

Interactive comment on “Modelling the partitioning of ammonium nitrate in the convective boundary layer” by J. M. J. Aan de Brugh et al.

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We thank the anonymous first reviewer for his or her useful feedback. Despite the overall positive comments, the reviewer had some remarks, which we will gratefully use to improve our manuscript for submission to ACP.

A major remark was that our manuscript lacks discussion about the interaction between nitric acid and other cations instead of just ammonium. Initially, this discussion was left out because our database of hourly MARGA data lacked data for sodium for both 6 and 8 May. We think that the sodium concentration was quite low, because the mean wind direction during our simulated days was from the east. Nonetheless, the reviewer is right in that the effect of sodium on the partitioning of nitrate may still be non-negligible. Along the same line, reviewer 2 remarked that the presence of organic

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material may influence the water content and thereby the nitrate partitioning. As this is also an interaction with a component that is not in the model, we will include a general discussion about both issues in Sect. 2.2

Sea salt (sodium chloride) is known to react with nitric acid, resulting in sodium nitrate and hydrochloric acid. In other words, chloride works the same as nitrate, but it is more volatile. In general, aerosol chloride will be replaced by gas phase nitrate. It is important to correctly represent the mixing state of sodium and sulphate, because they cannot 'communicate' with each other via the gas phase. If sodium and sulphate are not internally mixed (on different aerosols), sulphate takes up ammonium and sodium takes up nitrate, adding ammonium and nitrate to the aerosol phase. However, if they are internally mixed, they neutralise one another and the ammonium nitrate may remain in the gas phase depending on the ambient temperature.

Since ISORROPIA does not allow the simulation of organics and the water uptake of different organic molecules is highly diverse (Varutbangkul et al., 2006), any treatment of these effects would be highly speculative and is considered beyond the scope of this research. Nevertheless, the following discussion will be added in Sect. 2.2

"Furthermore, the interaction between sulphate, ammonium and nitrate with other components is neglected. It should be noted that sea salt (sodium chloride) may displace nitric acid, becoming sodium nitrate and outgassing hydrochloric acid (Schaap et al., 2004; von Glasow, 2008), shifting the partitioning of nitrate towards the aerosol phase. However, the simulated days were dominated by easterly winds and therefore low sea salt concentrations. Additionally, soluble organic matter may also influence the activity of inorganic components by uptake of additional water. These effects are difficult to represent in our model and are considered beyond the scope of this study."

The second general comment of the reviewer is that it is not representative to pick one value for the temperature and humidity. This issue returns on page C13239 of the review as specific comment: P28298, L19–20: "ISORROPIA was run for an atmospheric

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column using surface temperature and RH and values that are more representative of the atmospheric columns resulting in better results. Would running ISORROPIA with the vertical temperature profile result in similar improvements?"

It should be realised here, that Sect. 3.4 was written from the perspective of a coarse-resolution model, for instance a GCM. The resolutions of these models are generally too coarse to resolve the processes described in our manuscript. Therefore, the idea of this section is to provide a cheap way for coarse-resolution models to match the the gas-aerosol partitioning at the surface more precisely by indirectly accounting for vertical mixing. Thus, we made an attempt to indirectly account for vertical mixing by calculating the gas-aerosol equilibrium with temperature and humidity on an altitude, which is higher during daytime than during night-time because there is more vertical mixing during daytime. We stuck to just a diurnal variability, although this altitude should also depend on parameters like stability, boundary layer height and wind shear. As these parameters were not taken into account, we conclusion has been nuanced with the text: "For clear days in May 2008," (P28300, L12). Since running ISORROPIA with temperature and humidity profiles instead of a representative values is not cheap, we will leave Sect. 3.4 as it is. However, we agree that the method discussed in Sect. 3.4 is insufficient to calculate the optical parameters of the aerosols, as the reviewer remarked with "Local conditions are . . . the boundary layer" (Page C13236–C13237).

P28275, L23–24: The reviewer remarks that it was already mentioned that the nitrate aerosol equilibrium depends on the relative humidity in L18–20. In L18–20, we discussed the RH-dependence of the water uptake, while in L23–24, we discussed whether ammonium nitrate itself remains in the aerosol phase or not, not the water. We will accentuate that L18–20 discuss the water and L23–24 are dealing with ammonium nitrate itself to prevent confusion.

