Manuscript Details

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Title	Using GLUE to pull apart the provenance of atmospheric dust
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Abstract

Identifying the sources of aeolian dust is a crucial step in mitigating the associated hazards. We apply a Generalized Likelihood Uncertainty Estimation (GLUE) model to constrain the uncertainties associated with sediment fingerprinting of atmospheric dust in the Sistan region on the Iran-Afghanistan border, one of the world's dustiest places. 57 dust samples were collected from the rooftop of the Zabol Department of Environmental Protection during a summer dusty period from June to October 2014, in addition to 31 surface soil samples collected from potential sources nearby, including cultivated land (n=8), uncultivated rangeland (n=7), and two dry lakes: Hamoun Puzak (n=10) and Hamoun Saberi (n=6). Dust and soil samples were analyzed for 24 tracers including 16 geochemical elements and 8 water-soluble ions. Five optimum composite fingerprints (Fe, Sr, Mn, Cr and Pb) were selected for discriminating sources by a two-stage statistical process involving a Kruskal-Wallis test and stepwise discriminant function analysis (DFA). Uncertainty ranges for source contributions of dust determined by the GLUE model showed that the dry lake Hamoun Puzak is the dominant source for all dust samples from Zabol and cultivated land is a secondary source. We found marked spatial variance in the importance of regional dry lake beds as dust sources, and temporal persistence in dust emissions from Hamoun Puzak, despite very large areas of adjacent lake beds drying during the study period. Aeolian sediment fingerprinting studies can benefit considerably from the constraints provided by modelling frameworks, such as GLUE, for quantifying the uncertainty in dust provenance data.

Keywords	Sediment fingerprinting; uncertainty; GLUE; atmospheric dust; Iran

Corresponding Author	Matt Telfer
Corresponding Author's Institution	Plymouth University
Order of Authors	Reza Dahmardeh behrooz, hamid gholami, Matt Telfer, John Jansen, Abolhassan Fathabadi
Suggested reviewers	Jan-Berend Stuut, Richard Brazier

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Data for: Using GLUE to pull apart the provenance of atmospheric dust

The full distributions of Generalized Likelihood Uncertainty Estimate (GLUE) modelling used in Behrooz et al (submitted, Aeolian Research, 2018)

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Using GLUE to pull apart the provenance of atmospheric dust

HIGHLIGHTS

- The GLUE model is applied to reveal the provenance of aeolian dust in Zabol, Iran.
- Quantitative estimates and uncertainties of four different dust sources are distinguished.
- One dry lake, Hamoun Puzak, is by far the dominant dust source, despite others nearby.
- The major dust source is insensitive to large changes in exposed local dry lake beds.
- Cultivated land is the second most important dust source, ahead of some playas.

Abstract

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KEY WORDS: sediment fingerprinting, uncertainty, GLUE, atmospheric dust, Iran

1. Introduction

Constraining the source of atmospheric dust particles circulating in the ancient past as well as the present-day is central to understanding the manifold implications of dust in the Earth system (Ridgwell, 2002; Goudie and Middleton, 2006; Shao et al., 2011). Ancient dust deposits are archives of long-term environmental change (Dietze et al., 2016); the best-known and longest being the >8 Myr loess record in China (Sun & Zhu 2010). Present-day dust storms trigger a series of negative off-site and on-site repercussions (Goossens, 2003). Off-site effects include respiratory disease in humans and non-humans, contamination of food and water supplies, and interference with traffic safety, machinery, and electronics. On-site effects include the loss of soil organic matter, nutrients, and overall agricultural productivity (Goudie and Middleton, 2006). From this perspective, identifying sources of dust and quantifying multi-source contributions and their uncertainties is a key step towards hazard mitigation, especially in drylands.

A diverse range of techniques have been employed for tracing sources of atmospheric dust, including isotopic ratios (e.g., Krom et al., 1999; Nakano et al., 2004; Grousset and Biscaye, 2005; Chen et al., 2007; Cao et al., 2008; Wang et al., 2005; Rio-Salas et al., 2012; Yang et al., 2009); mineralogical and chemical characteristics (Shen et al., 2009); meteorological data (Rezazadeh et al., 2013; Nabavi et al., 2016; Ge et al., 2016; Rashki et al., 2017); synthesis of isotopic and geochemical data (e.g., Aarons et al., 2017; Wei et al., 2017; Chavagnac et al., 2008); synthesis of trace element and water-soluble ion analyses (Dahmardeh Behrooz et al., 2017a,b); numerical simulation (Hamidi et al., 2014; Nabavi et al., 2017); satellite data (Long et al., 2016; Cherboudj et al., 2016; Schepanski et al., 2012); and multidisciplinary approaches (Yan et al., 2015; Cao et al., 2015). While most of the studies listed above are highly successful at inferring dust sources, we note that in many cases the uncertainties associated with ascribing provenance are not considered formally. We see this an important omission for two reasons: 1) airborne dust is commonly generated simultaneously from multiple populations and areas of fine-grained particles; and 2) these multiple populations are, in turn, typically an amalgam generated from different sources and mixed to differing degrees over timescales ranging from geological to individual storm events. In other words, dust provenance presents a diabolical mixing-problem and hence uncertainty is fundamental. These two points ultimately stem from geomorphic processes of fine-particle production, transport, deposition, and reworking.

