code}^*(\rho_G(x)) \leq \text{code}^*(\rho_G(y)) \) then
15:       if \( \text{code}(P(\rho_G(x), \rho_G(y); C)) \leq \text{code}(P(\rho_G(x), \rho_G(y); C_i), i \in [1, 3] \) then
16:         Answer := True
17:       end if
18:     end if
19: end if
20: end if
21: output Answer.
8 Intra-Duplication of Mono-block 2-Augmented Trees

The parent-child relationship helps us avoid inter-duplication, that is, generating isomorphic structures by adding edges to topologically different monocyclic graphs. However, the parent-child relationship is not sufficient to eliminate intra-duplications, since isomorphic children might occur from a single monocyclic graph $G$ by adding an edge between a pair of non-adjacent vertices. Henceforth, we treat a graph as a labeled one and use the information on the labeling, since there is no other way to distinguish isomorphic mono-block 2-augmented trees generated from a single monocyclic graph.

For a monocyclic graph $G$, two distinct non-adjacent vertex pairs $\{x, y\}$ and $\{x', y'\}$, and integers $p \in [1, \min\{\res(x), \res(y)\}]$ and $q \in [1, \min\{\res(x'), \res(y')\}]$, we examine under which conditions $G + p \cdot xy$ and $G + q \cdot x'y'$ are isomorphic.

**Theorem 14.** Let $G$ be a connected graph that contains exactly one cycle $C = (v_0, v_1, \ldots, v_{n-1}, v_0)$, and $\{x_1, y_1\}, i = 1, 2$, be two pairs of non-adjacent vertices in $G$ such that $x_1, x_2 \in V(C) \backslash \{v_0\}$, $y_1, y_2 \notin V(C)$ and $\rho_G(y_1) = \rho_G(y_2) = v_0$. Let $H_i, i = 1, 2$, denote the graph $G + x_iy_i$, and $c$ be a coloring of the graph $H_1 + x_2y_2$. Let $w_2$ denote the child of $v_0$ in the rooted tree $G(v_0)$ such that $G(w_2)$ contains $y_2$. Assume that $H_1$ and $H_2$ are isomorphic. Then one of the following holds.

(i) $y_1 \neq y_2$, $c(x_1y_1) = c(x_2y_2)$, and $G(v_0)$ has an automorphism $\xi$ such that $\xi(v_0) = v_0$ and $\xi(y_1) = y_2$;

(ii) $y_1 = y_2$, $c(x_1y_1) = c(x_2y_2)$, and $G$ has an automorphism $\xi$ such that $\xi(x_1) = x_2$ and $\xi(v_i) = v_{n-i \pmod n}$ for each vertex $v_i \in V(C)$; and

(iii) $y_1 = y_2$, $c(x_1y_1) = c(x_2y_2) = c(v_0w_2)$, $G(w_2)$ has an automorphism $\phi$ such that $\phi(w_2) = y_1$, and $G - G(w_2)$ has an automorphism $\xi$ with an integer $k \geq 1$ such that $\xi(x_1) = x_2$ and $\xi(v_i) = v_{i+k \pmod n}$, $v_i \in V(C)$.

**Proof.** For a subgraph $A$ of $H_i$, $i = 1, 2$, let $A^{(i)}$ denote the subgraph of $H_i$ induced by the vertices in $V(A)$ and $V(H_i(v))$, $v \in A$. Without loss of generality assume that $\{x_1, y_1\} \neq \{x_2, y_2\}$, $x_1 = v_{j_1}$, $x_2 = v_{j_2}$, and $1 \leq j_1 \leq j_2 \leq n - 1$. For each $i = 1, 2$, let $P_i, Q_i$ and $R_i$ denote the paths between vertices $x_i$ and $\rho_G(y_i)$ in $H_i$, where we assume that $y_i \in V(P_i)$, $v_i \in V(Q_i)$ and $v_{n-i} \in V(Q_2)$. Let $\psi : V(H_1) \rightarrow V(H_2)$ be an isomorphism between $H_1$ and $H_2$. Since $H_i$ for each $i = 1, 2$ has exactly one block $B_i$ with exactly two junction vertices $x_i$ and $\rho_G(y_i) = v_0$, each path between $x_1$ and $v_0$ in $H_1$ is mapped by $\psi$ to a path between $x_2$ and $v_0$ in $H_2$. This means that $c(x_1y_1) = c(x_2y_2)$, $\psi(\{x_1, v_0\}) = \{x_2, v_0\}$, $\psi(B_1) = V(B_2)$, $|V(B_1)| = |V(B_2)|$, and $\{\psi(P_i^{(1)}), \psi(Q_i^{(1)}), \psi(R_i^{(1)})\} = \{V(P_2^{(1)}), V(Q_2^{(1)}), V(R_2^{(1)})\}$. Note that $|V(B_i)| = |V(C)| + |V(P_i)| - 2$ for each $i = 1, 2$. Since $|V(B_1)| = |V(B_2)|$, we see that $|V(P_1)| = |V(P_2)|$ and $|V(Q_1)|, |V(R_1)| = |V(Q_2)|, |V(R_2)|$.

(i) Assume that $y_1 \neq y_2$. Let $z$ denote the deepest vertex in $V(P_1) \cap V(P_2)$ in the rooted tree $G(v_0)$, where possibly $z = v_0$. When $z \neq v_0$, let $Z$ denote the path from $w_2$ to $z$ in the subtree $G(v_0)$, where we regard $V(Z)$ as an empty set when $z = v_0$. For each
Let $i = 1, 2$, let $z_i$ denote the child of $z$ such that $y_i \in V(G\langle z_i \rangle)$ and $\overline{P}_i$ denote the subpath of $P_i$ between $z_i$ and $y_i$. Note that

$$\psi(z) \not\in \psi^{-1}(\overline{P}_2),$$

(1)

since otherwise $H_2\langle z \rangle$ would be isomorphic to a proper subgraph $H_2\langle t \rangle$ for the vertex $t = \psi(v_0) \in V(\overline{P}_2)$.

