Dust-Air Pollution Dynamics over the Eastern Mediterranean
acp-2015-109
Reply to Anonymous Referee #2

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We thank the anonymous referee for the constructive comments – hopefully addressed satisfactorily with this reply. Modifications of the manuscript are summarized in Table 1-5. Please also note the modifications in Table 1 in our reply to referee #1.

The manuscript attempts to pin down the importance of the chemical aging process of dust over the Eastern Mediterranean. Aging occurs when polluted air laden with acids (for ex: H2SO4, HNO3, HCl...) encounters parcels containing dust. This is a well chosen topic relevant to ACP, but the way the authors treat the results significance does not establish the proof that aging is the main cause for the observed decrease in dust lifetime and loading. To convince the reader with the results from model, the authors would have to show that it mimics the atmosphere behavior. That includes two points that I could not find in the paper:

1. Show that a majority from the air mass that encounters air pollution are aged. A number of factors relevant to this aging being omitted from the discussion (see below)

The EM is subject to intercontinental transport of air pollution from Europe, North America and Asia, which increases the loadings of aerosols and precursor gases during the usually dry summer period and the lack of sufficient precipitation. Especially the loading of acids such as H_2SO_4 and HNO_3 are enhanced, as a result of the high insulation and the resulting photo-oxidation of the ubiquitous gaseous air pollutants (e.g., NO_x and SO_2 of ship and road traffic and waste and biomass burning). This is confirmed by numerous EMAC modeling studies and observations (Lelieveld et al., 2002; Metzger et al., 2006; Joeckel et al., 2010; de Meij and Lelieveld, 2011; de Meij et al., 2012; Pozzer et al., 2012; Lyamani et al., 2015, among others). The current study is based on the EDGARv4 emissions set (Pozzer et al., 2012) which has been prepared in the framework of the CIRCE project with a focus on the Mediterranean region as shown in p. 7498 line 25 in the MS. All EMAC studies show a high aerosol loading over the EM that is needed for dust-air pollution interaction. To demonstrate this, Figure 1 will be added to the supplement of the revised MS. The upper panel shows an indicative air pollution loading, the lower panel the corresponding cross section passing through CUT-TEPAK station. The air pollution loading represents the vertical integral (burden) of the mass concentration $[mg\ m^{-2}]$ of (left) total inorganic acids ($HCl+HNO_3+H_2SO_4$) that are present in the gas phase, maintaining gas-aerosol equilibrium, (right) the corresponding lumped aerosol burden ($SO_4^{2-}+HSO_4^-+NO_3^-+NH_4^++H_2SO_4$); both average over 20th September - 1st October 2011.

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The pollution loading basically covers the northern part of the EM and decreases eastwards. Figure 1 is consistent with the back trajectories (Fig. 14) shown in the MS, which support the north-southeast air pollution gradient that seems to be typical for the Mediterranean basin.

2. That the hygroscopicity of the dust exposed to acids is well represented.

First of all, to clarify the complexity of the EMAC model used in this study and to answer the additional question raised by referee #1, we will modify the MS to include the following paragraph in the model description section (details are shown in the table of modifications):

