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composition of  
rainwater at MCOH**

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# Chemical composition of rainwater at Maldives Climate Observatory at Hanimaadhoo (MCOH)

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## Abstract

Water soluble inorganic components in rain deposited at the Maldives Climate Observatory Hanimaadhoo (MCOH) were examined to determine seasonality and possible source regions. The study, which is part of the international Atmospheric Brown Cloud (ABC) project, covers the period June 2005 to December 2007. Air mass trajectories were used to separate the data into situations with transport of air from the Asian continent during winter (December–April) and those with southerly flow from the Indian Ocean during the monsoon season (June–September). The concentrations of  $\text{nss-SO}_4^{2-}$ ,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$  and  $\text{H}^+$  were more than a factor of 4 higher in winter than during the monsoon season. This shows a pronounced influence of continental pollutants during winter. The average rainwater pH was significantly lower in winter (4.7) than during the monsoon (6.0). The lower pH in winter is probably due to a more rapid decrease in the alkaline components than in the acidifying components as air is transported southwards over the Indian Ocean. The moderately high loadings of  $\text{nss-SO}_4^{2-}$  during the monsoon season, supported by our measurements of Methane sulphonate (MSA), indicate that Dimethyl sulphide (DMS) is likely to contribute substantially to the  $\text{nss-SO}_4^{2-}$  concentration during this season. The origin of the high concentration of  $\text{nss-Ca}^{2+}$  during the monsoon season – a factor of 4 to 7 higher than during the winter situations with trajectories from the continent – is unclear. We discuss various possibilities including long-range transport from the African or Australian continents, calcareous plankton debris and exopolymer gels emitted from the ocean surface. The occurrence of  $\text{NO}_3^-$  and  $\text{NH}_4^+$  during the monsoon season suggests emissions from the ocean surface. Part of the  $\text{NO}_3^-$  could also be associated with lightning over the ocean. Despite the fact that the concentrations of  $\text{nss-SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$  were highest during the winter season their wet deposition was at least as high during the monsoon season reflecting the larger amount of rainfall in this season. The annual wet deposition of these components was comparable to that observed in the eastern US and in India but substantially higher than what has been observed on Amsterdam Island in the Southern Indian Ocean.

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## 1 Introduction

The Indian Ocean Experiment (INDOEX) intense field campaign carried out during January to April 1999 illuminated and described the phenomenon of brown clouds over South Asia and Northern Indian Ocean due to long range transport of anthropogenic aerosol particles during the dry winter monsoon season. During this season the continental pollutants from Northern Hemisphere are transported by a north-easterly flow towards the inter-tropical convergence zone (ITCZ). The brown clouds consist of a mixture of anthropogenic sulfate, nitrate, organics, black carbon, dust, fly ash and natural aerosol particles such as sea salt and mineral dust. The anthropogenic particles contribute about 80% to the total aerosol load over most of south Asia and northern Indian Ocean (Ramanathan et al., 2001a; Guazzotti et al., 2003). The composition of brown clouds has significant implications for radiative forcing of climate, the hydrological cycle, agriculture and health (Ramanathan et al., 2001b; Guazzotti et al., 2001; Gabriel et al., 2002; Ramanathan et al., 2005; Stone et al., 2007; Corrigan et al., 2006; Ramanathan et al., 2007a, b; Corrigan et al., 2008).

In response to the findings of INDOEX the international Atmospheric Brown Cloud (ABC) project was designed to address the environmental impact of the brown clouds on regional and global scales. The project specifically aimed at monitoring radiative effects, climate change and atmospheric pollution in the Asia Pacific region (Ramanathan and Crutzen, 2003).

One of the key sites for observations related to the brown clouds in South Asia is the Maldives Climate Observatory at Hanimadhoo (MCOH) located at 6°46'N and 73°11'E in the northern part of Hanimadhoo island in the Republic of Maldives (Fig. 1). The island is situated in the upper north east corner of Maldivian archipelago. The weather is characterized by the dry winter monsoon season during November to April and the wet monsoon season during June to October. The north-easterly winds carry polluted air from the Asian continent, including the Middle east, out over the Indian Ocean during winter while the south-easterly winds during the wet monsoon season – turning

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south-westerly before they reach the continent – brings tropical remote marine air that provides India with most of its annual rainfall. The MCOH therefore serves as an ideal location for sampling air coming either from Asia or from the Southern Hemisphere and it is free from significant local sources of pollution. Further details of the monsoon system and pollution transport are described elsewhere (Krishnamurti et al., 1998; Verver et al., 2001; Ramachandran, 2005; Momin et al., 2005; Corrigan et al., 2006).

Scavenging by precipitation is an important mechanism for removing pollutants from the atmosphere. Measurements of the chemical composition of precipitation are therefore a fundamental prerequisite for estimating the cycling through the atmosphere of components occurring in aerosol particles and as soluble gases. Such measurements also provide information on the input of anthropogenic and natural components to terrestrial, aquatic and marine ecosystems. Precipitation chemistry studies in the South Asian region have been carried out mainly in India (Kulshrestha et al., 2005; Mouli et al., 2005; Momin et al., 2005; Tiwari et al., 2007; Rastogi and Sarin, 2007; Salve et al., 2008; Budhavant et al., 2009). No systematic long-term measurements have been carried out previously in the Maldives or at other locations in the northern parts of the Indian Ocean. The present paper describes the chemical composition of rainwater at MCOH collected during the period June 2005 to December 2007. The aim of the study is to characterize the seasonality of the inorganic components of the rain deposited at MCOH and to identify possible anthropogenic and natural sources. In a separate paper (Granat et al., 2010) a comparison is made between the chemical composition of rainwater and of aerosol particles collected in air at MCOH. The main emphasis of that paper was on the scavenging characteristics of light absorbing material (soot) compared to other components.

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## 2 Methods

### 2.1 Sampling equipment

Precipitation samples were collected during the period June 2005 to December 2007 on a daily basis at 6 a.m. – sometimes also a second time at 6 p.m. (local time) – using two wet-only collectors of MISU (Department of Meteorology, Stockholm University, Sweden) design located outside the railing at the top of a 15 m tower at MCOH. The collector consists of a cylindrical part with a polyethylene funnel (diameter ca 200 mm) and a bottle inside and with a polypropylene lid making a tight seal against the collector. The funnel has a screw cap moulded to the spout to give a watertight connection to the collecting bottle with no possibility for rain to enter from the side of funnel. A second wet-only collector of the same type but with funnel and bottle made of glass (Granat et al., 2010) was used primarily for sampling of soot but also as a quality check on sampling reproducibility for inorganic compounds. At least once a week and when found visibly contaminated the wet-only collector was cleaned as part of quality control scheme using a plastic brush and de-ionized water (18 M $\Omega$  cm resistivity) from a spray bottle in different combinations. The brush was kept in a plastic bag between each cleaning. Plastic gloves were used during collection of rain samples and at funnel washing. The samples were transferred to 50 cm<sup>3</sup> polyethylene bottles for shipping and analysis by ion chromatography (IC) during clean room conditions at MISU. The amount of each sample collected was determined by weighing on an electronic balance. To prevent biological degradation in the rainwater samples during storage and transport, a preservative was added in advance at MISU to the empty transport bottles (to give 400 mg Thymol per dm<sup>3</sup> (Gillett and Ayers, 1991)). By this procedure any spill of Thymol in the station environment was eliminated. All samples were stored in a refrigerator (4 °C) at the observatory and after arrival at MISU.