Sediment fingerprinting is widely used to quantify source contributions of fluvial sediments (e.g., Collins et al., 1997; Walling, 2005; Stone et al., 2014; Zhou et al., 2016a; and Manjoro et al., 2017) and its application to aeolian problems is growing (e.g., Liu et al., 2016; and Gholami et al., 2017a,b). Moreover, the uncertainties involved with this method are gaining increased attention (Walling, 2013). In order to manage and quantify the uncertainty in fluvial sediment fingerprinting, some studies have applied a Monte Carlo simulation framework (e.g., Motha et al., 2003; Collins et al., 2012; Voli et al., 2013; Smith and Blake, 2014; Stone et al., 2014; Sherriff et al., 2015; and Vale et al., 2016). Similarly, Bayesian approaches are also applied to fingerprinting aeolian sands (Gholami et al., 2017b) and fluvial sediments (e.g., Massoudieh et al., 2013; Cooper et al., 2014; Cooper et al., 2015; Stewart et al., 2015; and Abban et al., 2016). Yet, several challenges remain in adequately capturing the uncertainty associated with diverse aeolian dust sources and pathways (Walling, 2013) and we suggest that techniques developed in other disciplines may offer a way forward (Gholami et al., 2017b).

First proposed for hydrological modelling by Beven and Binley (1992), GLUE (Generalised Likelihood Uncertainty Estimation) has gained much favour as a tool for evaluating uncertainty estimates (e.g., Hassan et al., 2008; Zhou et al., 2016b; Viola et al., 2009; Mantovan and Todini, 2006; and Gong et al., 2011). Here, we apply GLUE to the problem of dust provenance in the Sistan Hamoun region on the Iran-Afghanistan border. Since it constitutes a major dust source for south-west Asia, Sistan has been the focus of numerous previous investigations, (e.g., Goudie and Middleton, 2006; Rashki et al., 2012, 2013 a,b, 2015; Alizadeh Choobari et al., 2014). Recent work has generated an important geochemical dataset of dust samples from a meteorological station at Zabol which has been analyzed geochemically to provide qualitative estimates of source (Dahmardeh Behrooz et al., 2017a) and the temporal variability of dust emissions (Dahmardeh Behrooz et al., 2017b). Here we provide the first attempt to formally quantify aeolian dust provenance and associated uncertainties with this dataset using GLUE (Dahmardeh Behrooz et al., 2017a; 2017b).

2. Study area

The Sistan-Hamoun study area (Fig. 1) straddles the border between Afghanistan and the Sistan and Baluchestan province of south-eastern Iran (30˚5′ to 31˚28′ N and 61˚15′ to 61˚50′ E) (Rashki et al., 2012, 2013a). The Hamoun lakes complex comprises three main lakes: Hamoun Hirmand, Hamoun Saberi, and Hamoun Puzak, which are recharged primarily from Afghanistan by the Hirmand (Helmand) River with smaller contributions from streams to the north and west (Esmaeili and Omrani, 2007). Following exceptionally high runoff, the lakes form a single body of water ~5700 km^2 in area and ~13 Mm^3 in volume (Sharifikia, 2013), though such events have become rare in recent decades while dust emissions have grown correspondingly in magnitude (Goudie and Middleton, 2006; Rashki et al., 2012).

