

## **Comments by Reviewer #1, including some remarks by the editor.**

### 1) Distinguishing the three categories

a) The reviewer still questions the representativeness of the three categories. As the main characteristic is the ammonia and nitrate content, evaporation of these compounds might bias the conclusion on which category the samples belong to.

Can you comment on this possible bias?

b) The reviewer criticizes that the expression 'adequate separation' in l. 92 is too vague. Please clarify.

### 2) Assumptions on emissions

The reviewer asks why emissions for the year 2008 were used as they may not be representative for the following years. As you explain that data from the year 2006 were extrapolated to the year 2008, I (the editor) wonder if the same methodology could be applied in order to obtain emission scenarios for the subsequent years, too.

### 3) Importance of dust intensity

The reviewer still questions the statement of the low importance of the dust intensity. Can you comment on the statement that  $\text{Ca}^{2+}$  should be dependent on the dust event intensity?

### 4) Category 3

I agree with the reviewer that Category 3 should be mentioned in the abstract. In addition, I am missing a discussion of Category 3 in Section 4.1.

### 5) Inorganic nitrogen

a) The reviewer asks why the focus of the study was inorganic nitrogen. Can you estimate any possible contribution of organic nitrogen in the particles?

b) I think 'inorganic nitrogen' is a too broad term. Only at one place in the manuscript it is mentioned that nitrite is excluded. I suggest being explicit and replacing 'inorganic nitrogen' by ' $\text{NH}_4^+$  and  $\text{NO}_3^-$ ' throughout the manuscript.

### 6) Median vs Mean

Please correct the contradiction of l. 205 and Table S1 (cf reviewer comment)

## **Additional editor comments**

l. 22: Do you mean 'externally mixed', i.e. in separate particles?

l. 26: What does  $< 3$  refer to here?

l. 57/8: This sentence is not clear. Please reword.

l. 113: remove 'the'

l. 173: Are the emissions modeled, i.e. predicted based on assumptions of sources or are they an input to the model?

l. 204/5: This is ambiguous. As it is written, the text suggests that each individual sample pair exhibited a net increase of 82-1303%. Is this true? Or was this large range the range that was determined based on all samples?

l. 239: This sentence needs to be improved. Do you mean 'the absolute increase...'? I don't understand what is meant by 'complex for the interactions'. It is very vague and grammatically wrong.

l. 259 and throughout the manuscript: What is meant by 'exterior sample'? Do you mean an outlier? How was this determined?

l. 262: 'It was commonly believed' should be changed here. What evidence was this assumption based on? Could it be concluded based on more than one study?

l. 267: Do you mean 'may be externally mixed'?

l. 270/1: I don't understand this. How does the dilution effect affect particle composition and what chemical reaction(s) is/are referred to here?

l. 275 and throughout the manuscript: Is there any evidence in previous studies that metal ions form stable salts in particles? References? Are these all salts or would also metal-sulfato-complexes be possible?

Section 4.3: I got a bit lost in this Section? Which part is based on measurements and which based on model results? Please clarify.

l. 351: 'totally off' is very colloquial. Is there any explanation for this discrepancy?

l. 373: I cannot follow here. Why is ammonium excluded in Category 3? Isn't that a contradiction as you mention in the following sentence that you discuss here  $N(NH_4^+ + NO_3^-)$ ?

l. 374: 'A larger decrease' than what? Please clarify.

l. 378 ff: Again, it is not clear whether the following text is based on observations or measurements. Please clarify.

Final comment: What are the main conclusions of your study? They should be summarized in a separate conclusion section after Section 4.4.