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## 2.2 Analysis of the rain samples

After filtration the rainwater samples were analyzed for major cations (ammonium:  $\text{NH}_4^+$ , sodium:  $\text{Na}^+$ , potassium:  $\text{K}^+$ , magnesium:  $\text{Mg}^{2+}$  and calcium:  $\text{Ca}^{2+}$ ) and anions (fluoride:  $\text{F}^-$ , chloride:  $\text{Cl}^-$ , formate:  $\text{HCOO}^-$ , acetate:  $\text{CH}_3\text{COO}^-$ , methane sulfonate, MSA:  $\text{CH}_3\text{SO}_2\text{O}^-$ , nitrate:  $\text{NO}_3^-$ , bicarbonate:  $\text{HCO}_3^-$ , and sulfate:  $\text{SO}_4^{2-}$ ) by ion chromatography (Dionex ICS-2000). pH and conductivity was analysed in an unfiltered fraction of each sample. The anions were analyzed with Dionex AG11/AS11 columns and an ASRS-II auto suppressor mode, eluent potassium hydroxyl 0.2 mM at  $0.5 \text{ cm}^3 \text{ min}^{-1}$  for weak anions (MSA), gradually increasing to 25 mM to elute strongly retained ions. In samples collected before February 2007 the analysis of cations was made with Dionex CG12A/CS12A columns (column temperature  $20^\circ\text{C}$ ) and a CSRS-II auto suppressor with external water mode and 18.5 mM MSA eluent (isocratic) with a flow of  $0.25 \text{ cm}^3 \text{ min}^{-1}$ . From February 2007 the analyses of cations were made with Dionex CG16/CS16 columns (column temperature  $40^\circ\text{C}$ ) with 30 mM MSA using an eluent generator with Milli-Q water. pH was measured with Orion model 720 pH meter with combination glass electrode Ross sure flow (model for low ionic strength samples). Conductivity was measured with Orion conductivity meter 120.

All samples collected contained a certain amount of sea salt. To be able to apportion the measured quantities to different sources it is necessary to estimate the non-sea salt (nss) concentration of the various components. This was calculated using the observed rain-water concentrations of  $\text{Na}^+$  as the reference element and assuming that all  $\text{Na}^+$  is of marine origin (Keene et al., 1986). The non sea salt concentration of any particular component "X" is calculated based on the known sea water ratios with respect to  $\text{Na}^+$ :

$$[\text{nss-X}] = [\text{X}_{\text{rain}}] - [\text{Na}^+_{\text{rain}}] \left\{ \text{X}/\text{Na}^+ \right\}_{\text{seawater}}$$

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## 2.3 Data quality

A quality check of the analyses was done based on laboratory produced test samples and certified reference samples. Random error, measured with a synthetic sample in each batch, given as one standard deviation, was 10% for cations and 3% for anions (excluding  $\text{HCO}_3^-$ ). Systematic errors as evaluated by an inter-laboratory test (organised by EANET, 2008) with concentrations similar to the present ones were less than 2% (with exception for  $\text{Mg}^{2+}$  with less than 3%) for all components except  $\text{HCO}_3^-$  that was not included in the test. The quality of the chemical analyses was also assessed by the ion balance (sum of anions versus sum of cations, Fig. 2) and a comparison between measured and calculated conductivity (Fig. 3). The average difference between anions and cations ( $A-C/(A+C)$ ) was 2%, well below the limits recommended by Mohnen et al. (1994) for precipitation chemistry analyses. The  $R^2$  values for the ion and conductivity balance was close to 0.98.

The sampling quality in terms of a random error added to measured concentrations was estimated from samples simultaneously collected with the two wet-only collectors. 103 samples from the glass wet-only collector were also analysed and the results compared with those from the plastic collector, intended for determination of soluble components. For sea salt ( $\text{Na}^+$ ) there was a good agreement between the two collectors, close to the analytical reproducibility, in 87 out of the 101 samples. In 13 samples the  $\text{Na}^+$  concentration was substantially higher in the glass collector while in one case the concentration was higher in the plastic collector. The likely explanation is that the lid opening/closing mechanism failed in some cases for the glass collector and in one for the plastic collector and left the collector open also during a dry periods during which sea salt was deposited in the open collector. For  $\text{nss-SO}_4^{2-}$  there was a good agreement for all samples (slope 0.99 and  $r^2=0.98$ ). Obviously, the  $\text{nss-SO}_4^{2-}$ , with most of the mass in the fine particle mode, did not deposit to an appreciable amount during exposed, dry periods. The lid, when closed, was found to prevent any entry of sea salt in the collector even during extended dry periods.

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The agreement of  $\text{NH}_4^+$  concentrations between collectors was quite poor. This is explained by the uncertainty in determining low concentrations of  $\text{NH}_4^+$  in the presence of high concentrations of  $\text{Na}^+$ . The standard deviation of  $\text{NH}_4^+$  for individual samples relative to average for a single collector was 0.4. In the case of nss- $\text{K}^+$  it was even larger (1.1), an obvious effect of analytical uncertainty when random error for the total  $\text{K}^+$  concentration is related to the much smaller concentration of nss- $\text{K}^+$ .

## 2.4 Trajectories

Daily 10 days receptor-oriented trajectories were calculated for MCOH using the Hysplit 4.8 model from the National Oceanic and Atmospheric Administration (NOAA) (Draxler and Rolph, 2003). Arriving heights were set at 50, 1000 and 2000 m and time at 12:00 GMT (5 p.m. local time). The trajectories were classified in four groups:

1. *Marine*: trajectories at all arrival heights arriving from the west-south-southeast sector with no land contact during the last 10 days (Fig. 4a). Most of these trajectories originated south east of MCOH reaching the site after a clockwise turn during the last few days.
2. *Arabian Sea*: Trajectories traversing the eastern parts of the Arabian Sea arriving at MCOH after possible land contact with western India or other mostly arid regions surrounding the Arabian Sea (Fig. 4b).
3. *Indian*: Trajectories having spent at least 2 of the last 10 days over polluted parts of south Asia and with at most 4 days over the ocean before reaching MCOH (Fig. 4c).
4. *Mixed*: Trajectories which do not belong to either of the above three groups (Fig. 4d). Because of its heterogeneity this trajectory group is not analysed in detail.

The air flow over the Maldives is dominated by the winter circulation during November–April bringing dry air from the Asian continent arriving at the Maldives from

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a north-easterly or northerly direction (c.f. Fig. 4b and c) and the monsoon circulation from late June to October bringing moist air from the southern Indian Ocean (Fig. 4a).

## 2.5 Statistical analysis

To search for statistical, and possibly causal, relationships between two or more of the observed ionic data (daily averaged) correlations were calculated using STATISTICA 6.0. This was done separately for the trajectory groups Marine, Arabian Sea and Indian, c.f. Tables 2, 4 and 6. In these tables correlation coefficients significant at 95% level have been highlighted bold.

A principal component analysis (PCA) was also applied to the data. This is a simplified representation of the data, which identifies the relationship among the variables and provides indications of their source types based on the nature of factors obtained (Seto et al., 2000; Topcu et al., 2002; Semionov et al., 2003; Astel et al., 2004). In principle, PCA produces a linear combination of the variables, which explains a large fraction of the variance of the data. These linear combinations represent factors that are obtained by computing a correlation matrix between the variables. Varimax rotated mode of PCA was applied in this study. The software package used in this study is STATISTICA 6.0. The statistically significant rotated factor loadings ( $\geq 0.70$ , marked bold) and the explained variance for the three trajectory groups are presented in Tables 3, 5 and 7.

