Answers to the comments of Anonymous Referee #1

We would like to thank the referee for the detailed comments (marked blue in the following text).

1) Section 2.3: The authors should present a table in which all the emitted and simulated PM species will be presented in detail for each model. The existing Table 2 cannot explain how sea salt is speciated in model simulations (is it simulated explicitly as Na and Cl?). Similar is the comment for fire PM species. Usually, fire PM emissions are simulated as OC and EC emissions and I would expect these emissions to have a contribution to OC and EC levels. How is this issue addressed in the different models? If fire PM emissions were speciated as OC and EC then how was it possible to distinguish between anthropogenic OC/EC and fire-related OC/EC? The models simulate OC or OA? Also in Table 2, the PM size is not presented (PM10 versus PM2.5).

- LOTOS-EUROS and CMAQ calculate the sea salt components separately, in SILAM and EMEP sea salt is not speciated and is emitted and transported as whole. Standard sea salt composition is assumed when comparing the total sea salt concentration with Na observations with 30.8% sodium content by dry mass.

- The fire PM originated from IS4FIRES (Sofiev et al., 2009), which provides unspeciated PM_{10} and $PM_{2.5}$ emissions. In SILAM, EMEP and LOTOS-EUROS the emitted PM was transported as a separate field of unspeciated particulates, in CMAQ the fine fraction was included in primary OA and the coarse fraction in coarse primary PM. The fire OA in CMAQ cannot be distinguished from the anthropogenic OA and fire EC was not included in that model. In the other models the fire PM can be further speciated as post-processing following Akagi et al., (2011) or Andreae and Merlet, (2001). On average these papers suggest roughly 5% EC and 50% OC content for fire emitted aerosol, the rest mainly consisting of non-carbon atoms in the organic compounds and some inorganics (up to 5%).

- CMAQ provided total organic aerosol mass; EMEP model calculates both OC and OA.

Table 2 in the paper has been changed to include the speciation and sizes of the aerosol components computed by each model. Clarifications have been added to Section 2.5 (Model measurement comparison). Wild-land fire emitted contributions to OC and EC have been added to comparisons and discussion of them has been extended.

2) Section 2.4: Please present in Table 3 the size of the PM measured (PM10 versus PM2.5).

The inorganic species were measured mostly in total aerosol without size limits; concentration in PM_{10} was used where available. In 2005 EC and OC were measured in both $PM_{2.5}$ and PM_{10}. The 2002-2003 campaign observed EC and OC in PM_{10}.

Table 3 has been updated to include this information.

3) Section 2.5: The method for the estimation of nss-Ca levels from the model results is not clear. Are the simulated nss-Ca concentrations estimated as the sum of the 10% of dust concentrations plus 3.5% of the unspeciated other primary PM concentrations? The authors should also make clear in the whole manuscript (e.g. in Table 6) that it is not the mineral dust model results that are evaluated but the nss-Ca values (including both the desert dust and the anthropogenic contribution).

The simulated nss-Ca concentrations are indeed estimated as the sum of the 10% of dust concentrations plus 3.5% of the unspeciated other primary PM concentrations. *Clarifications were added.*

4) Section 3.1, page 9, lines 7-18: The authors explained the bad model performance in Schauinsland Mountain station as the result of model spatial resolution and high station altitude. The position and altitude of the stations should be provided in the supplement. Are there other stations of similar altitude in which the models performance is not as bad in the Schauinsland Mountain station?

There are other high altitude stations, located in Alps and in Spain. The Schauinsland station was pointed out in the paper, because it was the only station where the models consistently overestimated the PM concentration. Also the temporal correlations between the models and observations are the lowest there for both PM_{10} and $PM_{2.5}$. The models' performance does degrade also in the other high stations - there is a strong negative correlation between the station altitude and the models' temporal correlation coefficients for both PM_{10} and $PM_{2.5}$. However, the bad model performance is caused not only by the altitude difference between the station and the model grid cell average, but also other inhomogeneities, such as strong emission sources in the area. The Schauinsland station, for instance, is located about 10 km from Freiburg city, and about 1 km above it. In the models both the city and the station are covered with one uniformly mixed grid cell, while in reality the winter time low boundary layer traps the pollution below the station altitude. Indications of wintertime overestimation are also visible for some of the other high stations, though with smaller magnitude, as not all the high stations are located at extreme points of the terrain, such as mountain summits, and not all of them have strong emission sources in the immediate vicinity. Opposite problems arise for sites located in narrow valleys, where the models cell-mean altitude is higher than the station and the models overspread the pollution that in reality can be trapped in the valley.

The stations' locations have been be added to the supplement. The discussion about the station representativity has been extended.

5) Section 3.1, page 10, lines 2-4: Explain the reasons for models' different ability in simulating PM2.5 and PM10 in summer and winter.

For most of the models, temporal correlations and factor-two agreements are better in winter than in summer for both PM fractions, and the spatial correlation of $PM_{2.5}$ is also better in winter. The only score with opposite behaviour is the spatial correlation for PM_{10}. The summertime worse scores are probably due to the highly uncertain components that dominate the summer aerosol - wind-blown dust, wild-land fires, and biogenic secondary organic aerosols. In summer the PM_{10} pattern over Europe is formed by the inflow of Saharan dust and wild-land fires in Portugal and Spain creating a strong south to north gradient. This gradient is reproduced by the models, although with smaller magnitude (Figure 1, lower right panel). As LOTOS-EUROS misses the dust and fire contribution, it does not reproduce this pattern. As the species contributing to this summer time south-north gradient are desert dust and wild-land fires, which by nature are episodic and hard to model, the temporal correlation and factor-two agreement are still generally lower and bias is larger in summer. In winter the particulate matter is dominated by the anthropogenic emissions, forming a more complex pattern, and thus the spatial correlation is worse.

Explanations have been added to the section.

