Characterizing and Modeling of Particle Size Distributions from Propellant Open Burning or Open Detonation (OB/OD)

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Abstract

The US Department of Defense (DoD) demilitarization activities conducted at the Utah Test and Training Range (UTTR) include destruction of ballistic missile motors, which contain solid propellants, by open burning or open detonation (OB/OD). Detonations and burns of these solid propellants generate alumina-based particulate matter (alumina particulate) as an emission. Airborne particulate matter (PM) generated by OB/OD, such as PM2.5 (any PM with a diameter less than or equal to 2.5 microns) and PM10, is regulated as a pollutant emission. The data historically used for permitting activities at the UTTR do not provide test-based particle size information (fraction of PM2.5 and PM10), and no modeling approach is known for the prediction of particle size distributions (PSDs) or PM emission rates.

A predictive particle model was developed with two primary components: the Cheetah combustion model, and a MATLAB stochastic particle model. The predictive particle model uses an incremental time step approach to address nucleation, coagulation, and surface growth of alumina particulate in the detonation plume.

Laboratory work has been performed to validate sampling methodologies for measurement of pollutant emissions and PSDs from solid propellant OB/OD at the Utah Test and Training Range. Preliminary PSD and PM emission rate data have been acquired for small-scale burns of a surrogate propellant. Sub-scale detonation data will be used to refine and validate the particle prediction model.

These initial project phases are directed towards development of a data set to support a model of OB/OD phenomena that is acceptable to regulators and is sufficient to predict environmental emissions.

Introduction

The Utah Test and Training Range (UTTR) is the only installation on US soil permitted to perform open detonation (OD) of munitions with greater than 10,000 lbs net explosive weight (NEW) and open burn (OB) of much larger items. The OB/OD mission is one of the major emission events on the UTTR. The airborne PM released from these events is a regulated pollutant.

Historical data for PM emissions from OB/OD events is based on Bang Box emission factors. These factors do not provide PSDs. No modeling approach is known that can provide emission factors and PSDs for OB/OD events.

This paper will describe a modeling approach that provides predictive information about PSDs and emission rates from OB/OD events. The model consists of two primary

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components: the Cheetah combustion model, and a MATLAB particle model. Cheetah is a kinetic combustion and detonation dynamics model developed by Lawrence Livermore National Laboratory (LLNL). The MATLAB particle model provides particle size information by stochastically solving the general dynamics equation (GDE) of particle formation.

Laboratory work has been performed following the methodology used in previous work (Palmer, et al, 2008). A Grimm 1.107C particle monitoring system was used to measure PSDs from small scale burns of a surrogate propellant at the University of Utah Combustion Research Center.

Modeling Procedure

The two component modeling approach separates the detonation process into chemical reactions and particulate formation steps. This separation contrasts with typical soot formation models, which simultaneously simulate chemical reactions and particle processes. However, the complexity of developing a traditional soot formation model precluded this approach. Instead, the Cheetah combustion model and a MATLAB particle model were connected in series. The model components are connected as shown in Figure 1.
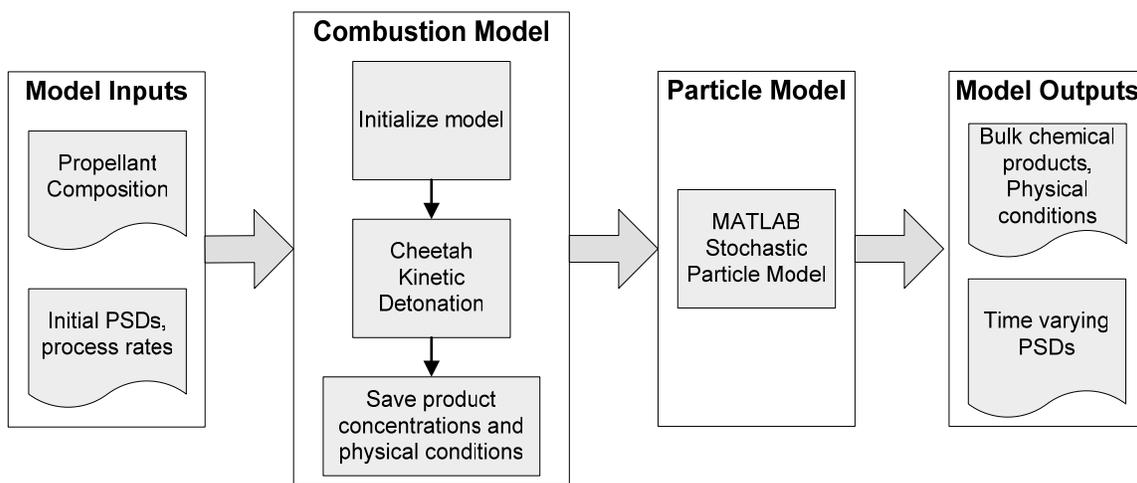