## 3 Results and discussion

### 3.1 An overview of the precipitation chemistry data

Figure 5 shows the temporal development of all data at a monthly resolution, based on volume weighted mean (vwm) values for each month. For obvious reasons the seasonal distribution of the number of observations is uneven with few measurements

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5 during the period December through May (no rainfall events at all for February, Fig. 5a). The components  $\text{NH}_4^+$  (Fig. 5c),  $\text{NO}_3^-$  (Fig. 5g),  $\text{nss-SO}_4^{2-}$  (Fig. 5h) and  $\text{H}^+$  (from pH, Fig. 5b) all show highest mean concentrations during the winter season (December–April), indicating an impact of continental pollutants. The concentration of  $\text{nss-Ca}^{2+}$  (Fig. 5f), on the other hand was more abundant during the monsoon season (June and July). Possible reason for this seasonality will be discussed in Sect. 3.3. The concentration of both  $\text{K}^+$  (Fig. 5d) and  $\text{Mg}^{2+}$  (Fig. 5e) showed a less clear-cut seasonal distribution.

### 3.2 Data separated into trajectory groups

10 To evaluate possible source regions for the precipitation collected the MCOH observatory, the volume weighted mean (vwm) values of the data were separated into the three well defined trajectory groups (Marine (1), Arabian Sea (2) and Indian (3)) plus a fourth, less well defined group, see Table 1.

15 The major sea salt components  $\text{Na}^+$  and  $\text{Cl}^-$  were found to be strongly correlated in all groups (Tables 2, 4 and 6). They were also the major rainwater components in all trajectory groups. The strong correlation between  $\text{Na}^+$  and  $\text{Cl}^-$ , and the similarity between the observed ratio  $\text{Cl}^-/\text{Na}^+$  (1.14, 1.12 and 1.12 in the trajectory groups respectively) to that of sea water (1.17) shows that both  $\text{Na}^+$  and  $\text{Cl}^-$  ions were primarily derived from marine sources (Khemani, 1998; Hegde et al., 2007).

20 The high values of sea salts during the monsoon season are related to the prevailing winds, which activate aerosol production from sea surface through bubble bursting (Blanchard, 1963; Norman et al. 2003). The contribution of  $\text{nss-SO}_4^{2-}$  to the total  $\text{SO}_4^{2-}$  in this season is around 25%. This moderately high loading of  $\text{nss-SO}_4^{2-}$  in PCA factor 1 during the monsoon season (Table 3) points towards a marine biogenic sulfur source as suggested by Norman et al. (2003) and Leck et al. (2002). A number of possible mechanisms have been suggested to explain the association of  $\text{nss-SO}_4^{2-}$  with marine aerosol particles. Gaseous sulfur dioxide ( $\text{SO}_2$ ) can participate in the  $\text{nss-SO}_4^{2-}$

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production under the conditions existing in the Marine boundary layer (MBL). Oxidation of  $\text{SO}_2$  in presence of ozone (Arndt et al., 1997) and hydrogen peroxide (Chameides and Stelson, 1992) contributes to  $\text{nss-SO}_4^{2-}$  concentration in cloud and rain water. The concentration of MSA was about  $0.05 \mu\text{eqdm}^{-3}$  in the Marine trajectory group and at least 30 times lower in the two groups with flow from the Asian continent, including the Middle East (Table 1). These values will be used in Sect. 3.4 to estimate the amount of  $\text{nss-SO}_4^{2-}$  contributed by dimethyl sulfide (DMS) produced by marine phytoplankton (Leck et al., 1990).

The mean concentrations of  $\text{NH}_4^+$ ,  $\text{NO}_3^-$  and  $\text{nss-SO}_4^{2-}$  were more than a factor of four larger in the Indian trajectory group compared to the Marine group. The anthropogenic origin of these components is also indicated by the PCA analysis (Table 5). In the Indian group the most loaded factor (48%) is clearly anthropogenic in nature (combustion processes for nitrogen oxides ( $\text{NO}_x$ ) and  $\text{SO}_2$  and agriculture for ammonia ( $\text{NH}_3$ )) and includes  $\text{H}^+$ ,  $\text{NH}_4^+$ ,  $\text{nss-K}^+$ ,  $\text{NO}_3^-$  and  $\text{nss-SO}_4^{2-}$  whereas in the Arabian group the most loaded factor (58%) seems to carry a signal including a mixture of anthropogenic and crustal sources ( $\text{NH}_4^+$ ,  $\text{nss-K}^+$ ,  $\text{nss-Mg}^{2+}$ ,  $\text{nss-Ca}^{2+}$  and  $\text{nss-SO}_4^{2-}$ ).

It can be seen in Table 1 that the  $\text{nss}$  fractions of  $\text{K}^+$  and  $\text{Mg}^{2+}$  in all trajectory groups represent a small part of the total concentrations, which make it difficult to draw any conclusions concerning their origin. In trajectory groups 2 and 3  $\text{nss-K}^+$  values are likely to be influenced by anthropogenic biomass burning and combustion processes (Andreae, 1983; Norman et al., 2003) as well as crustal sources.

In the Marine and Arabian Sea trajectory groups the  $\text{nss-Ca}^{2+}$  is more than half of the total  $\text{Ca}^{2+}$  clearly indicating significant sources other than sea salt. The mean value of  $\text{nss-Ca}^{2+}$  in the Marine group is substantially larger than in the Indian and Arabian Sea groups. This surprising feature will be further discussed in Sect. 3.3.

The frequency distributions of pH in the different groups are shown in Fig. 6. The rainwater pH varies systematically between the three trajectory groups with the lowest volume weighted mean value ranging from 4.7 in the Indian group (Fig. 6b) to 6.0 in the Marine group (Fig. 6a). A pH value as low as 4.0 was recorded on one occasion with air

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flow from the Indian continent. The pH values are determined by a balance between acids – as seen from  $\text{nss-SO}_4^{2-}$  and  $\text{NO}_3^-$  – and neutralizers such as carbonate and  $\text{NH}_3$  as seen from the concentration of  $\text{nss-Ca}^{2+}$  and  $\text{NH}_4^+$  in the collected samples. It should be noted that the number of rainfall days is much smaller in the two groups with northerly flow than in the Marine group. A comparison with pH values measured in rain falling in the Indian subcontinent during winter season – e.g. typical values  $>6.0$  at rural sites in India (Kulshrestha et al., 2005) – reveals that pH has a tendency to decrease as air is transported out over the Indian Ocean. The main reason is probably that the concentration of  $\text{CaCO}_3$  from soil dust – which occurs mainly in the coarse particle mode, Granat et al. (2010) – decreases more rapidly than the concentration of the acidifying components sulfuric and nitric acids of which at least the sulfuric acid is predominantly associated with the fine particle mode. A similar observation has been made previously by Kulshrestha et al. (2001), Granat et al. (2002) and Norman et al. (2003).

### 3.3 Sources of $\text{nss-Ca}^{2+}$

As pointed out above the concentration of  $\text{nss-Ca}^{2+}$  in the Marine trajectory group was surprisingly high – on average four times higher than in the Arabian Sea trajectory group and seven times higher than in the Indian group. If continental soil dust were the main source one would have expected the highest concentrations associated with the northerly trajectories.