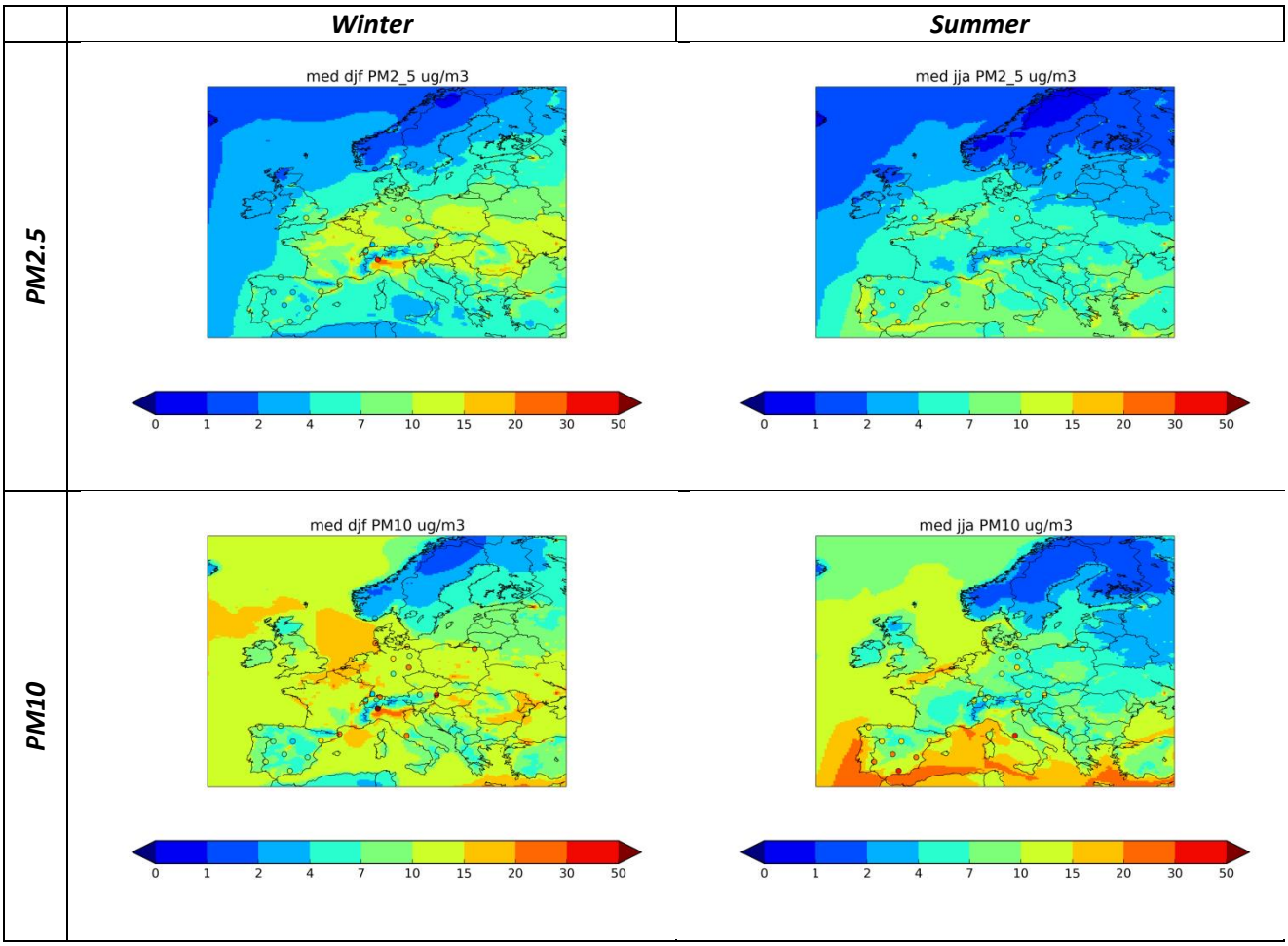

Figure 1 Seasonal average observed and ensemble median PM concentrations for winter and summer.

6) Section 3.1, page 10, line 11: The authors state: "The medianComp fully includes SOA, desert dust and fire-induced PM." This is not true since according to Table 2, SOA are not included in LOTOS-EUROS and in SILAM. Also BCs dust is not included in LOTOS-EUROS.

MedianComp is the sum of the ensemble medians of all the individual PM components. Although not all components are provided by all four models, when computing the median field for every component only those models are used which provided a valid field for that component. Thus, medianComp will include the median SOA of EMEP and CMAQ and median dust of SILAM and EMEP. As these models have provided valid fields for these components, the medianComp PM also includes valid fields of SOA and dust.

The description of MedianComp model has been extended

7) Section 3.1, page 10, lines 16-18: The authors explain that the differences between median and medianComp are due to desert dust. However, according to Table 2, it is only the fire originated PM that were not included in total PM while BCs dust was included all CMAQ, EMEP and SILAM.

That is correct. However, in CMAQ the dust and fire contributions are very low and LOTOS-EUROS does not have them at all, so the median total PM is based on half of the models with zero or very low dust concentration. MedianComp is based only on the valid dust fields of SILAM and EMEP and thus includes noticeably higher dust contribution.

Explanations have been added.

8) Section 3.2, Table 6: Which are the size bins of the PM species validated (comment mostly for sea salt and mineral dust). The words "mineral dust" should be replaces with "nss-Ca".

- The Ca and Na observations in EMEP network are made mostly in whole aerosol without size limits.

- Dust in the EMAC model is emitted in two log-normal modes with the size distribution parameters from (Dentener et al., 2006): accumulation mode with 0.42 µm median diameter and standard deviation 1.59, and coarse mode with 1.30 µm median diameter and standard deviation 2.00. These modes are selected to cover the whole dust distribution relevant for atmospheric transport (Ginoux et al., 2004, 2001). When the EMAC boundary conditions are projected to the model size bins in EMEP and SILAM, both of the size modes are assumed to be fully inside PM_{10}, which agrees reasonably well with the observations of (Dubovik et al., 2002). The sum of these two modelled modes is used when comparing with the nss-Ca observations.