Figure 1. Model Components

The two component modeling framework unites the disparate timescales inherent in the detonation process. Cheetah provides chemical and detonation dynamics information (on the order of microseconds), while the MATLAB component models particle dynamics occurring on the order of milliseconds or seconds. By using Cheetah's output as an input to the MATLAB model, these timescales are united into a single modeling approach.

Cheetah

Cheetah 4.0 models the combustion and dynamics of the detonation process. Cheetah's combustion solver uses chemical kinetics to give time-varying product concentrations. Detonation dynamics are modeled according to the 2D Wood-Kirkwood (WK) theory, which considers a cylindrical charge of infinite length expanding radially from the centerline (Fried, et al, 1997).

A WK kinetic detonation was run in Cheetah. The reactant composition was 90% cyclotetramethylene-tetranitramine (HMX) and 10% aluminum (Al) by weight. All other parameters were set to their default values. The Cheetah reactant composition does not align well with the composition of the surrogate propellant used in the laboratory. The 90% HMX, 10% Al composition was used as an example case in this work. Future work will include the appropriate propellant composition.

MATLAB Particle Model

The MATLAB particle model stochastically simulates particle formation. Modeling begins with an initial PSD. The initial distribution for this work, shown in Figure 2, is a bimodal distribution fit to preliminary laboratory data. The model creates an initial number of simulation particles (N_{s0}) within an initial simulation volume (V_{s0}). These simulation particles are assumed to represent the entire particulate distribution.
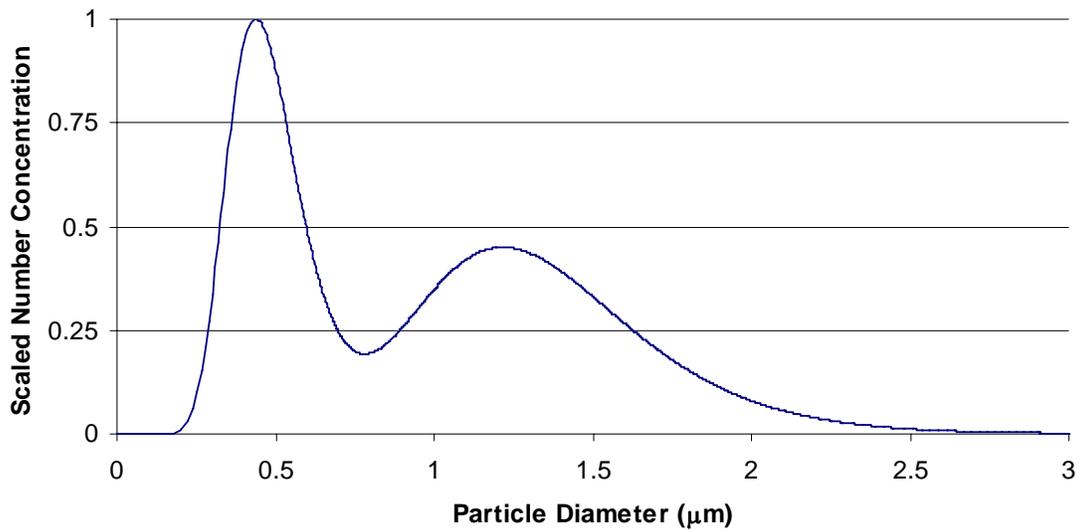

Figure 2. Initial PSD for MATLAB particle model.

