

**Chemical impact of transboundary PM on Japanese air quality**

T. Moreno et al.

# Daily and hourly chemical impact of springtime transboundary aerosols on Japanese air quality

T. Moreno<sup>1</sup>, T. Kojima<sup>2</sup>, F. Amato<sup>3</sup>, F. Lucarelli<sup>4</sup>, S. Nava<sup>4</sup>, J. de la Rosa<sup>5</sup>, G. Calzolari<sup>4</sup>, M. Chiari<sup>4</sup>, A. Alastuey<sup>1</sup>, X. Querol<sup>1</sup>, and W. Gibbons<sup>6</sup>

<sup>1</sup>Inst. of Environmental Assessment & Water Research (ID/EA-CSIC), Jordi Girona 18, 08034 Barcelona, Spain

<sup>2</sup>Dept. of Earth & Environmental Sciences, Kumamoto University, Kurokami, Kumamoto 860-8555, Japan

<sup>3</sup>TNO Climate, Air and Sustainability, Princetonlaan 6, P.O. Box 80015, 3508 TA Utrecht, The Netherlands

<sup>4</sup>Dept. of Physics and Astronomy, University of Florence, and INFN, Sesto Fiorentino, Florence 50019, Italy

<sup>5</sup>Center for Research in Sustainable Chemistry (CIQSO), University of Huelva, Campus de El Carmen, s/n, 21071 Huelva, Spain

<sup>6</sup>AP 23075, Barcelona 08080, Spain

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	



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Correspondence to: T. Moreno (teresa.moreno@idaea.csic.es)

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## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Abstract

The regular eastward drift of transboundary aerosol intrusions from the Asian mainland into the NW Pacific region has a pervasive impact on air quality in Japan, especially during springtime. Analysis of 24-h filter samples (ICP-AES and ICP-MS) and hourly Streaker (PIXE) samples of particulate matter collected continuously for six weeks reveal the chemistry of successive waves of natural mineral desert dust (“Kosa”) and metalliferous sulphatic pollutants arriving in Western Japan during spring 2011. The main aerosol sources recognised by PMF analysis of Streaker data are mineral dust and fresh sea salt (both mostly in the coarser fraction  $PM_{2.5-10}$ ), As-bearing sulphatic aerosol ( $PM_{0.1-2.5}$ ), metalliferous sodic PM interpreted as aged, industrially contaminated marine aerosol, and ZnCu-bearing aerosols. Whereas mineral dust arrivals are typically highly transient, peaking over a few hours, sulphatic intrusions build up and decline more slowly, and are accompanied by notable rises in ambient concentrations of metallic trace elements such as Pb, As, Zn, Sn and Cd. The magnitude of the loss in regional air quality due to the spread and persistence of pollution from mainland Asia is especially clear when cleansing oceanic air advects westward across Japan, removing the continental influence and reducing concentrations of the more undesirable metalliferous pollutants by over 90 %. Our new chemical database, especially the Streaker data, demonstrates the rapidly changing complexity of ambient air inhaled during these transboundary events, and implicates Chinese coal combustion as the main source of the anthropogenic aerosol component.

## 1 Introduction

The spectacular growth of the Chinese economy in recent years has been accompanied by an equally impressive deterioration in regional air quality (Liu and Mauzerall, 2007; Ohara et al., 2007; Chan and Yao, 2008; Aikawa et al., 2010). The problem is on such a scale that a plume of particulate matter (PM) rich in secondary inorganic

## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



compounds (SIC) regularly contaminates millions of square kilometres across the NW Pacific region and beyond (Prospero et al., 2003; Liu et al., 2008; Fairlie et al., 2010; Moreno et al., 2012). The Japanese archipelago and the Korean Peninsula are especially affected by these transboundary aerosol intrusions. A common scenario is for stagnant anticyclonic conditions over Central China concentrating pollutants which later become transported oceanward, sometimes mixing with Gobi desert dust blown in from the NW (Guo et al., 2004; Ma et al., 2004; Uno et al., 2004; Wang et al., 2004; Chung and Kim, 2008; Zhang et al., 2010; Takahashi et al., 2010). The exportation of the resulting aerosol cocktail (Fig. 1) is so frequent as to create in Japan what has been described as a “quasi-permanent” state of regional atmospheric pollution (Lasserre et al., 2008) and has led inevitably to concerns over possible health effects on the Japanese population (e.g. Ichinose et al., 2005; Ueda et al., 2010; Watanabe et al., 2011; Onishi et al., 2012). However there is still a relative paucity of detailed information published on the variations in chemical concentrations of the aerosols people are inhaling during these transboundary pollution events. Many chemical data provide only partial analyses of PM size fractions and/or analyse Total Suspended Particulate Matter rather than strictly the inhalable fraction.

With the above observation in mind, in 2010 we conducted a pilot campaign to analyse 24-h PM<sub>10</sub> filters collected during three transient transboundary pollution episodes crossing Western Japan. Our results confirmed the pronounced bimodality and inhomogeneity between natural and anthropogenic PM in east Asian transboundary aerosol intrusions (Moreno et al., 2012). Furthermore, although the number of filters analysed was relatively small, it was nevertheless enough to demonstrate the highly metalliferous and chemically complex nature of sulphatic plumes arriving from industrial China. Such plumes drift across Japan, creating regional pollution clouds that dissipate only slowly due to the dominance of atmospherically persistent submicron accumulation mode PM. It was this aspect of the chemical data, rather than the already well-characterised nature of the “natural” Gobi-desert derived “Kosa” intrusions, which seemed to us in more urgent need of further study. In this context this present manuscript moves for-

## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



ward by presenting a new database collected during a 6-week period of continuous hourly and daily measurements in Kumamoto, SW Japan. The study adopts an unusually multi-analytical approach by integrating results from Particle Induced X-ray Emission (PIXE), Inductively Coupled Plasma Mass Spectrometry/Atomic Emission spectroscopy (ICPMS/AES), chromatography and thermal-optical transmission methods, allowing comparison between hourly (Streaker) and 24-h (filter) data. Such data are unprecedented in the chemical detail they offer on Japanese air chemistry during transboundary aerosol inflows. The Streaker data in particular provides over 1000 hourly measurements of major and trace elements for each of the coarse and fine size fractions, a very large number which allows us to perform a positive matrix factorization (PMF) analysis and so separate differently sourced components within the aerosol mixture.

## 2 Methodology

Data were obtained in March and April 2011 at the top of a nine-storey building within the Kumamoto University precinct on the island of Kyushu in Western Japan (Fig. 1) approximately midway between Tokyo (ca. 1000 km ENE) and Shanghai (ca. 1000 km WSW). Kumamoto city is not impacted by any nearby heavy industrial point sources, making it an excellent location to observe the arrival of transboundary aerosol intrusions. The monitoring site can be classified as an urban background site influenced to a limited extent by road traffic emissions from a city centre arterial road 1400 m to the west and a minor two-lane road crossing the University area.