- For all models the Na concentration for model-measurement comparison was computed from sea salt in PM_{10}. As the models already overestimate Na concentration, if taking into account also the particles larger than 10 µm the real overestimation would be even larger than what is shown. However, comparing Na in PM_{10} with Na in whole sea salt in SILAM (size range 0.01 to 30 µm), the changes are minor for majority of the EMEP stations that observed Na in 2005: below 5% for 65% of the stations, below 10% for 77%, and below 20% for all stations. The concentration changed more than 10% only in the stations located directly at seaside.

Table 3 has been updated to include the aerosol sizes for the observations. Reference to the dust size distribution has been added to EMAC model description. Explanations have been added to the results section about the sea salt size distributions.

Why evaluation is presented explicitly only for SO2 and not for other PM precursor gas compounds like NH3 or NOx?

- Observations of NH_3 and HNO_3 are more prone to artefacts(e.g. Chang et al., 2002; Schaap et al., 2011), thus the less uncertain NH_3+NH_4 and HNO_3+NO_3 observations were originally chosen for model evaluation. Those were also available from larger number of stations. The contribution of HNO_3 and NH_3 to the sum is shown on **Figure 2** with dark shading. On annual average level the gas phase fraction is in both cases relatively well reproduced by most of the models, only SILAM underestimates HNO_3 and overestimates NH_3 contributions. However, the models do not reproduce well the seasonal variations in HNO_3 and NH_3 concentrations – EMEP and LOTOS-EUROS overestimate the seasonal variability of HNO3, CMAQ strongly overestimates the autumn NH3 concentrations and so does with smaller magnitude also SILAM. Temporal correlation coefficients and factor-2 agreements are noticeably worse for NH_3 and HNO_3 compared with NH_4 and NO_3 aerosols or the gas-aerosol sums. In addition to the uncertainties in the observations, there are other reasons for model errors in these species. Especially for NH_3 the timing of the emissions as used in the models (fixed temporal profiles) can deviate substantially from real world emission timing which is largely controlled by meteorology (Backes et al., 2016; Hamaoui-Laguel et al., 2014; Hendriks et al., 2016). Meteorology also influences the total amount of emitted NH_3 but the strongest influence is on the timing. The timing of agricultural activities, such as manure spreading has also a strong impact on the emissions (Hendriks et al., 2016). This mismatch may translate into episodes where the models predict high concentrations but due to unfavorable meteorology very little was emitted and vice versa, partly explaining why the models do

not properly reproduce the seasonal fluctuations but on annual average are quite OK. This is something that clearly needs to be improved on the model input side.

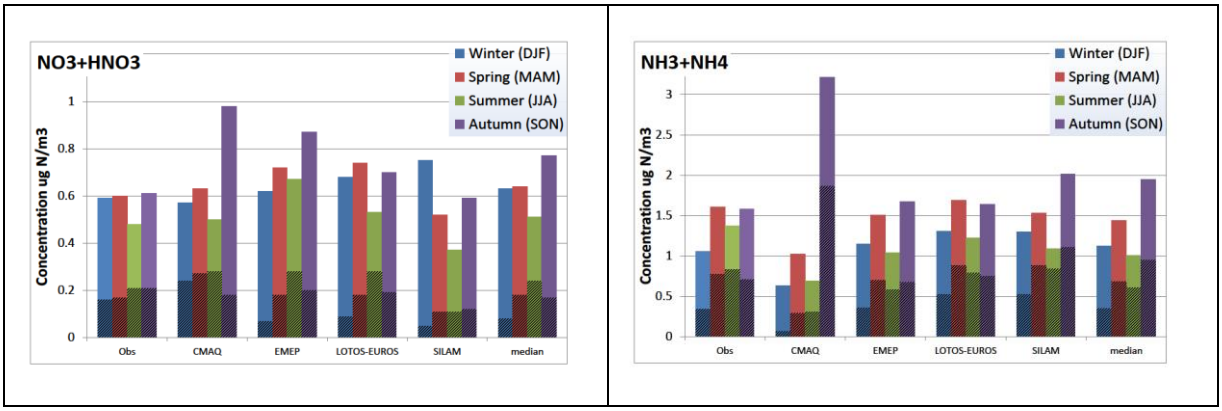

Figure 2 Observed and predicted seasonal concentrations of HNO_3+NO_3 and NH_3+NH_4, mean over the EMEP stations [µg N m^{-3}]. Shaded part shows the concentration of the gas phase species HNO_3 and NH_3. Only when at least two of the gas, aerosol and their sum were observed simultaneously so that the gas phase fraction could be estimated, the data point was included in the averaging.

Regarding NOx, NO was measured by only one EMEP station and thus no thorough evaluation is possible. The model scores and seasonal developments of NO_2 concentration are shown on Table 1 and Figure 3. The seasonal variations of NO_2 are well reproduced, but all models apart from CMAQ overestimate NO_2 all the seasons. This can be one of the reasons for the overestimation of the sum of NO_3^- and HNO_3 seen on **Figure 2**, left panel.

Table 1 Model skill scores for the gas phase precursors of NO_3 and NH_4, average over the EMEP stations that observed these species.

