Response to Referee 1's comments

The authors thank the reviewer (Anonymous Referee #1) for their helpful comments and suggestions which will help improving the original paper. Please find our itemized responses to comments in bold italic text below.

Comments:

The Introduction section contains a lot of information on chemical species, their sources, and chemical reactions in the atmosphere. I would find a Table to be helpful in presenting this information.

Response:

We will follow your suggestions adding a new Table 1 which shows the main natural and anthropogenic sources and formation pathways for the major secondary particles (sulphate, nitrate, chloride and ammonium):

Table 1: Summary of the main natural and anthropogenic sources, and formation pathways for the major inorganic constituents of secondary $PM_{2.5}$.

Species	Formation Processes	Reactions	Natural Sources	Anthropogenic Sources
SO_4^{2-}	1) Gas phase oxidation of SO_2[(*)] followed by condensation H_2SO_4 and gaseous NH_3 neutralizing acids	$SO_2 + OH \rightarrow HSO_3$ $HSO_3 + O_2 \rightarrow SO_3 + HO_2$ $SO_3 + H_2O \rightarrow H_2SO_4$ $H_2SO_4 + NH_3 \rightarrow NH_4HSO_4$ $NH_4HSO_4 + NH_3 \rightarrow (NH_4)_2SO_4$	Volcanic and fire activities, marine phytoplankton degradation of dimethyl sulfide (DMS)	Biomass burning, fossil fuel combustion, gas to particle conversion
	2) SO_2 dissolution in cloud, fog, or rain water[(**)] and subsequent aqueous phase oxidation to H_2SO_4	$SO_2 + H_2O \rightarrow H_2SO_3$ $H_2SO_3 \rightarrow SO_3^{2-} + 2H^+$ $SO_3^{2-} + O_3$ (or H_2O_2) $\rightarrow$ sulphates		
NO_3^-	3) Gas phase oxidation of NO_x[(*)] and gaseous NH_3 neutralizing acids	$NO_2 + OH \rightarrow HNO_3$ $HNO_3 + NH_3 \leftrightarrow NH_4NO_3$	Soil, lighting	Biomass burning, fossil fuel combustion, vehicle exhaust, gas to particle conversion
	4) Gas phase oxidation of NO_x[(***)] and gaseous NH_3 neutralizing acids	$NO_2 + O_3 \rightarrow NO_3 + O_2$ $NO_2 + NO_3 \leftrightarrow N_2O_5$ $N_2O_5 + H_2O \rightarrow 2HNO_3$		
Cl^-	5) Gaseous NH_3 neutralizing acids	$HCl + NH_3 \leftrightarrow NH_4Cl$	Volcanic activity	Biomass burning, fossil fuel combustion, gas to particle conversion
NH_4^+	6) Gaseous NH_3 neutralizing acids	$H_2SO_4 + NH_3 \rightarrow NH_4HSO_4$ $NH_4HSO_4 + NH_3 \rightarrow (NH_4)_2SO_4$ $HNO_3 + NH_3 \leftrightarrow NH_4NO_3$ $HCl + NH_3 \leftrightarrow NH_4Cl$	Ocean, microbial activity, crop, soils, wild animals, and vegetation	Biomass burning, fossil fuel combustion, sewage, fertilized lands, vehicle exhaust, gas to particle conversion

[(*)] means "day"
[(**)] means "day and night"
[(***)] means "night"

In addition, it will be necessary to change the numeration of all Tables in the original paper.

There are several instances of the word “Indeed” that begin sentences used to support a claim made in the prior sentence (i.e., page 17152, lines 17&21). In this, and other cases, the word “Indeed” is being used incorrectly. Also, the word “Besides” (page 17151, line 5) is being used incorrectly in this case.

Response:

We will follow your suggestions deleting the words “Indeed” and “Besides” in all paper.

We will delete the word “Indeed” in the following sentences:

P17139, line 26;

P17140, lines 16 and 27;

P17142, lines 20 and 25;

P17145, lines 12 and 14;

P17147, lines 22 and 24;

P17148, line 27;

P17149, lines 7 and 15;

P17151, line 22;

P17152, lines 17 and 21;

P17153, lines 21 and 26.