We measured hourly element concentrations continuously from 17 March to 28 April using a Streaker sampler collecting hourly aerosol samples in two size ranges (0.1–2.5  $\mu\text{m}$  and 2.5–10  $\mu\text{m}$ ) at an air flow rate of 1 l min<sup>-1</sup> that were then analyzed by Particle Induced X-Ray Emission (PIXE, see Lucarelli et al., 2011 for details) at the LABEC-INFN facility in Florence (based on a 3 MV Tandatron accelerator), where an external beam set-up is fully dedicated to atmospheric aerosol studies. For daily samples we

### Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

used a SIBATA HV-1000F high volume PM sampler ( $60\text{ m}^3\text{ h}^{-1}$ ) which excluded particles larger than  $10\text{ }\mu\text{m}$ , obtaining 24 h filter samples from 22 March until 28 April (from 12:00 p.m. local time). Quartz fibre filters (ADVANTEC QR-100) were conditioned (30–40 % relative humidity over 48 h) and weighed before and after sampling to determine 24 h  $\text{PM}_{10}$  concentrations by standard gravimetric procedures. Once the gravimetric determination was performed the filters were treated and analysed for the determination of the chemical composition of  $\text{PM}_{10}$ . One quarter of each filter was acid digested (HF :  $\text{HNO}_3$  :  $\text{HClO}_4$ , with a mixture of 2.5 : 1.25 : 1.25 ml, kept at  $90\text{ }^\circ\text{C}$  in a Teflon reactor during 6h, driven to dryness and re-dissolved with 1.25 ml  $\text{HNO}_3$  to make up a volume of 25 ml with water) for the chemical analysis using ICP-AES and ICP-MS. To assure the quality of the analytical procedure 5 mg of the NIST-1633b (fly ash) reference material loaded on a 1/4 quartz micro-fibre filter were also analysed. Detection limit and accuracy of the techniques were estimated as  $0.18\text{ ng m}^{-3}$  and 1–3 % respectively for ICP-AES, and  $0.007\text{ ng m}^{-3}$  and 0–7 % respectively for ICP-MS. The detection limits for Zr and Hf are higher ( $0.05\text{ ng m}^{-3}$ ). Another 1/4 of each filter was water leached (6 h at  $60\text{ }^\circ\text{C}$ , preceded by incubation in an ultrasound bath for 10 min, in 50 ml sealed PVC bottles) for the determination of soluble ion concentrations by ion chromatography (sulphate, nitrate and chloride) and ion selective electrode (ammonium), allowing an average detection limit for the analysed components of  $25\text{--}30\text{ ng m}^{-3}$ . A portion ( $1.5\text{ cm}^2$ ) of the remaining half of each filter was also used for the determination of organic and elemental carbon (OC and EC, respectively) by a thermal–optical transmission technique (Birch and Cary, 1996) using a Sunset Laboratory OC-EC Analyser with the EUSAAR-2 standard temperature programme. The accuracy of the equipment is in the range of 5–10 %, depending on the relative quantities of OC and EC on the filter, and the detection limit  $0.2\text{ }\mu\text{g m}^{-3}$  for both OC and EC. The sum OC + EC is  $\text{C}_{\text{total}}$ . The OM + EC (organic matter plus elemental carbon) value was obtained after applying a 1.6 factor to the OC concentrations (Turpin et al., 2000).

$\text{SiO}_2$  and  $\text{CO}_3^{2-}$  were indirectly determined on the basis of empirical factors ( $\text{Al} \times 1.89 = \text{Al}_2\text{O}_3$ ,  $3 \times \text{Al}_2\text{O}_3 = \text{SiO}_2$  and  $1.5 \times \text{Ca} + 2.5 \times \text{Mg} = \text{CO}_3^{2-}$ , see Querol et al., 2001).

Blank field filters were used for every stock purchased for sampling and analysed in the same batches of their respective filter samples. The corresponding blank concentrations were subtracted from each sample.

A Positive Matrix Factorization (PMF, Paatero and Tapper, 1994) was performed on the two data matrices of concentrations and uncertainties of hourly Streaker samples values. This provides a reliable estimation of the main sources contributing to the measured PM by weighting each data point by its analytical uncertainty and solving the following equation:

$$\mathbf{x}_{ij} = \sum_{h=1}^p \mathbf{g}_{ih} \mathbf{f}_{hj} + \mathbf{e}_{ij} \quad (1)$$

The model uses the least squares method where the indices  $i$ ,  $j$  and  $h$  refer to the number of samples, chemical components and factors respectively, while the matrices  $\mathbf{x}$ ,  $\mathbf{g}$  and  $\mathbf{f}$  refer to the concentration data, factor contribution (or factor scores) and factor profiles (or factor loading), respectively. The matrix  $\mathbf{e}$  is the matrix of residuals defined as:

$$\mathbf{e}_{ij} = \mathbf{x}_{ij} - \sum_{h=1}^p \mathbf{g}_{ih} \mathbf{f}_{hj} \quad (2)$$

The matrices  $\mathbf{g}$  and  $\mathbf{f}$  are found by minimizing the loss function  $Q$  defined as the sum of the squared residuals weighted by the uncertainty  $u_{ij}$  associated with the each data point:

$$Q = \sum_i \sum_j \left[ \frac{\mathbf{e}_{ij}}{u_{ij}} \right]^2 \quad (3)$$

Individual estimates of the concentration errors were calculated following the methodology described by Amato et al. (2009). The uncertainty estimate provides a basis to

## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



separate species which retain a significant signal from the ones dominated by noise. This principle is based on the signal-to-noise  $S/N$  ratio described by Paatero and Hopke (2003). However, due to the sensibility of  $S/N$  to sporadic values much higher than the level of noise, the percentage of data above detection limit (ADL) was used as a complementary criterion (Amato et al., 2009).

The transport pathways of air-masses into the Kumamoto area during the monitoring period were assessed using the HYSPLIT-model (Draxler and Rolph, 2003), with vertically modelled transport back-trajectories being calculated for 5 days at 750, 1500 and 2500 m a.s.l. In addition dust and sulphate maps forecasted by the Chemical Weather Forecasting System (CFORS) were obtained from the website of the National Institute for Environmental Studies (<http://www-cfors.nies.go.jp/~cfors/>) from 24 March onwards (before this date the system had been disrupted by the Great Eastern Japan earthquake on 11 March). CFORS numerically calculates distributions of Asian dust and anthropogenic sulphate aerosols every three hours, the results being uploaded on the website every day (Uno et al., 2003; Satake et al., 2004), and is widely referred to as a source of real-time information on movements of dust and pollution plumes over Asia. Finally, weather conditions (wind velocity and direction, precipitation, relative humidity and ambient temperature) were obtained from the Kumamoto Meteorological Observatory, located about 2 km west of the sampling site.

### 3 Results

Our PMF analysis of > 1000 analyses obtained from the Streaker coarse fraction ( $PM_{2.5-10}$ ) data allows us to detect 4 main source factors (Fig. 2a) these being:

1. *mineral dust* with major elements Si, Al, Ca, Fe, K, Mg and a range of trace elements that includes several metals such as Ti, Mn, Cu;
2. *metalliferous sodic aerosol* (aged sea salt), accounting for 55 % of Na and significant proportions of S, Mg, Ca, Cu, Zn and Sr;

## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





3. *fresh marine aerosol*, explaining almost 90 % of Cl concentrations and including also Na and smaller amounts of Mg;
4. *ZnCu aerosol* (road traffic) tentatively attributed to traffic emissions and associated with mineral elements (Si, Al, Mg, Ca, K, Ti) related to dust resuspension processes.