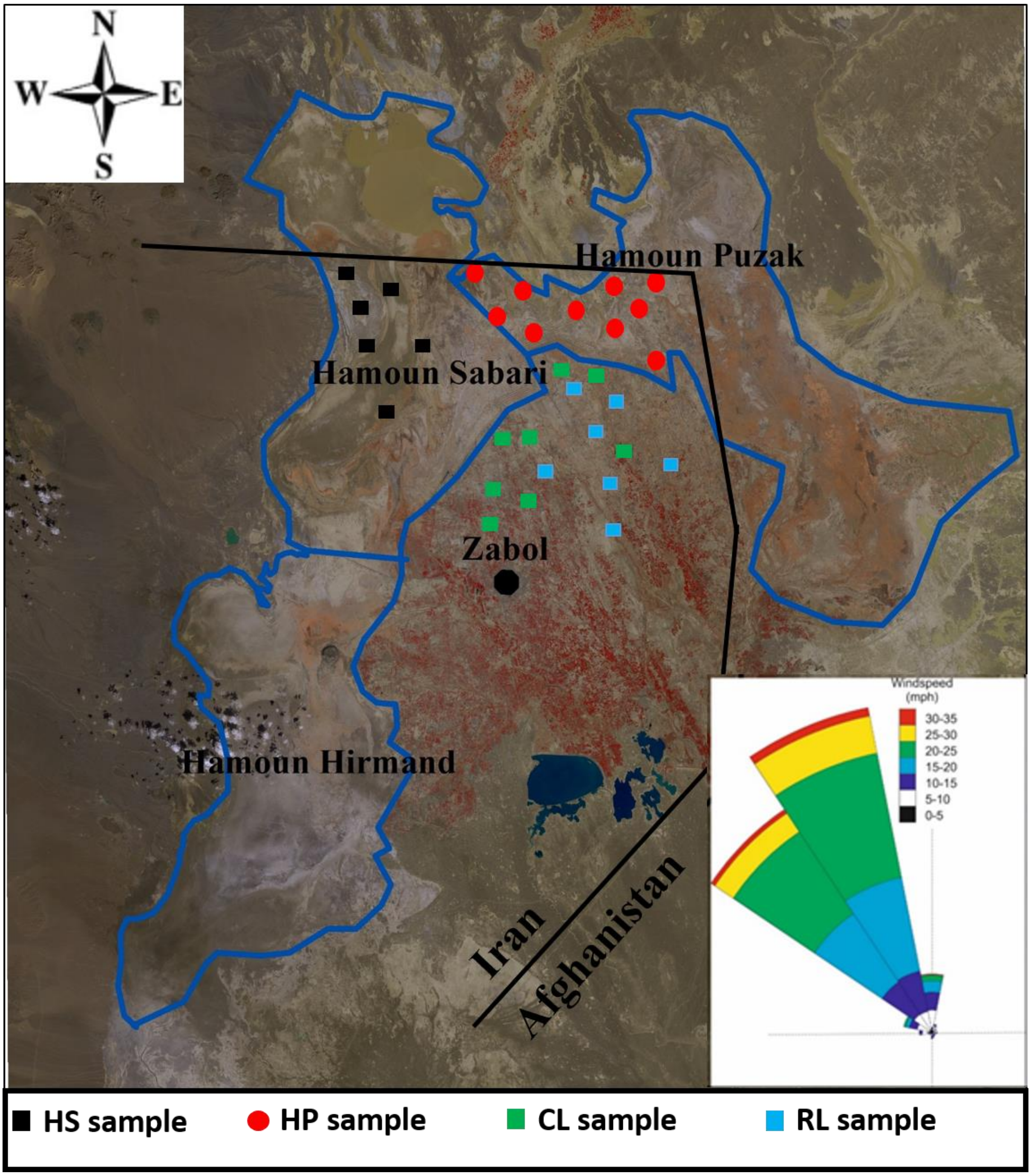

Figure 1: Sampling sites in the Sistan region: the dry-bed of Hamoun Puzak (HP); the dry-bed of Hamoun Saberi (HS); uncultivated range land (RL); and cultivated land (CL). Inset shows the hourly averaged wind regime for the period June-October 2014 (data source: National Climatic Data Centre, Climate Data Online).

The climate the[illegible]tan region is arid to hyper-arid, and land-use is chiefly linked to agriculture and fishing. At Zabol meteorological station (Fig. 1), mean rainfall is 55 mm/y and mean evaporation is

>4000 mm/y (Moghaddamnia et al., 2009). The prevailing wind is the notorious "Wind of 120 Days" from the north, which in the summer is accelerated into a Low-Level Jet (LLJ) by a persistent high-pressure system over the Hindu Kush and the channeling effect of the surrounding topography (Alizadeh-Choobari et al., 2014). As a result, the city of Zabol and its ~135,000 inhabitants experience dust storms of catastrophic proportions, resulting in Zabol ranking as the world's most polluted city for particulate matter less than 2.5 µm (PM2.5) in size (World Health Organisation, 2016).

3. Methods

3.1 Field sampling

We set out to characterise the soil materials for four different potential sources of atmospheric dust emissions to the north of Zabol city (Fig. 1): 1) the dry lake-bed of Hamoun Puzak (Fig. 2a); 2) the dry lake-bed of Hamoun Saberi (Fig. 2b); 3) cultivated arable farmland generally without crop-cover in summer (Fig. 2c); and 4) bare land surfaces with sparse to negligible natural vegetation cover (Fig. 2d). A total of 31 surficial soil samples (<5 cm depth in a 30 cm^2 area) were collected from the four potential sources (Table 1). We sieved the soil samples with a 400-mesh sieve, retaining particles with a nominal geometric diameter of < 38.5 µm, which is equivalent to the aerodynamic diameter of dust (Cao et al., 2008). After sieving we retained about 5 g of dust-sized material from each sample.