Our model version distinguishes aerosol particles in 7 modes, 4 Soluble (nucleation, aitken, accumulation, coarse) and three INSoluble modes (aitken, accumulation, coarse) with the complexity of the aerosol thermodynamics as investigated in Metzger et al. (2006), by considering case F4 since ISORROPIA-II used here does not include organic salt compounds in the gas/aerosol partitioning and aerosol neutralization framework. Within EMAC, the dust particles are emitted online following Astitha et al. (2012) (e.g., governed by model dynamics, precipitation and soil moisture) in either the INSoluble accumulation and/or coarse mode and only upon aging and transport they can be transferred to the respective Soluble accumulation and/or coarse modes. The aging depends on the available condensable compounds calculated within the chemistry scheme (Sander et al., 2005). In addition, via coagulation and hygroscopic growth the size-distribution can change and small particles are transferred to larger sizes, i.e., for dust from accumulation to coarse, whereby hygroscopic growth of bulk dust and dust salt compounds is only allowed in the soluble modes. For the latter we explicitly account for the water uptake of various major mineral salt compounds, i.e., $CaSO_4$, $Ca(NO_3)_2$, $CaCl_2$, $MgSO_4$, $Mg(NO_3)_2$, $MgCl_2$, Na_2SO_4, $NaNO_3$, $NaCl$, K_2SO_4, KNO_3, KCl, whereby the mineral cations Mg^{2+}, Na^+ and K^+ are only considered as tracers for the online calculated sea salt emissions, while K^+ is additionally used for biomass burning emissions being emitted here only in the insoluble aitken mode. Thus, the dust particles can be present in our set-up in four modes, each represented by various calcium compounds that chemically characterize the bulk dust emissions depending on the level of aging. Note that we have limited the dust neutralization reactions in this work to calcium to be able to separate the dust associated water uptake and associated aging from sea salt effects. Since our set-up is flexible, the level of aerosol neutralization complexity can/will be changed for other application tasks. For the current modeling study though, this set-up represents the dust air-pollution dynamics over the Eastern Mediterranean well.

Note that the water uptake of aged dust particles is based in this study on ISORROPIA-II and on the full gas-liquid-solid partitioning following (Metzger et al., 2006). Since ISORROPIA-II is widely used in atmospheric air pollution modeling, we can assume that the hygroscopicity and associated water uptake of dust particles exposed to acids is represented well. Furthermore, there are not many studies which explicitly resolve the thermodynamics of such a complex system of aerosol and precursor gases, thermodynamics and aerosol dynamics. This study considers calcium as dust tracer and $HCl, HNO_3, and H_2SO_4$ for acidic uptake by calcium. The calcium salt compounds, $CaSO_4$, $Ca(NO_3)_2$, $CaCl_2$, are subject to water uptake, however, the $CaSO_4$ is negligible, due to the relative high deliquescence humidity which is close to unity.

To clarify the treatment of the dynamical uptake of gases on dust aerosols in our EMAC study, we add the following paragraph being part of the model description section:

Within GMXe, the aging of dust aerosols depends on the total particle surface area and on the concentrations of the aerosol precursor gases (Pringle et al., 2010). The uptake of gases is

kinetically limited considering random motion and diffusion processes that govern the condensation. The rate constant for the condensation on dust particles is given by Eq. 1

$$D_{flux} = \frac{4\pi D_f^2 r_w}{\frac{4D_f}{\nu r_w a_i} + \frac{r_w}{r_w + z_{f1}}} \tag{1}$$

where r_w is the ambient (wet) radius, D_f the temperature dependent diffusion coefficient defined by $D_f = 0.073\, P \left(\frac{T}{T_{ref}}\right)^{\frac{3}{2}}$, P the pressure, T the temperature and T_{ref} the reference temperature (298.15 K). ν denotes the particle mean velocity, defined by $\sqrt{\frac{8R_g T}{\pi M_g}}$, with R the gas constant ($8.31 J\ mol^{-1}K^{-1}$), M_g the molar mass, z_{f1} the mean free path length of the kinetic regime, and a_i the accommodation coefficient (Fuchs and Davies, 1989; Seinfeld and Pandis, 2006). In the current setup we use the accommodation coefficients 0.1, 0.01, 0.01 for sulfuric, hydrochloric and nitric acid, respectively. These values have been empirically determined by a comprehensive modeling analysis, which will be presented separately. The uptake of acids is calculated for each particles size, i.e., for dust for the insoluble accumulation and coarse mode.

Both points require a careful comparison with observations from either or both laboratory and field studies. Even when dust is exposed for a long duration to H2SO4 in the laboratory, it hardly becomes hygroscopic. I did not see a review of the work that has been done on this topic. Studies such as the one of Tobo et al. (2009) indicate that in the presence of H_2SO_4, dust gets hardly hydrated except if chloride is present. Another important limiting factor in the uptake of acids and hence in the aging of dust is its calcite content. Fairlie et al. (2010) have treated this limitation but no discussion is made of how this is considered in the present work. Since these factors are paramount to the effect studied here, much more effort should be put to assess whether the model is considering them adequately and this is missing from this work.

