For the two parts below, please address the reviewers' comments in a more informative way.

1. The revised text for the response to Review #1 - comment #1 is still unclear. Do you mean all primary emissions use the same size distribution in the model? If so, please provide the size distribution in supporting information as the reviewer requests.

The default parameters for the size distribution of the emitted particles into the CMAQ model are given below. These values originate from paragraph 14 of Binkowski and Roselle 2003 but were updated in 2014 by Kathleen Fahey to reflect those in Elleman and Covert 2010 table 5. These values are located within the CMAQ source code in the “AERO_DATA” module (https://github.com/USEPA/CMAQ/blob/5.2.1/CCTM/src/aero/aero6/AERO_DATA.F).

Parameter	Aiken	Accumulation	Coarse
dgvem (nm)	60	280	6000
def_diam (nm)	15.0	80.0	600.0
min_diam_g (nm)	1.0	30.0	120.0
max_diam_g (nm)	80.0	500.0	100.0
sgem	1.7	1.7	2.2
def_sigma_g	1.70	2.0	2.2
min_sigma_g	1.05	1.05	1.05
max_sigma_g	2.5001	2.5001	2.5001

Where:

- dgvem = geometric mean diameter by volume,
- def_diam = default background mean diameter for each mode,
- min_diam_g = minimum geometric mean diameter for each mode,
- max_diam_g = maximum geometric mean diameter for each mode,
- sgem = geometric standard deviation of emitted particles in each mode,
- def_sigma_g = default background geometric standard deviation for each mode,
- min_sigma_g = minimum geometric standard deviation for each mode, &
- max_sigma_g = maximum geometric standard deviation for each mode.

The revised text at line 99 has been revised further to read, “Primary aerosol emission rates are provided by the 2011 National Emissions Inventory, which characterizes emissions based on source type and location. Within CMAQ, all primary aerosol emissions, independent of source type, are parameterized with modal size distributions per Elleman and Covert 2010 (see SI)." for further clarification.

The table and explanation are also included in a separate document as supplementary information for the reader should they be specifically interested in the details of the size distribution for primary emissions, as the reviewer requested.

2. The reviewer #1 also asked about how the model scheme for nucleation and growth may affect the modeled size distribution in comment #2, which hasn't been addressed in the response. Please add descriptions about it. For example, how sensitive the modeled size distribution depends on

the parameters used. If other studies have addressed this, the authors may cite as references and summarize briefly their results here to assist the discussion.

As noted previously, the default new particle formation and nucleation parameterization in CMAQ is based on a classical binary sulfuric acid-water homogeneous nucleation from Kulmala et al. (1998). To elaborate on the impact of the model scheme for nucleation and growth on the modeled size distribution, we refer to a comparison of parameterizations for ternary nucleation and nucleation mode processes evaluated by Elleman and Covert (2009). In addition to the binary nucleation parameterization that is utilized within CMAQ, Elleman and Covert investigated a ternary ammonia-sulfuric acid-water parameterization called the Napari parameterization, based on classical nucleation theory with nucleation rates several orders of magnitude higher than the default binary parameterization. The Napari parameterization was utilized in the base case as well as with nucleation mode processing to the Aiken mode to include the number of nucleated particles which survive growth to 10nm and addition to the existing Aiken mode without being lost by coagulation. These parameterizations and the resultant size distributions were compared to observations of size distributions from the coordinated Pacific Northwest 2001 and Pacific 2001 field campaigns. The main impact of changing nucleation parameterization within CMAQ is the large impact of the size distribution below 200nm, where "Adding Napari ternary nucleation increases the prominence of the Aiken mode, but Napari w/Processing better reproduces distinct Aitken and accumulation modes, the peak of the Aitken mode, and the prominence of the Aitken mode relative to the accumulation mode. Above 200nm, changes to nucleation have no effect on the size distribution." However, including ternary nucleation does not solve the issue of reproducing observed size distributions with CMAQ modeling, as nucleation is only one component. The impact of the size distribution from nucleation and growth is evident by this sensitivity study, but the default binary parameterization has not been updated within CMAQ and the addition of nucleation is more complex than simply updating the parameterization.

Though not a direct sensitivity study on the size distribution, Zhang et al. (2010) compared 12 nucleation parameterizations including 7 binary, 3 ternary, and 2 power laws, for their nucleation rates as compared to observations. It is noted that the Napari ternary parameterization (without processing to the Aitken mode) "grossly overpredicts the observed nucleation rates", and the default Kulmala et al. parameterization used in CMAQ has technical mistakes in the formula regarding the kinetic treatment of hydrate formation, reducing nucleation rates. Updating the nucleation parameterization within CMAQ to a more accurate parameterization should be done to improve our prediction of nucleation of particles but is well outside of the scope of this study.

I am also wondering how differ the current understanding about nucleation and growth from the model scheme and whether the difference would affect the modeled results. This is perhaps not the focus of the paper. But it is an important information for readers to understand the results and the interpretation herein. This is somewhat related to the reviewer #2's comment #2(b). But the revised text in line 362 is too brief to strength the discussion.

It is noted by Elleman and Covert 2009 when investigating nucleation parameterizations that "The Kulmala et al. [1998] binary nucleation theory in the standard version of CMAQ v4.4 does not reflect current knowledge of particle nucleation and processing. More recent versions of CMAQ update portions of its aerosol science but do not change the nucleation code.” It is known that the binary sulfuric acid-water theory of nucleation does not fully explain nucleation since the measured sulfuric acid concentrations are not always high enough to produce the observed nucleation rates alone, and there are most likely other processes and species that impact nucleation and need to be further investigated. One of the biggest drawbacks to the default binary parameterization is that there is not a nucleation mode included within CMAQ. The Aitken mode normally is between 10-40nm, while nucleating particles are well below the 10nm threshold, and adding the nucleated particles to the Aitken mode would change the definition of that smallest mode without accounting for loss within particle growth to the traditional Aitken mode. As Elleman and Covert note, “Including nucleation in CMAQ requires both an updated aerosol nucleation theory as well as including nucleation mode dynamics separate from CMAQ’s three mode structure.”, which has not been implemented at present. Not including nucleation and particle growth of nucleated particles to the Aitken mode impacts the accuracy of the size distributions utilized in CMAQ and therefore the number, surface area, and volume of particles that are predicted. This may explain part of the issue with surface area that was investigated in this paper, though the issue is too complex to pinpoint without further investigation outside of the scope of this study.

The text at line 362 has been updated to clarify and expand on the nucleation and particle growth in CMAQ and now reads, “It is also important to acknowledge that some of the model-measurement disagreement could be due to processes not considered in the model such as phase separation, viscosity changes of aerosols, and direct modeling of clouds impacting cloud processing of aerosols, though the impacts of these processes are not investigated further in this work. The lack of a fourth mode below the Aitken mode for nucleation of particles and growth to the Aitken mode also impacts the accuracy of the size distribution within CMAQ and may explain a portion of the model-measurement disagreement, though it is known that improving the default parameterization does not reduce all errors to the size distribution (Elleman and Covert, 2009b).”

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