With regard to the fine aerosol fraction ( $PM_{0.1-2.5}$ ) once again the best PMF solution is obtained with 4 factors (Fig. 2b), although with distinct differences from those of the coarser fraction:

1. *As-bearing sulphatic aerosol* (China plume), dominated by S and As but also with smaller amounts of many other elements (K, Se, Sr, Ti, Mn, Fe, Zn);
2. *metalliferous sodic aerosol* (aged sea salt), which in this size fraction accounts for > 80 % of Na, associated with Se, Sr, Mg, Cu, Ca and S;
3. *mineral dust*, including Al (Si not analysed), Ca, Fe, K, and Mg;
4. *Zn-Cu aerosol* again attributed at least in part to traffic and explaining most of Zn and Cu (as seen in the coarse fraction), but also associated with Mn, K, Se, Fe, S and Sr suggesting a more mixed source for this very fine metalliferous component.

The chemical results for the 37  $PM_{10}$  filter samples reveal considerable daily variation in PM concentrations. The full analyses are provided in the Supplement on Table S1 but a selected sample group representing the main chemical variation is provided in Table 1. The ICP-AES and MS database confirms predictions made by CFORS and HYSPLIT data (Fig. 3) and identifies two extended periods when transboundary sulphatic air pollution was most prominent (28 March–3 April and 9–18 April), separated by a cleansing episode induced by the advection of oceanic air across Japan. Levels of  $nss-SO_4^{2-}$  rose to peaks exceeding  $15 \mu g m^{-3}$  during the two pollution events (samples 290311, 150411 and 170411 on Table 1), but fell to a minimum of  $< 2 \mu g m^{-3}$  during

## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



the intervening clean period (040411 and 190411 Table 1). The data also indicate the presence of elevated levels of mineral dust in several samples (e.g. 220311, 280311, 090411, 100411, 210411 Table 1, Fig. 3), recorded by increased concentrations of typically “geological” major elements such as Al, Ca, Fe. Levels of Ti, a reliable tracer for mineral dust, rise above  $55 \text{ ng m}^{-3}$  during these “Kosa” events (Table 1).

The Streaker data are summarized in Fig. 4a–d and also clearly identify the two main pollution episodes (280311–030411 and 09–180411) separated by a phase of oceanic advection. Increased levels of fine sulphate (represented by  $\text{S PM}_{0.1-2.5}$  on Fig. 4a) are typically accompanied by higher concentrations of the more toxic metallic elements (e.g. Pb, As on Fig. 4b). A somewhat contrasting pattern of fluctuations in the natural mineral dust component is represented by concentrations of Al and Ca (hourly Streaker data) plotted in Fig. 4c, revealing a series of transient peaks of these crustal elements as waves of Gobi dust crossed Kyushu during the 6-week sampling period. With reference to Fig. 4 we now consider these air quality events in more detail by subdividing the results from the sampling period into five distinct phases.

### 3.1 Phase 1: 17–27 March

The first 10 days of the Streaker campaign were characterized by NW winds feeding in transient, dilute waves of aerosols from the Asian mainland. PM levels stayed below  $40 \mu\text{g m}^{-3}$ , with both  $\text{nss-SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations being confined to a narrow range ( $3\text{--}6 \mu\text{g m}^{-3}$ ), and amounts of mineral dust ( $5\text{--}14 \mu\text{g m}^{-3}$ ) varied depending on the timing of the arrival of Gobi intrusions. Whereas the first desert dust event recorded (19 March) was relatively uncontaminated, later peaks (220311, 240311 Table 1) coincide with peaks in  $\text{S}_{0.1-2.5}$  (Fig. 4a, c) and  $\text{SIC} (\text{SO}_4^{2-} + \text{NO}_3^- + \text{NH}_4^+ > 10 \mu\text{g m}^{-3})$ . The cleanest conditions were produced by rainfall (afternoon of 20 March), and five short-lived NaCl hourly peaks occurred during periods of increased wind speed blowing sea spray into the island (Fig. 4d). The passing waves of aerosols arriving into Kyushu from offshore were interspersed with periods of light winds and low temperatures when

## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



traffic-derived local pollutants produced high  $\text{NO}_3^-$  relative to  $\text{SO}_4^{2-}$  and concentration spikes in several metals (Cr, Co, Ni, Cu, Zn, Sb) and C (240311 Table 1).

### 3.2 Phase 2: 28 March–3 April

During this phase a sulphatic transboundary intrusion, driven by an anticyclone initially persisting in East Central China then moving east, degraded air quality over much of the NW Pacific (Fig. 1). Peak daily average  $\text{PM}_{10}$  concentrations at Kumamoto reached  $66 \mu\text{g m}^{-3}$  (290311 Table 1), an increase mainly attributable to a sharp rise in SIC ( $< 10 \mu\text{g m}^{-3}$  to  $> 20 \mu\text{g m}^{-3}$ ). On some days there were also increases in Gobi desert dust as cold high level air sourced from Mongolia raised daily mineral dust levels back to  $> 10 \mu\text{g m}^{-3}$  (Fig. 4c, 280311 Table 1). Streaker data demonstrate these “Kosa” peaks to have been highly transient in nature, rising and falling over half a day or less (Fig. 4c). In contrast, metalliferous sulphatic pollutants build up more gradually to successive peaks which tend to arrive slightly later than the “mineral dust” peak, and linger in the atmosphere as fine grained, atmospherically persistent particles (Fig. 4b). Another chemical characteristic of this phase is relatively high levels of  $\text{NO}_3^-$  which over the first four days rise above  $8.0 \mu\text{g m}^{-3}$ . Such concentrations are attributed to the mixing of cold, humid air with industrial pollutants, inhibiting thermal dissociation of ammonium nitrate and so favouring high levels of particulate  $\text{NH}_4\text{NO}_3$  which attained a campaign maximum on 29–30 March (290311 Table 1) during cold ( $< 10^\circ\text{C}$ ), misty, and wet conditions in Kumamoto. Subsequent average daily temperatures rose into double figures and such elevated levels of  $\text{NO}_3^-$  were not repeated.

### 3.3 Phase 3: 3–8 April

Advection of cleansing marine air across Kyushu removes the Asian mainland influence and results in a series of NaCl peaks recorded by the Streaker data (Fig. 4d; 080411 Table 1). Ambient  $\text{PM}_{10}$  levels fall to around  $20 \mu\text{g m}^{-3}$  (samples 060411 and 070411, Table S1), and levels of contaminants  $\text{C}_t$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$ , Sc, V, Ni, Cu, As, Cd, 25897

## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Sn, Pb, and Bi all decline to a campaign minimum. In fact, by the end of this phase, concentrations of the most toxic elements such as Pb, Bi, As, and Cd are a mere 5–8% of their maxima reached during Phase 2 the previous week. Ambient levels of mineral dust are subdued to within the narrow range of 5–8  $\mu\text{g m}^{-3}$ , with the exception of a transient peak on 7 April (Fig. 4c) which we attribute to a local resuspension event under unusually strong afternoon winds ( $6 \text{ ms}^{-1}$ ).