Several studies have attributed  $\text{Ca}^{2+}$  enrichment in marine aerosol and precipitation to transported crustal dust from distant continents and/or locally produced wind-blown calcareous shell debris. A model study over Indian Ocean (Adhikary et al., 2007) suggested that aerosol mass composition at Hanimadhoo during the monsoon season is dominated by mineral dust in both coarse and fine modes. The dust was transported eastwards from Africa and then brought into the Asian monsoon circulation. In the present study, the 10 days trajectories arriving at Hanimadhoo do not show any recent

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contact with land. We extended trajectories for days with particularly high concentrations of  $\text{Ca}^{2+}$  in rain to 20 days and saw a few cases where they originate close to Australia.

Direct emission of calcareous ( $\text{CaCO}_3$ ) plankton debris in association with marine aerosol production from bursting bubbles has been suggested to be a significant in situ source in the marine atmosphere (Sievering et al., 2004). Another possibility of  $\text{Ca}^{2+}$  enrichment in marine air could be through its association with organic matter (Keene et al., 2007). One further explanation that extends the conclusion by Keene et al., is the possibility that the nss- $\text{Ca}^{2+}$  originates from exopolymer gels or marine gels (Decho, 1990) consisting of very hydrophilic polysaccharides inter-bridged with divalent ions, preferable  $\text{Ca}^{2+}$ , present in the ocean surface layer. Such gels have been shown to be an important component of the tropical marine aerosols (Leck and Bigg, 2008 and Bigg and Leck, 2008). A final possibility to explain the high values of nss- $\text{Ca}^{2+}$  might be contamination from very local emissions on the Hanimaadhoo island. But if that were the case one would have expected a larger influence in aerosols than in rainwater since rainwater collects much of its aerosol material at cloud levels where local contamination is much less likely. According to Granat et al. (2010), such larger influence was not observed.

### 3.4 Sources of nss- $\text{SO}_4^{2-}$ during the monsoon season

There are strong indications that most of the nss- $\text{SO}_4^{2-}$  in the both the Arabian Sea and Indian trajectory groups is causally related to anthropogenic pollution carried to the Maldives by the northerly or north-easterly winds during the winter season. This conclusion is supported both by correlations with other pollution components and by the factor analysis.

In order to estimate if the moderately high loading nss- $\text{SO}_4^{2-}$  in Marine group could be derived from DMS, we calculated the molar ratio of MSA to nss- $\text{SO}_4^{2-}$  in the collected rainwater. Several field studies have investigated the factors controlling the airborne

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particulate molar ratio of MSA to  $\text{nss-SO}_4^{2-}$  and found a negative temperature dependence (Bates et al., 1992; Ayers et al., 1996; Leck et al., 2002). The mean molar ratio value was around of 1.3% in this study, which is in agreement with the estimate of 1

5 This is somewhat lower than the 3% reported by Norman et al. (2003) from ship measurements in an area south of MCOH in air that had been over the ocean for at least 10 days. Given the uncertainties in the underlying data, both in this study and in Norman et al., we regard DMS being the major source for the  $\text{nss-SO}_4^{2-}$  observed in the Marine group. In trajectory group 2 (Indian) the molar ratio of MSA to  $\text{nss-SO}_4^{2-}$  ratio  
10 was only 0.003% indicating that DMS was a minor source with anthropogenic sources dominating.

### 3.5 Sources of $\text{NO}_3^-$ and $\text{NH}_4^+$

Both  $\text{NO}_3^-$  and  $\text{NH}_4^+$  occur in higher concentration in the two groups with northerly flow compared to the Marine group (Table 1). The ratio between the mean concentrations  
15 in Indian group to that in the Marine group is 7 and 5.5 for  $\text{NH}_4^+$  and  $\text{NO}_3^-$ , respectively. Both these ions have high loading in the “pollution factor” in the two northerly groups and also correlate well with  $\text{nss-SO}_4^{2-}$  in these groups, both facts pointing strongly at anthropogenic sources on the Asian continent. Substantial amounts of anthropogenic aerosol containing nitrogenous and sulphurous compounds are thus carried by the  
20 winds towards the study site in winter.

A strong correlation between  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and  $\text{nss-SO}_4^{2-}$  is observed also in the Marine trajectory group. The source of  $\text{NO}_3^-$  in this group is unclear. One possibility is oxidation of  $\text{NO}_x$  produced by lightning (Hauglustaine et al., 2001). Nitrogen is also a  
25 key element for algal metabolism because it is a main component of algal proteins and enzyme catalyst capacity, and often limits plant growth and biomass (e.g. Lapointe, 1987).

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Major sources of  $\text{NH}_3$  in remote marine surface water are degraded organic nitrogen compounds and excretion from zooplankton while sink pathways mainly include bacterial nitrification, uptake by phytoplankton and ventilation to the atmosphere (Bouwman et al., 1997; Norman and Leck, 2005). The correlation between  $\text{NH}_4^+$  and  $\text{nss-SO}_4^{2-}$  could be due to an emission of  $\text{NH}_3$  that is limited by the amount of sulphuric acid formed in the marine boundary layer, in agreement with earlier suggestions by Norman and Leck (2005).

### 3.6 Wet deposition

In this section we consider the amount of the various components in rainwater that is deposited each month, Table 8. The wet deposition is obtained as the product of the measured concentration in the rainwater and the amount of rain measured in the precipitation chemistry sampler.

By providing data on input of both nutrients – e.g. nitrogen – and potentially harmful components – e.g. acidity – this information can be of considerable importance for studies of Maldivian ecosystems. However, deposition associated with rainwater (wet deposition) is only one of the pathways by which ecosystems can receive input from the atmosphere. Other processes include direct uptake of gases and particulate matter from the air (dry deposition) and deposition of spray droplets from the surf on the windward side of the island. Our data do not allow reliable quantitative estimates of these additional deposition processes but rough estimates indicate that the dry deposition of gaseous and particle-bound sulfur and nitrogen compounds is much less important than wet deposition, at least on an annual basis.

A limited number of measurements of the concentration of  $\text{SO}_2$  and nitric acid ( $\text{HNO}_3$ ) at MCOH during the monsoon period makes it possible to estimate an approximate magnitude of the dry deposition of these gases. With a typical concentration for  $\text{SO}_2$  of  $1.5 \text{ nmol/m}^3$  and for  $\text{HNO}_3$  of  $0.4 \text{ nmol/m}^3$  and dry deposition velocities of 1 and 4 cm/s, respectively the dry deposition flux for these gases is two orders of magnitude smaller than the corresponding wet deposition values.

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Table 8 shows that the annual cycle of deposition for most components observed is characterized by a maximum during the monsoon season (June–September) obviously associated with the maximum in rainfall during this season. The deposition of  $\text{NH}_4^+$ ,  $\text{NO}_3^-$  and  $\text{nss-SO}_4^{2-}$  has a more even seasonal distribution, where higher concentrations during the winter season are balanced by lower rainfall amounts. Input of net acidity only occurs during the winter season (December–April). The alkaline nature of the rainwater (positive values of net  $\text{HCO}_3^-$ ) during the monsoon season seems to be due to alkali associated with  $\text{Ca}^{2+}$  and low concentration of acidifying components. The contribution from  $\text{NH}_3$ , as viewed from the  $\text{NH}_4^+$  concentration, is small during the monsoon season.

In Table 9 we compare the measured deposition values with data from some other regions. The MCOH values are comparable in magnitude to those in the US of America and only moderately lower than those in India. The values from Amsterdam Island, which has a more isolated location in the southern Indian Ocean, are substantially lower.