The particle model simulates three particle formation processes: particle inception (nucleation), particle growth by condensation (surface growth), and particle collision (coagulation). Kernels (size dependent process rates) are used to define the rates of these three processes over the course of the simulation. Constant, size independent kernels are used throughout this work.

The rate of a process determines its likelihood of occurring during the simulation time step. Rates and probabilities for each process are calculated as shown in Table 1. For each incremental time step, a process is randomly selected based on its probability of occurring. Particles are then randomly chosen for the process, the process is performed, and time is incremented by a rate dependent time constant, $\tau = (R_n + R_s + R_c)^{-1}$.

Table 1. Particle Formation Process Rates

Process	Process Rate (1/s)	Probability
Nucleation	$R_n = \dfrac{V_s}{N_s} \cdot \sum\limits_{m=1}^{Ns} J_m$	$P_n = \dfrac{R_n}{R_n + R_s + R_c}$
Surface Growth	$R_s = C_0 \cdot \sum\limits_{m=1}^{Ns} Ks_m$	$P_s = \dfrac{R_s}{R_n + R_s + R_c}$
Coagulation	$R_c = \dfrac{1}{V_s} \cdot \sum\limits_{m=1}^{Ns} \sum\limits_{n=m+1}^{Ns} B_{m,n}$	$P_c = \dfrac{R_c}{R_n + R_s + R_c}$

J, Ks, and B are the kernels for nucleation ($m^{-3}s^{-1}$), surface growth (m^3/s), and coagulation (m^3/s).

N_s is the number of particles in the simulation.

V_s is the simulation volume (m^3).

Because the number of simulation particles (N_s) fluctuates over the course of the simulation, it is necessary to adjust the simulation volume (V_s), by bounding N_s so that $\dfrac{N_{s0}}{2} \leq N_s \leq 2N_{s0}$. When N_s is too high, V_s is halved, and simulation particles are randomly discarded until $N_s = N_{s0}$. When N_s is too low, V_s is doubled, and the simulation particles are replicated. Although these volume changes lead to small errors, they are necessary to maintain an appropriate number of simulation particles.

Test Procedure

Using the laboratory setup of Palmer et al, 2008, a Grimm 1.107C particle monitoring system was used to measure PSDs from small scale burns of a surrogate propellant. Distributions were measured every six seconds with thirty one bins, with diameters ranging from 0.265-32 microns (μm).