To show that $G\langle v_0 \rangle$ has an automorphism $\xi$ such that $\xi(v_0) = v_0$ and $\xi(y_1) = y_2$, it suffices to prove that $H_2\langle z \rangle$ and $H_1\langle z \rangle$ are $(z, z)$-isomorphic.

(2)

Case 1. $\psi(z) \not\in G(V\langle v_0 \rangle)$: In this case, $\psi(P_1) = V(P_2)$ and “$\psi(v_0) = v_0$ or $z \neq v_0$.” Now $H_1\langle z \rangle$ and $H_2(\psi(z))$ are $(z, \psi(z))$-isomorphic. Hence if $z = v_0$ and $\psi(v_0) = v_0$, where $\psi(z) = z$, then (2) holds. Assume that $z \neq v_0$. We know that $H_1(\psi(z))$ and $H_2(\psi(z))$ are $(\psi(z), \psi^2(z))$-isomorphic. By (1), $\psi(z) \in V(Z)$ holds, implying $H_2(\psi(z)) = H_1(\psi(z))$. Since $\psi$ maps path $P_1 - \{v_0, x_1\}$ to path $P_2 - \{v_0, x_2\}$, we see that $\psi^2(z) = z$. Therefore $H_1(z)$ and $H_2(z)$ are $(z, z)$-isomorphic.

Case 2. $\psi(z) \in V(G\langle v_0 \rangle)$: In this case, “$\psi(P_1) = V(P_2)$, $\psi(x_1) = v_0$ and $z = v_0$” or “$\psi(P_1) \in \{V(Q_2), V(R_2)\}$.” Let $h, k \in [1, n - 1]$ denote the indices such that $v_h = \psi(z) \in \psi(P_1)$ and $v_k = \psi^{-1}(z) \in \psi^{-1}(P_2)$. Define subtrees $T(v), v \in V(C)$ to be $G\langle v \rangle$ if $v \neq v_0$ and $T(v_0) = H_2\langle v_0 \rangle$. For the subset $S_1 = V(C) \setminus \psi^{-1}(\overline{P}_2)$ of $V(C)$, define a function $f : S_1 \rightarrow V(G)$ such that

$$f(v) = \left\{ \begin{array}{ll} \psi(v) & \text{if } v \in S_1 \setminus \psi^{-1}(Z) \\ \psi^2(v) & \text{if } v \in \psi^{-1}(Z). \end{array} \right.$$

We here prove the following properties:

(a-1) For the subset $S_2 = V(C) \setminus \psi(\overline{P}_1)$ of $V(C)$, $f$ is a bijection from $S_1$ to $S_2$;

(a-2) Let $v^1 = v_k$ if $z \neq v_0$ and $v^1 = v_0$ if $z = v_0$. Then $f(v^1) = v_h$. For each vertex $v \in S_1 \setminus \{v^1\}$, $T(v)$ and $T(f(v))$ are $(v, f(v))$-isomorphic; and

(a-3) $f_p(v_0) \in S_2 \setminus \psi(\overline{P}_2) \subseteq S_1$ for any integer $p \geq 1$.

(a-1) Since $S_1 \setminus \psi^{-1}(Z) = V(C) \setminus \psi^{-1}(\overline{P}_2) \setminus \psi^{-1}(Z) = V(C) \setminus \psi^{-1}(V(P_2) \setminus \{v_0\})$, the set $S_1 \setminus \psi^{-1}(Z)$ is mapped by $\psi$ to $V(Q_2) \cup V(R_2) \subseteq V(C)$. Observe that $f(\psi^{-1}(Z)) = \psi(Z) \subseteq V(C)$ and $V(Z) \subseteq V(P_1) \setminus \{v_0\} \subseteq V(G) \setminus V(C)$. Since $S_1 \setminus \psi^{-1}(Z)$ and $V(Z)$ are disjoint and $\psi$ is a bijection from $V(G)$ to $V(G)$, we see that $f(S_1 \setminus \psi^{-1}(Z)) = \psi(S_1 \setminus \psi^{-1}(Z))$ and $f(\psi^{-1}(Z)) = \psi(Z)$ are disjoint. Since $V(\overline{P}_1)$ is disjoint with $(S_1 \setminus \psi^{-1}(Z)) \cup V(Z)$, this also means that $f(S_1) \subseteq V(C) \setminus \psi(\overline{P}_1) = S_2$. Therefore $f(S_1) = S_2$, since $|S_1| = |S_2|$ and $f$ is a bijection from $S_1$ to $f(S_1)$.

(a-2) We distinguish two cases.

Case of $z \neq v_0$, where $v^1 = v_k \in \psi^{-1}(z) \in \psi^{-1}(Z)$: Then $f(v^1) = f(v_k) = \psi^2(v_k) = \psi(z) = v_h$. Let $v \in S_1 \setminus \{v_k\}$. If $\psi(v) \in V(C)$, then $T(v) = H_1(\psi(v))$ and $T(f(v)) = H_2(\psi(v))$ are $(v, \psi(v))$-isomorphic. Note that for any vertex $u \in V(Z) \setminus \{z\}$, $H_1(\psi(u)) = H_2(\psi(u))$. Also $v \neq v_k$ means that $\psi(v) \neq z$. If $\psi(v) \in V(Z) \setminus \{z\}$, then $H_1(\psi(v))$
and $H_2(\psi(v))$ are $(v,\psi(v))$-isomorphic and $H_1(\psi(v))$ and $H_2(\psi^2(v))$ are $(\psi(v),\psi^2(v))$-isomorphic, implying that $T(v) = H_1(v)$ and $T(f(v)) = H_2(\psi^2(v))$ are $(v,\psi^2(v))$-isomorphic.

Case of $z = v_0$, where $v^1 = v_0$. In this case, $V(Z) = \emptyset$ and $f = \psi$. Then $f(v^1) = f(v_0) = \psi(z) = v_h$. Therefore for any vertex $v \in V(C) \setminus \{v_0\}$, $T(v) = H_1(v)$ and $T(f(v)) = H_2(\psi(v))$ are $(v,\psi(v))$-isomorphic.