Species	Model	Scaled bias	tCor	Fac2
NH_3	*CMAQ*	0.04	0.18	0.25
Ave obs:	*EMEP*	-0.07	0.30	0.36
0.75 µg N/m^3	*LOTOS-EUROS*	0.19	0.22	0.38
	SILAM	0.32	0.30	0.40
	median	0.05	0.31	0.39
HNO_3	*CMAQ*	0.21	0.34	0.43
Ave obs:	*EMEP*	-0.11	0.38	0.39
0.19 µg N/m^3	*LOTOS-EUROS*	0.00	0.38	0.40
	SILAM	-0.53	0.32	0.32
	median	-0.16	0.41	0.44
NO_2	*CMAQ*	-0.06	0.57	0.59
Ave obs:	*EMEP*	0.19	0.52	0.60
1.76 µg N/m^3	*LOTOS-EUROS*	0.26	0.45	0.58
	SILAM	0.39	0.54	0.57
	median	0.16	0.56	0.61

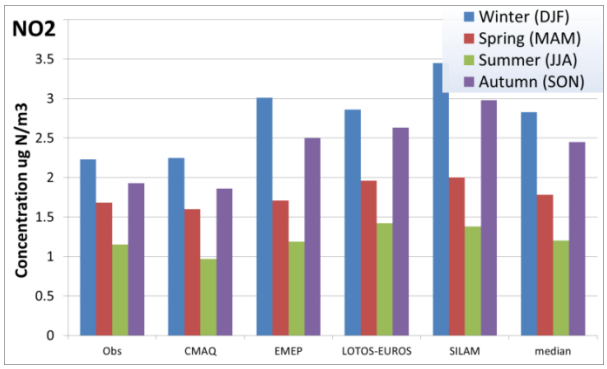

Figure 3 Observed and predicted seasonal concentrations of NO_2

NH_3 and HNO_3 have been added to Figure 4 of the manuscript, model scores for those species have been added to Table 7. Section on the comparison results has been extended. A figure with seasonal variations of NO_2 and a table with model scores for it have been added to the supplementary material.

9) Section 3.2.3: The performance of CMAQ, can it be explained by the fact that fire emissions are included in other PM?

CMAQ has the largest negative bias and lowest correlation for the carbonaceous aerosols. For EC EMEP is as biased as CMAQ, while being closer to the observations for OC. In CMAQ the fine mode fire emissions were actually included in the primary OA, although not to EC; only the coarse mode fire PM was included in the unspeciated coarse primary PM. For all other models the fire contribution was excluded from comparison with EC and OC. Thus, the missing fire contribution cannot explain the bias in CMAQ and other explanations are needed for the negative bias found for the carbonaceous aerosols.

The fire PM concentrations modelled by EMEP, LOTOS-EUROS and SILAM can be speciated as post-processing following Akagi et al., (2011) or Andreae and Merlet, (2001). On average these papers suggest roughly 5% EC and 50% OC content for fire emitted aerosol. The fire contribution to EC and OC calculated following this composition is shown on **Figure 4** with darker shading. SILAM only shows a small fire contribution to EC in spring and summer, while in EMEP and LOTOS-EUROS the contribution is larger and visible all year round. EMEP also predicts a noticeable fire contribution to OC for all seasons. For EMEP and LOTOS-EUROS, the fire contribution reduces the model bias for the carbonaceous species, while at the same time reducing the correlation with the measurements (Table 2). The SILAM EC prediction quality does not noticeably change.

Table 2 has been corrected to show that the fine particles from fires were included in CMAQ as OA. Fire contributions have been added to Figure 6 in the paper. For CMAQ the fire emitted OC is mixed with the primary anthropogenic part and cannot be shown separately and the fire emissions are excluded for EC. The section has been updated so that the carbonaceous aerosols of the other models include the fire contribution.

Table 2 Comparison of model scores for the carbonaceous species with and without the wildfire contribution

Species	Model	*Without firePM* Scaled bias	tCor	Fac2	*With firePM* Scaled bias	tCor	Fac2
EC in $PM_{2.5}$	CMAQ	-0.61	0.51	0.35	-0.61	0.51	0.35
Ave obs:	EMEP	-0.60	0.56	0.43	-0.56	0.53	0.4
1.08 µg C/m^3	LOTOS-EUROS	-0.42	0.58	0.45	-0.34	0.51	0.44
	SILAM	-0.17	0.61	0.41	-0.17	0.61	0.4
	median	-0.51	0.61	0.37	-0.45	0.6	0.38
OC in $PM_{2.5}$	CMAQ	-0.80	0.52	0.26	-0.80	0.52	0.26
Ave obs:	EMEP	-0.38	0.58	0.64	-0.25	0.54	0.6
3.61 µg C/m^3	median	-0.59	0.60	0.58	-0.52	0.54	0.61
EC in PM_{10}	CMAQ	-0.69	0.42	0.32	-0.69	0.42	0.32
Ave obs:	EMEP	-0.70	0.43	0.37	-0.66	0.46	0.35
1.32 µg C/m^3	LOTOS-EUROS	-0.53	0.43	0.45	-0.48	0.39	0.44
	SILAM	-0.36	0.43	0.37	-0.35	0.45	0.38
	median	-0.61	0.46	0.38	-0.58	0.49	0.37
OC in PM_{10}	CMAQ	-0.85	0.36	0.18	-0.85	0.36	0.18
Ave obs:	EMEP	-0.51	0.38	0.52	-0.37	0.46	0.52
4.78 µg C/m^3	median	-0.67	0.40	0.45	-0.61	0.46	0.48