We will delete the word “Besides” in the following sentences:

P17131, line 9;

P17151, line 5;

Use caution in claiming any relationship with an R2 value < 0.5 as “good”, perhaps consider changing to “significant” (i.e., page 17147, line 25).

Response:

Despite the fact that R^2 is a unitless statistic, there is no absolute standard for what is a "good" value. Jacob Cohen (1988) has written the most on this topic. In his well-known book he suggested the Cohen scale, for example, in which a correlation greater than 0.5 is large/strong

(0.50-1.00), 0.3 is weak/moderate (0.30-0.49) and 0.1 is small, right on the cusp of "medium/modest" (0.30-0.49). However these cutoff criteria are largely arbitrary and shouldn't be applied too strictly. The real answer is that the R^2 is only an indicator of the completeness of the regression model. Only the p-value of the coefficients should be used to determine the goodness or significance of a regression. If the p-value is less than 5% then you should consider the regression to have found a significant relationship.

However, we will follow your suggestion using the Cohen scale only the word "significant" and not "good" for the Cohen scale with a R2 value < 0.5:

P17145, lines 14-15;

P17147, line 25;

The phrasing "probably coal combustion" (page 17131, line 5) implies the authors aren't sure of the major source of HCl. Does the the scientific community, in general, have a fundamental lack of knowledge in these regards? A reference and flux estimate would be appropriate here.

Response:

HCl is emitted by biomass burning (Andreae et al., 1996), such as coal combustion and waste combustion (McCulloch et al., 1999), and also by the reaction of gaseous HNO_3 with NaCl in sea salt particles (Keene et al., 1996). About emission fluxes of 50 Tg Cl yr^{-1}, 6 Tg Cl yr^{-1}, 4.6 Tg Cl yr^{-1}, and 2 Tg Cl yr^{-1} for HCl are emitted globally to the atmosphere from dechlorination of sea salt aerosols (Graedel and Keene, 1995; Keene et al., 1996), biomass burning (Lobert et al., 1999), from coal combustion and waste burning (McCulloch et al., 1999), respectively. As Beijing is ~150 km from the sea, it has been demonstrated that the contribution to the aerosols from the sea could be ignored here (Yuan et al., 2004). Most inorganic Cl (particulate plus gas) is principally emitted in China from both coal combustion and biomass burning. In fact, estimated HCl emissions are 843 Gg Cl yr^{-1}, 480 Gg Cl yr^{-1} and 856 Tg Cl yr^{-1} from coal combustion, waste combustion (McCulloch et al., 1999), and biomass burning (Lobert et al., 1999), respectively. These HCl emissions contribute about 10% of the global source strengths. Both the chemical analysis of ambient PM2.5 and gas samples (Yao et al., 2002; Duan et al., 2006; Song et al., 2006) and source profiles measured in the laboratory (Zheng et al., 2005) indicated that the major source of HCl and Cl^- was coal combustion due to domestic heating activities and the presence of coal power plants in Beijing.

Thus, we will replace the sentence "The major source of HCl in the atmosphere is probably coal combustion" with the new sentence: "The major source of HCl in the atmosphere is biomass burning (Andreae et al., 1996), such as coal combustion and waste combustion (McCulloch et al., 1999), and also by the reaction of gaseous HNO_3 with NaCl in sea salt particles (Keene et al., 1996). About emission fluxes of 50 Tg Cl yr^{-1}, 6 Tg Cl yr^{-1}, 4.6 Tg Cl yr^{-1}, and 2 Tg Cl yr^{-1} for HCl are emitted globally to the atmosphere from dechlorination of sea salt aerosols (Graedel and Keene, 1995; Keene et al., 1996), biomass burning (Lobert et al., 1999), from coal combustion and waste burning (McCulloch et al., 1999), respectively. As Beijing is ~150 km from the sea, it has been demonstrated that the contribution to the aerosols from the sea could be ignored here (Yuan et al., 2004). Most inorganic Cl (particulate plus gas) is principally emitted in China from both coal combustion and biomass burning. In fact, estimated HCl emissions are 843 Gg Cl yr^{-1}, 480 Gg Cl $yr^{-1}$and 856 Tg Cl yr^{-1} from coal combustion, waste combustion (McCulloch et al., 1999) and biomass burning (Lobert et al., 1999), respectively. These HCl emissions contribute about 10% of the global source strengths. Both the chemical analysis of ambient $PM_{2.5}$ and gas samples (Yao et al., 2002; Duan et al., 2006; Song et al., 2006) and source profiles measured in the laboratory (Zheng et al., 2005) indicated that the major source of HCl and Cl˙ was coal combustion due to domestic heating activities and the presence of coal power plants in Beijing".