(a-3) By definition, $v_h = \psi(z) = \psi(P_1) \subseteq V(Q_2) \cup V(R_2) \subseteq V(C)$. By (1), $v_h \in V(C) \setminus \psi^{-1}(\overline{P_2}) = S_1$. By (a-2), $f$ maps a vertex $v \in S$ to a vertex $f(v)$ so that $T(v)$ and $T(f(v))$ are $(v,f(v))$-isomorphic. Hence for any integer $i \geq 1$, $f^i(v_h)$ is not a vertex in $\psi^{-1}(\overline{P_2})$, since otherwise $T(v)$ would be isomorphic to a tree $T(f(v))$ that is a proper subgraph of $T(v)$. Therefore $f^i(v_h)$, $i \geq 1$ is a vertex in $V(C) \setminus \psi^{-1}(\overline{P_2}) = S_1$.

We are ready to prove (2). By (a-3), it holds $f^0(v_h), f(v_h), f^2(v_2), \ldots, f^{S_1+1}(v_h) \in S_1$, and there is an integer $i \in [1, S_1]$ such that $f^i(v_h) = f^i(v_h)$ for some $j \in [0, i-1]$. Let $p$ denote the smallest such integer $i$, where $j = 0$ and $v_h = f^p(v_h)$ since $f$ is a bijection by (a-1). By (a-2), if $v_0 \neq z$ (resp., $v_0 = z$), then $f^{-1}(v_h) = f^{-1}(v_h) = v^1 = v_h$ (resp., $f^{p-1}(v_h) = f^{-1}(v_h) = v^1 = v_0$) and $T(v_h)$ and $T(v_h)$ are $(v_h, v_h)$-isomorphic (resp., $T(v_h)$ and $T(v_h)$ are $(v_h, v_0)$-isomorphic). This proves (2).

In the following, we assume that $y_1 = y_2$, from which it follows that $x_1 \neq x_2$, $j_1 = n-j_2$ and $|V(Q_1)| = |V(Q_2)| < |V(R_1)| = |V(R_2)|$. Hence $\psi(R_1(1)) \in \{V(P_2(2)), V(R_2(2))\}$. We first prove that

$$P_1^{(1)} \text{ and } P_2^{(2)} \text{ have an isomorphism } \eta \text{ such that } \eta(x_1) = x_2 \text{ and } \eta(v_0) = v_0.$$  

(3)

Recall that $c(x_1,y_1) = c(x_2,y_2)$. To prove (3), it suffices to show that $G(x_1)$ and $G(x_2)$ are $(x_1,x_2)$-isomorphic, which immediately holds when $\psi(x_1) = x_2$. When $\psi(x_1) = v_0$ and $\psi(v_0) = x_2$, we see that $G(x_1)$ and $G(x_2)$ are $(x_1,x_2)$-isomorphic, because $H_1(x_1) = G(x_1)$ and $H_2(v_0) = H_1(v_0)$ are $(x_1,v_0)$-isomorphic and $H_1(v_0)$ and $H_2(x_1) = G(x_2)$ are $(v_0,x_1)$-isomorphic. This proves (3).

We distinguish two cases.

(ii) Assume that $y_1 = y_2$ and “$\psi(x_1) = x_2$ or $\psi(P_1) \in \{V(Q_2), V(R_2)\}$.” For two indices $i, j \in [0, n-1]$ with $i < j$ (resp., $i > j$), let $G[i,j]$ denote the subpath of $G$ induced by the vertices $v_2$ with $\ell \in [i,j]$ (resp., $\ell \in [i,n-1] \cup [0,j]$). Note that $R_1 = G[j_1,0]$ and $R_2 = G[0,j_2]$. In this case of (ii), we prove that

$$G[j_1,0]^{(1)} \text{ and } G[0,j_2]^{(2)} \text{ admit an isomorphism } \eta \text{ such that } \eta(v_{j_1}) = v_{j_2} \text{ and } \eta(v_0) = v_0.$$  

(4)

When such an isomorphism $\eta$ exists, then $G$ has an automorphism $\xi$ such that $\xi(x_1) = x_2$ and $\xi(v_1) = v_{n-i} \mod n$ for each vertex $v_1 \in V(C)$.

In what follows, we prove (1). When $\psi(x_1) = x_2$ and $\psi(R_1^{(1)}) = V(R_2^{(2)})$, we see that (1) holds.

We first consider the case of $\psi(P_1^{(1)}) = V(R_2^{(2)})$, where $\psi(R_1) = V(P_2)$ and $\psi(P_1) = V(R_2)$. We see that $R_1^{(1)}$ and $P_2^{(2)}$ are $(v_0,v_0)$-isomorphic (resp., $(x_1,v_0)$-isomorphic) and $P_1^{(1)}$ and $R_2^{(2)}$ are $(v_0,v_0)$-isomorphic (resp., $(x_1,v_0)$-isomorphic) if $\psi(x_1) = x_2$ (resp., $\psi(x_1) = v_0$). By (3), this means that $R_1^{(1)}$ and $R_2^{(2)}$ are $(v_0,v_0)$-isomorphic, implying (1).
We next consider that $\psi(x_1) = v_0$ and $\psi(P_1(1)) = V(Q_2(2))$, where $\psi(Q_1) = V(P_2)$ and $\psi^{-1}(P_2) = V(G[0,j_1])$ and $\psi(P_1) = V(G[j_2,0])$. In this case, $G[0,j_1](1)$ and $G[j_2,0](2)$ are $(v_0, v_0)$-isomorphic. We observe that $G[j_1,0](1)$ (resp., $G[0,j_2](1)$) is a repetition of $G[0,j_1](1)$ (resp., $G[j_2,0](1)$) in the following sense. Let $a = \lceil \frac{j_2-j_1}{j} \rceil$ and $b = (j_2-j_1) - a \cdot j_1$. Since $G[0,j_1](1)$ and $G[j_1,2j](1)$ are $(v_0, v_{j_1})$-isomorphic under the isomorphism $\psi$, we see that for each integer $j \in [0, a-1]$, $G[0,j_1](1)$ and $G[j \cdot j_1, (j+1) \cdot j_1]$(1) are $(v_0, v_{j(j+1)j_1})$-isomorphic. When $b \geq 1$, $G[0,b](1)$ and $G[a \cdot j_1, a \cdot j_1+b](1)$ are $(v_0, v_{a,j_1})$-isomorphic. Symmetrically $G[j_2,0](2)$ and $G[n-2j_2,j-2j_2](2)$ are $(v_0, v_{n-j_2})$-isomorphic, and we see that $G[j_1,0](2)$ and $G[n-(j+1) \cdot j, n-j \cdot j_1](2)$ are $(v_0, v_{n-j_2})$-isomorphic for each integer $j \in [0, a-1]$, where for $b \geq 1$, $G[n-b,0](2)$ and $G[n-a \cdot j_1-b, n-a \cdot j_1](2)$ are $(v_0, v_{n-a,j_1})$-isomorphic. Recall that $G[0,j_1](1)$ and $G[j_2,0](2)$ are $(v_0, v_0)$-isomorphic. Hence $G[j_1,0](1)$ and $G[0,j_2](2)$, which are repetitions of $G[0,j_1](1)$ and $G[j_2,0](2)$, respectively, are $(v_0, v_0)$-isomorphic. This proves (ii).