Results

Laboratory Data

Data were collected for multiple burns of the surrogate propellant. Figure 3 is an example of the time-varying total suspended particulate (TSP), PM10, and PM2.5 curves for a single burn. Figure 4 provides mass concentration contours (μg/m3) for the same event.
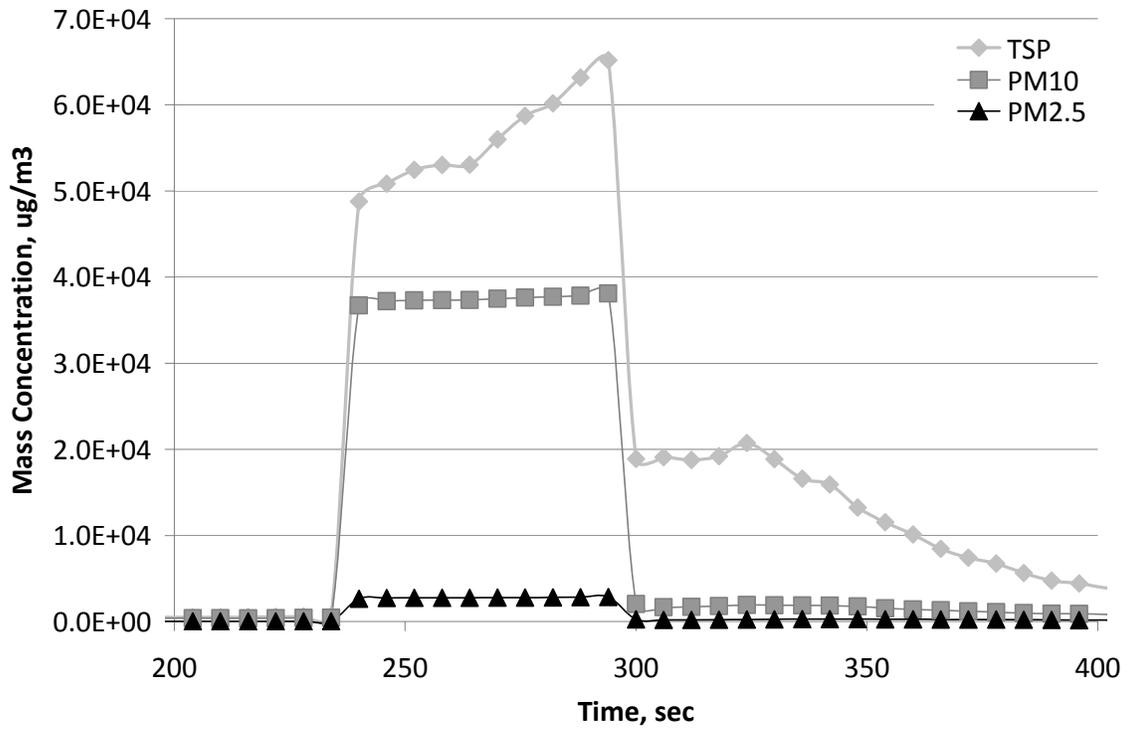
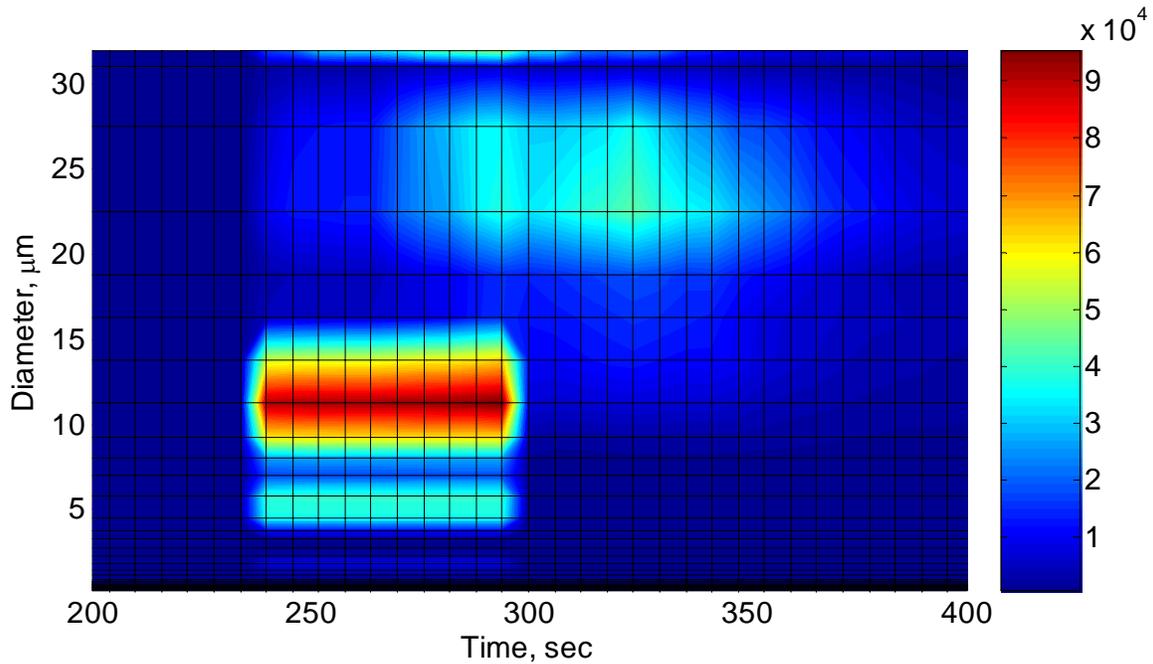**Figure 3.** Total suspended particulate (TSP), PM10, and PM2.5 for run 1 on June 6, 2008.

Figure 4. Mass concentration (μg/m3) of alumina over time for run 1 on June 6, 2008. The grid shows time and particle diameter sampling intervals.

