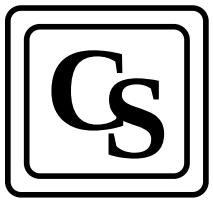

COMPUTER SCIENCE TECHNICAL REPORT

A Newton-Cotes Quadrature Approach for Solving the Aerosol Coagulation Equation

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Abstract

This paper presents a direct approach to solving the aerosol coagulation equation. Newton-Cotes formulas are used to discretize the integral terms, and the semi-discrete system is built using collocation. A semi-implicit Gauss-Seidel time integration method is employed. The approach generalizes the semi-implicit method of Jacobson.

Keywords: Aerosol dynamics, coagulation, Newton-Cotes integration.

1 Introduction

As our understanding expands, new processes are incorporated into air quality computer models. One example are the particulate matter (aerosol) processes, the importance of which is now widely recognized. Aerosols are a priority focus area in environmental science due to the leading role they play as a cause of adverse human health, and their ability to scatter and absorb incoming solar radiation and thus modify warming due to greenhouse gases and reduce visibility. To accurately study the effects of aerosols it is necessary to resolve aerosol number and mass distributions as a function of chemical composition and size. In this paper we develop a numerical approach to solving the aerosol coagulation equation. The method is of stationary sectional type: the number densities are computed at a predefined set of particle volumes (bin mean volumes). The discretization is based on approximating the integral terms by Newton-Cotes sums, and imposing that the resulting equations hold exactly at the node points (collocation). The resulting bilinear system of coupled ordinary differential equations can be advanced in time by the time-stepping algorithm of choice; here we use the semi-implicit formula solved with Gauss-Seidel iterations.

The paper is organized as follows: Section 2 gives an overview of aerosol dynamic equations; Section 3 presents the discretization technique while Section 4 shows numerical results obtained on a test problem with analytical solution. Conclusions and future work are highlighted in Section 5. Newton-Cotes integration is reviewed in the Appendix.

2 Continuous particle dynamics equations

In this paper the continuous particle size distributions are functions of particle volume (v) and time (t). For simplicity we consider single component particles; the technique can be generalized to multiple components. The size distribution function (number density) of a family of particles is be denoted by $n(v,t)$; the number of particles per unit volume of air with the volume between v and $v+dv$ is $n(v,t)dv$. Similar formulations can be given in terms of mass and surface densities [10], etc.

The aerosol population undergoes a series of physical transformations which change the number density according to [3]

$$\begin{aligned} \frac{\partial}{\partial t} n(v,t) &= -\frac{\partial}{\partial v}\left[I_v(v)\, n(v,t)\right] + \tfrac{1}{2}\int_0^v \beta_{v-w,w}\, n(v-w,t) n(w,t) dw \\ &\quad -n(v,t)\int_0^\infty \beta_{v,w}\, n(w,t) dw + S(v,t)\ , \\ n(v,0) &= n_0(v)\ . \end{aligned} \tag{1}$$

The different terms in equation (1) describe, in order, the modification in the number of particles due to growth, creation of particles of volume v by coagulation, loss of volume v particles due to coagulation, increase in particle number due to nucleation, emissions and depositions (sources and sinks). The equation is subject to a specified initial condition n_0.

2.1 Previous work

Representations of the particle size distribution. Three major approaches are used to represent the size distribution of aerosols: continuous, discrete and parametrized. In this paper we focus on continuous models; for computational purposes one needs to use finite-dimensional approximations. In the *sectional approach* the size domain $v \in [0,\infty]$ is divided into size bins $v \in [V_i^{\text{low}}, V_i^{\text{high}})$. In each size bin j there are n_j particles per unit volume, all of them having the same mean volume V_i. In the *full-stationary* structure the number of particles in each bin $n_j(t)$ is allowed to change in time, but the particle volumes in each bin (V_j) are not. Formally, the density function is $n(v,t) = \sum_{j=1}^{s} n_j(t)\,\delta(v-V_j)$, where $\delta(v)$ is the Dirac delta function*.

