Responses to the comments of anonymous referee #1

We thank the referee for the valuable comments that have greatly helped us to improve the manuscript. Please find below our responses (in black) after the referee comments (in blue). The changes in the revised manuscript are written in *italic*.

General comments

The paper "Sources of organic aerosols in Europe: A modelling study using CAMx with modified volatility basis set scheme" by Jiang et al. deals with a very interesting topic for modelling science. Indeed, the modelling reconstruction of Organic aerosol fraction remains a challenging issue due to the relevant number of species and processes involved. The paper can surely provide a very interesting contribution to the scientific knowledge in this field, particularly in the European context and therefore fits the scope of ACP. The paper is well written, with concise and clear statements, and it does not require any substantial review of syntax and language.

However, before publication, there are a few issues that should be addressed by the authors, that are detailed in the following:

1) One of the key aspect of the paper concerns the implementation of modified parameterizations in the 1.5 VBS scheme, however from section 2.2.3 is not clear what modifications have been actually introduced beyond the split of the original 5 basis sets into 11. From the text it seems that just two modifications were introduced: a) Setting SOA yield for DN to 0. b) Enabling oxidation of SOA from Biomass burning. Is it correct? If yes, the modifications introduced by authors are surely reasonable and interesting, but limited only to a few aspects of 1.5 VBS scheme and this should be better clarified in the text. Conversely if other modifications have been introduced they should be better describe it (maybe introducing a table comparing BASE and NEW VBS parameterizations)

The major modification in the code was extending and splitting the standard VBS scheme to enable source apportionment for POA and SOA, together with the adjustment of yields for the new diesel vehicles and oxidation rate of SOA from biomass burning. Since most of the changes are in the model framework (adding new species to the model species list, splitting reactions for precursors from different sources), it is difficult to display them in a table. We will however upload the modified model codes in a public data repository once published. Following similar comments of referee 2#, we added a more detailed description of the 1.5-D VBS and our modifications in this study which we called PSI-VBS in order to distinguish from other modified versions.

(Section 2.2.1 P6 L3 – L7) "The 1.5-D VBS framework in CAMx is based on the one-dimensional (1-D) VBS, in which the organic species are grouped only by their volatility (Donahue et al., 2006). The 1-D VBS was later extended to a second dimension (2-D) to include the oxidation state – specifically O:C ratio (Donahue et al., 2011). In order to reduce the high computational burden of the 2-D VBS when implemented in CTMs, the 1.5-D VBS was developed, which combines the 1-D VBS and the multiple reaction trajectories defined in the 2-D VBS space, it can therefore account for changes in both volatility and oxidation state (Koo et al., 2014). The default VBS scheme in CAMx…"

(Section 2.2.1 P6 L16 – L21) "The VBS scheme has been modified in previous studies to improve the performance of air quality models. For example, Shrivastava et al. (2013; 2015) treated SOA as a non-absorbing semisolid with low "effective volatility" and added the fragmentation reactions. Using this method in the regional model CHIMERE, it was found that fragmentation could effectively reduce the SOA formation when further aging of biogenic SOA was allowed, leading to a better agreement with observations (Cholakian et al., 2018). Instead of a major modification of the chemical mechanism, this study aims at modifying the 1.5-D VBS framework of CAMx to enable source apportionment of OA in Europe. As a first step to separate…"

(Section 2.2.1 P6 L25 – L26) "The schematic diagram of the VBS with the modified basis sets (it will be referred to as PSI-VBS thereafter) is shown in Fig. 1"

(Section 2.2.3 P7 L17 – L23) "...While the standard VBS of CAMx disables the aging of SOA for the basis set PBS (biomass burning and biogenic sources) to avoid overestimation of biogenic SOA, the separated sets for biomass burning (BB) and biogenic (BIO) sources allow us to implement individual parameterization schemes. Therefore, we kept the default parameterization (without aging of SOA) for BIO as a compromise for the lack of gas-phase fragmentation, and enabled the oxidation of secondary gases from biomass burning (see BB in Fig. 1) with a reaction rate of 4×10^{-11} cm^3 molec^{-1} s^{-1} according to previous studies (Ciarelli et al., 2017a; Ciarelli et al., 2017b; Denier van der Gon et al., 2015; Fountoukis et al., 2014; Murphy and Pandis, 2009; Theodoritsi and Pandis, 2019). For other basis sets, the default parameters of CAMx v6.3 were used."