This is a good point and the reason why we have explicitly considered the chemical speciation of mineral dust. Our treatment of using the calcium ion to determine the level of bulk dust neutralization through the reaction with inorganic acids such as H_2SO_4, HNO_3, and HCl reflects indeed the situation mentioned. In contrast to $Ca(NO_3)_2$ and $CaCl_2$, $CaSO_4$ does not contribute to water uptake, if the RH is below 99%. The reason is that the relative humidity of deliquescence (RHD) of calcium sulfate is close to unity by room temperature, while the RHD of calcium nitrate and calcium chloride is with $\approx$ 0.5 and 0.28, respectively, significantly lower. This is the actual reason for the fact that dust particles basically only become hygroscopic once exposed to nitric or hydrochloric acid. Especially, the coating by chlorides leads to hygroscopic growth and associated water uptake at almost all atmospheric conditions, particularly when dust plumes expand over oceans, such at the dust outflow over the EM considered in this study. With the additional model description we address this issue more clearly in the revised MS.

This being said the region of study and the cases chosen are very good choices to address the aging of dust. There is an inference in this work that I did not see well documented. The authors are convinced that in polluted air masses dust grows hygroscopically very rapidly compared to an airmass with dust in the absence of pollution. The simple observations of changes in altitude of the dust layer cannot suffice to prove it. Other factors such as dynamical or thermodynamical can bring the dust layers to a lower altitude, so why claim that this is caused by aerosol growth?

As outlined above, dust particles coated by inorganic acids such as HNO_3 and HCl, or other soluble compounds, can absorb water vapor, which basically depends on the relative humidity (available water mass) and the total mass loading of the coated (aged) dust. The

water uptake is more-or-less proportional to the mass loadings in most cases. In case of sufficient water vapor, the dust particles size increases with humidity until the dust particles either become activated as cloud droplets and potentially washed out by precipitation, or until the water evaporates again as a result of decreased ambient humidity (e.g., transport effects). All processes are considered here in sufficient detail. The aerosol dynamical processes are considered in the GMXe aerosol submodel (Pringle et al., 2010), the aerosol thermodynamics (dust neutralization and water uptake) by the thermodynamic gas-lqiuid-solid equilibrium partitioning module ISORROPIA II (Fountoukis and Nenes, 2007), and for the in- and below cloud scavenging we follow Tost et al. (2006). Note that thermodynamic equilibrium is considered here only for the gas-liquid-solid partitioning, but the uptake of gases on the aerosols is kinetically limited (see above). Additionally, only a small, size-dependent fraction of the bulk dust is used to chemically specify dust reaction in terms of calcium (see below). Both constraints significantly limit the equilibrium water uptake of bulk aerosols, but are somewhat empirical. The dry deposition is described in (Kerkweg et al., 2006) and explicitly depends on the ambient particle size, so that changes in particle radius, e.g., due to dust aging and associated water uptake directly increases the dry deposition.

However, due to lack of observations, we cannot absolutely quantify the effect of dust aging. Nevertheless, our sensitivity study can give an indication and estimates of the consequences of dust–air pollution interactions. Note that our sensitive study (labeled Noaging) does not consider any dust–air pollution interaction (i.e, the condensation of acids on the dust particle surface is not allowed). Both simulations are based on the same meteorological conditions and on exactly the same model set-up (except dust aging). The result of the "Noaging" case is that dust particles remain in the insoluble mode, where hygroscopic growth is generally omitted. This affects particle size, and therefore dry deposition, wet scavenging and the direct radiative forcing of dust particles. Note that the results of our sensitivity are summarized by Figs. 12 and 13 (of the MS). Interestingly, in all our (EMAC) simulations (not all are shown) the deposition has significantly increased in case of dust aging, which basically is a result of the particle hygroscopic growth, coagulation and the condensation of acids. Significant changes of the height of the dust layer is not predicted by the model. There might be an effect of the aged dust particles on the average transport height which might not be resolved by our model, since the model meteorology is nudged towards observations (ECMWF reanalysis) – which however includes air pollution. We may speculate that a pristine atmosphere without or with considerable less air pollution, dust transport might occur at a different (higher?) altitude. But to investigate this, a free running climate model with the same complexity as used here would be required for quantification. This is an interesting subject for a follow-up study but beyond the scope of this work.