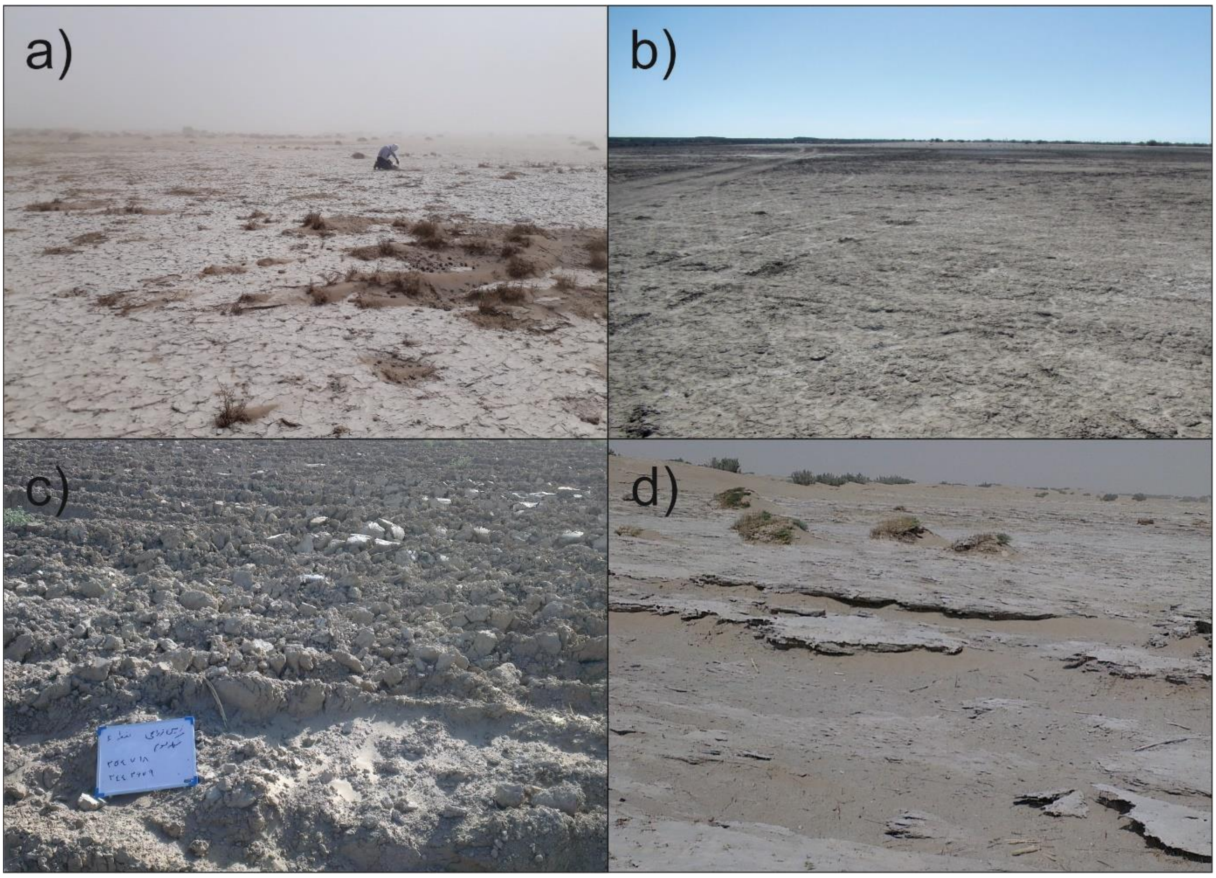

Figure 2. Typical examples of the land surfaces we sampled. a) Hamoun Puzak dry lake-bed, b) Hamoun Saberi dry lake-bed, c) close-up view of cultivated agricultural land surface during the summer months, and d) sparsely vegetated, uncultivated rangeland.

During an exceptionally dusty summer period in Zabol (23 June to 4 October 2014), 57 atmospheric dust samples were collected at one- to four day intervals (Table 1) with sampling apparatus fitted to the rooftop of the Department of Environmental Protection (5 m above ground level, 31˚N, 61.3˚E) in an outer suburban area with no major industrial activities nor local fugitive dust sources. Our two dust samplers (Model Chrono, Zambelli, Milan) were equipped with cyclones operating at a flow rate of 16.7 L/min as per the EU norms (Dahmardeh Behrooz et al, 2017a; 2017b). Total suspended-particle (TSP) samples were collected in Teflon filters (0.45 µm pore size and 47 mm diameter) and then desiccated for 24-hours at 25 °C. Dust mass concentrations were measured gravimetrically by weighing the Teflon filters before and after sampling using an analytical balance (Adam model) with ±0.1 mg precision. We refrigerated all dust samples at 4˚C until chemical analysis (Dahmardeh Behrooz et al., 2017 a).

3.2 Laboratory analysis of water-soluble ions and trace elements

We measured the concentrations of 8 water-soluble ions in our samples (viz., Na^+, NH^+_4, K^+, Ca^{2+}, Mg^{2+}, Cl^-, NO^-_3, NO^-_2). Three cations (Na^+, NH^+_4 and K^+) were measured with a Shim-pack IC-C1 (Shimadzu DGU-12A) using 5-mM HNO_3 solution as eluent. Three anions (Cl^-, NO^-_3 and NO^-_2) were measured with a Shim-pack ICA1 (Shimadzu DGU-12A), using 2.5–mM phthalic acid combined with 2.4-mM tris-(hydroxymethyl) aminomethane as eluent (Lin, 2002). Two cations (Ca^{2+} and Mg^{2+}) were measured via flame atomic absorption spectrometry (Philips, PU9400X, England).

After acid digestion, all samples were analyzed to determine the concentrations of 16 trace elements (viz., Al, As, Au, Co, Cr, Cu, Fe, Li, Mg, Mn, Ni, Pb, Pt, Sn, Sr, and Zn) via Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-OES, Perkine Elmer, Optima 2000, USA). Further details of sampling and laboratory procedures are given in Dahmardeh Behrooz et al. (2017a,b).

3.3 Two-stage method: Kruskal-Wallis test and discriminant function analysis

Our measurements of 8 water-soluble ions and 16 trace elements form the basis of the sediment fingerprinting method aimed at identifying the source contribution of the Zabol dust samples. We adopt a two-stage statistical procedure following the approach of Collins et al. (1997). In stage one we tested the primary ability of tracers to discriminate dust sources using the Kruskal-Wallis H test.

Tracers with critical values at the 95 % confidence levels or better were taken to the second stage in which we identified optimum composite fingerprints using a stepwise discriminant function analysis based on minimization of Wilk's lambda.

3.4 Generalised Likelihood Uncertainty Estimation (GLUE)

GLUE was first devised by Beven and Binley (1992) as a means of sensitivity analysis and uncertainty estimation in environmental model outputs. We use GLUE to quantify the uncertainty in the sediment fingerprinting results via the following five steps:

1) Random sampling of parameter sets (300,000 iterations) are conducted using the Latin Hypercube Sampling (LHS) method (Zhou et al, 2016b) and assuming source contributions from each source are non-negative and total contributions sum to unity. Due to the lack of prior information, we used a uniform distribution as the prior distribution for all parameters.