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Shrivastava, M., Easter, R. C., Liu, X. H., Zelenyuk, A., Singh, B., Zhang, K., Ma, P. L., Chand, D., Ghan, S., Jimenez, J. L., Zhang, Q., Fast, J., Rasch, P. J., and Tiitta, P.: Global transformation and fate of SOA: Implications of low-volatility SOA and gas-phase fragmentation reactions, J. Geophys. Res.-Atmos, 120, 4169-4195, doi: 10.1002/2014jd022563, 2015.

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2) Authors point out that one of added values of their work is the evaluation of model results over a long term period. However both meteorological and air quality model performance evaluation (for chemical species other than OA) is limited only to a winter and summer month. Though interesting, such analysis is not fully adequate to evaluate the CAMx performance over the whole year. Moreover, in most cases the selected months (February and July) do not overlap with the observation periods of OA measurements (see table 1). An yearly based analysis of AQ and meteorological model performance should be added. Moreover, considering the observation periods covered by OA measurements a seasonal based analysis could be added too. The latter would also be coherent with several results presented by authors in sections 3.1.2, 3.2, 3.3 and 3.4.

A whole-year model performance evaluation for both meteorological and air quality simulations was added into the revised manuscript. Statistical results for different seasons (December-January-February, March-April-May, June-July-August, and September-October-November) as well as for whole year were displayed in Tables S2 and S3. The numbers in section 3.1.1 were updated as well.

As the referee mentioned, actually we have already presented a season-based analysis for OA components in section 3.2. To be more coherent, we added the Table S5 presenting the seasonal statistical results as a supplement to Table 2 and Figure 3.

Table S5. *Seasonal statistical analysis of daily average organic aerosols at nine ACSM/AMS stations. MB: mean bias; ME: mean error; RMSE: root-mean-square error; MFB: mean fractional bias; MFE: mean fractional error. Spring: March-April-May, summer: June-July-August, autumn: September-October-November, winter: December-January-February.*