"There are two . . . higher relative humidities", (P28275, L17–26) becomes:
"Ammonium nitrate and ammonium sulphate behave differently with varying temperature and relative humidity. First, ammonium sulphate resides exclusively in the aerosol

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phase, while ammonium nitrate resides in both the gas and the aerosol phase, where the gas-aerosol equilibrium strongly depends on the temperature and relative humidity. Furthermore, the aerosol water uptake by ammonium nitrate aerosol depends more strongly on the relative humidity than the water uptake of ammonium sulphate (Tang, 1996). Because both the aerosol dry mass (ammonium nitrate) and the aerosol water content is strongly enhanced at lower temperature and high relative humidity, the interaction of ammonium nitrate aerosol with solar radiation is also more strongly increased at these conditions than that of ammonium sulphate aerosol."

P28279, L6: The reviewer remarks that other important ions, like chloride, sodium or calcium may have been observed by the MARGA. We described sulphate, ammonium and nitrate because these are the main components discussed in this article. We will rephrase the sentence in such a way that it does not seem that the mentioned ions are the only ones observed.

"Measured were the ... SO_4^{2-} and NH_4^+ " (P28279, L5–6) becomes:

"Among other inorganic components, the gases NH_3 and HNO_3 as well as the PM components NO_3^- , SO_4^{2-} and NH_4^+ were measured with hourly frequency."

P28279, L11: The reviewer asks to cite the references to which we refer in "We refer to abovementioned reference".

We will add a citation to the papers cited in this paragraph after this phrase. In this paragraph, citations to Slanina et al. (2001) and Trebs et al. (2004) have been added, of which the latter was proposed by the second reviewer. These are also added after this phrase.

P28279, L14–15: The reviewer asks if advection, emission and deposition are implicitly included in the observed concentrations. Our research addresses the partitioning of ammonium nitrate between the gas and aerosol phase avoiding budget considerations of total nitrate, total ammonium and sulphate. This prescription module defines boundary conditions in such a way that these budgets are out of the model's scope.

We will rephrase this sentence so that it is more clear that the processes (emission, deposition and advection) are not considered important and that we aim to correctly represent total-concentrations.

On advice of the second reviewer, we moved this paragraph to the end of Sect. 2.2.2. In Sect. 2.1, we retain the sentences in which we state that that we use the observations of the total-tracers to prescribe the model and that we use the observed partitioning to evaluate the model.

"In the model . . . effect of aerosols" (P28279, L12–23) is removed from Sect. 2.1 and substituted by:

"The concentrations of sulphate, total ammonium ($\text{NH}_3 + \text{NH}_4^+$) and total nitrate ($\text{HNO}_3 + \text{NO}_3^-$) are used to prescribe the model."

At the end of Sect. 2.2.2, the following text is added:

"The concentrations of the inert tracers: sulphate, total nitrate ($\text{NO}_3^- + \text{HNO}_3$) and total ammonium ($\text{NH}_4^+ + \text{NH}_3$) at the surface were prescribed equal to the time-interpolated observations at Cabauw. This allows us to represent the correct concentrations of these tracers without implementation of processes like advection, emission and deposition. When updating a concentration, the partitioning between gas and aerosol phase is set equal to that of the previous time step in the model.", after which the text "Although this prescription . . . effect of aerosols" (P28279, L16–23) from the original manuscript is added.

P28279, L20: The reviewer raised a question about how errors are introduced when advection varies with altitude.

We thank the reviewer for pointing out this incorrect formulation. Our prescription module prescribes the surface concentration, effectively adding an emission or deposition flux to match the surface observation. On clear summer days, turbulent mixing would normally cause a uniform concentration (mixing ratio) in the entire boundary layer. In fact, we treat the total-tracers as bottom-up tracers (only emission, deposition and ver-

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tical mixing), so we do not mimic any advection at higher altitudes. We expect low biases when the boundary layer is convective, because the real total-concentrations are expected to be well-mixed. Our simulations also show well-mixed total-concentrations.