(iii) Finally assume that $y_1 = y_2$, $\psi(x_1) = v_0$ and $\psi(P_1) = V(P_2)$. Since $P_1(1)$ and $P_2(2)$ are $(x_1, v_0)$-isomorphic, we see that $c(x_1 y_1) = c(x_2 y_2) = c(v_0 w_2)$, $G \langle w_2 \rangle$ has an automorphism $\phi$ such that $\phi(w_2) = y_1$. Let $k = |E(Q_1)|$. Now $R_1(1)$ and $R_2(2)$ are $(x_1, v_0)$-isomorphic and $Q_1(1)$ and $Q_2(2)$ are $(v_0, x_2)$-isomorphic. This means that $G - G \langle w_2 \rangle$ has an automorphism $\xi$ such that $\xi(x_1) = x_2$ and $\xi(v_1) = v_{i+k \mod n}$, $v_i \in V(C)$.

\[ \square \]

In addition, we have the following lemma.

**Lemma 15.** Let $G$ be a connected graph that contains exactly one cycle $C = (v_0, v_1, \ldots, v_{n-1}, v_0)$, let $x_1$ and $x_2$ be distinct vertices in $V(C) \setminus \{v_0\}$, where $x_1 = v_k$, and let $y$ be a vertex in $V(G \langle v_0 \rangle) \setminus \{v_0\}$. For $i = 1, 2$, let $p_i \in [1, \min\{\text{res}(x_i), \text{res}(y)\})$, and let $H_i$ denote $G + p_i \cdot x_i y$. If $H_1$ and $H_2$ are isomorphic, then it holds that $x_2 = v_{n-k}$.

**Proof.** Let $B_i$, $i = 1, 2$, denote the block in $H_i$, and let $P_1$, $P_2$, and $P_3$ be the paths in $H_1$ from $x_1$ to $v_0$, where $y \in V(P_1)$, $v_1 \in V(P_2)$ and $v_{n-1} \in V(P_3)$. Let $Q_1$, $Q_2$, $Q_3$ be the paths in $H_2$ from $x_2$ to $v_0$, where $y \in V(Q_1)$, $v_1 \in V(Q_2)$ and $v_{n-1} \in V(Q_3)$. We have $|V(C)| + V(P_1) - 2 = |V(B_1)| = |V(B_2)| = |V(C)| + V(Q_1) - 2$. Hence, we get $|P_1| = |Q_1|$ and $\{|P_2|, |P_3|\} = \{|Q_2|, |Q_3|\}$.

Suppose that $|P_2| = |P_3|$ and $|Q_2| = |Q_3|$ hold. Then we have $|P_2| = |P_3| = |Q_2| = |Q_3| = n/2$. Since the length of a path is an integer, in order to satisfy the condition, $n$ must be even, and then $x_1 = x_2 = v_{n/2}$ holds. However, this contradicts that $x_1$ and $x_2$ are distinct vertices. Therefore, we have $|P_2| \neq |P_3|$ and $|Q_2| \neq |Q_3|$. Next, we see that $|P_2| = |Q_2|$ would again imply that $x_1 = x_2$, and therefore it holds that $|P_2| = |Q_2|$ and $|P_3| = |Q_2|$. Let $k \in [1, n-1]$ be an integer such that $x_1 = v_k$. In order to satisfy the condition $|P_2| = |Q_3|$, $x_2$ must be $v_{n-k}$, as required.

\[ \square \]

Let $G$ be a monocyclic graph and $C = (v_0, v_1, \ldots, v_{n-1}, v_0)$ denote the cycle in $G$. If $G$ admits an automorphism $\xi$ such that $\xi(v_i) = v_{n-i \mod n}$ for each vertex $v_i \in V(C)$, as in Theorem 4(ii), then we say that $G$ admits an axial symmetry $\xi$. Further, for a vertex $y \in V(C) \setminus \{v_0\}$, let $q$ be the child of $v_0$ such that $G(q)$ contains $y$. If there exists an automorphism $\phi$ on $G(q)$ such that $\phi(q) = y$, and an automorphism $\xi$ on $G - G(q)$ such
Figure 4: Graphs augmented from $G$ by adding an edge $x_iy_i$, $i = 1, 2$: (a) $H_1 = G + x_1y_1$ with $z \neq v_0$, (b) $H_2 = G + x_2y_2$ with $z \neq v_0$, (c) $H_1 = G + x_1y_1$ with $z = v_0$, (d) $H_2 = G + x_2y_2$ with $z = v_0$. 
that $\xi(v_i) = v_{i+k \mod n}$, $v_i \in V(C)$ and $k \geq 1$, as in Theorem 13(iii), then we say that the pair $(G, q)$ admits a rotational symmetry $(\xi, \phi)$ for $k \geq 1$.