### 3.4 Phase 4: 8–19 April

A return to anticyclonic atmospheric stagnation over Central China creates a pollution plume which is invaded from the NW by a Gobi dust intrusion and both masses move eastward into Japan (Fig. 3). The dilute southern edge of the  $\text{SO}_4^{2-}$  plume arrives in Kyushu on the afternoon of 8 April (Fig. 4a), followed that evening by Gobi dust which peaks the following midday (Fig. 4c). This is the biggest “Kosa” event in the campaign (sample 090411 Table 1,  $45 \mu\text{g PM}_{10} \text{ m}^{-3}$ ) and is followed by further mineral dust peaks over the next two days (Figs. 3a and 4c) as the dust intrusion recirculates over Kyushu producing campaign maxima in Ca, Al, Fe, K, Mg, Li, Be, Sc, Ti, Mn, Co, Rb, Sr, REE, Ta, U (samples 090411, 100411 Table 1). Sulphur  $\text{PM}_{0.1-2.5}$  recorded by the Streaker also peaks at midday 9 April but then does not decline significantly (unlike the mineral dust: Fig. 3b).

The final part of this prolonged regional pollution event was especially interesting. Whereas all previous arrivals of SIC contaminants had travelled out from China via a NW corridor typically involving transport over the Yellow Sea, by 15 April a concentrated wave of industrial pollutants was travelling directly east from the Shanghai coast into Western Japan. This fresh new wave of transboundary aerosols produced the most pronounced pollution event during the campaign ( $18 \mu\text{g SO}_4^{2-} \text{ m}^{-3}$ ). Beginning with the arrival around midnight on 15 April of a brief peak of aluminous dust accompanied by SIC (Fig. 4a, c), levels of anthropogenic contaminants were to hit new maxima on the 16 and 17 April (Fig. 4b; samples 150411, 160411, 170411 Table 1). The trace element content accompanying this sulphatic and carbonaceous aerosol intrusion is again very

## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



metalliferous (high V, Cd, Sn, Bi, K, Pb, Sb, As, Zn, W) but the chemical mix is subtly different from those recorded previously, with unusually high levels of total carbon, V, Cd, Sn and differences in element ratios (e.g. higher As/Se). This multiple pollution event was finally terminated on the evening of 18 April with the arrival of strong, clean NNW winds carrying enough sea spray to mark a marine event (Fig. 4d).

### 3.5 Phase 5: 19–28 April

The northerly advection event which swept the Phase 4 pollution plume south of Japan during 18–19 April was initially accompanied by an uncontaminated “Kosa” event which briefly raised crustal dust levels as SIC concentrations continued to fall rapidly (compare Fig. 4a, c, 18 April). The following day, still dominated by a strong northerly Russian airflow moving into Kyushu via the Korean Peninsula and Sea of Japan, provided the cleanest 24 h filter of the entire sampling campaign, with PM<sub>10</sub> levels dropping to 18.6 µg m<sup>-3</sup>, mineral dust to 3 µg m<sup>-3</sup> and SIC to 4 µg m<sup>-3</sup> (190411 Table 1). However this rapid and thorough atmospheric cleansing event provided only brief respite from the Chinese pollution plume, which continued to recirculate widely across the NW Pacific and returned into Japan on 20 April. For the remainder of the campaign this diluted but persistent SO<sub>4</sub><sup>2-</sup> haze recirculated over Japan, occasionally supplemented by influxes of fresh mainland contaminants and Gobi dust to produce several minor S<sub>0.1–2.5</sub> and Al<sub>2.5–10</sub> peaks (Fig. 4a, c; 240411 Table 1). Comparison between the coarser and finer PM fractions in the Streaker data indicate an increase in the coarser sulphate particles ( $S_{PM_{0.1-2.5}}/PM_{2.5-10}$  drops from 20.4 in Phase 4 to 9.9 in Phase 5), suggesting a relative coarsening by coagulation and chemical interaction of PM with gaseous precursors with time as the regional pollution plume ages and persists across the NW Pacific region (Fig. 4a).

## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## 4 Discussion and conclusions

The chemical data summarized in this paper allow us to view both hourly and daily fluctuations in transboundary aerosols affecting Japanese ambient air quality over a 6-week period in spring, and provide a clearer idea of the reality of what people are inhaling at that time of the year. Such data highlight the limitations of legal air quality standards based only on PM mass without considering the complex chemical mixture of inhalable aerosols involved. The PMF analysis of Streaker hourly data confirms that natural PM predominate in the coarser PM fraction ( $PM_{2.5-10}$ ), with continental mineral dust dominant over fresh marine aerosol. In the finer fraction ( $PM_{0.1-2.5}$ ) however the mineral dust is much less abundant and instead the dominant component is As-bearing sulphatic aerosol. The metalliferous sodic PM component recognised in both size fractions is suggested to represent dechlorinated, sulphate-enriched aged sea salt aerosols contaminated by industrial emissions during long-distance transport.

No extreme “Kosa” events (when average daily  $PM_{10}$  concentrations in Japan can rise dramatically to well above  $100\mu g m^{-3}$ ) were recorded during the campaign, but instead our sampling interval was typical of what these days may be considered normal springtime conditions. Under such conditions prevailing NW winds bring frequent but normally brief intrusions of Gobi dust into Western Japan so that during the combined 4-week period of phases 1, 2 and 5 our data recorded nine such “natural” events when concentrations of Al exceeded  $1\mu g m^{-3}$  (Fig. 4c) for a few hours. A notably heavier loading of mineral dust occurred during the pollution episode of Phase 4, but even here the peaks remained highly transient, with concentrations of Al rising from around  $1\mu g m^{-3}$  to double or triple over 2 or 3 h but then falling back equally rapidly. In contrast the anthropogenic component of suspended particulate matter in the atmosphere usually builds up more slowly and does not decline as rapidly as the mineral dust. Once  $S_{0.1-2.5}$  levels rose above  $2\mu g m^{-3}$  during the pollution episodes of Phases 2 and 4 they stayed high, reflecting the atmospherically persistent nature of this extremely fine sulphatic PM. The same behaviour can be seen in the metalliferous components, well

### Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



displayed by Pb, Zn, Sn and As on Fig. 4a, b, which consistently correlate more with the sulphatic component than with the natural mineral dust. Sulphatic plumes dissipate only slowly, due to the dominance of submicron accumulation mode aerosols which are atmospherically persistent, although interestingly our Streaker data for Phase 5 suggest that the toxic metal loading may decrease as the air mass ages and coarsens by coagulation (Fig. 4a, b).