Cheetah Kinetic Detonation

Cheetah models both thermodynamic and chemical aspects of the detonation. Figure 5 shows the pressure and temperature decay over the course of the detonation. Figure 6 shows the conversion of aluminum to aluminum oxide over the course of the detonation.
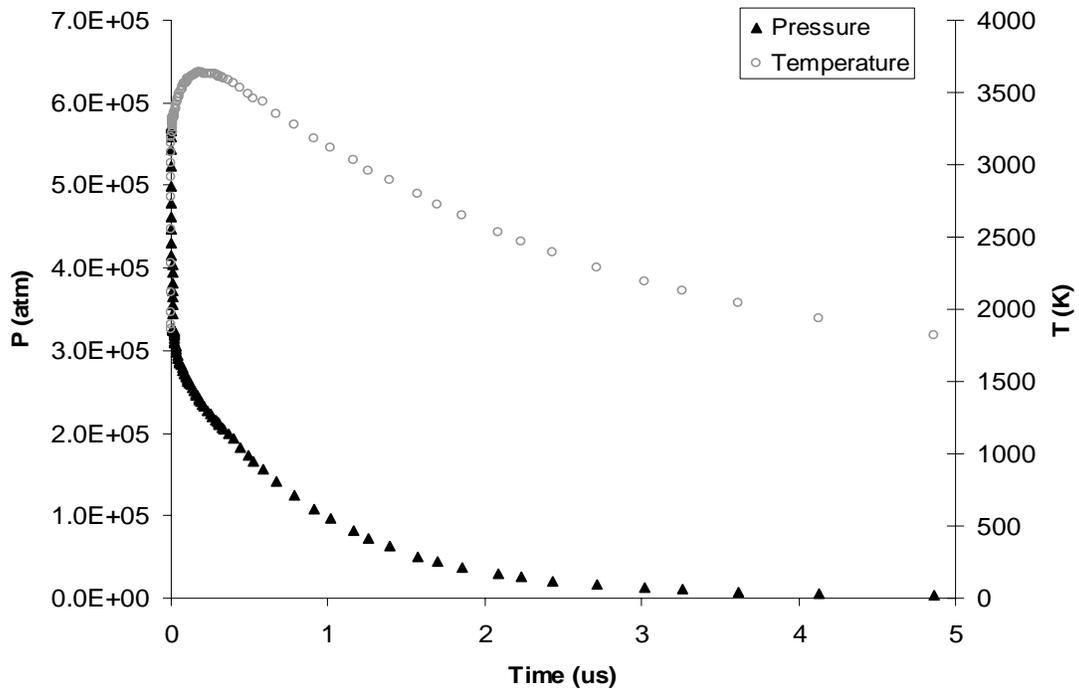

Figure 5. Pressure and temperature over time for Cheetah WK detonation of a charge composed of 90% HMX, 10% Al.
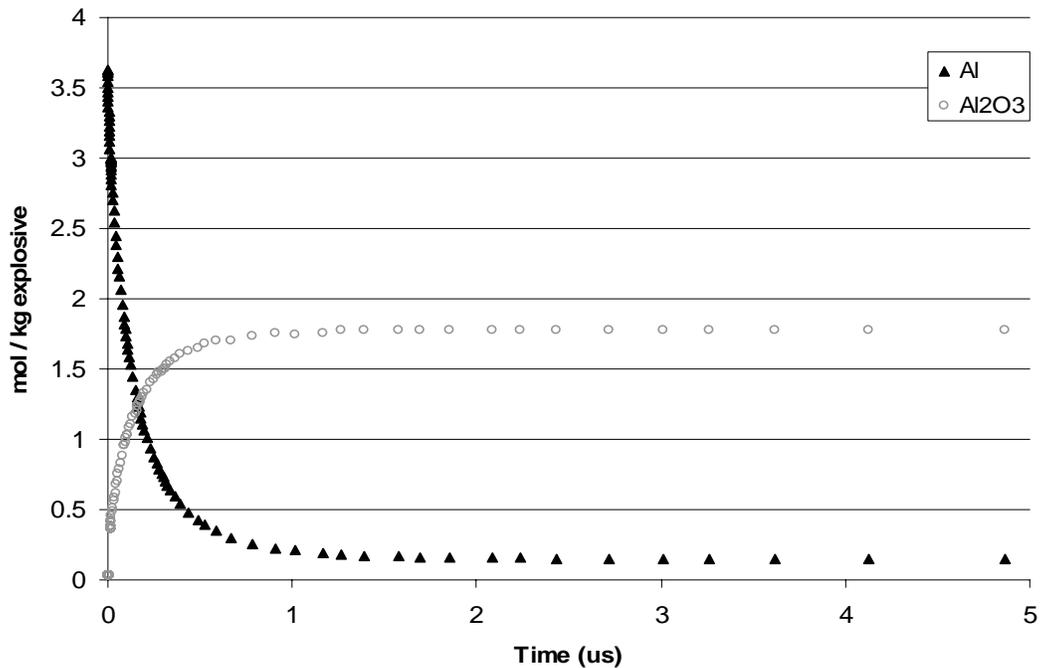