In order to judge the ecological significance of the wet deposition values measured at MCOH, information is needed on the status of the Maldivian ecosystems and the characteristics of the soils. Any negative effects due to eutrophication (over-fertilization) through excessive nitrogen deposition are unlikely. Even if an additional contribution from dry deposition of nitrogen compounds is included the total nitrogen input to Maldivian ecosystems is likely to be well below the so called “critical load” for nitrogen deposition which in many European and North American terrestrial and aquatic ecosystems has been estimated to be above  $500 \text{ mgN/m}^2/\text{yr}$  (Bobbink et al., 2010). Still, the wet deposition of nitrogen measured at MCOH clearly indicates a substantial nutrient input to Maldivian terrestrial ecosystems, with a large part of it being due to natural sources of nitrogen.

## 4 Conclusions

This paper summarizes the results of 30 months of data on the chemical composition of precipitation at the MCOH site in the northern Indian Ocean. The measurement period includes both the monsoon season (June–October) with air arriving from more southerly parts of the Indian Ocean and the winter season (November–April) with air flow from the Asian continent. For  $\text{nss-SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and  $\text{H}^+$  the concentrations during the winter season were more than four times higher than during the monsoon season, indicating a strong anthropogenic influence during the former season.

Rainwater pH was substantially lower in air emanating from the Asian continent (mean pH=4.7 in trajectory group “Indian”) than in the monsoon air (6.0 in trajectory group “Marine”). On one occasion in the “Indian” group the pH was as low as 4.0. This is much lower than what is normally recorded in rainfall on the Indian Subcontinent. A systematic decrease in pH seems to occur in air transported out over the Indian Ocean during the winter season possibly caused by a more rapid decrease in the concentration of alkaline soil dust (coarse mode particles) than of the acidifying sulphur and nitrogen compounds (fine mode).

During both seasons  $\text{Cl}^-$  and  $\text{Na}^+$  were the most abundant components. These ions were also highly correlated and their ratio was close to that found in sea water. A comparison with  $\text{Na}^+$  showed that also for several other components an appreciable part of the concentration was derived from sea-salt. The concentration of the non sea-salt  $\text{SO}_4^{2-}$  ( $\text{nss-SO}_4^{2-}$ ) was lower during the monsoon season than during winter, but only by a factor of two to four. The PCA analysis indicated that a marine biogenic source is involved during the monsoon season. This conclusion is supported by our measurements of MSA which indicated that DMS from the ocean is likely to have made a substantial contribution to the concentration of  $\text{nss-SO}_4^{2-}$  in this season.

Non-negligible amounts of  $\text{NO}_3^-$  and  $\text{NH}_4^+$  were found also in the “Marine” trajectory group. Whereas at least some of the  $\text{NO}_3^-$  may be derived from  $\text{NO}_x$  emissions during lightning most of the  $\text{NH}_4^+$  is likely to have been emitted as  $\text{NH}_3$  from the ocean.

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The high concentration of nss-Ca<sup>2+</sup> during the monsoon season – a factor of four to seven higher than during winter days with trajectories in groups “Arabian Sea” and “Indian” – is intriguing. The source of this nss-Ca<sup>2+</sup> is unclear. Long-range transport of soil dust, as suggested by Adhikary et al. (2007) is one possibility although it is not well supported by our trajectory analysis. We discuss other possibilities, including calcareous (CaCO<sub>3</sub>) plankton debris and exopolymer gels emitted from the ocean surface.

The annual wet deposition of NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup> and nss-SO<sub>4</sub><sup>2-</sup> at MCOH is comparable to that observed in India and in eastern parts of North America. The corresponding values at the remote Amsterdam Island in the southern Indian Ocean are roughly a factor of ten lower. This shows that the chemical climate of the Maldives is clearly affected by pollution sources on the Asian continent during the winter season. The atmospheric deposition of nitrogen is likely to represent an important input of nutrients to marine and terrestrial ecosystems in the Maldives.

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**Table 1.** Volume weighted annual mean values (in  $\mu\text{eq/l}$ ) of the measured quantities separated into trajectory groups.

vwm	Marine	Arabian Sea	Indian	Mixed
net $\text{H}^+$	0.0	6.3	10	0.0
net $\text{HCO}_3^-$	22	0.0	0.0	14
$\text{Na}^+$	170	43	146	82
$\text{NH}_4^+$	2.1	9.5	15	2.5
$\text{K}^+$	4.1	1.7	4.3	1.9
nss- $\text{K}^+$	0.3	0.8	1.1	0.1
$\text{Mg}^{2+}$	40	11	39	20
nss- $\text{Mg}^{2+}$	1.3	1.6	5.9	0.9
$\text{Ca}^{2+}$	27	7.0	9.4	16
nss- $\text{Ca}^{2+}$	20	5.1	2.9	12
MSA *100	4.5	0.0	0.1	7.1
$\text{Cl}^-$	195	49	164	93
nss- $\text{Cl}^-$	-3.5	-1.8	-6.3	-2.3
$\text{NO}_3^-$	3.3	9.1	18	3.2
$\text{SO}_4^{2-}$	27	21	47	18
nss- $\text{SO}_4^{2-}$	6.8	16	29	8.3

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**Table 2.** Correlation coefficients for rainwater ions in Marine trajectory group.

Marine	H <sup>+</sup>	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	nss-K <sup>+</sup>	nss-Mg <sup>2+</sup>	nss-Ca <sup>2+</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	HCO <sub>3</sub> <sup>-</sup>	nss-SO <sub>4</sub> <sup>2-</sup>
H <sup>+</sup>	1.00	-0.12	0.05	0.05	0.07	-0.16	-0.13	-0.06	-0.10	-0.06
Na <sup>+</sup>	-0.12	1.00	-0.05	0.07	<b>0.21</b>	<b>0.19</b>	<b>1.00</b>	<b>0.23</b>	0.04	0.08
NH <sub>4</sub> <sup>+</sup>	0.05	-0.05	1.00	<b>0.49</b>	0.05	0.03	-0.04	<b>0.67</b>	-0.03	<b>0.56</b>
nss-K <sup>+</sup>	0.05	0.07	<b>0.49</b>	1.00	0.02	0.09	0.06	<b>0.56</b>	-0.02	<b>0.49</b>
nss-Mg <sup>2+</sup>	0.07	<b>0.21</b>	0.05	0.02	1.00	<b>0.35</b>	<b>0.21</b>	0.12	<b>0.20</b>	0.08
nss-Ca <sup>2+</sup>	-0.16	<b>0.19</b>	0.03	0.09	<b>0.35</b>	1.00	<b>0.19</b>	<b>0.34</b>	<b>0.58</b>	<b>0.61</b>
Cl <sup>-</sup>	-0.13	<b>1.00</b>	-0.04	0.06	<b>0.21</b>	<b>0.19</b>	1.00	<b>0.24</b>	0.05	0.09
NO <sub>3</sub> <sup>-</sup>	-0.06	<b>0.23</b>	<b>0.67</b>	<b>0.56</b>	0.12	<b>0.34</b>	<b>0.24</b>	1.00	0.14	<b>0.73</b>
HCO <sub>3</sub> <sup>-</sup>	-0.10	0.04	-0.03	-0.02	<b>0.20</b>	<b>0.58</b>	0.05	0.14	1.00	<b>0.30</b>
nss-SO <sub>4</sub> <sup>2-</sup>	-0.06	0.08	<b>0.56</b>	<b>0.49</b>	0.08	<b>0.61</b>	0.09	<b>0.73</b>	<b>0.30</b>	1.00

\* Bold font indicates that the correlation is significant at 95% level.

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**Table 3.** A summary of the factor loadings and total variance identified in the Marine data set.