It is clear that transboundary aerosol intrusions arriving into Japan are chemically inhomogeneous. As we have observed in our precursor pilot study of “Kosa” events in Kyushu, some waves of Gobi dust are more calcareous than others, presumably depending on their geological source area (Yuan et al., 2008; Zhang et al., 2005; Shao et al., 2008; Moreno et al., 2012). The Streaker data reinforce this observation of natural chemical variables in transboundary events, with later mineral dust peaks in phases 4 and 5 registering as more aluminous than earlier, more calcareous ones (Fig. 4c). Furthermore, our data reveal distinct differences in the chemical signature of the Asiatic mainland pollution plume, even after a journey of around 1000 km or more from source. The best example is provided by the unusually high levels of  $\text{SO}_4^{2-}$ , V, Cd, Sn, Bi, Cu, As, Sb, and Pb in response to the arrival of pollution directly from East China into Kyushu on 15 April. Yet another confounding factor for those trying to determine the possible health effects of transboundary aerosols is the way peak concentration arrivals of mineral dust are commonly not synchronous with anthropogenic pollution peaks. A good example of this is provided by Phase 4 when the initial arrival of a “Kosa” PM wave was accompanied by a much slower build up to a sulphate peak (Fig. 4a and c). Indeed, the high levels of natural dust at the beginning of Phase 4 contrast greatly with the later peaks in anthropogenic contaminants as the sulphatic plume was repeatedly recharged by mainland-sourced pollution without any additional influence of Gobi desert dust. Finally, while still on the theme of chemical variation within the East Asian pollution plume, our data show considerable differences in ambient PM nitrate concentration (from 1 to  $16 \mu\text{g m}^{-3}$ ), depending not just on local versus exotic sources but also on temperature controlling the volatility of atmospheric ammonium nitrate.

## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Chemical impact of transboundary PM on Japanese air quality**

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The magnitude of the loss in air quality over Japan due to the 21st century spread of the mainland Asiatic pollution plume is emphasized by our Phase 3 record of 5 days when winds brought air from the oceanic rather than continental sector. As the origin of these winds crossing SW Japan rotated clockwise from north to east to south we see proof of how little regional industrial air pollution these days originates from the Japanese archipelago. Average levels of anthropogenic pollutants elements fall abruptly, in some cases by > 90 %, as the continental source is removed. This is especially true of the most undesirable pollutants: Pb from > 60 to 3 ngm<sup>-3</sup>, As from 6 to 0.3 ngm<sup>-3</sup>, Cd from 1.1 to 0.09 ngm<sup>-3</sup>. Things, of course, were not always this way, with extreme levels of Japanese air pollution accompanying rapid industrialization in the mid 1950s to mid-1960s, leading to the introduction of the Basic Law for Environmental Pollution Control in 1967 (Kawamoto et al., 2011). The modern problem in Japan thus has to be placed in perspective, and is clearly less severe than impacts on air quality in many parts of urban China (Kan et al., 2007; Okuda et al., 2008; Zhao et al., 2008; Saikawa et al., 2009; Zheng et al., 2011). However, even given the more dilute character of the transboundary plumes reaching neighbouring receptor countries, their atmospheric persistence and highly respirable nature create reasonable cause for concern. Much of this concern has focussed more on the pulmonary toxicity and corresponding acute effects of PM inhalation, especially on asthmatic patients (e.g. Ichinose et al., 2005; Ueda et al., 2010; Watanabe et al., 2011), or on the potential transport of active bioaerosols between countries (e.g. Chen et al., 2010). However, a greater health problem is more likely to be related to premature deaths from cardiovascular and respiratory diseases resulting from long-term exposure to elevated levels of atmospherically persistent, respirable, metalliferous PM (Chan et al., 2008; Liu et al., 2009). Our 6-week continuous database from Kumamoto demonstrates the constantly changing chemical complexity of this pernicious problem.



Supplementary material related to this article is available online at:  
[http://www.atmos-chem-phys-discuss.net/12/25887/2012/  
acpd-12-25887-2012-supplement.pdf](http://www.atmos-chem-phys-discuss.net/12/25887/2012/acpd-12-25887-2012-supplement.pdf).

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## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Chemical impact of transboundary PM on Japanese air quality**

T. Moreno et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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**Chemical impact of  
transboundary PM on  
Japanese air quality**

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Chemical impact of transboundary PM on Japanese air quality**

T. Moreno et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

**Table 1.** Daily, average, maximum, and standard deviation values for elemental concentrations analysed in selected PM<sub>10</sub> gravimetric filters collected in Kumamoto March–April 2011 (see text for details), including average values for the full dataset from 22 March to 28 April. Each sample bears the date when 24-h sampling started at midday and therefore includes the first half of the following day. Mineral = CO<sub>3</sub><sup>2-</sup> + SiO<sub>2</sub> + Al<sub>2</sub>O<sub>3</sub> + Ca + Fe + K + Mg; OM + EC = Organic matter + elemental carbon; SIC = NH<sub>4</sub><sup>+</sup> + NO<sub>3</sub><sup>-</sup> + SO<sub>4</sub><sup>2-</sup>; Marine = Na + Cl. n.a.: not analysed