Coagulation Equation. The integro-differential coagulation equation is difficult to solve accurately, due to the quadratic terms under the integral. The standard discrete version of the coagulation equation uses a monomer size distribution (the volume of the particles in bin each i is a multiple of the smallest volume, $V_i = i\,V_1$, $i = 1,2,\cdots$). Jacobson [8, Section 16] proposed the semi-implicit scheme to solve the discrete coagulation equation . The differential equation is discretized in time using backward Euler formula, and the quadratic terms $n_{j-\ell}(t) n_\ell(t)$ are replaced by the "linearized" version $n_{j-\ell}(t) n_\ell(t-h)$. The scheme can be adapted to general size distributions, and admits a volume-conserving formulation.

Lushnikov [9] uses generating functions to solve analytically the coagulation equation for particles consisting of monomers of two kinds, under the assumptions of a constant coagulation rate β and particular initial distributions.

3 A Direct Discretization of the Coagulation Equation

The theoretical coagulation equation for single-component particles is [8, Section 16]

$$\frac{\partial}{\partial t} n(v,t) = \tfrac{1}{2}\int_0^v \beta_{v-w,w}\, n(v-w,t) n(w,t) dw - n(v,t)\int_0^\infty \beta_{v,w}\, n(w,t) dw\ . \tag{2}$$

In practice the coagulation equation is restricted to particles having volumes in a finite range, $0 < V_{\min} \le v \le V_{\max} < \infty$:

$$\frac{\partial}{\partial t} n(v,t) = \tfrac{1}{2}\int_{V_{\min}}^{v-V_{\min}} \beta_{v-w,w}\, n(v-w,t)\, n(w,t)\, dw - n(v,t)\int_{V_{\min}}^{V_{\max}} \beta_{v,w}\, n(w,t)\, dw\ . \tag{3}$$

*Recall that $\delta(x) = 0$ for $x \neq 0$, $\delta(0) = \infty$, and $\int_{V_j-\epsilon}^{V_j+\epsilon} f(x)\delta(x-V_j)\,dx = f(V_j)$.

Particle volumes are distributed over several orders of magnitude; to better capture the distribution logarithmic volume coordinates are frequently employed

$$\begin{array}{rcl} \frac{\partial}{\partial t} n(\log v, t) & = & \frac{1}{2} \int_{\log V_{\min}}^{\log(v-V_{\min})} \beta_{v-w,w}\, n(\log(v-w), t)\, n(\log w, t)\, w\, d(\log w) \\ & & -n(\log v, t) \int_{\log V_{\min}}^{\log V_{\max}} \beta_{v,w}\, n(\log w, t)\, w\, d(\log w) \ . \end{array} \tag{4}$$

Consider s size bins of volumes $V_{\min} = V_1 < V_2 < \cdots < V_s = V_{\max}$. The direct approach for solving (3) is based on discretizing the integrals by a quadrature formula with nodes $V_1, \cdots, V_s$

$$\int_a^b f(v) dv \approx \sum_{i=1}^{s} \xi_i^{[a,b]} f(V_i) \ . \tag{5}$$

The bin volumes (the nodes V_i) are given, but we need to specify the appropriate weights ξ_i.

Replacing the integrals in (3) by (5) gives

$$\frac{\partial}{\partial t} n(v,t) = \frac{1}{2} \sum_j \xi_j^{[V_1, v-V_1]} \beta_{v-V_j, V_j}\, n(v-V_j, t) n(V_j, t) - n(v,t) \sum_j \zeta_j^{[V_1, V_s]} \beta_{v,V_j}\, n(V_j, t) \ , \tag{6}$$

where we allow different weights (ξ, ζ) on different intervals. For example, if β is symmetric ($\beta_{v,w} = \beta_{w,v}$ for all v, w) then the integrand $\beta_{v-w,w}\, n(v-w,t) n(w,t)$ is periodic on $[V_{\min}, v - V_{\min}]$. Consequently, the first term in (3) is the integral of a periodic function on a period, and choosing ξ_j the trapezoidal rule weights should provide a very accurate discretization.