Marine	Factor1	Factor2	Factor3
H <sup>+</sup>	0.05	−0.15	−0.24
Na <sup>+</sup>	0.06	<b>0.98</b>	0.12
NH <sub>4</sub> <sup>+</sup>	<b>0.84</b>	−0.09	−0.11
nss-K <sup>+</sup>	<b>0.78</b>	0.05	−0.10
nss-Mg <sup>2+</sup>	0.02	0.23	0.46
nss-Ca <sup>2+</sup>	0.23	0.07	<b>0.88</b>
Cl <sup>−</sup>	0.06	<b>0.98</b>	0.13
NO <sub>3</sub> <sup>−</sup>	<b>0.88</b>	0.18	0.18
HCO <sub>3</sub> <sup>−</sup>	0.01	−0.10	<b>0.83</b>
nss-SO <sub>4</sub> <sup>2−</sup>	<b>0.79</b>	−0.03	0.44
% Tot Variance	28	21	20

\* Bold font indicates that the factor loading is significant at 95% level.

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**Table 4.** Correlation coefficients for rainwater ions in the Indian trajectory group.

Indian	H <sup>+</sup>	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	nss-K <sup>+</sup>	nss-Mg <sup>2+</sup>	nss-Ca <sup>2+</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	HCO <sub>3</sub> <sup>-</sup>	nss-SO <sub>4</sub> <sup>2-</sup>
H <sup>+</sup>	1.00	<b>0.59</b>	<b>0.74</b>	<b>0.95</b>	0.32	<b>0.75</b>	0.54	<b>0.90</b>	-0.23	<b>0.90</b>
Na <sup>+</sup>	<b>0.59</b>	1.00	0.38	<b>0.62</b>	<b>0.69</b>	<b>0.60</b>	<b>1.00</b>	<b>0.63</b>	-0.11	0.51
NH <sub>4</sub> <sup>+</sup>	<b>0.74</b>	0.38	1.00	<b>0.81</b>	0.11	0.51	0.32	<b>0.78</b>	0.04	<b>0.83</b>
nss-K <sup>+</sup>	<b>0.95</b>	<b>0.62</b>	<b>0.81</b>	1.00	0.25	<b>0.61</b>	0.57	<b>0.84</b>	-0.26	<b>0.96</b>
nss-Mg <sup>2+</sup>	0.32	<b>0.69</b>	0.11	0.25	1.00	0.36	<b>0.70</b>	0.32	0.01	0.19
nss-Ca <sup>2+</sup>	<b>0.75</b>	<b>0.60</b>	0.51	<b>0.61</b>	0.36	1.00	0.56	<b>0.92</b>	0.21	0.56
Cl <sup>-</sup>	0.54	<b>1.00</b>	0.32	0.57	<b>0.70</b>	0.56	1.00	<b>0.58</b>	-0.13	0.46
NO <sub>3</sub> <sup>-</sup>	<b>0.90</b>	<b>0.63</b>	<b>0.78</b>	<b>0.84</b>	0.32	<b>0.92</b>	<b>0.58</b>	1.00	0.10	<b>0.81</b>
HCO <sub>3</sub> <sup>-</sup>	-0.23	-0.11	0.04	-0.26	0.01	0.21	-0.13	0.10	1.00	-0.11
nss-SO <sub>4</sub> <sup>2+</sup>	<b>0.90</b>	0.51	<b>0.83</b>	<b>0.96</b>	0.19	0.56	0.46	<b>0.81</b>	-0.11	1.00

\* Bold font indicates that the correlation is significant at 95% level.

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**Table 5.** A summary of the factor loadings and total variance identified in the Indian data set.

Indian	Factor1	Factor2	Factor3
H <sup>+</sup>	<b>0.91</b>	0.31	0.14
Na <sup>+</sup>	0.38	<b>0.89</b>	0.06
NH <sub>4</sub> <sup>+</sup>	<b>0.89</b>	0.03	-0.06
nss-K <sup>+</sup>	<b>0.92</b>	0.27	0.22
nss-Mg <sup>2+</sup>	0.02	<b>0.88</b>	-0.05
nss-Ca <sup>2+</sup>	<b>0.67</b>	0.45	-0.39
Cl <sup>-</sup>	0.32	<b>0.91</b>	0.08
NO <sub>3</sub> <sup>-</sup>	<b>0.88</b>	0.36	-0.24
HCO <sub>3</sub> <sup>-</sup>	-0.06	-0.06	<b>-0.96</b>
nss-SO <sub>4</sub> <sup>2-</sup>	<b>0.93</b>	0.15	0.11
% Tot Variance	48	29	12

\* Bold font indicates that the factor loading is significant at 95% level.

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**Table 6.** Correlation coefficients for rainwater ions in the Arabian Sea trajectory group.

Arabian Sea	H <sup>+</sup>	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	nss-K <sup>+</sup>	nss-Mg <sup>2+</sup>	nss-Ca <sup>2+</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	HCO <sub>3</sub> <sup>-</sup>	nss-SO <sub>4</sub> <sup>2-</sup>
H <sup>+</sup>	1.00	-0.26	0.27	0.38	0.01	0.03	-0.26	0.29	-0.05	0.30
Na <sup>+</sup>	-0.26	1.00	-0.04	-0.02	0.16	0.24	<b>1.00</b>	0.18	0.07	0.13
NH <sub>4</sub> <sup>+</sup>	0.27	-0.04	1.00	<b>0.96</b>	<b>0.86</b>	<b>0.90</b>	-0.06	<b>0.91</b>	-0.44	<b>0.95</b>
nss-K <sup>+</sup>	0.38	-0.02	<b>0.96</b>	1.00	<b>0.87</b>	<b>0.89</b>	-0.03	<b>0.91</b>	-0.35	<b>0.95</b>
nss-Mg <sup>2+</sup>	0.01	0.16	<b>0.86</b>	<b>0.87</b>	1.00	<b>0.98</b>	0.14	<b>0.86</b>	-0.37	<b>0.86</b>
nss-Ca <sup>2+</sup>	0.03	0.24	<b>0.90</b>	<b>0.89</b>	<b>0.98</b>	1.00	0.22	<b>0.88</b>	-0.32	<b>0.90</b>
Cl <sup>-</sup>	-0.26	<b>1.00</b>	-0.06	-0.03	0.14	0.22	1.00	0.16	0.08	0.11
NO <sub>3</sub> <sup>-</sup>	0.29	0.18	<b>0.91</b>	<b>0.91</b>	<b>0.86</b>	<b>0.88</b>	0.16	1.00	<b>-0.61</b>	<b>0.93</b>
HCO <sub>3</sub> <sup>-</sup>	-0.05	0.07	-0.44	-0.35	-0.37	-0.32	0.08	<b>-0.61</b>	1.00	-0.35
nss-SO <sub>4</sub> <sup>2-</sup>	0.30	0.13	<b>0.95</b>	<b>0.95</b>	<b>0.86</b>	<b>0.90</b>	0.11	<b>0.93</b>	-0.35	1.00

\* Bold font indicates that the correlation is significant at 95% level.

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**Table 7.** A summary of the factor loadings and total variance identified in the Arabian Sea data set.