The word "rapidly" (p 7513 line 25) is removed since indeed, it cannot be accurately determined so the wording of rapid aging is not really justified.

In the abstract, the sentence: Our results show the importance of chemical aging and deposition of the dust during transport. Needs to be completed by saying for what aspect of dust is this chemical aging important.

The sentence is changed to:

Our results show the importance of chemical aging of dust, which increases particles size, dust deposition and scavenging efficiency during transport, overall reducing the life-time relative to non-aged dust particles.

The following sentence p 7496, lines 11 to 13: "Since Cyprus lows are often associated with precipitation, the residence time of dust particles in the atmosphere can be relatively short (approximately one day).", is vague and the estimation of the residence time is not substantiated.

This comment refers to the dust outflow into the EM based on satellite observations during autumn (Dayan et al., 1991), which indicated a residence time of dust by one day. Since the meteorological condition and level of air pollution that govern this study may not be identical, the residence time may also be different. To clarify, the sentence is modified to:

Since Cyprus lows are often associated with precipitation, the residence time of dust particles in the atmosphere can be relatively short. In case of a barometric trough penetrating from the Red Sea into the EM, it can be as short as one day (Dayan et al., 1991).

Page 7496 lines 24-28 : Especially the interaction between dust and anthropogenic pollution from eastern and western Europe (Levin et al., 2005) in addition to intercontinental air pollution transports from North America and Asia (Lelieveld et al., 2002) deserves attention, being the focus of this study., needs to be reworked

The sentence is changed to:

Especially the interaction of atmospheric dust particles with air pollution from eastern and western Europe and long-range transport from North America and Asia (Lelieveld et al., 2002; Levin et al., 2005, among others) remains to be scrutinized – the focus of this study.

Page 7497, lines 19 to 20 : the fraction of water on dust is very much debated, some authors argue that dust can seldomly uptake dust. You need to review the experimental and field evidence to backup this claim: "The latter includes in- and below cloud scavenging, and depends on the chemical composition of the dust surface, which can include a large fraction of water".

Assuming the referee means "dust can seldomly uptake water", we agree that "can include a large fraction of water" needs to be put in context. We believe that we have clarified this aspect with our above explanations and additional description that will be part of the MS.

To be more precise, the sentence is changed to:

..., which can include a large fraction of water in case dust particles are coated by hydrochloric or nitric acids and/or are exposed to high relative humidity.

Note, although dust particles are insoluble there is a soluble fraction which depends on the dust mineralogy. Calcium rich particles preferentially form calcium sulfate, when coated with sulfuric acid. But this compound is insoluble (as mentioned above) and therefore does not contribute to water uptake, if the relative humidity is below 99%. In contrast, uptake of hydrochloric acids on mineral dust can yield calcium chloride, which can absorb water at RH 28% and therefore increases the hygroscopicity of dust particles (Tobo et al., 2009). But for both cases, the water uptake strongly depends on the available water vapor. For instance, Levin et al. (1996) performed a field study over the EM, showing that dust particles of 20 μm diameter coated with sulfate can grow up to 40 μm under supersaturated conditions as a result of heterogeneous reactions within clouds.

On the other hand, Osada (2013) showed no hygroscopic growth of dust particles in case of sub-saturated conditions. The study of Twohy et al. (2009) further showed dust particle hygroscopicity over the eastern Atlantic ocean only. Mixtures of dust and sea salt particles may contribute to the particle hygroscopicity, depending on the air pollution levels which affects

both the sea salt and mineral dust composition – effects that are explicitly considered in our model study (implicitly by the complexity of the model set-up).