2) Selection of a likelihood function and behavioral parameter thresholds. Here, we adopt the Nash–Sutcliffe coefficient (ENS) as the likelihood function (Jin et al, 2010):

$$ME = 1 - \frac{\sum(O_{obs} - O_{sim})}{\sum(O_{obs} - \hat{O}_{obs})} = 1 - \frac{\sigma_i^2}{\sigma_{obs}^2} \qquad \text{(eq.1)},$$

where $\hat{O}_{obs}$ is the mean value of the observed tracer concentration; O_{sim} is the simulated tracer concentration; O_{obs} is the observed tracer concentration; σ^2_i is the error variance for the *i*th model (i.e., the combination of the model and the *i*th parameter set) and σ^2_{obs} is the variance of the observations.

3) Sampled parameter sets from step 1 are input to the mixing model (equation 2) and the likelihood function is calculated for each parameter set as:

$$C_{dust} = C_{Sources} \times P \qquad \text{(eq. 2)}$$

where P is an m dimensional column vector of sources contribution (sampled parameter sets), C_{dust} is an n-dimensional column vector of element concentration in sediment sample, $C_{Sources}$ is an n×m-dimensional matrix representing mean tracer concentration in sources (each row represents mean tracer concentration in each source), where n is the number of optimum composite fingerprints (n=5) and m is the number of dust sources (m=4).

4) Parameter sets are divided into behavioural and nonbehavioural types with respect to a threshold value (Zhou et al, 2016). In this step, those parameter sets that have likelihood functions greater than a threshold value were classified as behavioural parameter sets. For the next step, nonbehavioural parameter sets were discarded.

5) For behavioural parameter sets, likelihood weights are rescaled such that they sum to one, then each parameter is sorted and we calculate cumulative distributions for each parameter. Quintiles and uncertainty intervals are calculated via the cumulative distributions.

3.5 Geospatial analysis and climate data

Landsat data were downloaded from the United States Geological Survey's Earth Explorer, and all analysis was conducted within ArcGIS 10.3. Quantitative analysis of water extent was conducted using a modified Normalized Difference Water Index (NDWI), based on the green (Band 3) and short-wave infrared (Band 6) bands of Landsat 8 data (Xu, 2006). Climate data are taken from the Hourly Global Surface Data (DS3505) dataset for the Zabol station (World Meteorological Organization ID: 40829), accessed via the legacy Climate Data Online (CDO) portal of the National Oceanic and Atmospheric Administration's (NOAA) National Climatic Data Centre (NCDC) online (available at https://www7.ncdc.noaa.gov/CDO/dataproduct).

4. Results

4.1 Kruskal-Wallis test and discriminant function analysis

The results of the Kruskal-Wallis tests (Table 2) indicate that among the twenty-four measured properties (8 water-soluble ions and 16 element concentrations), thirteen trace elements (Mg, Sr, Li, Fe, Cr, Cu, As, Ni, Pb, Mn, [illegible] and Sn) and one ion (Ca^{2+}) show statistically significant differences at the 95 %-level between our four potential dust sources (the two dry lake beds, cultivated farmland, and areas of natural vegetation cover). Trace elements clearly out-performed water-soluble ions for tracking spatial sources of dust. We passed the thirteen trace elements to stage-two for stepwise discriminant function analysis (DFA). The DFA yielded five trace elements (Fe, Sr, Mn, Cr and Pb) with optimum composite fingerprints that correctly discriminate 87 % of our source samples (Table 2 and Fig. 3).

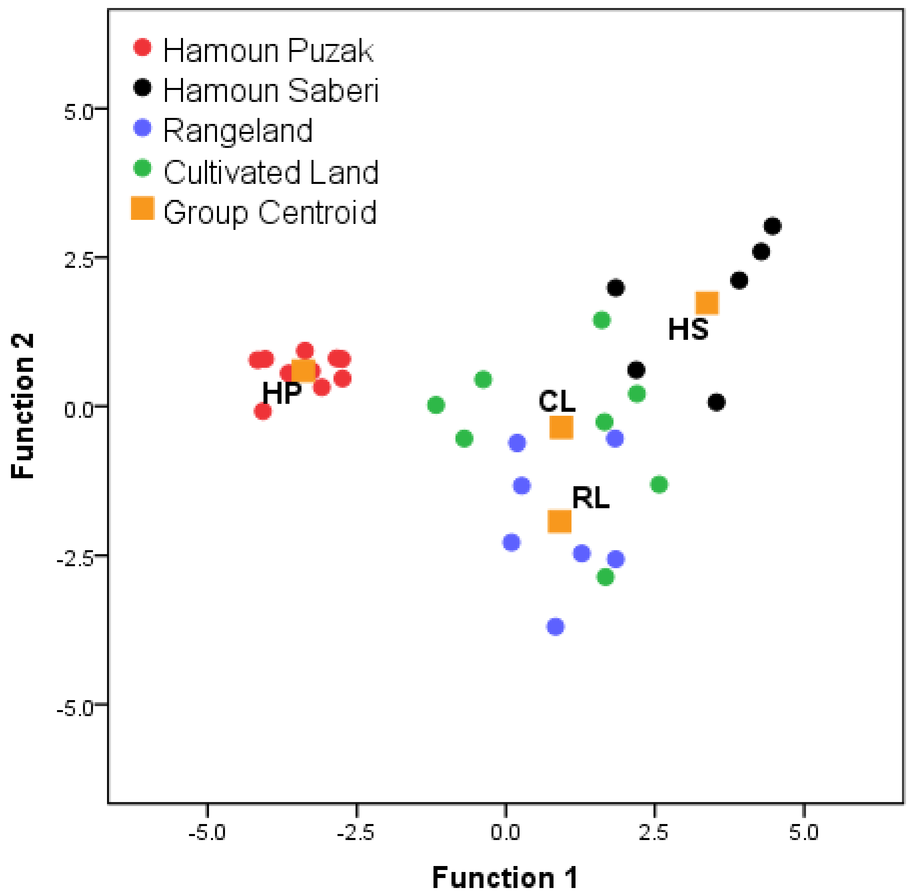

Figure 3. Scatterplot constructed from the first and second functions derived from a stepwise DFA for the source groups including the four land (i.e. Hamoun Puzak (HP), Hamoun Saberi (HS), cultivated land (CL) and uncultivated rangeland (RL). Five optimum fingerprints (Fe, Sr, Mn, Cr and Pb) were used to construct the scatterplot and 87% of the source samples are discriminated, correctly.