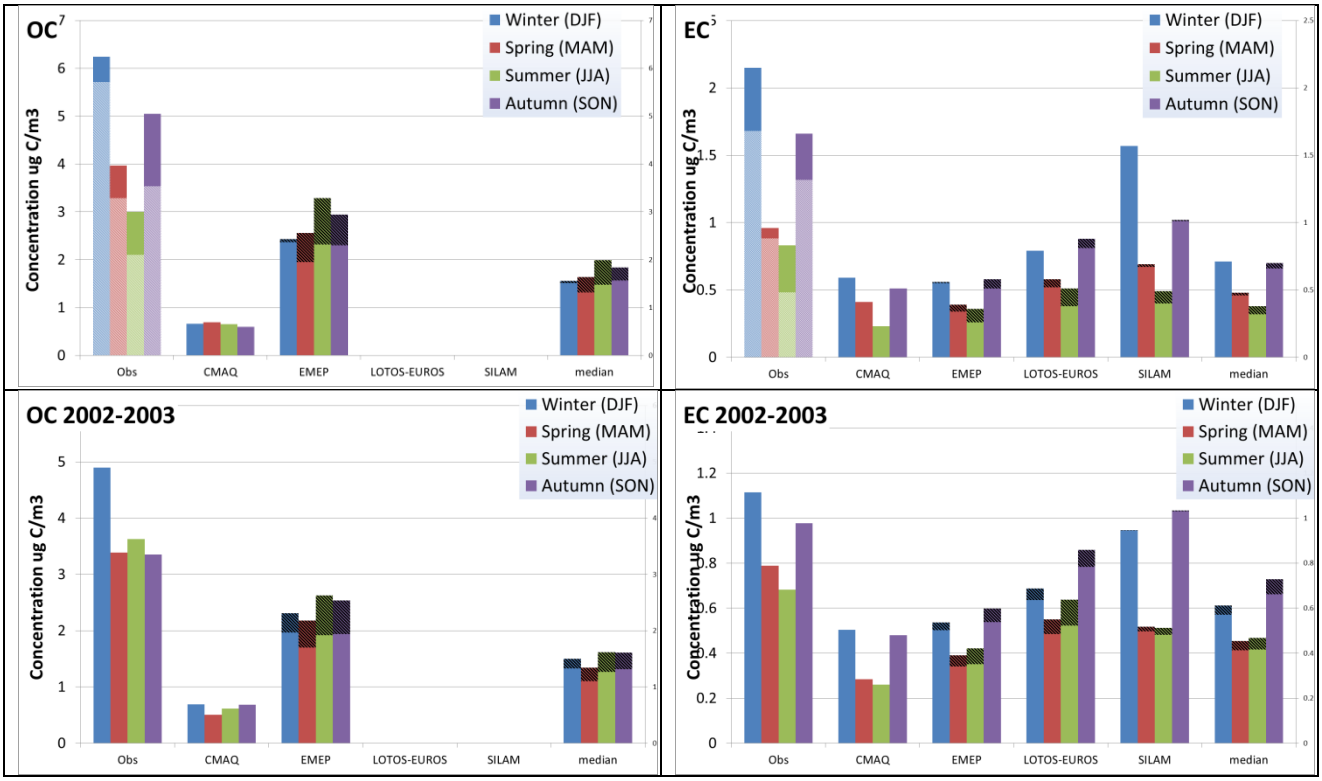

Figure 4 Observed and predicted seasonal concentrations of carbonaceous aerosols, mean over the EMEP stations [µg m-3]. The panels on the left-hand and right-hand sides represent OC and EC, respectively. The upper row: 2005, data from 4 stations, for the observations the lighter shading marks the concentration in $PM_{2.5}$, whole column the concentration in PM10; the lower row: EMEP 2002-2003 campaign, observations of OC and EC in PM_{10}. Dark shaded part shows the contribution from wild land fires (not separated for CMAQ).

Note: One station in Portugal (Braganca, PTR0001R) was excluded from the comparison with the 2002-2003 OC and EC due extremely high wild land fire contribution in the models, making that station not representative of average conditions – 2005 and 2003 were both record high wild land fire years in Portugal, but closer to average in rest of Europe. However, the 2002-2003 EMEP measurement period ends in the beginning of July 2003 and thus does not cover the 2003 Portuguese fires, which mostly took place in August.

10) Section 3.2.5: Any comments on OA comparison with observations should be based on CMAQ and EMEP results since the other models include OC in other PM. Similar remark for dust straightforwardly included only in EMEP and SILAM.

The text has been changed according to the reviewer's suggestion

11) Section 3.2.5., page 15, line 7: Correct OA with OC.

Done

12) Section 3.2.5., page 15, line 11: Please add that also the OC is for some models included in primary aerosols (as the case for LOTOS-EUROS and SILAM)

Done

13) Section 3.2.5., Figure 8: a) suggestion: add the simulated SOA and POA and compare them with the observed TOA, b) why fire PM are presented separately and not speciated in OC and EC so as to allow better comparison with observations? (see also comment 1). In the legend of Figure 8 the PM species are not presented in the same order as they appear in the plots.

a) As total OC is measured the observations on Figure 8 are shown as total OA. The modelled OA has been shown as POA and SOA separately, to demonstrate how the contribution of these varies between the models and the stations.

b) Apart from CMAQ that included the fire emissions in OA, the fire emissions were modelled as unspeciated particulates. The concentration can be speciated following the average composition from Akagi et al., (2011) and Andreae and Merlet, (2001), giving ~90% OM, ~5% EC and ~5% non-carbonaceous compounds.

Figure 8 has been updated to show the fire contribution to OC and EC. The legend has been fixed.

14) Section 4.1: This section provides a theoretical description of the possible reasons for overestimations or underestimations in the model results. However, there is no quantification of the uncertainties. For example, how much is the uncertainty introduced in the model validation because of the assumption that dust and anthropogenic mineral aerosols have a 10% and 3.5 Ca content respectively? Clarifications also are necessary in the last paragraph of this section in which the authors discuss about the absence of fire emissions from the computations while in Table 2 fire originated PM are presented as included in the simulations of all models.

As the models did not include any other dust sources than the inflow from the boundaries, underestimation of dust concentration is expected and comparison with nss-Ca confirms it, while not providing its exact magnitude. Various sources give Saharan dust Ca content from <5% to >15% and for the anthropogenic emissions the variations are even larger, depending on the source sector and local soil, so the uncertainty in aerosol Ca content can be expected to be a few times. However, with the 10 and 3.5% Ca content, the EMEP model underestimated the nss-Ca by 75% and SILAM by 58%, so even assuming twice the calcium content, the nss-calcium concentrations would still be underestimated by the models. This uncertainty does not influence the accuracy of the predictions and evaluation of PM_{10} and $PM_{2.5,}$ as primary PM and dust were modelled as totals and the 3.5% and 10% factors were only applied when comparing with the nss-Ca observations.