Date	220311	230311	240311	280311	290311	040411	080411	090411	100411	150411	160411	170411	190411	210411	240411	average	max	std. dev
µgm <sup>-3</sup>																		
PM <sub>10</sub>	39.65	22.94	36.39	47.69	65.73	22.94	27.72	45.07	52.34	54.86	63.99	54.73	18.60	47.58	36.34	37.62	65.73	12.41
OM + EC	7.03	6.93	11.23	11.03	11.78	4.53	4.26	8.72	10.42	9.01	18.16	13.40	5.13	11.67	9.05	8.96	18.16	3.14
OC	3.99	3.96	6.13	6.24	6.80	2.60	2.49	5.41	6.14	5.17	10.33	7.88	2.78	6.78	5.20	5.12	10.33	1.81
EC	0.84	0.54	1.21	1.02	0.76	0.42	0.39	0.52	0.80	0.71	1.30	0.76	0.55	0.89	0.55	0.72	1.30	0.24
CO <sub>3</sub> <sup>2-</sup>	2.19	0.86	1.65	1.83	1.00	0.94	1.13	2.41	2.11	1.25	1.44	1.33	0.60	1.89	0.45	1.20	2.41	0.47
SiO <sub>2</sub>	6.78	2.44	4.97	5.52	3.73	4.66	4.25	8.33	7.24	3.60	4.43	4.15	1.40	8.46	1.38	4.10	8.46	1.64
Al <sub>2</sub> O <sub>3</sub>	2.26	0.81	1.66	1.84	1.24	1.55	1.42	2.78	2.41	1.20	1.48	1.38	0.47	2.82	0.46	1.37	2.82	0.55
Ca	0.79	0.30	0.68	0.80	0.44	0.39	0.36	0.85	0.80	0.42	0.48	0.45	0.20	0.77	0.17	0.45	0.85	0.18
Fe	0.81	0.32	0.68	0.77	0.59	0.61	0.49	0.99	0.94	0.50	0.59	0.55	0.19	0.90	0.24	0.52	0.99	0.19
K	0.54	0.23	0.37	0.53	0.52	0.19	0.33	0.65	0.70	0.55	0.67	0.59	0.13	0.55	0.37	0.40	0.70	0.17
Na	0.61	0.38	0.32	0.36	0.25	0.40	0.30	0.51	0.46	0.88	1.09	1.02	0.61	0.61	0.25	0.55	1.11	0.25
Mg	0.40	0.16	0.25	0.25	0.14	0.14	0.23	0.46	0.37	0.25	0.29	0.26	0.12	0.29	0.08	0.21	0.46	0.08
Cl	0.80	0.17	0.47	0.36	0.79	1.22	n.a.	0.19	0.10	0.43	0.65	0.64	0.47	0.21	0.38	0.48	1.22	0.28
NO <sub>3</sub> <sup>-</sup>	4.40	3.87	5.98	8.13	16.12	1.77	1.31	4.21	4.58	6.15	4.42	3.74	2.02	2.91	5.03	4.58	16.12	2.92
SO <sub>4</sub> <sup>2-</sup>	5.10	3.15	3.73	9.21	16.56	1.90	4.06	4.57	7.03	18.00	11.21	16.99	1.97	7.25	10.46	6.98	18.00	4.10
nss-SO <sub>4</sub> <sup>2-</sup>	5.03	3.10	3.70	9.16	16.53	1.85	4.03	4.51	6.97	17.89	11.08	16.87	1.90	7.18	10.43	6.92	17.89	4.10
NH <sub>4</sub> <sup>+</sup>	0.58	0.54	0.65	1.54	4.31	0.31	0.86	0.75	1.13	1.72	1.79	1.68	0.20	0.72	1.54	0.98	4.31	0.77
ngm <sup>-3</sup>																		
Li	0.92	0.31	0.69	0.97	0.92	0.40	0.55	1.15	1.13	0.70	0.76	0.72	0.15	0.83	0.30	0.57	1.15	0.25
Be	0.06	0.04	0.03	0.04	0.03	0.04	0.04	0.06	0.07	0.04	0.04	0.04	0.02	0.06	0.03	0.04	0.07	0.01
Sc	0.84	n.a.	0.82	0.03	0.38	2.69	0.24	2.61	4.12	0.10	1.00	0.73	0.42	1.02	1.79	1.10	4.12	0.86
Ti	58.08	28.18	55.06	60.66	43.66	50.45	43.59	87.21	77.30	41.64	49.82	44.50	17.98	91.01	17.87	45.10	91.01	16.15
V	4.72	3.16	3.44	3.71	3.52	2.55	3.44	4.37	5.14	8.57	4.65	5.19	1.34	3.65	2.83	3.59	8.57	1.34
Cr	2.82	1.84	4.02	4.80	4.90	< d.l.	1.49	2.71	3.06	5.63	2.28	1.93	0.42	1.90	1.11	2.15	5.63	1.46
Mn	29.19	14.38	30.58	35.45	32.31	19.23	20.97	30.85	36.86	27.50	25.90	23.57	8.13	29.51	12.85	21.61	36.86	7.91
Co	0.36	0.20	0.34	0.38	0.31	0.25	0.23	0.42	0.42	0.28	0.27	0.25	0.08	0.35	0.11	0.24	0.42	0.08
Ni	2.68	2.05	4.33	4.58	4.46	3.12	1.85	2.19	2.78	4.39	2.99	3.12	1.18	1.63	1.47	2.81	4.90	1.20
Cu	5.68	4.60	7.64	8.93	9.69	4.24	3.42	5.68	9.84	10.69	7.29	6.46	3.26	6.71	5.82	5.96	11.12	2.28
Zn	67.15	73.18	88.72	120.21	143.00	34.60	30.03	58.59	112.05	130.90	97.85	87.91	23.14	56.53	70.14	71.53	151.49	38.14
Ga	0.63	0.31	0.52	0.84	1.24	0.29	0.40	0.63	0.75	0.68	0.67	0.64	0.09	0.54	0.38	0.47	1.24	0.23
As	3.41	2.02	2.76	5.89	5.33	1.75	1.10	2.62	3.21	5.31	5.06	4.48	0.52	2.18	1.95	2.67	5.89	1.52
Se	0.77	0.36	0.80	1.53	0.96	0.53	0.69	0.98	1.61	0.59	0.76	0.53	0.35	0.85	0.18	0.76	1.69	0.37
Rb	2.48	0.97	1.76	2.47	2.54	0.91	1.34	3.09	3.24	2.56	2.52	2.29	0.38	2.24	1.44	1.72	3.24	0.79
Sr	6.15	2.25	4.02	4.79	3.33	2.41	3.67	7.40	6.26	3.68	3.71	3.71	1.05	5.31	1.44	3.27	7.40	1.41

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Table 1. Continued.