Season	Site	MB ($\mu g\ m^{-3}$)		ME ($\mu g\ m^{-3}$)		RMSE ($\mu g\ m^{-3}$)		MFB (%)		MFE (%)	
		BASE	NEW	BASE	NEW	BASE	NEW	BASE	NEW	BASE	NEW
OA											
spring	MHD	-0.1	0.1	0.7	0.8	1.6	1.7	-86	-74	117	114
	MRS	-9.4	-7.6	9.4	7.6	9.7	8.0	-163	-116	163	116
	ZRH	-1.7	0.8	3.5	3.7	4.7	5.0	-34	8	64	53
	SMEARII	0.0	0.7	0.4	0.8	0.6	1.2	-10	58	54	70
summer	MHD	0.0	0.0	0.3	0.3	0.5	0.6	-91	-83	119	115
	MSA	-2.4	-2.0	2.6	2.3	3.2	2.8	-89	-73	97	83
	ZRH	-1.0	0.1	3.2	3.3	4.8	5.0	-21	3	65	59
autumn	BLQ	-8.4	0.5	8.7	5.5	10.8	7.0	-62	10	67	35
	FKL	-1.1	-0.1	1.5	1.4	2.2	1.9	-53	-17	70	58
	MHD	-0.1	-0.1	0.2	0.2	0.5	0.5	-90	-83	116	113
	MSA	-1.4	-0.6	1.9	1.7	2.4	2.2	-42	-12	72	59
	SIRTA	-7.6	-4.3	7.7	5.4	11.0	8.5	-108	-43	111	63
	SPC	-2.7	5.7	5.3	8.0	6.9	9.6	-14	52	59	68
	ZRH	-0.6	2.2	3.6	4.4	4.7	6.1	-16	17	54	53
winter	BLQ	-21.4	-16.2	21.4	16.4	23.4	19.0	-149	-98	149	99
	MHD	0.0	0.1	0.3	0.3	0.6	0.6	-4	7	91	89
	MRS	-7.8	-5.3	7.8	5.8	11.0	9.3	-109	-55	111	65
	MSA	0.1	0.5	0.6	0.9	0.9	1.3	-10	10	90	92
	SIRTA	-3.6	-1.7	3.6	2.8	5.5	4.4	-110	-31	117	71
	ZRH	-4.1	-0.5	4.6	3.7	5.9	4.9	-68	-3	82	55
HOA											
spring	MRS	-1.4	-0.8	1.4	1.2	2.1	1.7	-107	-33	137	99
	ZRH	-0.3	0.4	0.4	0.7	0.6	0.9	-41	52	81	81
summer	MSA	-0.3	-0.2	0.3	0.2	0.3	0.3	-157	-102	158	111
	ZRH	-0.2	0.3	0.3	0.5	0.4	0.6	-29	61	80	83
autumn	BLQ	-1.6	0.2	1.7	1.4	2.6	1.9	-66	35	82	65
	MSA	-0.2	-0.2	0.2	0.2	0.3	0.2	-147	-84	149	92
	SIRTA	-0.7	0.1	0.8	0.8	1.4	1.3	-76	21	98	73
	SPC	-2.3	-1.1	2.3	1.4	2.7	1.8	-129	-42	130	58
	ZRH	-0.3	0.6	0.5	0.9	0.7	1.1	-26	65	81	86
winter	BLQ	-3.4	-2.2	3.4	2.6	4.9	4.1	-134	-53	137	82
	MRS	-1.2	-0.5	1.3	1.1	2.1	1.8	-88	-3	116	83
	SIRTA	-0.3	0.1	0.3	0.3	0.5	0.5	-70	30	101	80
	ZRH	-0.1	1.0	0.4	1.2	0.6	1.5	3	92	80	105
BBOA											
spring	MRS	-3.8	-3.0	3.8	3.0	4.6	3.9	-164	-104	164	104
	ZRH	-0.4	0.4	0.6	0.8	0.8	1.2	-57	37	85	74
summer	ZRH	-0.4	-0.2	0.4	0.3	0.6	0.5	-108	-28	120	75
autumn	BLQ	-4.0	1.0	4.0	2.9	5.1	3.5	-83	23	86	46
	SIRTA	-2.0	0.0	2.1	1.9	3.4	2.8	-70	22	103	76
	SPC	-1.0	4.1	2.9	5.3	4.3	6.5	35	104	115	118
	ZRH	-0.8	0.2	0.8	0.8	1.2	1.2	-74	18	94	67
winter	BLQ	-4.8	-1.6	4.8	3.2	6.0	4.2	-121	-29	121	59
	MRS	-3.6	-2.6	3.7	3.1	6.9	6.4	-118	-37	126	80
	SIRTA	-1.0	0.5	1.2	1.5	2.1	2.1	-46	46	100	85
	ZRH	-0.5	1.3	0.8	1.7	1.2	2.2	-21	71	82	96
OOA											
spring	MRS	-3.9	-3.5	3.9	3.5	3.9	3.6	-158	-135	158	135
	ZRH	-0.3	0.6	2.9	3.0	3.8	4.0	-15	6	69	64
	SMEARII	-0.2	0.0	0.4	0.4	0.6	0.6	-57	-23	81	65
summer	MSA	-1.1	-0.9	1.5	1.3	1.9	1.7	-62	-48	82	72
	ZRH	0.4	0.8	2.5	2.5	3.5	3.6	5	16	64	62

Season	Site	MB (µg m⁻³)		ME (µg m⁻³)		RMSE (µg m⁻³)		MFB (%)		MFE (%)	
		BASE	NEW	BASE	NEW	BASE	NEW	BASE	NEW	BASE	NEW
autumn	BLQ	-0.8	1.4	2.3	2.9	2.9	3.5	-2	33	49	54
	FKL	-1.3	-0.7	1.6	1.3	2.3	1.9	-62	-37	75	61
	MSA	-1.0	-0.5	1.3	1.1	1.7	1.5	-43	-23	61	49
	SIRTA	-3.2	-2.6	3.2	2.9	4.8	4.4	-97	-72	104	86
	SPC	1.1	3.2	2.9	4.0	3.3	4.6	54	82	84	93
	ZRH	1.5	2.4	3.0	3.4	4.2	4.9	17	31	58	60
winter	BLQ	-9.3	-8.5	9.3	8.5	10.4	9.8	-144	-126	144	126
	MRS	-2.5	-1.8	2.6	2.2	3.4	2.9	-71	-43	84	65
	SIRTA	-1.3	-1.3	1.4	1.3	2.0	2.0	-138	-127	142	133
	ZRH	-3.8	-3.1	4.2	3.8	5.8	5.4	-78	-55	98	86