We now impose that the equation (6) holds exactly at the node points (collocation). This gives:

$$\begin{array}{rcl} \frac{\partial}{\partial t} n(V_i, t) & = & \frac{1}{2} \sum_{j=1}^{i} \xi_j^{[V_1, V_i - V_1]} \beta_{V_i - V_j, V_j}\, n(V_i - V_j, t) n(V_j, t) \\ & & -n(V_i, t) \sum_{j=1}^{s} \zeta_j^{[V_1, V_s} \beta_{V_i, V_j}\, n(V_j, t) \ , \quad 1 \le i \le s \end{array} \tag{7}$$

For a discrete coagulation equation we must express (7) only in terms of number density at the node points. In general

$$V_k \le V_i - V_j < V_{k+1} \ , \quad k = k(i,j)$$

with k depending on i and j, and therefore $n(V_i - V_j, t)$ must be approximated by polynomial interpolation (of the same order as the underlying Newton-Cotes formula).

For example, if the evaluation of the first integral uses trapezoidal rule, $n(V_i - V_j, t)$ can be computed by linear interpolation without losing the approximation order,

$$\alpha_{k(i,j)} = \frac{V_{k(i,j)+1} - V_i + V_j}{V_{k(i,j)+1} - V_{k(i,j)}} \ , \quad n(V_i - V_j, t) = \alpha_{k(i,j)}\, n_{k(i,j)}(t) + (1 - \alpha_{k(i,j)})\, n_{k(i,j)+1}(t) \ .$$

With this the semi-discrete coagulation equation becomes a system of s coupled, bilinear ordinary differential equations:

$$\begin{array}{rcl} \frac{\partial n_i(t)}{\partial t} & = & \frac{1}{2} \sum_{j=1}^{i} \xi_{ij} \beta_{V_i - V_j, V_j} \left(\alpha_{k(i,j)}\, n_{k(i,j)}(t) + (1 - \alpha_{k(i,j)})\, n_{k(i,j)+1}(t) \right) n_j(t) \\ & & -n_i(t) \sum_{j=1}^{s} \zeta_j \beta_{V_i, V_j}\, n_j(t) \ , \quad 1 \le i \le s \end{array} \tag{8}$$

Here ξ_{ij} denotes the j-th trapezoidal weight (node V_j) for the interval $[V_1, V_i - V_1]$, and ζ_j the j-th Newton-Cotes weight for the interval $[V_1, V_s]$.

3.1 The semi-implicit method

The semi-implicit method of Jacobson [8, Section 16.2] can be viewed as a particular Newton-Cotes approach, using the rectangular integration formula. Let $N(V_i,t)$ denote the total number of particles in size bin i, i.e. particles having volumes between V_{i-1} and V_i. $N(V_i,t)$ are related to the density $n(v,t)$ by

$$N(V_1,t) = V_1\, n(V_1,t)\ , \qquad N(V_i,t) = (V_i - V_{i-1})\, n(V_i,t)\ , \quad 2 \le i \le s\ .$$

The solution at t^{k+1} is computed by

$$\begin{aligned} &\text{Do} \quad i = 1:s \\ &\quad N(V_i,t^{k+1}) = \frac{N(V_i,t^k) + \frac{h}{2}\sum_{j=1}^{i} \beta_{V_i - V_j, V_j}\ N(V_i - V_j, t^{k+1}) N(V_j,t^k)}{1 + h\sum_{j=1}^{s} \beta_{V_i,V_j}\ N(V_j,t^k)} \end{aligned} \tag{9}$$

$N(V_i - V_j, t^{k+1})$ is obtained from $\{N(V_k,t)\}_k$ by linear interpolation.

3.2 Time integration

The semi-discrete system (8) could be integrated in time with an explicit or with an implicit time-stepping formula. For implementing an implicit formula the Jacobian of the derivative function is needed; the Jacobian is easy to derive analytically, due to the bilinear function form; we do not give more details here.