"Finally, the vertical . . . varies with altitude" (P28279, L20) becomes:

"Finally, prescription of the total-concentrations at the surface may result in slight biases when advection occurs in the upper convective boundary layer. Due to the strong vertical mixing during daytime, we think that these biases are generally small."

P28280, L8: The reviewer asks us to specify the upper size limit mentioned here.

The impactor enables correction for multiple charge. A particle larger than the largest size bin (520 nm) with more than one elementary charge could be counted by the DMA. This particle will be counted as if it is a smaller particle with a single elementary charge. This could be prevented with an impactor nozzle. Our SMPS used an 0.0457-centimetre impactor nozzle, with a 50% cutoff on about 800 nm. We will keep this issue short in our manuscript. We also add a reference with information about the SMPS.

A citation to Zieger et al. (2011) is added in the list of citations after "(SMPS)" (P28279, L26)

"The impactor upstream . . . to be counted" (P28280, L7–10) is changed into: "Particles with diameter larger than about 800 nm are removed by a 0.0457-centimetre-diameter orifice aerosol inlet in order to facilitate multiple charge correction."

P28281, L15–16: The reviewer asks if our results reflect the hypothesis that coarse aerosols reach equilibrium more slowly than fine aerosols. We think that our results indeed reflect this hypothesis. This issue has been discussed in Sect. 3.2.3 in the phrase: "In contrast to . . . equilibrated the quickest." (28293, L13–16). In the conclusion: "The situation at . . . approach described here." (28301 L5–9) also refers to the same issue. This resulted in the recommendation of continuing with a size-resolved aerosol scheme. In this conclusion paragraph, we should have stressed the aerosol size range (PM_{10} and $PM_{0.7}$) to accentuate that the link between the different outcomes from the

surface observations and from the aircraft observations to this hypothesis. We therefore modified the conclusion by adding "(PM₁₀)" between "surface" and "is" (P28301, L5) and "(PM_{0.7})" between "profiles" and "are" (P28301, L6).

P28282, L7: The reviewer proposed a more adequate formulation here.

"Evaporation is parameterised . . . the crop resistance," (P28282, L7) becomes:
"Evapotranspiration is parameterised by calculating the canopy resistance,"

P28283, L9: The reviewers asks to quantify the humidity in the Netherlands. The issue here is only to defend why we stick to metastable aerosols. This is valid when the aerosols are not dehydrated. Modelled relative humidities reach just below 40% at the surface on 8 May (and higher aloft or at other times), which is extremely low for the Netherlands. We will add the word 'enough' and a rough humidity range:

", the humidity is generally high," (P28283, L9) becomes:
", the humidity is generally high enough (40% and higher),"

The reviewer noted that Schaap et al. (2010) should be updated to the final revised paper. That is done.

P28290, L5: On this line, the phrase "does never reach" is changed to "never reaches"

P28290, L18: The reviewer asks for some quantitative numbers to judge our representation of the temperature and the relative humidity.

We added the root mean square errors of the entire days, and those of just the afternoon hours, because these hours are of most interest in this study.

after "In Fig. 3 . . . is represented reasonably" (P28290, L18–29), we add:
"The root of the mean square error in the temperature is 0.44, 0.77 and 0.54 °C for 6, 8 and 21 May respectively and during the afternoon hours (the period of interest, 12–18 UTC) only 0.18, 0.21 and 0.34 °C. The root of the mean square error in the relative humidity is 7.5, 7.8 and 6.2 percent (3.4, 4.7 and 2.6 percent during afternoon) for 6, 8

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and 21 May respectively."

The next sentence, "We underestimate the . . . in the evening" (P28290, L19–20) will start on a new paragraph. Note that this sentence is slightly changed (see next comment).

P28290, L19–20: The underestimation and overestimation of the relative humidity should be quantified. This is around 10 to 14 percent for both the underestimation and the overestimation. We will add this.

"We underestimate the . . . in the evening" (P28290, L19–20) becomes:

"We underestimate the humidity in the morning by 10–14% and we overestimate the humidity in the evening by an equal amount."