3) The "NEW" simulation includes two main modifications, the first one concerning the VBS scheme and the second one related to the estimation of SVOC emissions. It would be very interesting introducing an intermediate simulation where only one the two modifications is implemented (e.g. only the modified VBS scheme either only the chance in SVOC emissions). This could help in better quantifying the contribution of every change to the total concentration variation. The results of such analysis could be introduced in Table 2 as well as in figure 2 and 3.

The intermediate simulation only changing the SVOC emissions (referred to as 3POA) can be seen in the Figure 1 below. The modelled land-average SOA from biomass burning in winter increased by 76% and 28% using NEW and 3POA compared to BASE, respectively. However, the influence of each modification varies depending on both location and time period. For the measurement sites in this study, NEW generally leads to a better model performance than 3POA, but the difference between 3POA and NEW is small (see Table 2 below). However, the difference is higher at locations where biomass burning is the dominant source. That is why we decided to present only BASE and NEW in Table 2 and Figure 2, 3 to avoid any misunderstanding such as further aging of biomass burning SOA is not important.
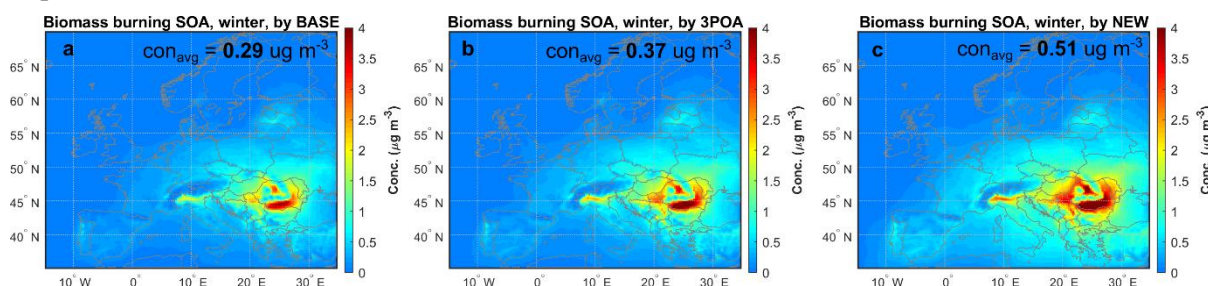
Figure 1 Comparison of modelled winter (December-January-February) SOA from biomass burning by BASE (a), 3POA (b) and NEW (c) parameterization. The con_{avg} values indicate the average concentration for the land.

Table 2 Statistical results for modelled winter OA and OOA by different OA scheme.

Site	MB (µg m⁻³)			ME (µg m⁻³)			RMSE (µg m⁻³)			MFB (%)			MFE (%)		
	BASE	3POA	NEW	BASE	3POA	NEW	BASE	3POA	NEW	BASE	3POA	NEW	BASE	3POA	NEW
OA															
BLQ	-21.4	-16.6	-16.2	21.4	16.8	16.4	23.4	19.3	19.0	-149	-101	-98	149	102	99
MHD	0.0	0.1	0.1	0.3	0.3	0.3	0.6	0.6	0.6	-4	7	7	91	89	89
MRS	-7.8	-5.8	-5.3	7.8	6.1	5.8	11.0	9.6	9.3	-109	-64	-55	111	72	65
MSA	0.1	0.4	0.5	0.6	0.8	0.9	0.9	1.1	1.3	-10	7	10	90	90	92
SIRTA	-3.6	-1.7	-1.7	3.6	2.8	2.8	5.5	4.4	4.4	-110	-31	-31	117	71	71
ZRH	-4.1	-0.9	-0.5	4.6	3.7	3.7	5.9	4.9	4.9	-68	-8	-3	82	56	55
OOA															
BLQ	-9.3	-8.9	-8.5	9.3	8.9	8.5	10.4	10.1	9.8	-144	-133	-126	144	133	126
MRS	-2.5	-2.2	-1.8	2.6	2.4	2.2	3.4	3.2	2.9	-71	-60	-43	84	76	65
SIRTA	-1.3	-1.3	-1.3	1.4	1.3	1.3	2.0	2.0	2.0	-138	-129	-127	142	134	133
ZRH	-3.8	-3.4	-3.1	4.2	4.0	3.8	5.8	5.6	5.4	-78	-64	-55	98	91	86