Of interest is the direct generalization of the semi-implicit method of Jacobson; the time-stepping idea can be extended to a full Gauss-Seidel approach, which (in the context of Newton-Cotes formulation) reads

$$\begin{aligned} &\text{Do} \quad i = 1:s \\ &\quad n(V_i,t^{k+1}) = \frac{n(V_i,t^k) + \frac{h}{2}\sum_{j=1}^{i-1} \xi_j^{[V_1,V_i]} \beta_{V_i - V_j, V_j}\ n(V_i - V_j, t^{k+1}) n(V_j,t^{k+1}) + \frac{h}{2}\xi_i^{[V_1,V_i]} \beta_{0,V_i}\ n(0,t^{k+1}) n(V_i,t^k)}{1 + h\sum_{j=1}^{i-1} \zeta_j^{[V_1,V_s]} \beta_{V_i,V_j}\ n(V_j,t^{k+1}) + h\sum_{j=i}^{s} \zeta_j^{[V_1,V_s]} \beta_{V_i,V_j}\ n(V_j,t^k)} \end{aligned} \tag{10}$$

4 Numerical Results

Test problem. For the numerical experiments we consider the test problem from [4]. Let N_{tot} be the total initial number of particles and V_{mean} the mean initial volume. The initial number distribution is exponential, and the coagulation rate is constant:

$$n_0(v) = (N_{\text{tot}}/V_{\text{mean}})\, e^{-v/V_{\text{mean}}}\ , \quad \beta(v,w) = \beta_0\ .$$

This test problem admits the analytical solution [4]:

$$n(v,t) = \frac{4N_{\text{tot}}}{V_{\text{mean}}(N_{\text{tot}}\beta_0 t + 2)^2} \exp\left(\frac{-2v}{V_{\text{mean}}(N_{\text{tot}}\beta_0 t + 2)}\right)\ .$$

We solve this coagulation equation for $V_{\text{mean}} = 0.05\ \mu m^3$, $N_{\text{tot}} = 10^4\ particles/cm^3$, and

$$\beta_0 = \left.\frac{8k_BT}{3\eta_a}\right|_{T=298K} = 6.017e-10\ \frac{cm^3}{sec\cdot particle}\ .$$

(η_a is the atmospheric dynamic viscosity). The integration time interval is 24 hours, and the integration time step $h = 0.2$ seconds is chosen small enough to ensure that time discretization errors are negligible when compared to volume discretization errors. For discretization of the integral terms we choose the fourth order repeated Newton-Cotes algorithm (Boole formula).

The initial and final number distributions are shown in Figure 1.

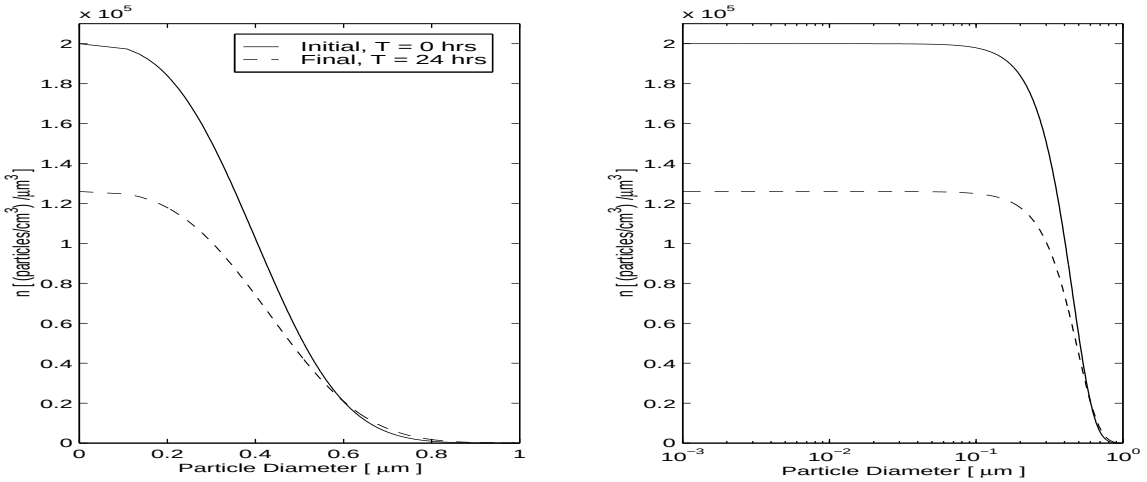

Figure 1: Exact initial and final distributions, in linear (left) and logarithmic (right) coordinates.