P28291, L2: The reviewer asks how good the correspondence of the simulated boundary layer height is with the backscatter data. As we stated in Sect. 2.1, the backscatter data provides a qualitative indication of the boundary layer height, so it is hard to quantify the correctness of the simulated boundary layer height. By interpreting the backscatter picture compared to our modelled boundary layer height, we estimate the mismatch in our model to be around 100 metres. It should be realised that this "100 metres" is not based on a quantitative analysis.

We will substitute the sentence: "These values are . . . the backscatter data" (P280291, L2) by: "With the aid of the provided backscatter data, we estimate that the mismatch with the real boundary layer heights is in the order of a hundred metres."

P28292, L7: The reviewer asks to quantify the improvement of the two-hour-timescale run for 6 May. In Fig. 5, it should be clearly visible that the purple line is much closer to the blue observations than the red ISORROPIA-only line, especially during the interesting afternoon hours. To mention a number, we calculated the reduction factor of the root mean square error, which is 2.1 over the entire day and even 5.9 during the afternoon.

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The part between brackets, "(see Fig. 5)" (L28292, L7) is expanded to:
"(compared to ISRROPIA-only, the root mean square error is reduced by factor 2.1 over the entire day and by 5.9 over the afternoon hours: 12–18 UTC, see Fig. 5)"

P28292, L10: The reviewer asks what disturbed the diurnal cycles in the observed nitrate partitioning on 8 and 21 May. We suggest advection. This suggestion is also confirmed in the next sentence: "Our model, not able to simulate advection . . .". If there was no advection, the partitioning would show a smooth diurnal cycle, because all other driving factors: temperature, humidity and boundary layer dynamics are smooth diurnal cycles. We also discussed in the sentence "Our model, not able to simulate advection . . ." that a smooth diurnal cycle is a logical result of having no advection.

After ". . . May are disturbed" (P28292, L10), we add ", very likely by advection"

We first discuss the last comment (on Fig. 7), because it is relevant for the upcoming comments.

As the reviewer proposed, the scatter plot of the observations in Fig. 7 is replaced by a statistical overview like in Fig. 8. To remain consistent, we decided to draw the boxes of the same shape as in Fig. 8. In Fig. 7, the box represents the median and the quartiles, the cross is the mean and the whiskers represent the lowest and highest measurement in that altitude bin.

P28293, L11: The reviewer asked us to quantify the correspondence of the model with the observations. Even with the updated version of Fig. 7, quantitative statistics is hard for several reasons. First, a different quantity is modeled (PM_{10}) than the observations ($PM_{0.7}$). Second, the model and observations refer to different altitude layers. Nevertheless, the green (20 s) simulation sticks out as being the best, merely because the 1200-altitude-bin shows the highest concentration, like the measurements. We rewrote this section to highlight this issue further.

"For 8 May . . . equilibrated the quickest." (P28293, L11–16) becomes:

"For 8 May, we clearly simulate higher nitrate concentrations than observed. This discrepancy can be explained by the fact that the model is based on PM₁₀ observations (MARGA), while the aircraft observations (AMS) are PM_{0.7}. Apart from this expected overestimation, we can conclude that the simulation with short (20 s) partitioning timescale represents the vertical profile best, with highest concentrations around 1200 metres altitude. The fact that this short-timescale simulation performs best can partly be explained by the fact that the aircraft AMS measures the finest particles, which are equilibrated quickly."

P28293, L20–23: The following phrases were asked to be quantified: "Captured very well" "not as large" and "remarkably low".

With 'captured very well', we mean that the model correctly simulates the absence of any vertical gradient between 500 metres and 1200–1300 metres. There is only a few percent concentration change in both the model and the observations, mainly due to air dilution. The 'not as large' is derived from Morgan et al. (2010), where the nitrate profiles of 21 May were not shown 'as the SLRs had reduced vertical coverage compared with B366' (page 8160 of Morgan et al., 2010). Here 'B366' means the flight of 8 May. Actually, this sentence is not needed in our manuscript, since the statistics bars now show the vertical coverage in the figure. Though it is indeed unfortunate that this is the case, we will leave out this sentence. The 'remarkably low' has already been quantified (almost 1 g N m⁻³ lower than. . .) after the comma.