Date	220311	230311	240311	280311	290311	040411	080411	090411	100411	150411	160411	170411	190411	210411	240411	average	max	std. dev
Cd	0.66	0.31	0.39	0.77	1.11	0.26	0.33	0.44	0.65	1.13	0.89	0.88	0.15	0.31	0.40	0.47	1.13	0.28
Sn	1.28	0.84	1.58	2.40	3.02	0.81	0.96	1.42	2.32	3.88	2.50	2.44	0.51	1.56	1.29	1.55	3.88	0.77
Sb	1.13	0.95	1.84	2.42	2.81	0.62	0.92	1.51	2.69	3.01	2.45	2.02	0.87	1.69	1.53	1.68	6.86	1.13
Cs	0.34	0.14	0.24	0.38	0.43	0.17	0.16	0.42	0.48	0.41	0.33	0.31	0.04	0.23	0.25	0.25	0.48	0.13
Ba	13.95	8.97	12.89	19.39	11.15	5.04	8.83	17.62	19.88	10.29	9.92	9.09	4.20	15.31	7.79	10.22	23.48	4.41
La	0.77	0.33	0.63	0.68	0.37	0.24	0.49	0.93	0.94	0.48	0.58	0.58	0.15	0.70	0.25	0.45	0.94	0.20
Ce	1.47	0.62	1.11	1.31	0.69	0.63	0.96	1.87	1.88	0.90	0.97	0.94	0.40	1.39	0.55	0.87	1.88	0.38
Pr	0.17	0.07	0.11	0.14	0.06	0.06	0.11	0.22	0.19	0.07	0.08	0.08	0.01	0.15	0.03	0.08	0.22	0.05
Nd	0.56	0.24	0.33	0.43	0.17	0.15	0.37	0.71	0.64	0.27	0.32	0.30	0.08	0.55	0.12	0.28	0.71	0.16
Sm	0.13	0.06	0.03	0.05	0.01	< d.l.	0.08	0.15	0.13	0.06	0.07	0.06	0.02	0.13	0.03	0.06	0.15	0.04
Eu	0.14	0.03	0.01	0.01	< d.l.	0.01	0.04	0.04	0.04	0.02	0.02	< d.l.	0.02	< d.l.	0.02	< d.l.	0.04	0.01
Gd	0.13	0.07	0.01	0.04	< d.l.	< d.l.	0.09	0.15	0.14	0.06	0.07	0.07	0.02	0.13	0.02	0.07	0.15	0.04
Tb	0.02	0.01	< d.l.	< d.l.	< d.l.	< d.l.	0.01	0.02	0.02	< d.l.	< d.l.	< d.l.	< d.l.	0.02	< d.l.	0.01	0.02	0.01
Dy	0.11	0.08	< d.l.	< d.l.	< d.l.	< d.l.	0.08	0.13	0.12	0.02	0.03	0.02	< d.l.	0.12	0.02	0.06	0.13	0.04
Ho	0.02	0.01	< d.l.	< d.l.	< d.l.	< d.l.	0.01	0.02	0.02	< d.l.	0.01	< d.l.	< d.l.	0.02	< d.l.	0.01	0.02	0.01
Er	0.05	0.03	< d.l.	< d.l.	< d.l.	< d.l.	0.04	0.06	0.06	0.01	0.01	< d.l.	< d.l.	0.06	0.01	0.02	0.06	0.02
Tm	0.02	0.02	< d.l.	< d.l.	< d.l.	< d.l.	0.02	0.02	0.02	< d.l.	< d.l.	< d.l.	< d.l.	0.01	< d.l.	0.01	0.02	0.01
Yb	0.05	0.03	< d.l.	< d.l.	< d.l.	< d.l.	0.03	0.06	0.06	0.01	0.01	< d.l.	< d.l.	0.05	0.01	0.02	0.06	0.02
Lu	0.02	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	< d.l.	0.02	0.02	< d.l.	< d.l.	< d.l.	< d.l.	0.02	< d.l.	0.02	0.02	0.00
Hf	0.49	0.63	0.47	0.57	0.48	0.42	0.56	0.51	0.56	0.34	0.37	0.35	0.32	0.64	0.56	0.48	0.64	0.09
Ta	< d.l.	0.05	0.02	0.04	< d.l.	< d.l.	< d.l.	0.03	0.09	< d.l.	< d.l.	< d.l.	< d.l.	0.04	< d.l.	0.04	0.10	0.03
W	0.66	0.86	1.08	0.83	1.30	0.60	0.40	0.79	0.95	1.35	1.68	0.96	0.56	1.04	0.44	0.79	2.06	0.40
Tl	0.14	0.07	0.13	0.28	0.39	0.12	0.08	0.18	0.37	0.33	0.34	0.34	0.02	0.13	0.18	0.17	0.39	0.11
Pb	27.65	14.45	24.37	45.34	62.50	6.84	13.76	28.87	44.71	57.39	44.36	40.47	4.44	14.46	28.96	24.63	62.50	16.26
Bi	0.51	0.25	0.64	0.96	1.38	0.24	0.73	0.55	0.95	1.65	0.99	0.97	0.19	0.41	0.74	0.57	1.65	0.36
Th	0.17	0.09	< d.l.	0.09	< d.l.	0.05	0.04	0.23	0.22	< d.l.	< d.l.	< d.l.	< d.l.	0.29	0.08	0.13	0.29	0.07
U	0.17	0.15	< d.l.	< d.l.	< d.l.	< d.l.	0.15	0.21	0.18	0.04	0.04	0.05	< d.l.	0.19	0.06	0.10	0.21	0.06
$\mu\text{g m}^{-3}$																		
Mineral	13.76	5.11	10.25	11.55	7.67	8.49	8.20	16.46	14.57	7.77	9.36	8.72	3.11	15.70	3.15	8.25	16.46	3.16
OM + EC	7.03	6.93	11.23	11.03	11.78	4.53	4.26	8.72	10.42	9.01	18.16	13.40	5.13	11.67	9.05	8.96	18.16	3.14
SIC	10.08	7.56	10.36	18.88	36.99	3.99	6.23	9.53	12.73	25.86	17.42	22.41	4.18	10.88	17.03	12.54	36.99	7.03
Marine	1.40	0.56	0.79	0.72	1.04	1.62	2.07	7.70	0.56	1.31	1.74	1.66	1.08	0.81	0.62	1.06	2.33	0.43
Trace metals	0.24	0.18	0.25	0.33	0.34	0.15	0.15	0.28	0.35	0.33	0.27	0.25	0.07	0.27	0.18	0.21	0.37	0.08

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

⏴

⏵

Back

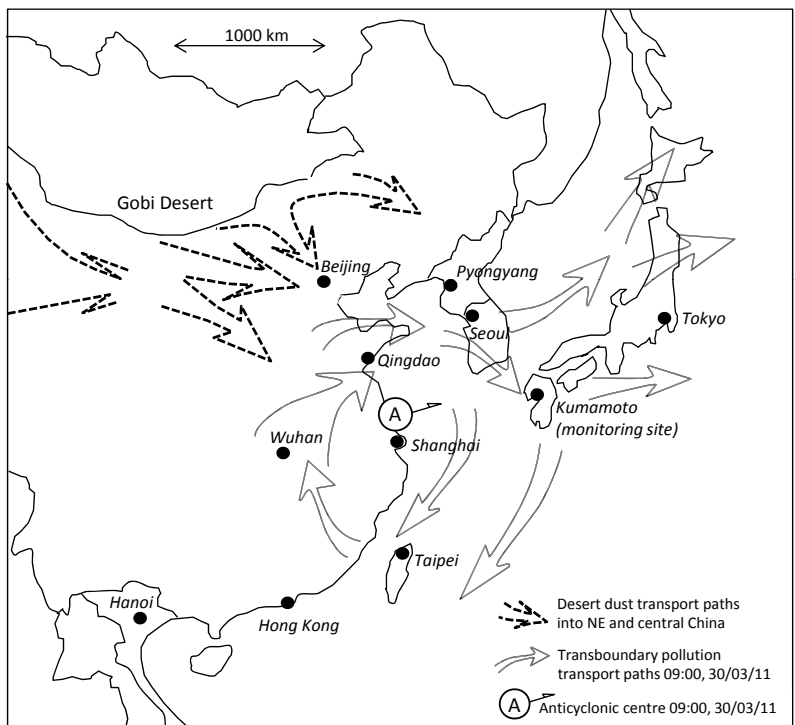
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Full Screen / Esc

Printer-friendly Version

Interactive Discussion





**Fig. 1.** Location map overviewing the area affected by the transboundary expulsion of anthropogenic aerosols from China into the NW Pacific region. Arrows depict typical atmospheric circulation patterns driving the pollution eastward out from the mainland, in this case by an anticyclone centred near Shanghai during the sampling period at the end of March 2011. The monitoring site in Kumamoto lies on the island of Kyushu in SW Japan, in the area most frequently visited by transboundary PM outflows from the Asian mainland. The main source areas of springtime desert dust intrusions into the area are also shown (Wang et al., 2004).