Of course, the hygroscopicity of the dust particle depends on many factors, including morphology, particle size, concentration of acids such as sulfuric, nitric or hydrochloric acid. All of them are difficult to measure and therefore only indirectly accessible. Therefore, we have performed a sensitivity study and compared the model results to multi-platform observations that more-or-less accurately resolve the dynamics of the dust-air pollution interactions. Despite the underlying uncertainties, our model results are at least consistent with different observations.

Page 7500, lines 7 to 11 : We consider the calcium cation (Ca2+) as a chemically reactive tracer on the dust, being emitted in the insoluble accumulation and insoluble coarse modes as a fraction of the dust emission flux (25 and 5% for the accumulation and coarse mode, 10 respectively). These are critical numbers for this paper, the 25% in the accumulation mode is a very large number compared to the composition of dust observed, how was it chosen ? What are the measurements to back it up ?

Calcium is used here only as a proxy for the overall chemical reactivity of the dust particles, which are otherwise (usually) only treated as bulk particles (without any reactivity). The fractions of 25% and 5% have been chosen to represent a different reactivity of fine and coarse mode dust particles, assuming that smaller particles contain eventually a higher amount of soluble ions (implicitly assuming differences in the size-dependent reactivity). Both numbers have been empirically derived from a comprehensive sensitivity study to match the calcium concentrations with observations, with the constraint that at the same time the best comparison with other aerosol species (ammonium, sodium, nitrate, sulfate, chloride, etc) as well as PM and AOD is achieved with respect to various (independent) observations (that include EMEP, CASTNET and AERONET, various satellite and LIDAR data). Part of our analysis is shown by Figure 3, 4, 10-12, of the MS. However, the comprehensive model evaluation will be presented separately. Of course, these numbers depend on the overall model set-up and might be subject to change for a different model complexity.

Page 7508 lines 13 to 16: For this case, both the model and the CALIPSO results show that dust was removed during transport from the atmosphere by wet and dry deposition, since the height of the dust plume decreases from about 4000m over the central Mediterranean on 21 September to about 2000m over the EM on 22 September.. How do you infer directly from a change in height of a dust plume that deposition has occurred?

Our conclusion is based on the dust outflow dynamics (Fig. 8, MS p. 7528) and on the deposition fluxes (Fig. 11, p. 7531), which show that the dust loading has significantly reduced after the frontal system has passed, while the dust deposition has increased during the presence of the frontal system (20 September to 24 September).

The sentence on p.7508, lines 13 to 16, is changed to:

For both cases, the model and CALISPO results show that the dust concentration decreased during the transport and the height of the dust plume decreased from about 4000m over the central Mediterranean on 21 September to about 2000m over the EM on 22 September. The model results showed that the dust loading has significantly reduced after the frontal system has passed (Fig. 8) which enhanced the dust deposition (see next section).

Figure 12 is a consequence of the properties you imposed to aged dust in the model. What evidence do you have that for an air parcel with the same characteristics the size distribution is profoundly changed between a parcel that encounters pollution and one that does not?

The only way to disentangle this question is a model study that represents two identical cases, which only differ with respect to the treatment of aging. This is what has been presented here. With the additional explanation given in this reply, and in the reply to referee #1, we believe to have clarified this point. Again, briefly, there would not be identical size-distributions. The level of aging determines the water uptake, which in turn determines the particle size. Strictly spoken, identical size distributions would yield identical results, since all model feedbacks considered (dry and wet deposition and radiation) are sensitive only to the particle size, but our particle size critically depends on the chemistry, which in turn depends on the dust-air pollution dynamics over the Eastern Mediterranean.

You state page 7512 line 26 that aged dust proxy increases from 1 to 13. What measurements or other evidence back up this number?

This aged dust proxy (ADP) is specific to our model set-up. The number represents (for a particular mode) a ratio of soluble to insoluble dust concentration. A high value indicates a high level of dust aging, since all dust is initially emitted in the insoluble modes and in our EMAC set-up only transferred upon aging to the respective soluble modes. Thus, high ADP value requires a high level of air pollution that suffices to coat the primary dust particles. But more important than the absolute ADP value is its relative change, which nicely reflects the interaction of dust–air pollution dynamics. Unfortunately there are no measurements to compare with. Maybe our results could motivate further studies in this direction.

