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Comment

## ***Interactive comment on “Change in global aerosol composition since preindustrial times” by K. Tsigaridis et al.***

**Anonymous Referee #1**

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Review of

**“Change in global aerosol composition since preindustrial times ”**  
by  
K. Tsigaridis et al.

This manuscript studies the change in global aerosol composition and burden since the preindustrial period. The types of aerosol included are sulfate, ammonium, nitrate, black carbon (BC), primary organic aerosol (POA), and secondary organic aerosol (SOA). The model uses bulk equilibrium calculations for sulfate-ammonium-nitrate-water aerosol. For primary carbonaceous aerosols BC and POA, the method here

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is similar to those that are published in the literature, except, instead of assuming constant exponential decay lifetime for conversion of hydrophobic to hydrophilic aerosol, this study assumes that the conversion rate is governed by heterogeneous reaction with ozone. For SOA, this study categorizes emissions of SOA precursors into a few classes of compounds and uses a lumped two-product model with dependence on VOC:NO<sub>x</sub> ratio for SOA yield. The results of this study is important for the atmospheric science community. Description of the model in the manuscript needs to be expanded; some aspects of the model can also be improved. Specific comments about the manuscript are provided below.

**(1)** This manuscript is not the only study on aerosol chemical composition changes since preindustrial times considering SOA with all other main aerosol components. In particular, LiaoSeinfeld05 studied the change in global distribution and radiative forcing of sulfate, ammonium, nitrate, BC, POA, and SOA from preindustrial period to 2100 using a unified chemistry-aerosol-climate model. Mineral dust and sea salt were also included in that study (global burdens were reported in Liaoetal04). This manuscript should cite LiaoSeinfeld05 and compare the results of this study with those of LiaoSeinfeld05.

**(2)** Even though much of the model details of this study was provided in TsigaridisKanakidou03, more details should still be provided in this manuscript to make this manuscript more self-contained. Examples of things that need to be addressed include: What is the gas-phase mechanism used? What is the model time step? How are emissions of monoterpenes and ORVOCs speciated between  $\alpha$ -pinene and  $\beta$ -pinene? What percentages of emitted BC and POA are assumed to be hydrophobic?

**(3)** Does the model used for the current study consider SOA formation from further oxidation of the higher volatility products shown in Equation 1 as in TsigaridisKanakidou03? To which simulation scenario of TsigaridisKanakidou03 does the SOA model used in this study correspond? The manuscript should provide more detailed discussion of differences (model and results) between this study and TsigaridisKanakidou03.

What is the preindustrial and present-day annual global SOA production rates determined by this study?

**(4)** The manuscript is not clear on how the dependence of SOA yield parameters on  $\text{VOC}:\text{NO}_x$  is modeled. There are three sets of SOA yield parameters for  $\alpha$ -pinene. What values of  $\text{VOC}:\text{NO}_x$  ratio are considered high  $\text{VOC}:\text{NO}_x$  conditions? At what range of  $\text{VOC}:\text{NO}_x$  ratio do APINp1H and APINp2H form? How about APINp1 & APINp2 and APINp1N & APINp2N? Modeling of SOA yield dependence on  $\text{VOC}:\text{NO}_x$  is very new. How sensitive are the model results to this dependence?

**(5)** For SOA modeling, this study categorizes the biogenic monoterpene and ORVOC emissions into  $\alpha$ -pinene and  $\beta$ -pinene. Because of the uncertainties involved in SOA yields and computational efficiency, classifying SOA precursors into groups is in general a good idea. However, the manuscript does not explain why  $\alpha$ -pinene and  $\beta$ -pinene were chosen as surrogates and current understanding of SOA formation suggests that more categories are needed to properly represent the wide range of SOA yield parameters. For example, sesquiterpenes generally have higher SOA yield than monoterpenes Griffinetal99b. Also, Ngetal06 suggest that the number of double bonds in the parent VOC determines whether SOA formation is rate-limited by the first oxidation or later oxidation steps.

**(6)** It has been determined that products of isoprene oxidation also lead to SOA formation (e.g. Claeysetal04, Krolletal05, Krolletal06). HenzeSeinfeld06 indicate that including isoprene as a source of SOA causes substantial increases in predicted SOA concentrations, particularly in the free troposphere and remote marine environments. How would inclusion of isoprene oxidation as a source of SOA affect the results of this study?

**(7)** This study assumes that conversion of hydrophobic BC and POA to hydrophilic BC and POA is determined by ozone oxidation and that the globally averaged turnover time is approximately 1 day. How does this turnover time vary geographically and vertically?

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The value of 1 day is most likely an upper bound estimate because condensation of hygroscopic material such as sulfuric acid can also convert hydrophobic aerosol to hydrophilic aerosol. Looking at functional groups using scanning transmission x-ray microscopy, Mariaetal04 estimated the oxidation rate is approximately 24% per day for BC and 13-21% per day for OC; these values correspond to turnover time of about 5 days. Can the authors provide insights as to why the modeled turnover time is much smaller?

**(8)** Since the modeled aerosol optical depth is derived from aerosol composition and size, the section “Aerosol optical depth” should be placed after section “Aerosol Composition”.

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