In EMEP and LOTOS-EUROS, using the IS4FIRES v1 emissions resulted in degradation of model scores for $PM_{2.5}$ and PM_{10} and thus these models excluded the fire PM from their total $PM_{2.5}$ and PM_{10} fields, while still providing it as a separate field. In SILAM, a newer version of the emission data was used (IS4FIRES v2, (Soares et al., 2015), together with dynamic emission vertical profiles of (Sofiev et al., 2012), while in other models, the IS4FIRES v1 emission data was spread evenly to the first 1000m. Mainly due to the vertical profiles that release most of the smoke high aloft, the ground level concentrations of fire PM were substantially lower in SILAM and thus the fire PM does not significantly affect the model performance for PM, demonstrating that the quality of the fire emission data is essential for simulating the particulate matter concentrations.

Clarifications have been added.

15) Section 4.3: It is a very interesting section. The authors should present results on the models' improved performance when water is accounted for (even in the four stations providing a complete set of PM measurements; after all section 3.2.5 was based on the measurements in these few stations).

Based on the dry PM mass and speciation provided by the models, aerosol thermodynamic model ISORROPIA2 (Fountoukis and Nenes, 2007) was applied to evaluate the equilibrium water content at the conditions where the filters were weighted (20°C, 50% relative humidity). ISORROPIA2 was run in the reverse mode, where the input quantities were the soluble inorganic components (SIA, sea salt, Ca) in the aerosol phase. The aerosols were assumed to be internally mixed. Both stable and metastable states were computed, corresponding to the lower and upper branches of the deliquescence hysteresis loop, the latter one describing the case when the aerosol has been exposed to more humid environment and crystallization has not occurred. In this way lower and upper limits of the aerosol bound water amount can be estimated.

ISORROPIA2 was applied to estimate the water contribution at all the EMEP sites that measured $PM_{2.5}$ or PM_{10}. In the stable case, the annual mean $PM_{2.5}$ water content, average over all EMEP stations, stayed between 4 and 9% depending on the model, and between 11 and 17% for PM_{10}. For $PM_{2.5}$, the models predicted annual average water content above 10% for only a few stations. For PM_{10}, CMAQ and EMEP predict majority of the stations to have less than 10% of water content, while LOTOS-EUROS and SILAM predict the majority to be between 10 and 20%; annual average water contents of more than 25% were predicted for some stations. The water content of PM_{10} computed in the metastable mode was on average about twice higher (~25%); ~20% water content was predicted for $PM_{2.5}$.

As seen from Table 3, adding the aerosol-bound water reduces noticeably the model bias for both PM_{10} and $PM_{2.5}$. For $PM_{2.5}$ the correlation coefficients are not much affected, while for PM_{10}, they are noticeably reduced. The factor-2 agreements improve due to the bias reduction. The worsening correlations could be related to the models overestimating the sea salt concentrations that can lead to overestimation of the water content in PM_{10}, as it is the most hydrophilic of the considered aerosol components.

Also other uncertainties exist estimating of the PM water content. Firstly, ISORROPIA2 computes the water content based on the inorganic part of aerosol – SIA, sea salt, calcium; it does not take into account the water related to the hydrophilic part of the organic aerosol. Secondly, the aerosols were assumed fully internally mixed, which lowers the deliquescence humidity compared to external mixtures and might lead to overestimation of water uptake.

The water contribution for the four stations with most complete aerosol composition observations can be seen on Figure 5. Adding the aerosol bound water in metastable state closes the gap between the observed total PM_{10} and the sum of the individual components in all stations (in Montseny the PM_{10} estimate based on nearby stations can be inaccurate). In Ispra and Birkenes the observed PM is exceeded, which could indicate that the aerosol on the filters is in crystallized state or be due to inaccuracies in other observed species. In Ispra, errors were suspected in the observations of the carbonaceous components, while in Birkenes, the sea salt observations are taken from all aerosol, not PM_{10}. The models predict very high aerosol water content for Birkenes and noticeably overestimate the PM_{10} there, as the overestimated sea salt concentration leads to very high water uptake of the aerosol.

The water contribution has been added to figures 3 and 8 and Table 6 has been divided to two parts, the first of those comparing the model scores for the dry PM to the water equilibrated PM. The discussion regarding the aerosol-bound water has been extended according to the reviewers suggestion.

Table 3 Model skill scores for dry PM and PM equilibrated at the filter weighting conditions.

		Dry			***50% relative humidity, 20° C, stable***			***50% relative humidity, 20° C, metastable***		
Species	**Model**	**Scaled bias**	**tCor**	**Fac2**	**Scaled bias**	**tCor**	**Fac2**	**Scaled bias**	**tCor**	**Fac2**
$PM_{2.5}$ *Ave obs: 11.78 µg/m³*	*CMAQ*	-0.47	0.50	0.47	-0.44	0.50	0.50	-0.34	0.49	0.58
	EMEP	-0.33	0.62	0.69	-0.30	0.62	0.71	-0.17	0.62	0.77
	LOTOS-EUROS	-0.40	0.46	0.51	-0.34	0.43	0.54	-0.26	0.45	0.58
	SILAM	-0.26	0.59	0.58	-0.18	0.57	0.61	-0.08	0.57	0.64
	median	-0.38	0.63	0.61	-0.35	0.63	0.63	-0.26	0.63	0.70
	medianComp	-0.30	0.60	0.62	-0.28	0.60	0.64	-0.17	0.60	0.71
PM_{10} *Ave obs: 17.09 µg/m³*	*CMAQ*	-0.49	0.46	0.49	-0.40	0.42	0.53	-0.29	0.41	0.59
	EMEP	-0.31	0.57	0.69	-0.21	0.51	0.70	-0.09	0.51	0.72
	LOTOS-EUROS	-0.44	0.40	0.53	-0.32	0.29	0.57	-0.25	0.32	0.61
	SILAM	-0.34	0.54	0.54	-0.24	0.50	0.58	-0.16	0.51	0.60
	median	-0.41	0.59	0.59	-0.33	0.53	0.63	-0.23	0.54	0.68
	medianComp	-0.35	0.57	0.63	-0.26	0.53	0.66	-0.17	0.54	0.70