Let $G$ be a monocyclic graph and $C = (v_0, v_1, \ldots, v_{n-1}, v_0)$ denote the cycle in $G$ such that the pendent tree $G(v_0)$ has the maximum number of vertices over all pendent trees in $G$ and is represented as a left-heavy tree. Let $\text{copy} : V(G(v_0)) \to \{0, 1\}$ be a function such that for $v \in V(G(v_0))$ it holds that $\text{copy}(v) = 1$ (resp., $\text{copy}(v) = 0$) if $v$ has a left sibling $u$ and for the parent $q = p(v) = p(u)$ it holds that $G(q, v) \approx G(q, u)$ (resp., $v$ does not have a sibling on its left, or for the sibling $u$ on its left it holds $G(q, v) \not\approx G(q, u)$) \cite{24}, and let $Y = \{y \in V(G(v_0)) \setminus \{v_0\} \mid \text{copy}(v) = 0 \text{ for } v \in V(P(v_0, y))\}$. We define the potential edge set $S(G)$ of non-adjacent vertex pairs in $G$ as follows:

**Case (i):** The pendent tree $G(v_0)$ is not exceeding, or $G$ has more than one pendent tree with at least $|V(G(v_0))|$ vertices. Then, we define $S(G) \triangleq \emptyset$.

**Case (ii):** The pendent tree $G(v_0)$ is exceeding and there is no other pendent tree of $G$ with at least $|V(G(v_0))|$ vertices.

**Case (ii)(a):** $G$ admits an axial symmetry. Then we define

$$S(G) \triangleq \{\{x, y\} \mid x \in \{v_i \mid i \in [n/2]\}, y \in Y\}.$$ 

**Case (ii)(b):** $G$ does not admit an axial symmetry. Then we define

$$S(G) \triangleq \{\{x, y\} \mid y \in Y, \text{ for the child } q \text{ of } v_0 \text{ such that } y \in V(G(q)) \text{ \ "(G, q) admits a rotational symmetry (}\xi, \phi\text{) with } k \geq 1\text{\" or} \text{ \ "(G, q) does not admit a rotational symmetry and } \xi : V(C) \to V(C) \text{ is an identity mapping,"}$$

$$x \in \{v_i \mid i \in [1, [n/2]]\}$$

$$\cup \{v_{n-i \mod n} \mid i \in [1, [n/2]], \xi(v_i) \neq v_{n-i \mod n}\}.$$ 

Then, we have the following lemma.

**Lemma 16.** For a monocyclic graph $G$, the potential edge set $S(G)$ is proper.

**Proof.** Let $C = (v_0, v_1, \ldots, v_{n-1}, v_0)$ denote the unique cycle in $G$, such that the pendent tree $G(v_0)$ has the maximum number of vertices among all pendent trees in $G$, $G(v_0)$ is represented as a left-heavy tree, and let $\text{copy} : V(G(v_0)) \to \{0, 1\}$ be a function such that for $v \in V(G(v_0))$ it holds that $\text{copy}(v) = 1$ (resp., $\text{copy}(v) = 0$) if $v$ has a left sibling $u$ and $G(p(v), v) \approx G(p(u), u)$ (resp., $v$ does not have a sibling on its left, or for the sibling $u$ on its left it holds $G(p(v), v) \not\approx G(p(u), u)$) \cite{24}.

**Case (i).** If $G(v_0)$ is not exceeding, or $G$ has more than one pendent tree with maximum number of vertices, then $S(G) = \emptyset$ is proper for $G$ since by Lemmas 9 and 10 $G$ has no children.
Case (ii). By Theorem 14(i), for two non-adjacent vertex pairs \( \{x_i, y_i\} \), \( i = 1, 2 \), in \( G \) with \( x_1, x_2 \in V(C) \) and \( y_1, y_2 \in V(G(v_0)) \setminus \{v_0\} \) intra-duplication occurs if \( G(v_0) \) admits an automorphism \( \xi \) such that \( \xi(y_1) = \xi(y_2) \). By choosing vertices \( y \in V(G(v_0)) \setminus \{v_0\} \) such that \( \text{copy}(v) = 0 \) holds for each \( v \in V(P(v_0, y)) \), we know that no two vertices \( y_1 \) and \( y_2 \) will be chosen such that \( G(v_0) \) admits an automorphism \( \xi \) with \( \xi(y_1) = y_2 \). Next we consider the case when for vertices \( x_1, x_2 \in V(C) \) and \( y \in V(G(v_0)) \setminus \{v_0\} \), and integers \( p_i \in [1, \min\{\text{res}(x_i), \text{res}(y)\}] \), \( i = 1, 2 \), it holds that \( G + p_1 \cdot x_1 y \) is isomorphic to \( G + p_2 \cdot x_2 y \).

Case (ii)(a). By Theorem 14(ii), \( G \) admits an automorphism \( \xi \) such that for \( i \in [1, [n/2]] \) it holds \( \xi(v_i) = v_{n-i \mod n} \), and therefore it suffices to consider vertices \( v_i, i \in [1, [n/2]] \), for the choice of \( x \), thereby for each \( x' \in \{v_i \mid i \in [\lceil n/2 \rceil, 1, n-1]\} \) there exists an \( x \) such that \( \xi(x) = x' \) and therefore \( \text{res}(x) = \text{res}(x') \), and for \( p \in [1, \min\{\text{res}(x), \text{res}(y)\}] \) \( G + p \cdot x \) and \( G + p \cdot x' \) are isomorphic. On the other hand, by Lemma 15 for \( x_1, x_2 \in \{v_i \mid i \in [1, [n/2]]\} \) and \( p_i \in [1, \min\{\text{res}(x_i), \text{res}(y)\}] \), \( i = 1, 2 \) \( G + p_1 \cdot x_1 y \) and \( G + p_2 \cdot x_2 y \) are not isomorphic, satisfying the conditions for a proper set.