Experiment I. In the first experiment we consider particles with diameters between $D_{\min} = 0\ \mu m$ and $D_{\max} = 1\ \mu m$ (i.e. volumes in the range $[V_{\min} = 0\ \mu m^3, V_{\max} = 5.236E-1\ \mu m^3]$, and use equidistant size bin volumes,

$$\Delta v = \frac{V_{\max} - V_{\min}}{s-1}\ ; \qquad V_i = V_{\min} + (i-1)\,\Delta v\ , \quad 1 \le i \le s\ .$$

The computed solutions and the error distribution (versus particle diameter) at the end of the 24 hours interval are shown in Figure 2. The fourth order Newton-Cotes discretization (Boole formula) provides more accurate solutions than the semi-implicit method for the same number of discretization points. Note that using the trapezoidal discretization for the first integral term (with a linear interpolation for $n(v-w,t)$) gives better results than the Boole formula with order 4 polynomial interpolation (for reasons of periodicity, as explained before).

Experiment II. In the second experiment we consider particles with diameters in between $D_{\min} = 1E-3\ \mu m$ and $D_{\max} = 1\ \mu m$ (i.e. volumes in the range $[V_{\min} = 5.236E-10\ \mu m^3, V_{\max} = 5.236E-1\ \mu m^3]$. The distribution of the bin volumes is logarithmic-uniform,

$$\rho = \left(\frac{V_{\max}}{V_{\min}}\right)^{\frac{1}{s-1}}\ ; \qquad V_i = V_{\min}\ \rho^{i-1}\ , \quad 1 \le i \le s\ .$$

We actually solve the form (4) of the coagulation equation, with the interpolation also performed on logarithmic scale. Linear interpolation works better than higher order interpolation.

The computed solutions and the error distribution (versus particle diameter) at the end of the 24 hours interval are shown in Figure 3. Boole formula again provides more accuracy, but the difference is less dramatic. For a large number of size bins the semi-implicit method is slightly better toward the high-volume end of the interval. This is probably due to the fact that the log-uniform grid has insufficient resolution at high particle volumes (diameters).

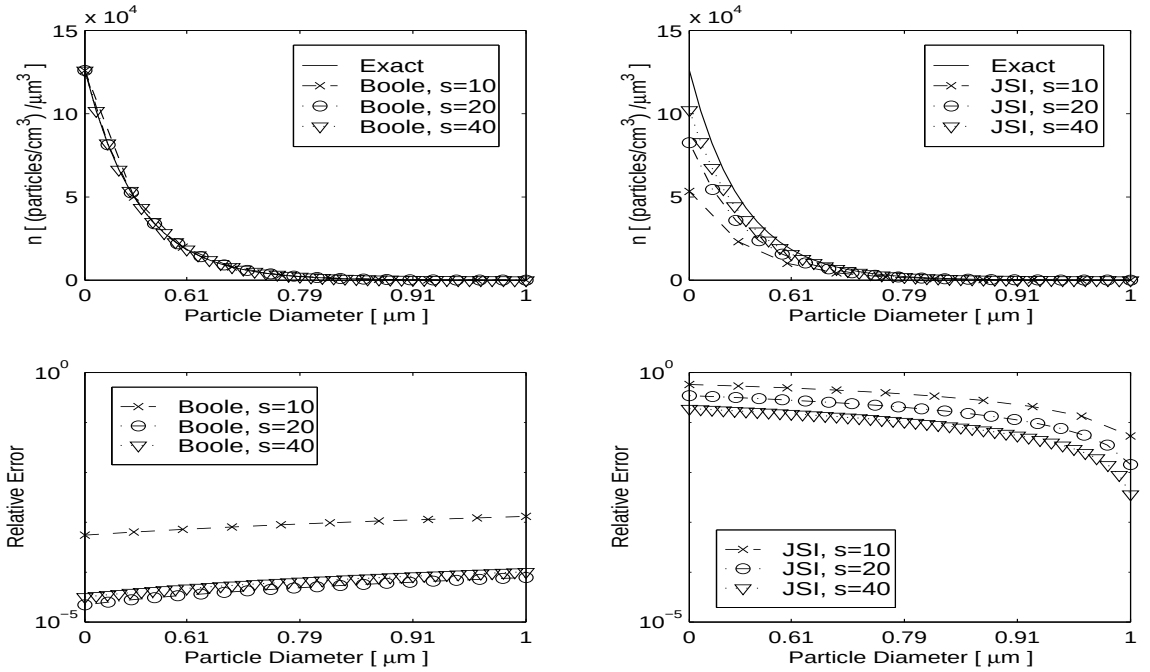

Figure 2: Distribution of solution errors in Experiment I after 24 hours. Compared are Jacobson's original method, and Boole discretization.