Page 7513, line 25: you mention that dust is rapidly aged, please give a time it takes for the model in hours/days to age dust with respect to the concentration(s) of acid(s) present and indicate what observations it can be compared to

The only time estimate we can give is that shown in the lower panel of Fig. 14 (p. 7534 of the MS). For instance, for the dust outflow-2, where the dust from Arabian desert enters the Mediterranean basin, the ADP changes from below 1 to more that 6 in less than 5 hours (model output frequency). This is expressed by the sharp change in the trajectory color from dark blue to cyan, indicating that the concentration of the soluble dust must have increased by a factor of 6 in less than 5 hrs. But this number depends on the atmospheric dynamics and cannot be generalized. The word "rapidly" (p 7513 line 25) is removed since indeed, it cannot be accurately determined so the wording of rapid aging is not really justified (see above).

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Table 1: Revised manuscript – Modifications for referee#2. Please also note the modifications in Table 1 in our reply to referee #1.

no.	P. no.	L. no.	Before	Correction
1	7494	13	Our results show the importance of chemical aging and deposition of the dust during transport.	Our results show the importance of chemical aging of dust, which increases particles size, dust deposition and scavenging efficiency during transport, overall reducing the life-time relative to non-aged dust particles.
2	7496	11	Since Cyprus lows are often associated with precipitation, the residence time of dust particles in the atmosphere can be relatively short (approximately one day)	Since Cyprus lows are often associated with precipitation, the residence time of dust particles in the atmosphere can be relatively short. In case of a barometric trough penetrating from the Red Sea into the EM, it can be as short as one day (Dayan et al., 1991)
3	7496	13	Dayan et al. (1991, 2008)	Dayan et al. (2008)
4	7496	24	Especially the interaction between dust and anthropogenic pollution from eastern and western Europe (Levin et al., 2005) in addition to intercontinental air pollution transports from North America and Asia (Lelieveld et al., 2002) deserves attention, being the focus of this study	Especially the interaction of atmospheric dust particles with air pollution from eastern and western Europe and long-range transport from North America and Asia (Lelieveld et al., 2002; Levin et al., 2005, among others) remains to be scrutinized – the focus of this study.

no.	P. no.	L. no.	Before	Correction
5	7497	19	..., which can include a large fraction of water.	..., which can include a large fraction of water in case dust particles are coated by hydrochloric or nitric acids and/or are exposed to high relative humidity.
6	7499	11	After: ... (Fountoukis and Nenes, 2007).	Our model version distinguishes aerosol particles in 7 modes, 4 Soluble (nucleation, aitken, accumulation, coarse) and three INSoluble modes (aitken, accumulation, coarse) with the complexity of the aerosol thermodynamics as investigated in Metzger et al. (2006), by considering case F4 since ISORROPIA-II used here does not include organic salt compounds in the gas/aerosol partitioning and aerosol neutralization framework. Within EMAC, the dust particles are emitted online following Astitha et al. (2012) (e.g., governed by model dynamics, precipitation and soil moisture) in either the INSoluble accumulation and/or coarse mode and only upon aging and transport they can be transferred to the respective Soluble accumulation and/or coarse modes. The aging depends on the available condensable compounds calculated within the chemistry scheme (Sander et al., 2005). In addition, via coagulation and hygroscopic growth the size-distribution can change and small particles are transferred to larger sizes, i.e., for dust from accumulation to coarse, whereby hygroscopic growth of bulk dust and dust salt compounds is only allowed in the soluble modes.
7	7500	14	... and sulfate (SO_4^{2-})	... and sulfate (SO_4^{2-}) as tracers for the online calculated sea salt emissions.
8	7500	14	Added text before "This chemical speciation ..."	Additionally, K^+ is used for biomass burning emissions being emitted here only in the insoluble aitken mode.