Case (ii)(b). In case \( G \) does not admit an axial symmetry, for each choice of \( y \in V(G(v_0)) \setminus \{v_0\} \) such that \( \text{copy}(v) = 0 \) holds for all \( v \in V(P(v_0, y)) \), for the child \( q \) of \( v_0 \) such that \( G(q) \) contains \( y \), we check whether the pair \( (G, q) \) admits a rotational symmetry \( (\xi, \phi) \) for \( k \geq 1 \), as in Theorem 14(iii). In case there does not exist an automorphism \( \xi \) on \( G - G(q) \) such that \( \xi(v_i) = v_{i+k \mod n} \), \( v_i \in V(C) \) and \( k \geq 1 \), and an automorphism \( \phi \) on \( G(q) \) with \( \phi(y) = y \), we take trivial automorphism \( \xi(x) = x, x \in V(C) \). Again, by the automorphism \( \xi \), for two vertices \( x_1, x_2 \in V(C) \) either \( x_1 = \xi(x_2) \) and then \( \text{res}(x_1) = \text{res}(x_2) \), \( \{x_1, y\} \in S(G) \) and \( \{x_2, y\} \notin S(G) \) but for any \( p \in [1, \min\{\text{res}(x_1), \text{res}(y)\}] \) it holds that \( G + p \cdot x_1 y \approx G + p \cdot x_2 y \), or \( x_1 \neq \xi(x_2) \), in which case \( \{x_1, y\} \notin S \) and \( \{x_2, y\} \in S \), but for any \( p_i \in [1, \min\{\text{res}(x_i), \text{res}(y)\}] \), \( i = 1, 2 \) it holds that \( G + p_1 \cdot x_1 y \neq G + p_2 \cdot x_2 y \), as required.

We give a description of an algorithm to compute the potential edge set of a given monocyclic graph \( G \) as Procedure 5 GENERATEPOTENTIALEDGESET.

**Procedure 5** GENERATEPOTENTIALEDGESET\((G)\)

**Input:** A monocyclic graph \( G \) with a cycle \( C = (v_0, v_1, \ldots, v_{n-1}, v_0) \), such that \( G(v_0) \) has the maximum number of vertices over all pendent trees in \( G \), and a function \( \text{copy} : V(G(v_0)) \rightarrow \{0, 1\} \) such that \( \text{copy}(v) = 1 \) if \( v \) has a left sibling \( u \) and \( G(p(v), v) \approx G(p(u), u) \) and \( \text{copy}(v) = 0 \) otherwise.

**Output:** The potential edge set \( S(G) \) of \( G \).

1. \( S := \emptyset \);
2. if \( |V(G(v_0))| \geq |V(G)|/3 \) and for \( i \in [1, n-1], |V(G(v_0))| > |V(G(v_i))| \) then
   3. for each child \( c \) of \( v_0 \) such that \( \text{copy}(c) = 0 \) do
4:  for each $y \in V(G(c))$ such that $\text{copy}(v) = 0$ for $v \in V(P(v_0, y))$ do
5:    $S := S \cup \{(v_i, y) \mid i \in [1, \lfloor n/2 \rfloor]\}$
6:  if $G$ does not admit an axial symmetry then
7:    Let $q \in V(G(v_0))$ be the child of $v_0$ such that $y \in V(G(q))$;
8:    if $(G, q)$ admits a rotational symmetry $(\xi, \phi)$ for $k \geq 1$ then
9:      $S := S \cup \{(v_{n-i \mod n}, y) \mid i \in [1, \lfloor n/2 \rfloor], \xi(v_i) \neq v_{n-i \mod n}\}$
10:     else /* $(G, q)$ does not admit a rotational symmetry for $k \geq 1 */
11:       $S := S \cup \{(v_i, y) \mid i \in [\lfloor n/2 \rfloor + 1, n - 1]\}$
12:   end if
13:  end if
14: end for
15: end for
16: end if
17: output $S$ as $S(G)$.

9 Experimental results

To test the effectiveness of our algorithm for enumerating mono-block 2-augmented trees, we have implemented it and performed computational comparison with MOLGEN [12], a generator for chemical graphs.

In particular, we did experiments for two different types of instances, named EULF-L-A and EULF-L-P, by considering a set $P$ of colored sequences with length at most a given integer $N$, given lower and upper bounds, $g_a : P \rightarrow \mathbb{Z}_+$ and $g_b : P \rightarrow \mathbb{Z}_+$, respectively, on the path frequencies of the paths in $P$, and integers $L$ and $d$. For a given set $P$ of colored sequences and a graph $G$, let $f_P(G) : P \rightarrow \mathbb{Z}_+$ denote the number $\text{frq}(t,G)$ of rooted paths $P \subseteq G$ such that $\gamma(P) = t \in P$. Assuming that $g_a \leq g_b$, and in particular, that $g_a[t] = g_b[t]$ is satisfied for each colored sequence $t \in P \cap \Sigma^{0,d}$, each of the instance types EULF-L-A and EULF-L-P asks to enumerate chemical graphs $G$ such that $g_a \leq \text{frq}_P(G) \leq g_b$, and for any $P \subseteq G$ such that $\gamma(P) \notin \mathcal{P}$, it holds that $|P| > L$ and $|P| \leq L$, for instance types EULF-L-A and EULF-L-P, respectively.

We have chosen six compounds from the PubChem database which when represented as hydrogen-suppressed chemical graphs have mono-block 2-augmented tree structure, and constructed feature vectors based on the path frequencies of the paths in the chemical graphs. All compounds have 13 non-hydrogen atoms, maximum path length 11, and maximum bond multiplicity $d \in \{2, 3\}$. All compounds include the three chemical elements $C$ (carbon), $O$ (oxygen), and $N$ (nitrogen). The information on the chosen compounds, identified by their Compound ID (CID) number in the PubChem database is given in Table I.