"For 21 May . . . on 8 May (Morgan et al., 2010)" (P28293, L20–22) becomes:

"For 21 May, both the model and the observations show similar nitrate concentrations at 500 metres altitude and at 1200 or 1300 metres altitude."

P28293, L24–26: The reviewer asked an interesting question about deposition of nitric acid. Rapid deposition of nitric acid can be a key reason for the low observed concentration of nitric acid.

This could indeed be the case. In such a case, there would also be a signal of total-

nitrate deposition (with lower concentrations at the surface than in the boundary layer). As our model is based on surface total-nitrate observations, it would mean that these concentrations are lower than those representative for the boundary layer. This mismatch could compensate the PM_{10} - $\text{PM}_{0.7}$ discrepancy leading to roughly equal values for the model and the observations.

This deposition issue is interesting, because rapid deposition of HNO_3 enhances nitrate outgassing from the aerosols close to the surface. This way, aerosol nitrate is indirectly affected by HNO_3 deposition, which is another interesting research topic (Nemitz et al., 2009). In our manuscript, we mentioned 'surface processes affecting the fine mode'. This includes this process. We will add a citation to this presented paper by Nemitz and co-authors.

P28295, L4: The discussion of the gradients of the scattering coefficients should be supported with numbers.

Like the aerosol nitrate profiles, we compared the different partitioning timescales and the different days. 6 May results in the strongest gradient, 8 May is in between and 21 May results in the weakest gradients. For the timescales, we observe that the shorter the timescale, the stronger the gradients. The strength of the gradient can be expressed as the ratio of the scattering coefficients at the top of the boundary layer and at the surface. The strength of the gradient also varies with time (time instance of the day). Therefore, we think it is no use calculating numbers precisely, but we will add some typical numbers.

"As was the . . . gradients are weaker" (P28295, L3–5) becomes:

"Like for aerosol nitrate concentrations, our model calculates increasing scattering coefficients with altitude when the partitioning timescale is assumed short. These gradients are stronger on 6 May (almost a doubling of the scattering coefficient over the boundary layer) and weaker on 21 May (no gradient). On 8 May, the situation is in between, with a scattering coefficient increment of 50% over the boundary layer for short

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(20 s) partitioning timescale. Longer partitioning timescales result in small gradients in the scattering coefficient (at most 20% increase on 6 May with $\tau_p = 1800$ s)."

P28295, L12: The reviewer wonders how we expect that the lack of organic matter in our model is the main reason for the systematic underestimation of the scattering coefficient by the model.

The idea here is that an underestimation of the scattering coefficient can be expected because the model only takes secondary inorganic aerosols into account. The most abundant other aerosols species in Western Europe, are organic aerosols (Putaud et al., 2004). Therefore, we expect that the lack of organic aerosols is the largest contributor of the systematic bias of the model. We will reformulate this sentence in such a way that it is clear that we have no direct indication for the statement (only the indirect indication just explained). A citation to Putaud et al. (2004), also cited in the conclusion can be added.

Before formulating a change, we will first discuss the next comment, because it is in the same paragraph.

P28295, L14: The reviewer correctly remarks that the phrase "For 8 May . . . the general underestimation" (P28295, L12–14) should be supported with a quantitative analysis of the systematic bias and the model precision in capturing the general trends. Like the case of the aerosol nitrate profiles, we evaluated our model qualitatively where we tried to disregard a systematic bias. Like the previous case, this should be reformulated. The formulation in our manuscript is too positive. The message is that a gradient in the scattering coefficient is observed, and out of the simulations, only the one with $\tau_p = 20$ s produces a gradient.

To address the abovementioned issues, the paragraph was changed "For both 8 . . . the general underestimation" (P28295, 11–14) becomes: "For both 8 May and 21 May, our model underestimates the scattering coefficient by about a factor of two. An important issue is that the model only takes secondary inor-

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ganic aerosols into account. The lack of organic aerosols in the model can cause quite an underestimation of the scattering coefficient, because organic aerosols are a major component of particulate matter in Europe (Putaud et al., 2004). For 8 May, increasing scattering coefficients with altitude have been observed. Such a gradient is only reproduced by the simulation in which the partitioning timescale is set to 20 seconds."