Table 3: Continued.

no.	P. no.	L. no.	Be-fore	Correction
9	7500	19	Added text	The dust particles can be present in our set-up in four modes, each represented by various calcium compounds that chemically characterize the bulk dust emissions depending on the level of aging. We account for the water uptake of various major mineral salt compounds, i.e., $CaSO_4$, $Ca(NO_3)_2$, $CaCl_2$, $MgSO_4$, $Mg(NO_3)_2$, $MgCl_2$, Na_2SO_4, $NaNO_3$, $NaCl$, K_2SO_4, KNO_3, KCl. Thus, We have limited the dust neutralization reactions in this work to calcium to be able to separate the dust associated water uptake and associated aging from sea salt effects. Since our set-up is flexible, the level of aerosol neutralization complexity can/will be changed for other application tasks. Within GMXe, the aging of dust aerosols depends on the total particle surface area and on the concentrations of the aerosol precursor gases (Pringle et al., 2010). The uptake of gases is kinetically limited considering random motion and diffusion processes that govern the condensation. The rate constant for the condensation on dust particles is given by Eq. 1 $$D_{flux} = \frac{4\pi D_f^2 r_w}{\frac{4D_f}{\nu r_w a_i} + \frac{r_w}{r_w + z_{f1}}} \qquad (1)$$ where r_w is the ambient (wet) radius, D_f the temperature dependent diffusion coefficient defined by $D_f = 0.073 \, P \left(\frac{T}{T_{ref}}\right)^{\frac{3}{2}}$, P the pressure, T the temperature and T_{ref} the reference temperature (298.15 K), ν denotes the particle mean velocity, defined by $\sqrt{\frac{8R_g T}{\pi M_g}}$, with R the gas constant (8.31 $Jmol^{-1}K^{-1}$), M_g the molar mass, z_{f1} the mean free path length of the kinetic regime, and a_i the accommodation coefficient (Fuchs and Davies, 1989; Seinfeld and Pandis, 2006). In the current setup we use the accommodation coefficients 0.1, 0.01, 0.01 for sulfuric, hydrochloric and nitric acid, respectively. These values have been empirically determined by a comprehensive modeling analysis, which will be presented separately. The uptake of acids is calculated for each particles size, i.e., for dust for the insoluble accumulation and coarse mode. For the current modeling study though, this set-up represents the dust air-pollution dynamics over the Eastern Mediterranean well.

Table 4: Continued.

no.	P. no.	L. no.	Before	Correction
10	7506	19	... at this elevation.	... at this elevation and the dust loading is significantly reduced as the frontal system passed through.
11	7508	13-16	For this case, both the model and the CALIPSO results show that dust was removed during transport from the atmosphere by wet and dry deposition, since the height of 15 the dust plume decreases from about 4000 m over the central Mediterranean on 21 September to about 2000 m over the EM on 22 September.	For both cases, the model and CALISPO results show that the dust concentration decreased during the transport and the height of the dust plume decreased from about 4000m over the central Mediterranean on 21 September to about 2000m over the EM on 22 September. The model results showed that the dust loading has significantly reduced after the frontal system has passed (Fig. 8) which enhanced the dust deposition (see next section).
12	7508	16	This is also confirmed by the independent LIDAR ground observations at CUT-TEPAK station (Mamouri et al., 2013)	removed.
13	7513	25	dust is rapidly aged	dust is aged.
14	7516	1	Added reference	Fuchs, N. A. and Davies, C. N.: The mechanics of aerosols, Dover Publications, New York, 1989.
15	7516	24	Added reference	Kanitz, T., Ansmann, A., Foth, A., Seifert, P., Wandinger, U., Engelmann, R., Baars, H., Althausen, D., Casiccia, C., and Zamorano, F.: Surface matters: limitations of CALIPSO V3 aerosol typing in coastal regions, Atmospheric Measurement Techniques, 7, 20612072, doi:10.5194/amt-7-2061-2014, http://www.atmos-meas-tech.net/7/2061/2014/, 2014.