We construct instances of types EULF-L-A and EULF-L-P for different values of parameter $L$ in the following way. We take a set $\Sigma$ of colors to be $\Sigma = \{\text{C}, \text{O}, \text{N}\}$, such that $\text{val(}\text{C}) = 4$, $\text{val(}\text{O}) = 2$, and $\text{val(}\text{N}) = 3$. For each hydrogen suppressed chemical graph $G$ that corresponds to a chemical compound in Table I, we take $d \in \{2, 3\}$ to be the maximum bond multiplicity in the chemical graph, and for some choice of values for
\( N \geq 0 \) we construct a set of colored sequences \( \mathcal{P} \subseteq \Sigma^{\leq N,d} \) that consists of all colored sequences \( t \) with length \( |t| \leq N \) such that \( G \) contains a rooted path \( P \) with \( \gamma(P) = t \). Finally, for an integer \( s \in [0,2] \) we set lower and upper bounds, \( g_a \) and \( g_b \) on feature vectors as follows: for \( t \in \mathcal{P} \), if \( |t| \in \Sigma^{0,d} \) then \( g_a[t] = g_b[t] = \text{frq}(t,G) \), otherwise \( g_a[t] = \max\{0, \text{frq}(t,G) - s\} \) and \( g_b[t] = \text{frq}(t,G) + s \). The parameter \( s \) effectively serves to “relax” the path frequency specification.

On the other hand, we used MOLGEN \cite{12} without aromaticity detection by specifying the hydrogen suppressed formula, number of cycles to be two in enumerated structures - thereby enumerating chemical graphs with 2-augmented tree structures with a maximum allowed bond multiplicity. Note that there is no option in MOLGEN to specify whether the enumerated structures have a mono-block structure or not.

We implemented our algorithm in the C++ programming language, and compiled and executed on the Linux 14.04.6 operating system by the gcc compiler version 4.8.4 and optimization level O3. All experiments were done on a PC with Intel Xeon CPU E5-1660 v3 running at 3.00 GHz, with 32 GB memory.

### 9.1 Experimental Results for EULF-L-A

To test the behavior of our algorithm for instance types EULF-L-A, especially the effect the choice of problem parameters have on the running time and the number of enumerated chemical graphs, we choose values for parameter \( N \in [2,6] \), and we took values for the parameter \( L \in \{2, \lceil N/2 \rceil, N\} \).

The results from our experiments for EULF-L-A are summarized in Figs. 5 to 10. We observe that our algorithm has a clear advantage when we are given a path frequency specification for instances of type EULF-L-A over using MOLGEN to generate molecules with a specified formula. We also observe some trends over the values of the parameters \( N \), \( L \), and \( s \). Namely, the number of generated molecules, as well as the time it takes our algorithm, reduces as the length \( N \) of the longest path given in the set of paths, as well as the parameter \( L \) increase, but grows with an increasing value \( s \) that we choose to relax the path frequency specification.

| Molecular formula | \( d \) | CID       |
|-------------------|--------|-----------|
| \( C_9N_1O_3 \)   | 2      | 301729    |
|                   | 3      | 57320502  |
| \( C_9N_2O_2 \)   | 2      | 6163405   |
|                   | 3      | 131335510 |
| \( C_9N_3O_1 \)   | 2      | 9942278   |
|                   | 3      | 10103630  |
Figure 5: Plots showing the computation time and number of chemical graphs enumerated by our algorithm for instance type EULF-L-A, as compared to MOLGEN. The sample structure from PubChem is with CID 301729, molecular formula $C_9N_1O_3$, and maximum bond multiplicity $d = 2$. (a)-(c) Running time; (d)-(f) Number of enumerated chemical graphs.
Figure 6: Plots showing the computation time and number of chemical graphs enumerated by our algorithm for instance type EULF-L-A, as compared to MOLGEN. The sample structure from PubChem is with CID 57320502, molecular formula C₉N₁O₃, and maximum bond multiplicity \( d = 3 \). (a)-(c) Running time; (d)-(f) Number of enumerated chemical graphs.
Figure 7: Plots showing the computation time and number of chemical graphs enumerated by our algorithm for instance type EULF-L-A, as compared to MOLGEN. The sample structure from PubChem is with CID 6163405, molecular formula $C_9N_2O_2$, and maximum bond multiplicity $d = 2$. (a)-(c) Running time; (d)-(f) Number of enumerated chemical graphs.
Figure 8: Plots showing the computation time and number of chemical graphs enumerated by our algorithm for instance type EULF-L-A, as compared to MOLGEN. The sample structure from PubChem is with CID 131335510, molecular formula $C_9N_2O_2$, and maximum bond multiplicity $d = 3$. (a)-(c) Running time; (d)-(f) Number of enumerated chemical graphs.
Figure 9: Plots showing the computation time and number of chemical graphs enumerated by our algorithm for instance type EULF-L-A, as compared to MOLGEN. The sample structure from PubChem is with CID 9942278, molecular formula $C_9N_3O_1$, and maximum bond multiplicity $d = 2$. (a)-(c) Running time; (d)-(f) Number of enumerated chemical graphs.
Figure 10: Plots showing the computation time and number of chemical graphs enumerated by our algorithm for instance type EULF-L-A, as compared to MOLGEN. The sample structure from PubChem is with CID 10103630, molecular formula $C_9N_3O_1$, and maximum bond multiplicity $d = 3$. (a)-(c) Running time; (d)-(f) Number of enumerated chemical graphs.
In addition, to check the limits as to the maximum number of vertices in graphs that can be enumerated in a reasonable time, we conducted experiments over a range \( n \in [9, 40] \) for the number of vertices in a target chemical graph. For a fixed number \( n \) of vertices, we tested two types of instances, one with molecular formula \( C_n \), and the other with molecular formula \( C_{n-4}N_2O_2 \), and set an execution time limit of 3,600 seconds. The results are summarized in Fig. 11. From Fig. 11(a) and (b), we see that the time limit is quickly reached even when the number \( n \) of number of vertices is less than 15, but that the program still enumerates structures within the time limit up to \( n = 30 \), after which there are cases when not even a single graph is enumerated during the time limit.