4) The analysis of the obtained results allows the authors to conclude that the "NEW" introduces an overall improvement of CAMx performance. I fully agree with their conclusion, however SOA

performance still highlights a general underestimation, differently from HOA and BBOA that are reproduced fairly well in "NEW" run. Could this result be specifically related to a possible underestimation of some key precursors such as IVOC? Any comment/additional analysis of this issue? Although we tried to improve the model performance for SOA by adding the SVOC emissions and aging of biomass burning SOA, there are still a number of possible reasons for the underestimation: It could be the underestimated IVOC/SVOC emissions as the referee mentioned, or missing pathways of SOA formation such as aqueous-phase chemistry or uncertainties of the VBS parameters (yield, reaction rate, *etc.*) due to wall loss, etc. We added a paragraph in Section 3.2 (P13 L18 – L31) as a summary of the possible reasons and future work.

"Although the model performance was substantially improved by the NEW parameterization, some limitations still remain to be further improved. The winter OOA is still underestimated at most sites. It could come from the missing pathways of SOA formation such as the aqueous processing of water-soluble organics (Ervens et al., 2011), which was found to contribute up to ~20% of winter OA measured in Bologna (Gilardoni et al., 2016; Meroni et al., 2017). The uncertainties in the model parameters (reaction rate, yield, etc.) could also account for part of the underestimation. A recent study reported that the reaction rate of SVOC with hydroxyl radicals is highly uncertain without strong constraints of the whole set of parameters for various processes, such as the vapor wall loss in chamber experiments (Bertrand et al., 2018). Another potential limitation is related to the uncertainties in SVOC/IVOC emissions. We adopted a factor of 3 for the SVOC and POA ratio for the whole domain, however, the substantial spatial and temporal variability of the factor could lead to an over- or underestimation of SVOC emissions at site scale (Denier van der Gon et al., 2015) and therefore over- or underestimation of SOA, as well as of POA. It could also partially explain the differences in model performance for the temporal variation of HOA and BBOA for each site. To further improve the model performance, it is necessary to continuously update the chemical mechanism in models by introducing the missing processes and improving the parameterization based on the advanced knowledge; as well as to improve the emissions by including more site-specific sources, IVOC and SVOC estimates, and updated diurnal variation profiles."

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Specific comments
P7 R15-16 Were coarse PM emissions split into EC, Na and SO4=? The default CAMx aerosol scheme (CF) includes only CCRS and CPRM species for the coarse fraction. In case, what aerosol scheme was used?
We split coarse PM emissions into coarse POA, EC, Na^+, PSO_4^{2-} and coarse primary particles (CPRM) based on the TNO profile. However, only CPRM was taken as input to CAMx (CF scheme). We modified the text to avoid any misunderstanding (P7 L28 – L30).

"The particulate matter emissions were split into POA, elemental carbon (EC), sodium (Na⁺), particle sulfate (PSO₄²⁻), and other primary particles in the fine (FPRM) and coarse (CPRM) size fractions according to the TNO PM-splitting profile."

P8 R5-8 SVOC emissions play a key role on OA processes both in terms of total mass as well as with respect to their volatility distribution. Did the authors introduce also a different volatility distribution in the "NEW" run, beyond increasing the total emissions by a factor of 3?

We totally agree with the referee that the volatility distribution is important for the issue of SVOC. For this study, we mainly focused on the total mass, and therefore the default volatility distribution was used for "NEW" run. However, we have already constrained the volatility distribution for SVOCs based on recent experimental studies (Stefenelli et al., 2019) and it will be implemented in CAMx soon as the next step.