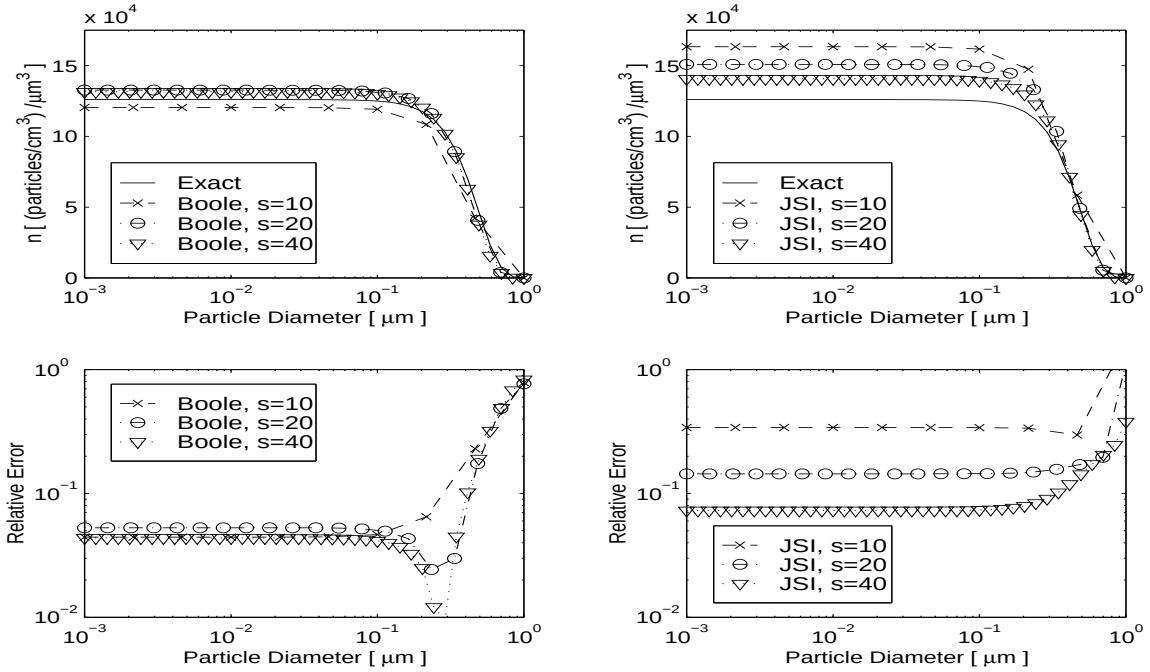

Figure 3: Distribution of solution errors in Experiment II after 24 hours. Compared are Jacobson's original method, and Boole discretization.

5 Conclusions and future work

In this paper we developed a direct method for the size discretion of aerosol coagulation equation. The number densities are computed at a predefined set of particle volumes (bin mean volumes). The coagulation rate integral terms are replaced by discrete approximations provided by Newton-Cotes formulas. The resulting equations are imposed to hold exactly at the node points (collocation). This semi-discretization in size results in a bilinear system of coupled ordinary differential equations. The system can be advanced in time by explicit, implicit or ad-hoc time-stepping techniques.

The discretization methods are inexpensive, since the weights can be calculated before the integration process and stored. For implicit time-stepping the Jacobian can be easily obtained; however it does not have any special structure (it is a full matrix) so solving the linear systems might prove costly for large number of bins. An extension of the semi-implicit time stepping method to a full Gauss-Seidel approach was used in the experiments for time integration.

Future work will focus on improving the accuracy of the quadrature discretizations for the logarithmic distribution of bin volumes.