Figure 11: Plots showing the computation time and number of chemical graphs enumerated by our algorithm for instance type EULF-L-A, over ranges for \( N \in [0, 2] \), \( L = N \), \( d = 3 \) and \( s = 0 \). (a), (b) Running time, T.O. stands for "Time Out"; (c), (d) Number of enumerated chemical graphs.
9.2 Experimental Results on EULF-L-P

We conduct similar computational experiments to test the performance of our algorithm for Problem EULF-L-P as in Section 9.1. We took values for $N \in [8, 10]$, and $L \in \{2, 3\}$.

The results from our experiments for instance type EULF-L-P are summarized in Figs. 12 to 17. Our results for instance type EULF-L-P indicate that there exist very few chemical graphs that satisfy the path frequency specification for our choice of a set $\mathcal{P}$ of colored paths obtained from the six compounds from the PubChem database, and parameter $L$. In fact, the only two instances where our algorithm enumerates any chemical graphs are for two of our chosen compounds; with CID 301729, molecular formula $C_9N_1O_3$ and bond multiplicity at most 2 (Fig. 12 (c) and (d)), and the compound with CID 10103630, molecular formula $C_9N_3O_1$ and bond multiplicity at most 3 (Fig. 17 (c) and (d)). This could be due to the very nature of mono-block 2-augmented structures, namely, due to the biconnectedness of a block, a single path frequency specification exhibits a strong influence on the structure of a chemical graph.

In addition, we observe that the running time of our algorithm, even when there are no enumerated chemical graphs, grows rapidly with the value of the parameter $L$. It is an interesting idea for future research to improve our algorithm in such a way that the non-existence of any chemical graphs that satisfy a given path frequency specification is determined much quicker.

10 Conclusion and Future Work

We formulated two problem settings of enumerating chemical graphs that satisfy given lower and upper bounds on path frequencies in a given set of paths, EULF-L-A, and EULF-L-P. The problem of enumerating chemical graphs has an important practical application in inverse QSAR/QSPR, and can be used as a part of a framework for inferring novel chemical structures [3, 9, 4] together with a method for solving the inverse problem on artificial neural networks based on linear programming due to Akutsu and Nagamochi [2].

We focused on enumerating chemical graphs with a mono-block 2-augmented tree structure. We designed a branch-and-bound algorithm for the problem by developing a new procedure to add edges between a pair of non-adjacent vertices of a monocyclic graph. Our procedure relies on a carefully chosen parent-child relationship between mono-block 2-augmented trees and monocyclic graphs to avoid inter-duplication, and a way of choosing a proper set of non-adjacent vertex pairs in a monocyclic graph, such that adding edges between each pair in the set will not cause intra-duplication, nor any possible mono-block 2-augmented trees to be omitted.

Experimental results reveal that our algorithm offers a big advantage in terms of running time and the number of generated structures for instance type EULF-L-A when we have a path frequency specification over using MOLGEN [12] to generate chemical graphs with a particular molecular formula. Namely, while MOLGEN may produce on the order of billions of chemical graphs with 2-augmented tree structure with a particular
Figure 12: Plots showing the computation time and number of chemical graphs enumerated by our algorithm for instance type EULF-L-P, as compared to MOLGEN. The sample structure from PubChem is with CID 301729, molecular formula \( C_9N_1O_3 \), and maximum bond multiplicity \( d = 2 \). (a), (b) Running time; (c), (d) Number of enumerated chemical graphs.

chemical formula and maximum bond multiplicity, for a given path specification our algorithm produces much fewer structures, and we also have the advantage to generate only mono-block structures.

However, for instance type EULF-L-P, the experimental results reveal that our algorithm takes much time to finish even when there are no chemical graphs that satisfy a given path frequency specification. It would be very interesting to equip our algorithm with a procedure that detects this situation much earlier in the computation process, or even design an algorithm based on a different idea - namely one that starts building a chemical graph from one of the paths with a non-zero lower bound in a given set.
Figure 13: Plots showing the computation time and number of chemical graphs enumerated by our algorithm for instance type EULF-L-P, as compared to MOLGEN. The sample structure from PubChem is with CID 57320502, molecular formula C₉N₁O₃, and maximum bond multiplicity $d = 3$. (a), (b) Running time; (c), (d) Number of enumerated chemical graphs (our algorithm detects that there are no chemical graphs that satisfy the given path frequency specification).

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Figure 14: Plots showing the computation time and number of chemical graphs enumerated by our algorithm for instance type EULF-L-P, as compared to MOLGEN. The sample structure from PubChem is with CID 6163405, molecular formula C₉N₂O₂, and maximum bond multiplicity \( d = 2 \). (a), (b) Running time; (c), (d) Number of enumerated chemical graphs (our algorithm detects that there are no chemical graphs that satisfy the given path frequency specification).

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Figure 15: Plots showing the computation time and number of chemical graphs enumerated by our algorithm for instance type EULF-L-P, as compared to MOLGEN. The sample structure from PubChem is with CID 131335510, molecular formula $C_9N_2O_2$, and maximum bond multiplicity $d = 3$. (a), (b) Running time; (c), (d) Number of enumerated chemical graphs (our algorithm detects that there are no chemical graphs that satisfy the given path frequency specification).

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Figure 16: Plots showing the computation time and number of chemical graphs enumerated by our algorithm for instance type EULF-L-P, as compared to MOLGEN. The sample structure from PubChem is with CID 9942278, molecular formula \( C_9N_3O_1 \), and maximum bond multiplicity \( d = 2 \). (a), (b) Running time; (c), (d) Number of enumerated chemical graphs (our algorithm detects that there are no chemical graphs that satisfy the given path frequency specification).

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Figure 17: Plots showing the computation time and number of chemical graphs enumerated by our algorithm for instance type EULF-L-P, as compared to MOLGEN. The sample structure from PubChem is with CID 10103630, molecular formula $C_9N_3O_1$, and maximum bond multiplicity $d = 3$. (a), (b) Running time; (c), (d) Number of enumerated chemical graphs.

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