Thus, we will add new references about HCl sources and emission such as:

Andreae M. O., Atlas E., , Harris G .W ., Helas G., de Kock A., Koppmann R., Maenhaut W., Manø S., P ollock W. H., Rudolph J., Scharffe D.,. Schebeske• G., Welling M.: Methyl halide emissions from savanna fires in southern Africa, J. Geophys. Res., 101, 23,603-23,613, 1996.

Graedel T.E., and Keene W. C.: The tropospheric budget of reactive chlorine, Global Biogeochem. Cycles, 9, 47-78, 1995.

Keene W.C., Khalil M. A. K., Erickson D. J., McCulloch A., Graedel T. E., Lobert J. M., Aucott M. L., Gong S.-L., Harpe D. B., Kleiman G., Midgley P., Moore R. A., Seuzaret C., Sturges W. T., Benkovitz C. M., Koropalov V., Barrie L. A., Li Y.-F.: Composite global emissions of reactive chlorine from anthropogenic and natural sources: The reactive chlorine emissions inventory, J. Geophys. Res. 104, 8429-8440, 1999.

Lobert J. M., Keene W. C., Logan J. A., Yevich R.: Global chlorine emissions from biomass burning: The reactive chlorine emissions inventory, J. Geophys. Res., 8373-8389, 1999.

McCulloch A., Aucott M. L., Benkovitz C. M., Graedel T. E., Kleiman G., Midgley P., Li Y.-F.: Global emissions of hydrogen chloride and chloromethane from coal combustion, incineration,

and industrial activities: The reactive chlorine emissions inventory, J. Geophys. Res. 104, 8417-8428, 1999.

Yuan C. S., Sau C. C., Chen M. C.: Influence of Asian Dusts On the Physiochemical Properties of Atmospheric Aerosols in Taiwan District Using the Penchu Islands as an Example. China Particuology 2(4): 144-152, 2004.

Technical corrections:

- P17129, line 16 add an "s" on the end of "source".

Response:

We will add an "s" to the word "source" (P17129, line 16).

- P17129, line 18 – missing a word after "gas"?

Response:

We will add the word "phase"after the word "gas" (P17129, line 18).

- P17129, line 23 – misspelling of the word "tipically", replace with "typically".

Response:

We will replace the word "tipically"with the word "typically" (P17129, line 23).

- P17130, line 6 – missing a word after "gases"?

Response:

We will add the word "such" after "gases" (P17130, line 6) and, thus, we will change the sentence "Ammonium nitrate (NH_4NO_3) and ammonium chloride (NH_4Cl) are formed via reversible phase equilibrium with precursor gases as NH_3, HNO_3 and HCl" with the sentence "Ammonium nitrate (NH_4NO_3) and ammonium chloride (NH_4Cl) are formed via reversible phase equilibrium with precursor gases such as NH_3, HNO_3 and HCl".

- P17130, line 23 – do the authors mean "OH radical"?

Response:

We will add the word "radical" after "OH" (P17130, line 23).

- P17130, line 24 – delete "s" on the end of the word "combines"

Response:

We will delete "s" on the end of the word "combines" (P17130, line 24).

- P17131, line 2 – is "attach" supposed to be "attached" or "attack"?

Response:

We will change the word "attach" with the word "attack" (P17131, line 2).

- P17131, line 8 – misspelling of the word "volcanoes".

Response:

We will change the word "vulcanoes" with the word "volcanoes" (P17131, line 8).