P28297, L16–22: The following phrases should be quantified as well: "very close to 1.0", "too high" and "lowest (i.e. worst)".

We decided to perform the same statistics for the observations as for the model results (mean \pm standard deviation). The b -values will also be turned into mean and standard deviations. With the observed a -values quantified, the 'too high' is also quantitative, because the modelled a -values themselves are already quantified (1.979 ± 0.058 , P28297, L21). For the 'lowest (i.e. worst)', we will add this lowest b -value.

"(all very close to 1.0)" (P28297, L16) becomes:

"($a = 1.033 \pm 0.085$)"

"(3.83 on 21 May to 7.15 on 12 May)" (P28297, L16–17) becomes:

"($b = 4.812 \pm 1.049$)"

After " $(b = 2.318 \pm 0.048)$." (P28297, L23), we add:

To compare, the lowest b -value for the ($\sim D^2$)-simulation is 2.356.

P28300, L26: Related to the added reference to Putaud et al. (2004), the reviewer raised a question about the word 'abundant'

Putaud et al. (2004) states: "Organic matter appears to be the major component of PM_{10} and $PM_{2.5}$, except at natural and rural background sites, where sulphate contribution may be larger." in the second paragraph of the abstract. This is also shown in Table 2 of their article. We think that this is enough evidence that out of the aerosol species that are not included in our model (everything except secondary inorganic aerosols), organic matter is likely the most important. We will remove the formulation 'quite abundant'.

"Organic aerosols are generally quite abundant in Europe (Putaud et al., 2004)" (P28300, L26–27) becomes:

"(in line with Putaud et al., 2004)"

directly connected to the previous sentence, which referred to Morgan et al. (2010). Quantitative mass or volume fraction of organic aerosols are not important in this discussion.

P28301, L2–4: The reviewer states that our conclusion in "With our optical analysis . . ." is made too general.

This is a good point, since whether ($\sim D^2$) or ($\sim D^3$) performs better may depend on the physical and chemical properties of the aerosols. The conclusion may be different if we move to a more urban location or to a more rural location. Also, the weather condition may influence the aerosol water content and thereby the aerosol viscosity. So we should limit this conclusion to the situation that we simulate (Cabauw, spring, fair-weather).

"With our optical . . . aerosol volume ($\sim D^3$)." (P28301, L2–4) becomes:

"With our optical analysis, we also found indirect evidence that for situations similar to the days simulated in this article, assuming condensation proportional to aerosol surface ($\sim D^2$) is more realistic than assuming condensation proportional to aerosol volume ($\sim D^3$)."

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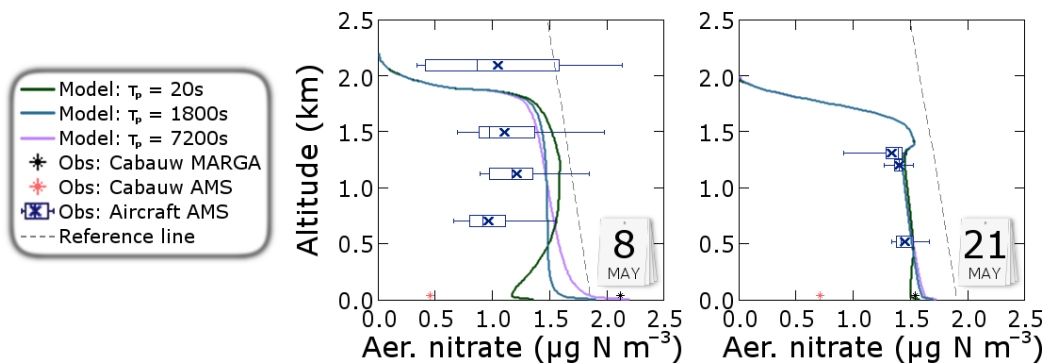


Fig. 1. Update of Fig. 7: Scatter plot of observations has been changed into statistics per altitude bin like in Fig. 8. The line thickness is increased, also in Figs. 4, 5, 8 and 10.

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