Arabian Sea	Factor1	Factor2
H <sup>+</sup>	0.25	-0.49
Na <sup>+</sup>	0.12	<b>0.96</b>
NH <sub>4</sub> <sup>+</sup>	<b>0.97</b>	-0.15
nss-K <sup>+</sup>	<b>0.96</b>	-0.15
nss-Mg <sup>2+</sup>	<b>0.93</b>	0.11
nss-Ca <sup>2+</sup>	<b>0.95</b>	0.18
Cl <sup>-</sup>	0.10	<b>0.97</b>
NO <sub>3</sub> <sup>-</sup>	<b>0.97</b>	0.03
HCO <sub>3</sub> <sup>-</sup>	-0.49	0.16
nss-SO <sub>4</sub> <sup>2-</sup>	<b>0.97</b>	0.00
% Tot Variance	58	22

\* Bold font indicates that the factor loading is significant at 95% level.

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**Table 8.** Wet deposition of net acidity and alkalinity (in  $\text{mmol/m}^2$ ) and other rainwater components (in  $\text{mg/m}^2$ ) by month and as annual total.

Month	mm rain	net H <sup>+</sup>	net HCO <sub>3</sub> <sup>-</sup>	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup> -N	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	MSA	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup> -N	SO <sub>4</sub> <sup>2-</sup> -S	nss-SO <sub>4</sub> <sup>2-</sup> -S
January	28	0.4	0	170	7	8	25	9	0	290	9	26	12
February	0												
March	15	0.1	0	29	3	2	5	7	0	49	4	9	7
April	176	1.3	0	89	23	8	13	16	0	150	22	45	37
May	16	0	0.1	50	5	3	5	7	0	90	4	9	5
June	250	0	4.5	1200	3	45	150	160	0	2100	9	120	26
July	527	0	23.6	1900	20	81	240	470	7	3500	32	260	94
August	443	0	2.5	850	12	36	110	110	2	1500	19	110	41
September	650	0	5.1	1800	17	71	220	170	2	3100	22	190	41
October	256	0	0.4	460	15	20	58	39	0	810	14	80	41
November	162	0	3.4	180	2	4	24	5	0	320	4	42	27
December	100	0.6	0	170	17	10	23	9	0	290	16	51	37
Annual Total	2622	2.4	39	6900	120	290	870	1000	12	12 120	150	940	370



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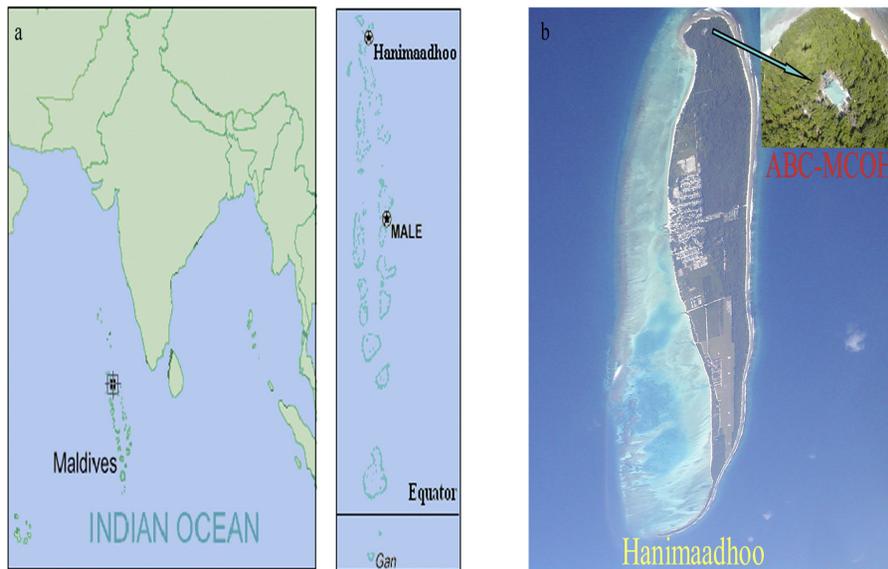
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**Table 9.** Measured annual wet deposition (in mg/m<sup>2</sup>/yr) of selected N and S compounds.

Stations	NH <sub>4</sub> <sup>+</sup> -N	NO <sub>3</sub> <sup>-</sup> -N	nssSO <sub>4</sub> <sup>2-</sup> -S	Reference
MCOH	120	150	370	This work
Amsterdam Island	30	10	60	Baboukas, 2000
India, rural median	140	250	700	Kulshrestha et al., 2005
Eastern US, rural	150–300	100–200	200–500	NADP, 2008



**Fig. 1.** Location of the Maldives Climate Observatory at Hanimaadhoo (MCOH).

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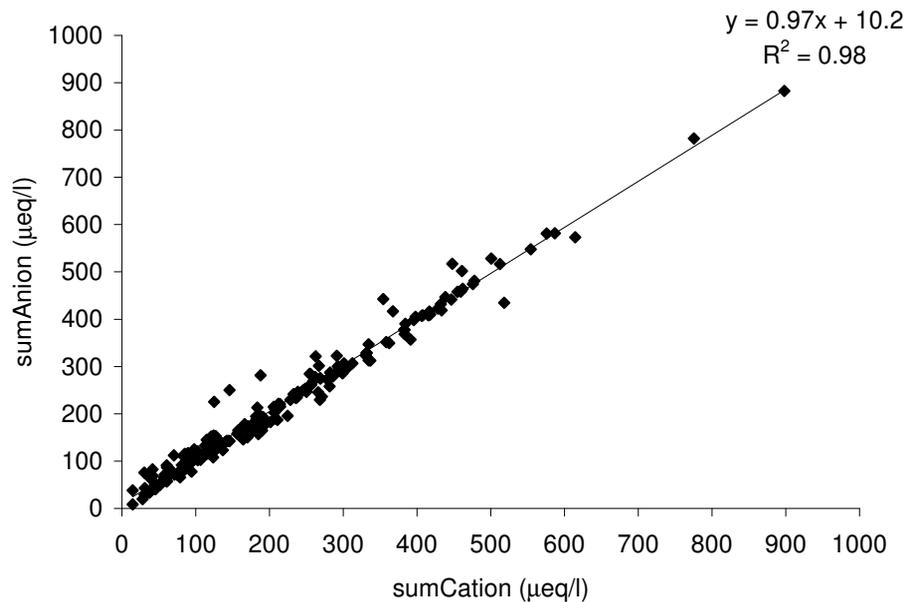
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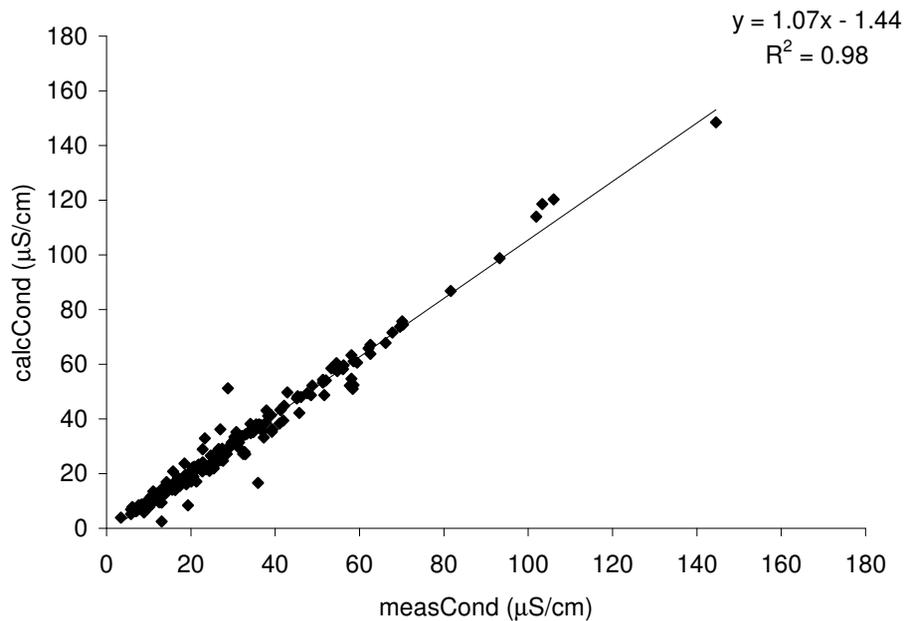
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**Fig. 2.** Comparison between sum of anions (in µeq/l) and sum of cations (in µeq/l) at MCOH.[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)



**Fig. 3.** Comparison between calculated conductivity (calcCond (in  $\mu\text{S}/\text{cm}$ )) and measured conductivity (measCond (in  $\mu\text{S}/\text{cm}$ )) at MCOH.