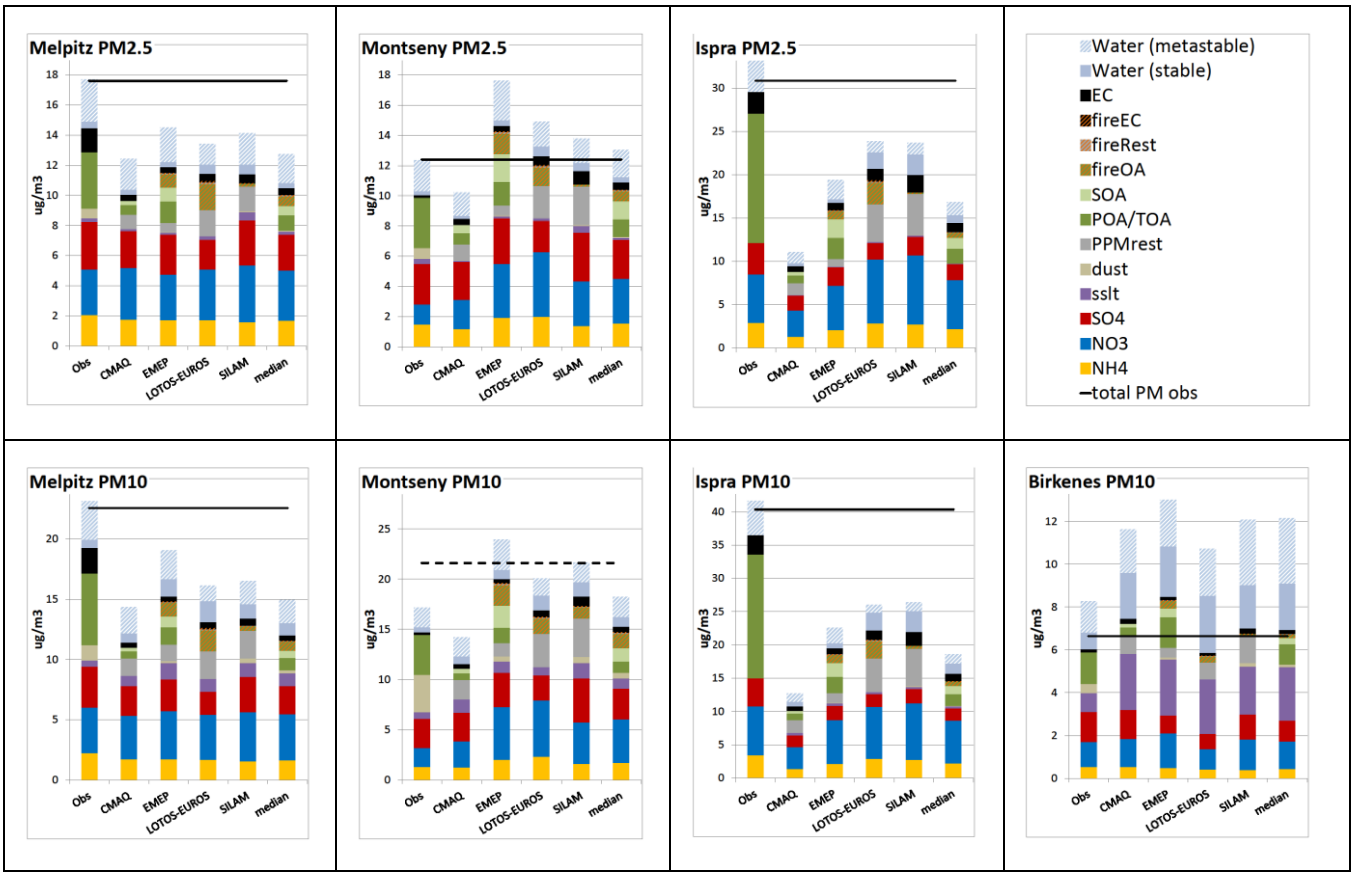

Figure 5 Aerosol chemical composition measured and modelled at four stations. Upper row – PM2.5, lower row – PM10

Water – aerosol water content at 50%RH and 20°C, computed with ISORROPIA2 based on observed or modelled aerosol composition.
metastable – particle is assumed to be in supersaturated liquid state if the relative humidity is below its deliquescence point
stable – particle is assumed solid if the relative humidity is below its deliquescence point
* Na observations were not available there in Ispra and were excluded from ISORROPIA input.
EC – elemental carbon from anthropogenic emissions
fireEC - elemental carbon from wild-land fire emissions, 5% of fire emitted PM
fireRest – mineral PM from wild-land fire emissions, 5% of fire emitted PM
fireOA– organic aerosol from wild-land fire emissions, 90% of fire emitted PM
SOA – secondary organic aerosol
POA/TOA – the primary part of organic aerosol for the models, total organic aerosol for the observations (OC * 1.6)
PPMrest – the unspeciated part of the modelled primary anthropogenic PM
Dust – modelled desert dust, observed non-sea-salt Ca^{2+} x 10
Sslt – sea-salt, observed $Na^{+} + Cl^{-}$
SO4, NO3, NH4 – secondary inorganic aerosols
Total PM obs – observed total $PM_{2.5}$ and PM_{10}.
* PM_{10} observations were not available for Montseny station. The dotted line marks an estimate calculated by averaging PM_{10} observations from the nearest EMEP stations (ES0010R, ES0014R).

