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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Nanoparticle formation in the exhaust of vehicles running onultra-low sulfur fuel" *by* Hua Du and Fangqun Yu

Anonymous Referee #2

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The paper presents an interesting analysis of nanoparticle formation in the exhaust of vehicles running on low sulfur fuels. Earlier simulation work predicted that nanoparticle formation would be negligible under these conditions but observations showed significant number concentrations of nanoparticles. Several mechanisms (enhanced sulfur conversion, storage/release, and the presence of non-volatile cores) are invoked to explain the observations. I have two significant concerns with the manuscript in current form.

1) Many of the conclusions appear to depend critically on input data whose values are not sufficiently justified. For example, a range of SREF values is assumed with little justification. This should be discussed more fully. Similarly, the result relating to non-volatile cores depends on the assumed number concentration. While a reference is

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given for their assumed size distribution, the assumed number concentration should be justified. Also, it needs to be made clear at what point (i.e. what dilution ratio) the quoted concentration refers. In some cases, additional sensitivity simulations are warranted (see below).

2) My other significant concern is that the work is not adequately documented. Parts of the methods appear to contradict each other, some critical input data is unclear, mathematical representation of various processes needs to be clarified, and the scenarios are described inadequately and in a scattered format. This makes the manuscript confusing to follow, and it is difficult to evaluate the model and how the main conclusions are reached. Examples of these problems are listed below.

I recommend that the manuscript be substantially clarified and somewhat reorganized to address the following points.

Equation 1 appears to have terms that account for evaporation to smaller size bins as well as coagulation. Condensation does not appear (there is no term for growth from bin i-1), yet it is clear that condensation of organics (and H2SO4?) are important. How is this treated mathematically?

Equation 3 partly answers the question about condensation of organics. However, the formulation seems not to grow particles from bin i to bin i+1. Why not and how does this impact the model predictions?

Are shape effects expected to influence the coagulation rates? Are these accounted for in the model (it appears no)? Is this justifiable? How will it impact the results?

Somewhere early in the methods, there should be a comprehensive list of species treated. The closest I can find is p. 2720, line 13: “H2SO4, H2O, and other species including organics, soot, and refractory cores.” Please clarify confirm whether this list is comprehensive of all species treated. Later it appears that “organics” is separated into a low-volatile and semi-volatile component.

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It would be better to have a complete list of species in one single place rather than have the reader piece it together from different parts.

Please write out mathematically how mixing and dilution are treated in the model.

Please clarify the size distribution of background aerosols used. Page 2721 line 19 gives a mean size and number concentration, but this does not specify the full size distribution. Is it assumed to be lognormal? If so, what is the sigma value for the distribution?

It appears that only one background aerosol size distribution is used. How sensitive are the results to this size distribution?

Page 2721 lines 20-21: “The mean size and concentration of soot particles is assumed to be 50 nm in diameter and 10^7 cm-3, respectively”. This sentence appears at the end of the paragraph about background aerosols. The number concentration given, however, is surely not meant to represent the ambient background. What does this refer to and how/where does is this input to the model? Also, please specify the full size distribution: lognormal? What is the sigma? Are the conclusions sensitive the this assumed value?

Please give the assumed vapor pressures of each organic component in the model. What value of organic surface tension is used when correcting for the Kelvin effect?

The treatment of the sulfur conversion efficiency is confusing. On p. 2723, lines 7-9, it says that the treatment of Giechaskiel et al (2007) is adopted by your study. Later (p. 2724, lines 9-11), however, you compare the sulfur conversion efficiency used in your simulations with the Giechaskiel results. This must be a contradiction? Shouldn’t the treatment of sulfur conversion by the model be discussed in the methods section?

A number of different scenarios are run with the model to reach different conclusions. It is difficult for the reader to keep track of which scenario goes with each figure and

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what are the corresponding input parameters. Much of the input data is scattered in different figure captions or appears scattered throughout the results section. This should be organized into one place more systematically for the benefit of the reader. Table 1 is a good start but documents only a subset of the simulations performed here. I recommend adding a section “2.3 Scenarios” to the methods section that describes all the scenarios in one place, preferably lays out a table with all the necessary data, and discusses the motivation behind each scenario.

Page 2728, line 26: “non-volatile particles are assumed to be in the range of 1.8 nm-3 nm in diameter”. Again, please specify the complete size distribution.

Minor comments

-Please define briefly in the introduction “sulfur storage/release effect”

p. 2719, line 26: “dilution ratio at plume age of 5 s varies from ˜5 to ˜10”: please clarify/confirm that this corresponds to the roadside and, therefore, the end of “stage 2”

Section 2.1: Please define precisely the upper and lower size boundaries for the sectional microphysics used as the manuscript currently has the vague “from molecular size to several micrometers”. How are the size sections spaced?

Section 2.1: How are the Nj values calculated?

Figure 1: The logarithmic scale makes it difficult to evaluate the agreement between observations and model. Please discuss in the text the total mass and number concentrations implied by each of these size distributions.

Figure 2a: the x-axis needs to be labeled and given units

Figure 3a: x-axis needs to be labeled and given units

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