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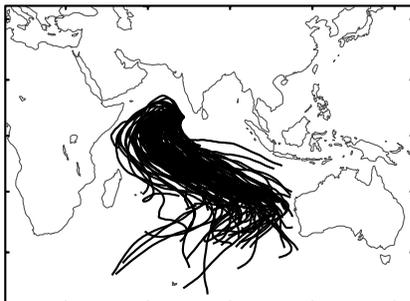
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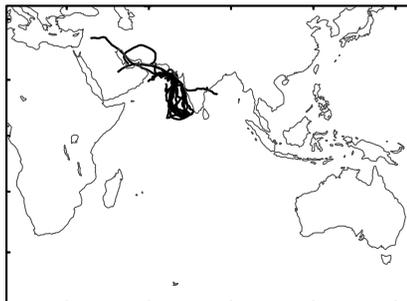
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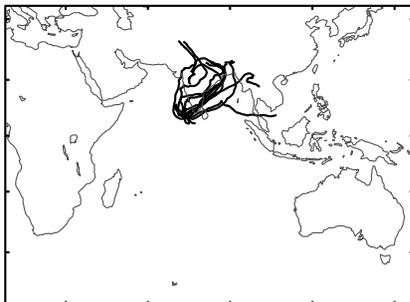
(a) Marine



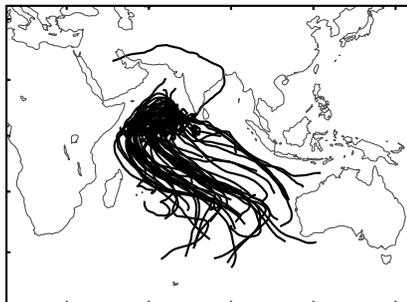
(b) Arabian Sea



(c) India



(d) Mixed



**Fig. 4.** Classification of trajectories arriving at MCOH, **(a)** Marine, **(b)** Arabian Sea **(c)** Indian and **(d)** Mixed. For definitions see text.

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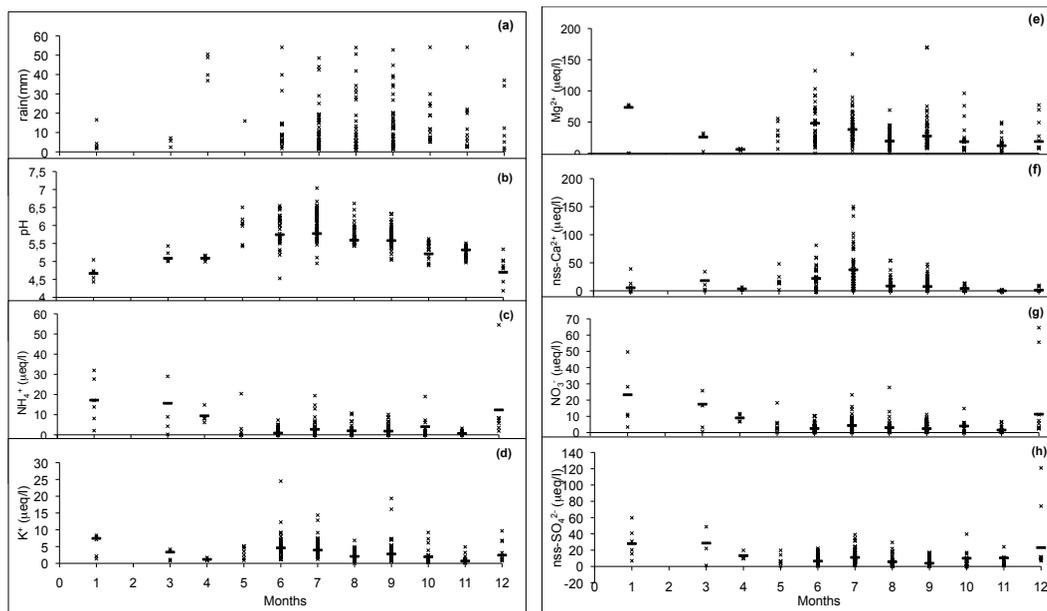
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**Fig. 5.** Summary by month of measured quantities **(a)** rainfall amount (in mm), **(b)** pH, **(c)**  $\text{NH}_4^+$ , **(d)**  $\text{K}^+$ , **(e)**  $\text{Mg}^{2+}$ , **(f)**  $\text{nss-Ca}^{2+}$ , **(g)**  $\text{NO}_3^-$  and **(h)**  $\text{nss-SO}_4^{2-}$ . Volume weighted mean values of ions (in  $\mu\text{eq/l}$ ) for each month are also shown as horizontal line.

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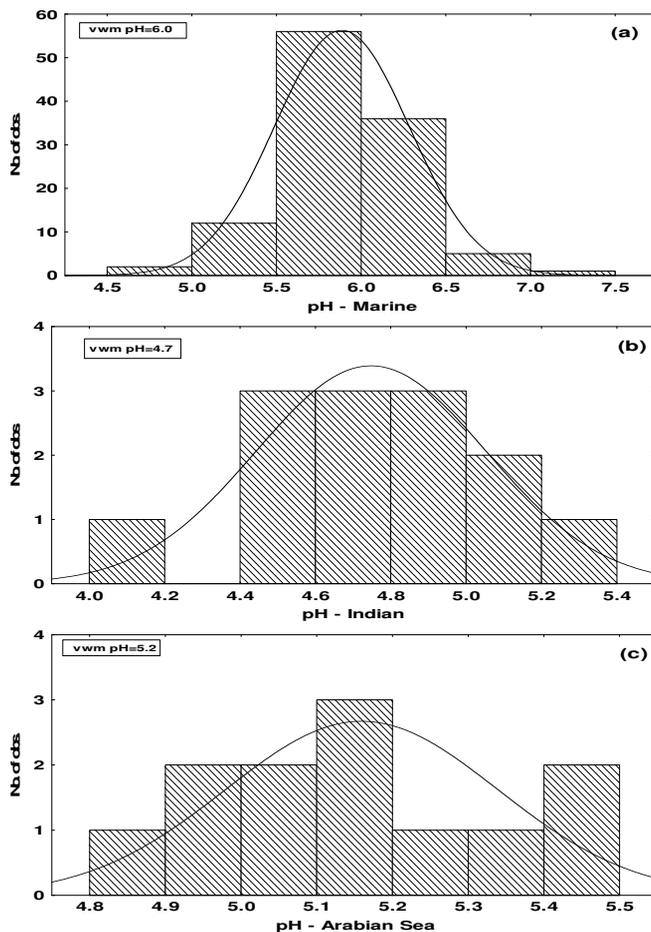
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**Fig. 6.** Frequency distributions of pH in **(a)** Marine, **(b)** Indian and **(c)** Arabian Sea trajectory groups.

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