16) Figure 6: Correct in the lower-right plot the OC to EC.

Done

17) Figure S6: Was CMAQ excluded from the average since dust in CMAQ was included in other PM?

Yes, the figures are based only on the models that included the shown component explicitly.

The figure captions have been updated to include this information.

18) Figure S7: Was LOTOS-EUROS and SILAM excluded from OA mean values since OA in these modes were included in other PM? The Figure is not commented in the text of the manuscript.

The OA figure is now based on EMEP model only, as SILAM and LOTOS-EUROS included primary OA in PM and CMAQ included in its OA the fire emissions.

The figure caption has been updated to include this information.

19) Figure S8: Please check comment 1. How fire emissions were simulated and consequently presented in the Figure for PM? If speciated to OC and EC emissions, how fire-related OC and EC concentrations were distinguished from anthropogenic OC and EC concentrations?

CMAQ emitted OC, which cannot be distinguished from the anthropogenic OC. EMEP, LOTOS-EUROS and SILAM emitted unspeciated $PM_{2.5}$ and PM_{10} and transported them as separate fields. Figure S8 shows the median of those fields.

The caption of Figure S8 has been updated

References

Akagi, S.K., Yokelson, R.J., Wiedinmyer, C., Alvarado, M.J., Reid, J.S., Karl, T., Crounse, J.D., Wennberg, P.O., 2011. Emission factors for open and domestic biomass burning for use in atmospheric models. Atmos. Chem. Phys. Discuss. 11, 4039–4072. doi:10.5194/acp-11-4039-2011

Andreae, M.O., Merlet, P., 2001. Emission of trace gases and aerosols from biomass burning. Global Biogeochem. Cycles 15, 955–966.

Backes, A.M., Aulinger, A., Bieser, J., Matthias, V., Quante, M., 2016. Ammonia emissions in Europe, part II: How ammonia emission abatement strategies affect secondary aerosols. Atmos. Environ. 126, 153–161. doi:10.1016/j.atmosenv.2015.11.039

Chang, K., Lu, C., Bai, H., Fang, G.C., 2002. A theoretical evaluation on the HNO3 artifact of the annular denuder system due to evaporation and diffusional deposition of NH4NO3-containing aerosols. Atmos. Environ. 36, 4357–4366. doi:10.1016/S1352-2310(02)00352-7

Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S., Hoelzemann, J.J., Ito, A., Marelli, L., Penner, J.E., Putaud, J.-P., Textor, C., Schultz, M.G., van der Werf, G.R., Wilson, J., 2006. Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom. Atmos. Chem. Phys. 6, 4321–4344.

Dubovik, O., Holben, B., Eck, T.F., Smirnov, A., Kaufman, Y.J., King, M.D., Tanré, D., Slutsker, I., 2002. Variability of Absorption and Optical Properties of Key Aerosol Types Observed in Worldwide Locations. J. Atmos. Sci. 59, 590–608. doi:10.1175/1520-0469(2002)059<0590:VOAAOP>2.0.CO;2

Fountoukis, C., Nenes, A., 2007. ISORROPIA II : a computationally efficient thermodynamic equilibrium model for K+-Ca2+-Mg2+-NH4+-Na+-SO42--NO3--Cl--H2O aerosols. Atmos. Chem. Phys. 7, 4639–4659.

Ginoux, P., Chin, M., Tegen, I., Prospero, J.M., Holben, B., Dubovik, O., Lin, S.-J., 2001. Sources and distributions of dust aerosols simulated with the GOCART model. J. Geophys. Res. 106, 20255. doi:10.1029/2000JD000053

Ginoux, P., Prospero, J.M., Torres, O., Chin, M., 2004. Long-term simulation of global dust distribution with the GOCART model: Correlation with North Atlantic Oscillation. Environ. Model. Softw. 19, 113–128. doi:10.1016/S1364-8152(03)00114-2

Hamaoui-Laguel, L., Meleux, F., Beekmann, M., Bessagnet, B., Génermont, S., Cellier, P., Létinois, L., 2014. Improving ammonia emissions in air quality modelling for France. Atmos. Environ. 92, 584–595. doi:10.1016/j.atmosenv.2012.08.002

Hendriks, C., Kranenburg, R., Kuenen, J.J.P., Van den Bril, B., Verguts, V., Schaap, M., 2016. Ammonia emission time profiles based on manure transport data improve ammonia modelling across north western Europe. Atmos. Environ. 131, 83–96. doi:10.1016/j.atmosenv.2016.01.043

Schaap, M., Otjes, R.P., Weijers, E.P., 2011. Illustrating the benefit of using hourly monitoring data on secondary inorganic aerosol and its precursors for model evaluation. Atmos. Chem. Phys. 11, 11041–11053. doi:10.5194/acp-11-11041-2011

Soares, J., Sofiev, M., Hakkarainen, J., 2015. Uncertainties of wild-land fi res emission in AQMEII phase 2 case study. Atmos. Environ. doi:10.1016/j.atmosenv.2015.01.068

Sofiev, M., Ermakova, T., Vankevich, R., 2012. Evaluation of the smoke-injection height from wild-land fires using remote-sensing data. Atmos. Chem. Phys. 12, 1995–2006. doi:10.5194/acp-12-1995-2012

Sofiev, M., Vankevich, R., Lotjonen, M., Prank, M., Petukhov, V., Ermakova, T., Koskinen, J., Kukkonen, J., 2009. An operational system for the assimilation of the satellite information on wild-land fires for the needs of air quality modelling and forecasting. Atmos. Chem. Phys. 9, 6833–6847. doi:10.5194/acpd-9-6483-2009