4.2 Using GLUE to constrain uncertainty in the source contributions of dust

Uncertainty intervals of source contributions estimated by our GLUE-mixing model at the 95 % confidence level are presented in Figure 4. These results show that the most important dust source is clearly Hamoun Puzak (FigS 4a and 5). Median contributions from this lake-bed span 29 to 88 % (samples 33 and 45, respectively). Hamoun Saberi is a less important source for our samples. Median contributions from this lake bed span 3 to 24 % (samples 11 and 33, respectively) (Figs. 4b). The sparsely vegetated rangeland is the least active dust source. Median contributions span 2 to 22 % (samples 45 and 34, respectively) (Fig. 4c). Cultivated farmland is recognized as the second-most important source for all of 57 samples. Median contributions from farmland span 4 to 25 % (samples 23 and 34, respectively) (Figs. 4d). We note that for most samples, the lower-limit of predicted uncertainty is zero for contributions from the three sources other than Hamoun Puzak (Figs. 4b-d).

Figure. 5 presents an overview of the source contributions with all samples plotted together as a frequency histogram.

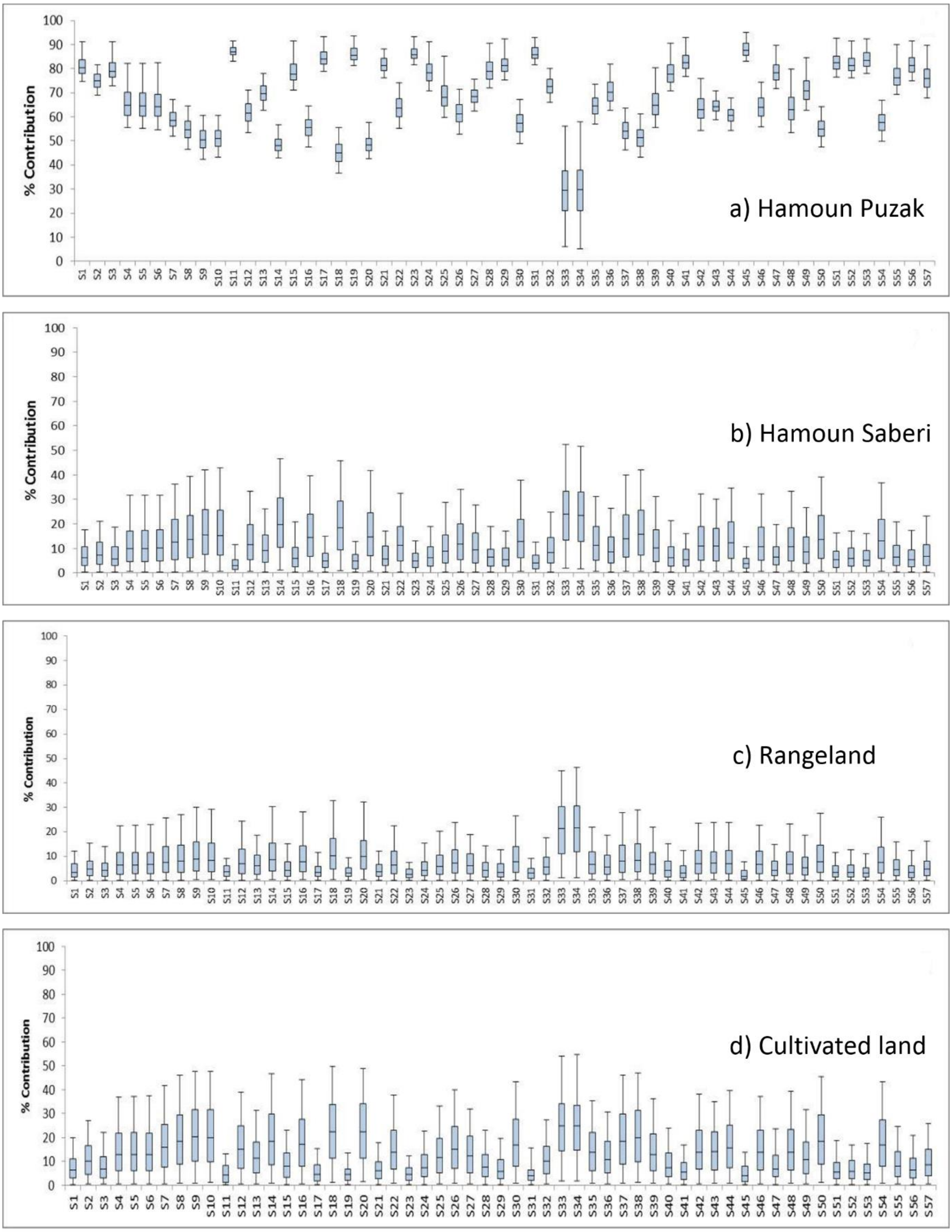

Figure 4. GULE results for dust source contributions yielding 95% confidence limits (with percentiles 2.5, 25, 50, 75 and 97.5). A) Hamoun Puzak; B) Hamoun Saberi; C) uncultivated rangeland; and d) cultivated land.