| no.	P. no.	L. no.	Before	Correction
16	7517	17	Added reference	Mamouri, R. E. and Ansmann, A.: Estimated desert-dust ice nuclei profiles from polarization lidar: methodology and case studies, Atmospheric Chemistry and Physics, 15, 34633477, doi:10.5194/acp-15-3463-2015, http://www.atmos-chem-phys.net/15/3463/2015/, 2015.
17	7517	17	Added reference	Mamouri, R. E. and Ansmann, A.: Fine and coarse dust separation with polarization lidar, At- mospheric Measurement Techniques, 7, 37173735, doi:10.5194/amt-7-3717-2014, http://www. atmos-meas-tech.net/7/3717/2014/, 2014.
18	7518	5	Added reference	Nisantzi, A., Mamouri, R. E., Ansmann, A., and Hadjimitsis, D.: Injection of mineral dust into the free troposphere during fire events observed with polarization lidar at Limassol, Cyprus, At- mospheric Chemistry and Physics, 14, 1215512165, doi:10.5194/acp-14-12155-2014, http://www.atmos-chem-phys.net/14/12155/2014/, 2014.
19	7519	12	Added reference	Seinfeld, J. H. and Pandis, S. N.: Atmospheric chemistry and physics : from air pollution to climate change, J. Wiley, Hoboken, N.J., 2006.
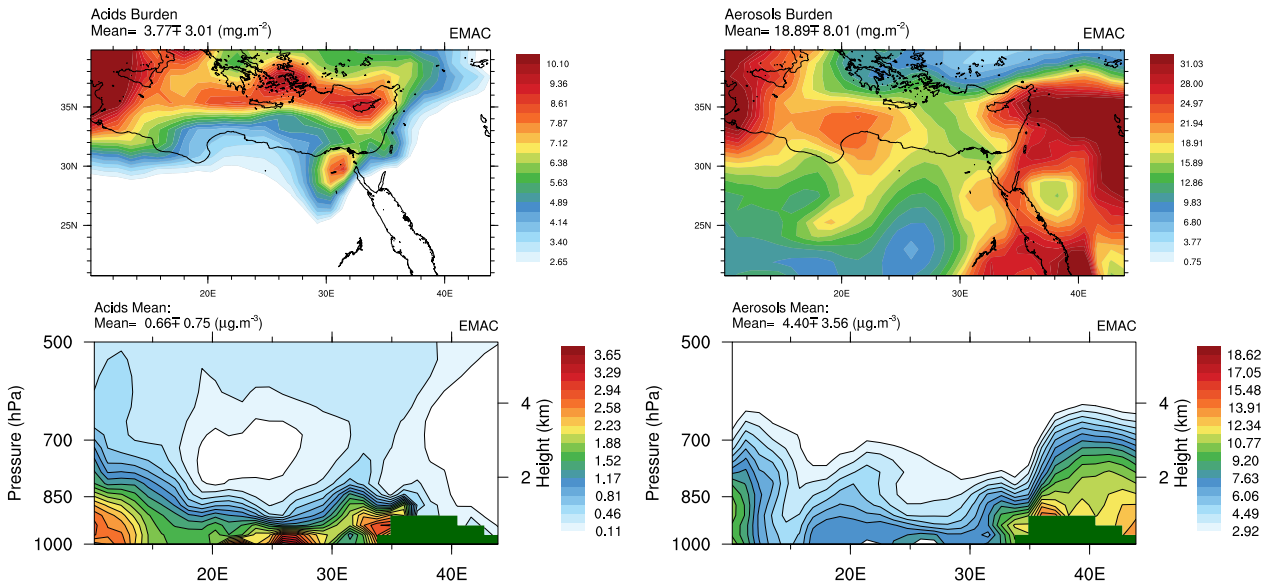
Figure 1: Air pollution loadings over the EM (average 20th September - 1st October 2011). The air pollution loading represents the vertical integral (burden) of the mass concentration [$mg\ m^2$] of (left) total inorganic acids ($HCl+HNO_3+H_2SO_4$) that are present in the gas phase, maintaining gas-aerosol equilibrium, (right) the corresponding lumped aerosol burden ($SO_4^{2-}+HSO_4^-+NO_3^-+NH_4^++H_2SO_4$); both average over 20th September - 1st October 2011. This figure is added to the supplement.