Appendix: Newton-Cotes integration

A Newton-Cotes formula of order n for evaluating $\int_a^b f(v)dv$ is based on a repeated application of the following elementary rule. The order n polynomial $p_n(v)$ that interpolates the function at nodes v_1 through v_{n+1} is expressed as $p_n(v) = \sum_{i=1}^{n+1} f(v_i)\,\ell_i(v)$, where ℓ_i are the degree n Lagrange interpolation polynomials, $\ell_i(v_j) = \delta_{ij}$. Then

$$\int_{v_1}^{v_{n+1}} f(v)dv \approx \int_{v_1}^{v_{n+1}} p_n(v)dv = \sum_{i=1}^{n+1} f(v_i) \int_{v_1}^{v_{n+1}} \ell_i(v)dv = \sum_{i=1}^{n+1} w_i\, f(v_i)\ .$$

To cover the whole interval $[a, b]$ the elementary rule is repeatedly applied on subintervals $[v_1, v_{n+1}]$, $[v_{n+2}, v_{2n+2}]$ etc. If the number of nodes is not a multiple of $n+1$ the integration of the last interpolation polynomial is restricted to the remaining subinterval. For convenience, we present the weights for orders 1, 2 and 4.

Trapezoidal rule (n=1):

$$w_1 = w_2 = \frac{v_2 - v_1}{2}\ .$$

Simpson rule (n=2):

$$w_1 = \frac{(v_3 - v_1)(2v_1 - 3v_2 + v_3)}{6(v_1 - v_2)}\ , \quad w_2 = \frac{(v_3 - v_1)^3}{6(v_1 - v_2)(v_2 - v_3)}\ , \quad w_3 = \frac{(v_3 - v_1)(v_1 - 3v_2 + 2v_3)}{6(-v_2 + v_3)}\ .$$

Boole rule (n=4):

$$\begin{aligned}
w_1 &= [(v_5 - v_1)(12v_1^3 - 15v_1^2v_2 - 15v_1^2v_3 + 20v_1v_2v_3 - 15v_1^2v_4 + 20v_1v_2v_4 \\
&\quad +20v_1v_3v_4 - 30v_2v_3v_4 + 9v_1^2v_5 - 10v_1v_2v_5 - 10v_1v_3v_5 + 10v_2v_3v_5 \\
&\quad -10v_1v_4v_5 + 10v_2v_4v_5 + 10v_3v_4v_5 + 6v_1v_5^2 - 5v_2v_5^2 - 5v_3v_5^2 - 5v_4v_5^2 + 3v_5^3)] \\
&\quad /[60(v_1 - v_2)(v_1 - v_3)(v_1 - v_4)]\ , \\
w_2 &= \frac{(v_5 - v_1)^3(3v_1^2 - 5v_1v_3 - 5v_1v_4 + 10v_3v_4 + 4v_1v_5 - 5v_3v_5 - 5v_4v_5 + 3v_5^2)}{60(v_1 - v_2)(v_2 - v_3)(v_2 - v_4)(v_2 - v_5)} \\
w_3 &= \frac{(v_5 - v_1)^3(3v_1^2 - 5v_1v_2 - 5v_1v_4 + 10v_2v_4 + 4v_1v_5 - 5v_2v_5 - 5v_4v_5 + 3v_5^2)}{60(v_1 - v_3)(-v_2 + v_3)(v_3 - v_4)(v_3 - v_5)}\ ,
\end{aligned}$$

$$
\begin{aligned}
w_4 &= \frac{(v_5-v_1)^3(3v_1^2-5v_1v_2-5v_1v_3+10v_2v_3+4v_1v_5-5v_2v_5-5v_3v_5+3v_5^2)}{60(v_1-v_4)(-v_2+v_4)(-v_3+v_4)(v_4-v_5)}\ , \\
w_5 &= [(v_5-v_1)(3v_1^3-5v_1^2v_2-5v_1^2v_3+10v_1v_2v_3-5v_1^2v_4+10v_1v_2v_4+10v_1v_3v_4 \\
&\quad -30v_2v_3v_4+6v_1^2v_5-10v_1v_2v_5-10v_1v_3v_5+20v_2v_3v_5-10v_1v_4v_5+20v_2v_4v_5 \\
&\quad +20v_3v_4v_5+9v_1v_5^2-15v_2v_5^2-15v_3v_5^2-15v_4v_5^2+12v_5^3)] \\
&\quad /[60(-v_2+v_5)(-v_3+v_5)(-v_4+v_5)]\ .
\end{aligned}
$$

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