**Chemical impact of transboundary PM on Japanese air quality**

T. Moreno et al.

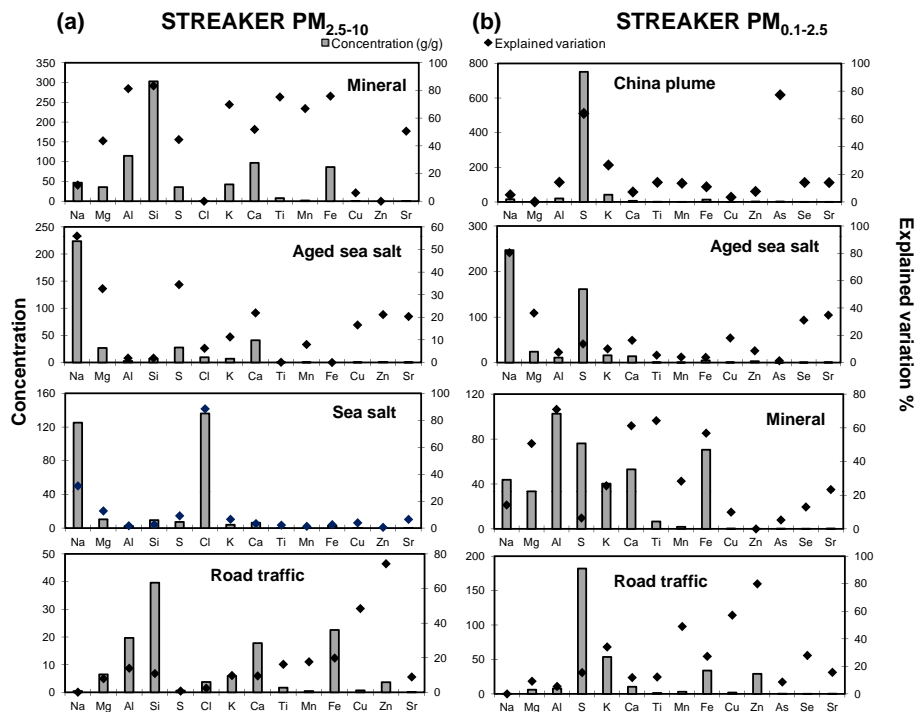
Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	





## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.



**Fig. 2.** Chemical profile for each factor identified by Positive Matrix Factorization (PMF) for hourly Streaker samples showing concentration histograms and the explained variation for each element.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

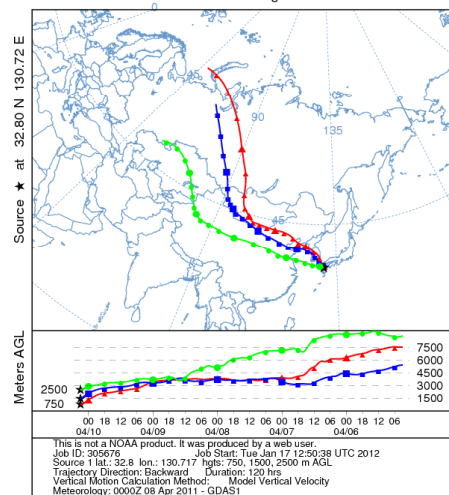
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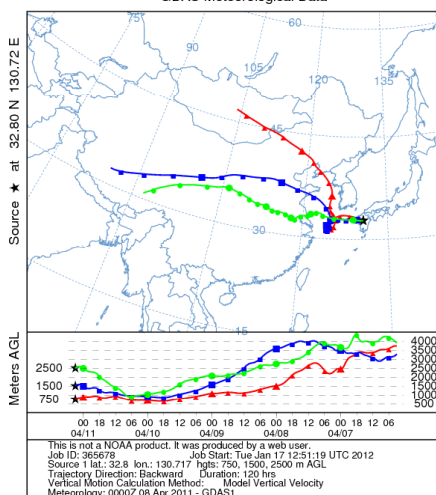
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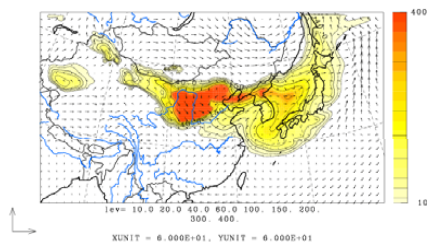
**(a)** NOAA HYSPLIT MODEL  
Backward trajectories ending at 0300 UTC 10 Apr 11  
GDAS Meteorological Data



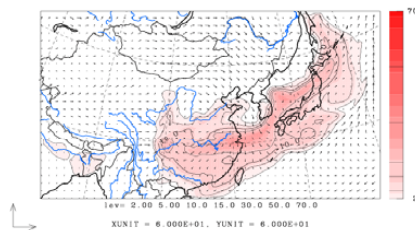
**(b)** NOAA HYSPLIT MODEL  
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GDAS Meteorological Data



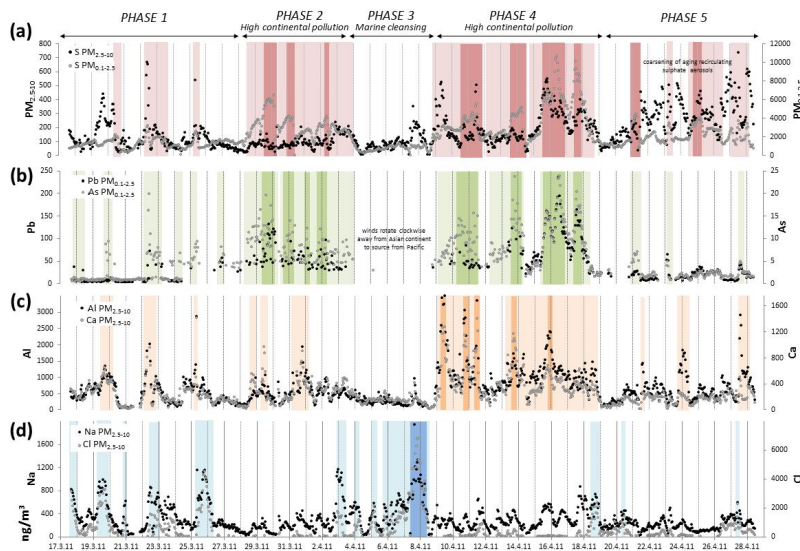
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U-V&Sulfate m/s&ug/m3 JST  
2011/04/11.09:00:00



**Fig. 3.** Examples of HYSPLIT backtrajectories (top) and CFORS maps (bottom) illustrating the arrival of air masses from the Gobi desert (**a** – 10 April 2011) and Asia mainland (**b** – 11 April 2011) resulting in a dust (**a**) and a sulphate (**b**) event in Kumamoto.



**Fig. 4.** Hourly selected elemental concentrations ( $\text{ng m}^{-3}$ ) obtained with the Streaker sampler for the five air quality phases identified during the monitoring campaign, with coloured bands highlighting main episodes of continental pollution and advection of salty marine air across Kumamoto. **(a)** shows both coarse ( $\text{PM}_{2.5-10}$ ) and fine ( $\text{PM}_{0.1-2.5}$ ) sulphatic aerosol intrusions. Whereas the finer fraction of these aerosols is always dominant (compare different scales), there is a clear tendency towards coarsening as the pollution plume introduced during Phase 4 is briefly forced south by northerly winds but brought back to age and recirculate over Japan during Phase 5. **(b)** uses Pb and As hourly concentrations to illustrate how toxic metallic aerosols accompany the sulphatic intrusions. **(c)** uses hourly concentrations of Al and Ca to reveal the rise and fall of Gobi desert PM introduced by NW winds crossing NE China and therefore usually mixed to some extent with anthropogenic pollutants. **(d)** uses hourly concentrations of Na and Cl to identify marine aerosol episodes, with Phase 1 recording several transient periods of strong NW winds accompanied by Gobi dust and sea spray sourced from the Yellow Sea, whereas Phase 3 shows a prolonged period of atmospheric cleansing under non-continental winds sourcing initially from the Sea of Japan and then from the Pacific Ocean.

## Chemical impact of transboundary PM on Japanese air quality

T. Moreno et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