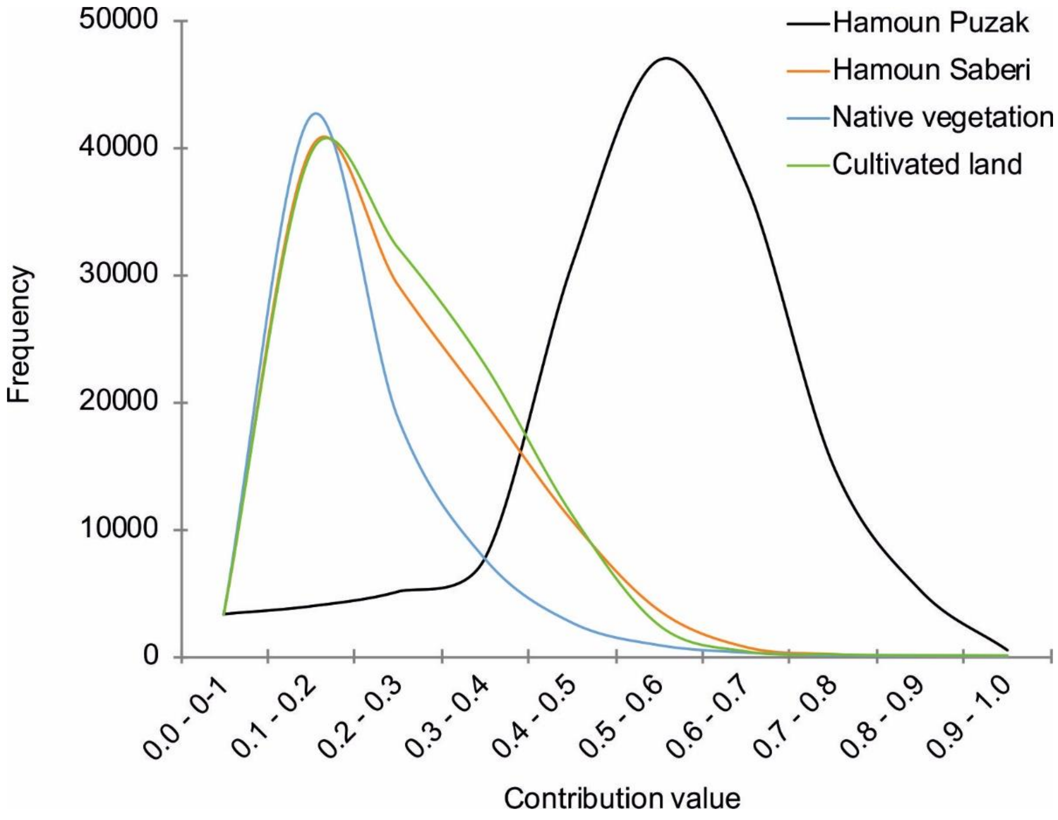

Figure 5. Summary of all source contributions plotted as a probability density function.

5. Discussion

Sediment fingerprinting is a highly effective technique for quantifying source contributions of fluvial sediments (e.g. Collins et al., 1997; Walling, 2005; Stone et al., 2014; Zhou et al., 2016a; Manjoro et al., 2017) and aeolian sands (e.g. Liu et al., 2016; Gholami et al., 2017a,b). Here we build upon this approach by exploring the potential of the GLUE methodology for distinguishing spatially proxima aeolian dust sources with similar underlying geology and geomorphology. We demonstrate its efficacy at formally quantifying the uncertainty distributions associated with aeolian dust fingerprinting due to spatial and temporal variation in the dust cycle, and use the method to reveal spatial complexity - alongside an unexpected lack of temporal complexity - in the nature of the dust sources.

5.1 Environmental context of dust emissions

The strong interannual correlation between dusty days and the surface area of exposed lake floors indicates that i) dust storms in the Sistan-Hamoun region are directly related to the dryness of the Hamoun lakes, and ii) these lake beds are the main source of dust emissions (Goudie and Middleton 2006; Rashki et al., 2012; 2013a; 2013b; 2015). Such relationships are not uncommon, as worldwide observations suggest that exposed dry lake beds can govern the frequency and intensity of dust storms; for example, at Owens Lake, USA (Reheis et al., 2009), Aral Sea, Uzbekistan (Breckle et al., 2012), Makgadikgadi pan complex; Etosha Pan, southern Africa (Prospero et al., 2002; Mahowald et al., 2003; and Washington et al., 2003); and Lake Eyre, Australia (Baddock et al., 2009).

The frequency and magnitude of dust emissions from the Hamoun Lakes has also been related to annual/decadal scale variations in the surface area of the lakes, which varies dramatically. At its maximum extent, observed following the 1998 spring-melt Hirmand River floods (Rashki et al., 2012a), the Hamoun lake complex forms a single body of water ~4500 km^2 in area. This is comprised of Hamoun Hirmand (~1400 km^2), Hamoun Saberi (~1400 km^2) and Hamoun Puzak (~1700km^2). During more typical lake-full episodes, these bodies of water are not conjoined; for instance, during the spring of 1996 (Figure 6a), Hamoun Saberi spanned ~815km^2 and Hamoun Puzak spanned 375km^2. Hamoun Hirmand lies mostly downwind of Zabol, and hence is not considered further. Between 1999 and 2010 a prolonged drought, likely related to the El Nino Southern Oscillation, resulted in the rapid and sustained desiccation of the Hamoun lakes, with a concomitant increase in the frequency of dusty days (Rashki et al., 2012; 2013). Since 2010, lake levels have been highly variable (Fig. 6), with returns to lake-full conditions experienced around 2011, but a subsequent return to large areas of exposed dry lake beds in recent years.