Figure 6. Al and Al_2O_3 concentrations over time for Cheetah WK detonation of a charge composed of 90% HMX, 10% Al.

MATLAB Particle Model

Particle model results are reported as mass concentration contours. As an example output, a ten-run average with constant process rates (coagulation: 10^{-10} m^3/s, nucleation: 10^{10} m^{-3}s^{-1}, surface growth: 0 m^3/s) is shown in Figure 7.
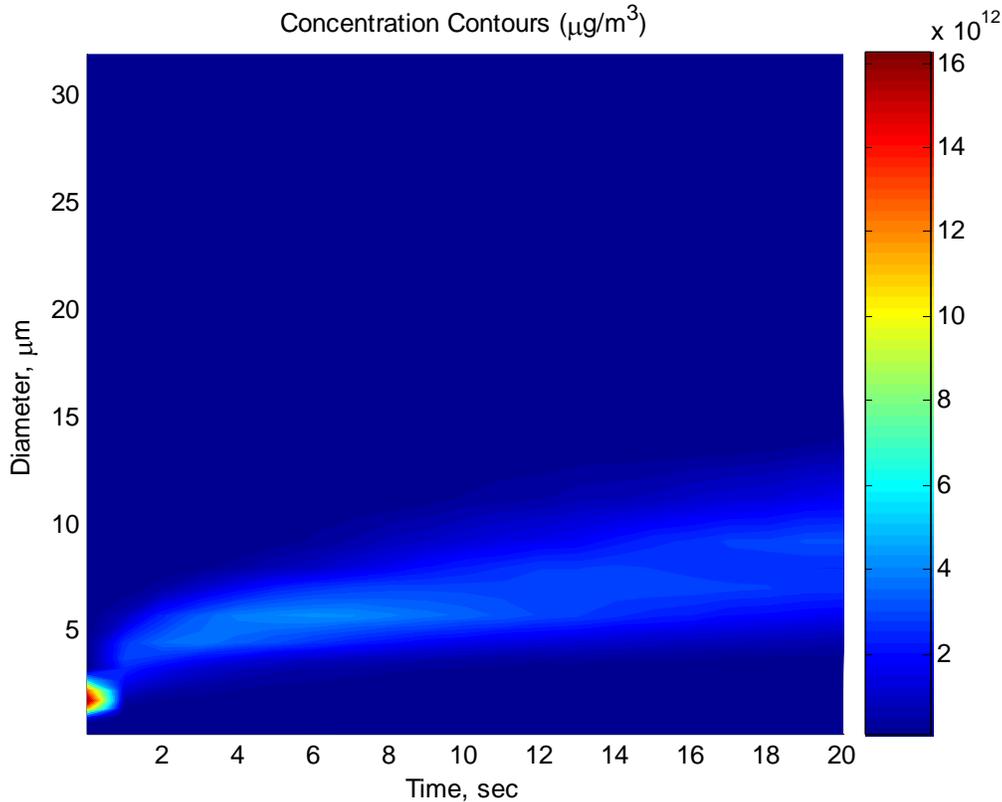

Figure 7. Average mass concentration (μg/m3) from ten particle formation model runs. (coagulation: 10^{-10} m^3/s, nucleation: 10^{10} m^{-3}s^{-1}, surface growth: 0 m^3/s)

Analysis

Laboratory particulate data (such as that presented in Figures 3 and 4) show high variability from run to run. Measures such as mean diameter of the distribution, maximum number and mass concentration, and total mass emitted, vary significantly between runs. Due to this variability, a more thorough experimental protocol and an improved understanding of the Grimm instrument are needed before laboratory data can be compared to model results.