Stefenelli, G., Jiang, J., Bertrand, A., Bruns, E. A., Pieber, S. M., Baltensperger, U., Marchand, N., Aksoyoglu, S., Prévôt, A. S. H., Slowik, J. G., and El Haddad, I.: Secondary organic aerosol formation from smoldering and flaming combustion of biomass: a box model parametrization based on volatility basis set, Atmos. Chem. Phys., 19, 11461–11484, https://doi.org/10.5194/acp-19-11461-2019, 2019.

P9 R22-24 Authors correctly point out that NO_2 is underestimated suggesting that the observed discrepancy could be related to a corresponding underestimation of NO_x emissions. As NO_x emissions are mostly related to road transport, could other emissions of the same sector be underestimated too? (e.g. NMVOC). Any possible influence on OM results?

One cannot generally assume that different components from the same sector are likely underestimated when this is shown for one of the components. Specifically for NMVOC and NO_x from traffic, the emission sources are due to different types of vehicles and different driving conditions. NO_x emissions are dominated by diesel vehicles during high load and high accelerations, while most NMVOC are due to gasoline emissions during cold start (Platt et al., 2017). At most sites, the contribution of traffic SOA is rather minor. However, we cannot rule out that especially under cold temperatures VOC emissions during cold start are especially high. Available ^{14}C measurements do not indicate large missing fossil organic carbon sources.

Platt, S. M., El Haddad, I., Pieber, S. M., Zardini, A. A., Suarez-Bertoa, R., Clairotte, M., Daellenbach, K. R., Huang, R. J., Slowik, J. G., Hellebust, S., Temime-Roussel, B., Marchand, N., de Gouw, J., Jimenez, J. L., Hayes, P. L., Robinson, A. L., Baltensperger, U., Astorga, C., and Prévôt, A. S. H.: Gasoline cars produce more carbonaceous particulate matter than modern filter-equipped diesel cars, Scientific Reports, 7, 4926, doi: 10.1038/s41598-017-03714-9, 2017.

P9 R24-30 CAMx performance in reproducing SO_2 are rather poor. Modelled concentrations are strongly overestimated (MFB is higher than 75% in February) and substantially uncorrelated to observed values (IOA is around 0.1). Considering that SO_2 emissions are mostly related to "Other anthropogenic sources" do authors think that such overestimation could influence also other species (e.g. NMVOC, PM) and, therefore, also the contribution of this sector to OM concentrations? (see for example conclusions P16 R5-9)

As we mentioned in P10 L10, the overestimation of SO_2 is most likely because we distributed all the emissions into the first layer. Unlike NO_x, PM and NMVOC, which are dominated by the area sources (road traffic for NO_x, biomass burning for PM, and solvent use for NMVOC), the major sources of SO_2 are point sources such as power plants (SNAP1) and industrial combustion (SNAP3 and 4) with stack emissions (Bieser et al., 2011). That is why a significant overestimation was only found for SO_2. For the organic aerosols, as most of the precursors and POA are from area emissions, the influence of the vertical distribution of emissions is small.

Bieser, J., Aulinger, A., Matthias, V., Quante, M., and Denier van der Gon, H. A. C.: Vertical emission profiles for Europe based on plume rise calculations, Environ. Pollut., 159, 2935-2946, doi: https://doi.org/10.1016/j.envpol.2011.04.030, 2011.

P10 R1-2 Table 2 is a bit misleading because it summarizes the model performance at all ACSM/AMS stations, which cover very different temporal periods. The different rows should be grouped somehow, for example separating stations covering the whole year from sites covering only winter periods,

We agree with the referee that showing seasonal differences of the statistical results are important, but grouping the stations is very complicated as some sites covering more than one season while some only cover a short period in a specific season. To avoid any confusion, we added a column in Table 2 showing the observation period and number of available observations. The seasonal-based statistical results were also added in Table S5 following the general comment (2).

Technical corrections P19 R18 Table 1?

Corrected (should be on P11 R18?). One more sentence was added in P11 L31-L32 to make it more clear.

"The modelled OA components were evaluated using AMS/ACSM measurements analyzed with PMF at different stations (Table 1). The statistical results are presented in Table 2."