- P17131, lines 27-29 – consider omitting the phrase "take about one third of the total coal consumptions" and change "emitting" to "emit".

Response:

We will omit the prase "take about one third of the total coal consumptions" and change "emitting" with "emit" (P17131, lines 27-29). Thus, the new sentence will be "Beijing's power plants emit 49% and 27% of the total SO2 and NOx emissions, respectively (He et al., 2003)".

- P17131, line 28 – end sentence after He et al., 2003 reference, as the next part of the sentence has already been mentioned.

Response:

We will delete the sentence "which contribute to formation of atmospheric inorganic fine particles (nitrates and sulphates), as said above" (P17131, line 28 – end sentence).

- P17132, line 16 – the word “simultaneous” is unnecessary.

Response:

We will delete the word “simultaneous” (P17132, line 16).

- P17132, line 28 – should the word “of” be “from”?

Response:

We will change the word “of” with the word “from” (P17132, line 28).

- P17135, line 8&18 – not sure what the authors mean with the phrase “ indicated with term of’. Needs to be re-phrased.

Response:

We will change the sentences “P17135, lines 7-26” with the new sentences “Total concentrations of ammonium salts in $PM_{2.5}$ are estimated as the sum of the measurements both on the Teflon filter for non-volatile fine particolate species (unevolved particulate species) and on the back-up filters for volatile fine particulate species (unevolved particulate species):

$$[NO_3^-]_{evolved} = [NO_3^-]_N \quad (1)$$

$$[Cl^-]_{evolved} = [Cl^-]_N \quad (2)$$

$$[NH_4^+]_{evolved} = [NH_4^+]_N + [NH_4^+]_{PA} \quad (3)$$

$$[NO_3^-]_{unevolved} = [NO_3^-]_T \quad (4)$$

$$[Cl^-]_{unevolved} = [Cl^-]_T \quad (5)$$

$$[NH_4^+]_{unevolved} = [NH_4^+]_T \quad (6)$$

$$[NO_3^-]_{fine} = [NO_3^-]_{unevolved} + [NO_3^-]_{evolved} \quad (7)$$

$$[Cl^-]_{fine} = [Cl^-]_{unevolved} + [Cl^-]_{evolved} \quad (8)$$

$$[NH_4^+]_{fine} = [NH_4^+]_{unevolved} + [NH_4^+]_{evolved} \quad (9)$$

where subscripts N, PA and T stand for the Nylon filter, phosphorous acid coated paper filter and Teflon filter, respectively. The concentration of evolved NH_4^+ is the sum of its amounts measured both on the Nylon filter, which absorbs a small fraction of evolved NH_3 (Masia et al., 1994), and on the coated paper filter. The remaining fine particulate species recovered on the

Teflon filter are indicated with term of unevolved particulate species. Thus, the total concentrations of the fine particulate Cl, NO_3^- and NH_4^+ were determined without disturbing the partition equilibrium existing in the atmosphere between gaseous NH_3 and the particulate components (NH_4Cl and NH_4NO_3).”

- P17139, line 25: change “to point” to “pointing”.

Response:

We will change the word “to point” with the word “pointing” (P17139, line 25).

- P17139, line 25-26 – for the phrase “in same time scale above that of”, do the authors mean “on the same times scale, higher than than that of: : :”? If so, a comma should appear after the word “always”.

Response:

We will change the sentence (P17139, lines 25-26) with the sentence “It is worth pointing out that [NH_3] in units of μmol m^3 was always in same time scale, higher than that of [HNO_3] + [HCl] in gas phase.

- P17146, line 3 – “An other” can be changed to one word “Another”.

Response:

We will change the “An other” with one word “Another” (P17146, line 3).

- P17146, line 4 – omit the word “solid”

Response:

We will omit the “solid (P17146, line 4).

- P17146, line 12: subscript the “4” in “NH4Cl”.

Response:

We will correct the word “NH4Cl” with subscript the “4” (P17146, line 12).

P17148, line 17: omit the word “Besides”.

We will omit the word “Besides” (P17148, line 17).