Cheetah has been shown to provide accurate predictions of detonation parameters in ideal and moderately non-ideal explosives (Lu, 2001). According to the LLNL website, Cheetah "has become the Department of Defense's preferred code for designing new explosives…and, to a lesser extent, propellants" (Heller, 1997). This usage of Cheetah validates its accuracy as a combustion and detonation modeling tool.

Sub-scale detonation data are needed to confirm Cheetah's applicability to the OD events considered in this modeling approach. These data, which will include high speed

pressure and temperature measurements, will be valuable for implementing and refining the Cheetah component of this model.

Stochastic methods have been effectively used to model time-varying PSDs. They have been applied to soot particles in premixed flames, aerosols in global weather prediction, and mineral dust particle size in mining operations. These applications highlight the computational advantage of stochastic models over deterministic models. The similarity of these applications to OB/OD activities supports the use of a stochastic model.

The MATLAB particle formation model component can be compared to deterministic solutions to the GDE for simple cases. For constant rate coagulation and nucleation (Maisels, et al, 2004), number concentration develops over time according to:

$$\frac{N}{N_0} = B\frac{1 + B\tanh(\tau/2)}{B + \tanh(\tau/2)}$$
(Equation 1)

As shown in Figure 8, the 10-run average output from the MATLAB particle model agrees with the theoretical value with a maximum error of 9%. While this error value is higher than that in the Maisels, et al, report, increased simulation accuracy in the particle model can be achieved by increasing the initial number of simulation particles (N_{s0}). This increase leads in turn to an increase in computational time.
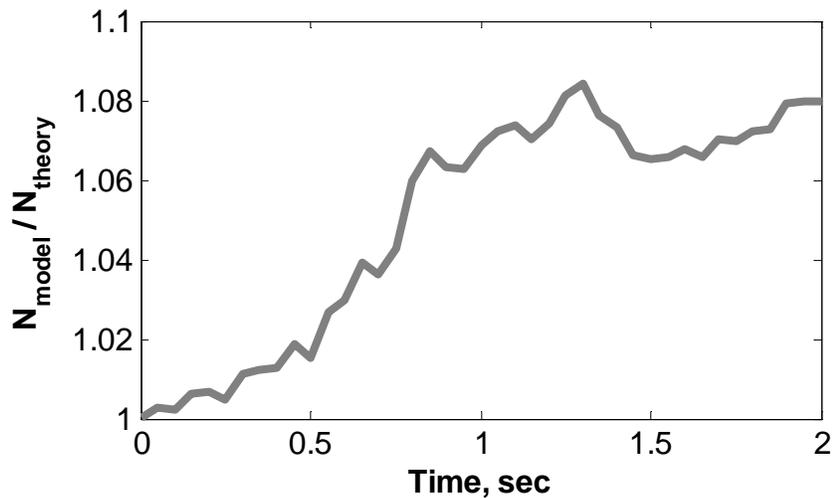

Figure 8. Comparing number concentration for theoretical and 10-run average stochastic particle model. (coagulation: 10^{-10} m^3/s, nucleation: 10^{10} m^{-3}s^{-1}, surface growth: 0 m^3/s)

Conclusion

A two component modeling approach has been developed for predicting particulate emissions from OD events. The Cheetah combustion model and MATLAB particle model components have been described. Cheetah predicts chemical products and detonation dynamics of a detonation. The MATLAB particle model uses a stochastic method to predict the

development of PSDs over time. Preliminary laboratory work has been performed to validate sampling methodologies.

More extensive laboratory and field work is needed to better validate this modeling approach. Improved experimental protocols will be developed to reduce run to run variability in the laboratory data. Sub-scale detonation data will be acquired to refine the particle prediction model. These data are critical for the validation of this modeling approach. Refinement and validation of this modeling approach will be important initial steps to support a model of OB/OD phenomena that is acceptable to regulators and is sufficient to predict environmental emissions.

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