The implications of such changes can develop rapidly. Within the timeframe of this study (June-October 2014), Hamoun Puzak and Hamoun Saberi lost around 295 km^2 and 640 km^2 of water surface area, respectively (i.e. 98.5% and 99.9% of their extent on June 14th) (Fig. 7). This desiccation affected Hamoun Saberi and Puzak proportionally at very similar rates, but Saberi's larger surface area at the start of this study led to greater absolute change in lake floor exposure area at Saberi. During this period, there was no rainfall recorded at the Zabol meteorological station, but the persistent 'Wind of 120 Days' blew from 327° ± 36° during June-October, with daily average windspeeds up to 35 mph, and 50 of the 138 days of the study period exceeding daily averages of 20 mph.

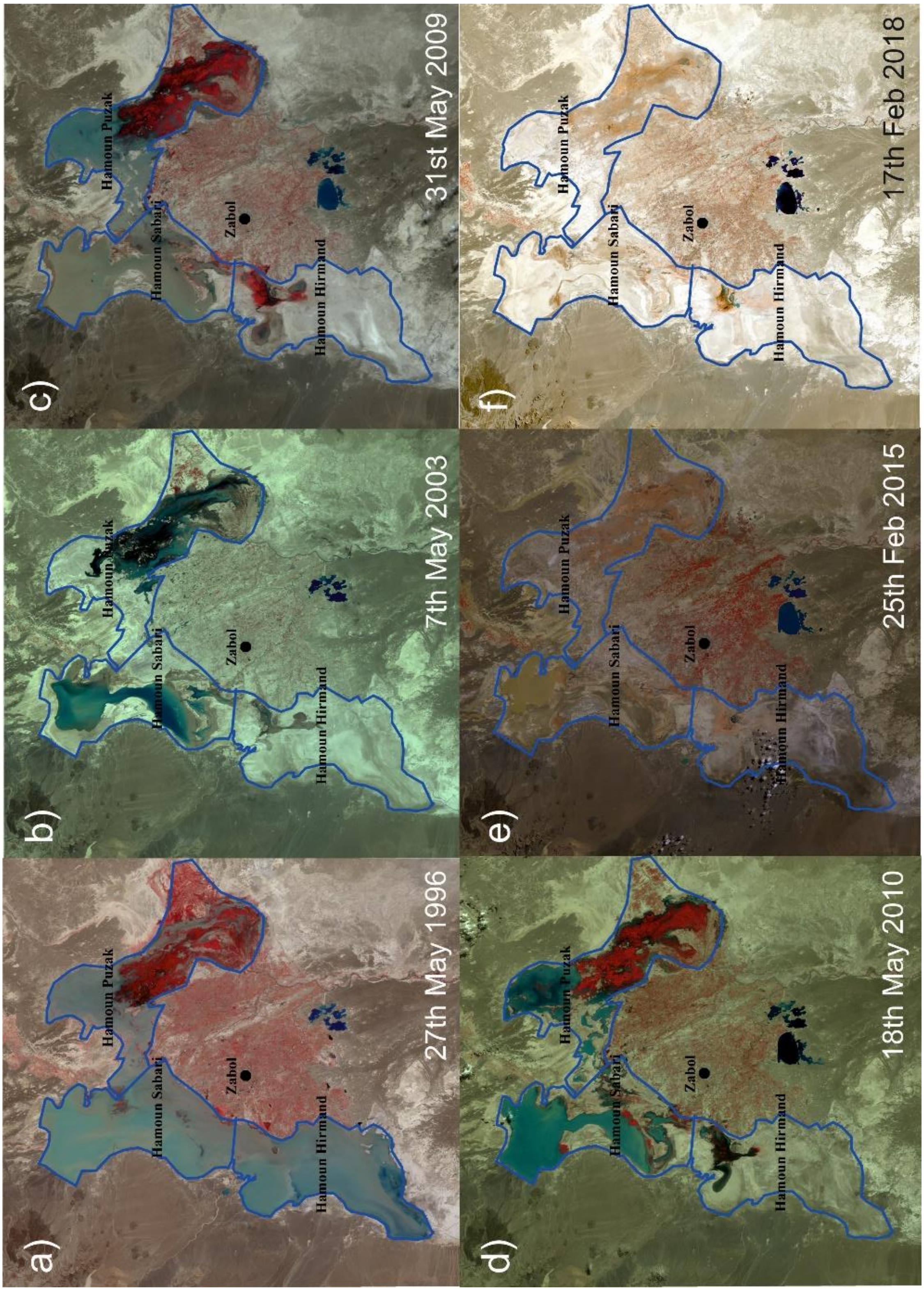

Figure 6. Decadal scale changes in the Hamoun Lakes. Following lake-full conditions in the late 1990s (a), a sustained decade of drought (b and c) resulted in the exposure of large areas of dry lake-beds and therefore potential dust sources. Since then, levels have varied and often changed rapidly (d, e and f). All images are infrared/red/green composites based on Landsat 5 and 8 imagery, using Bands 4/3/2 and 5/4/3 respectively. Vegetation is shown as red tones.

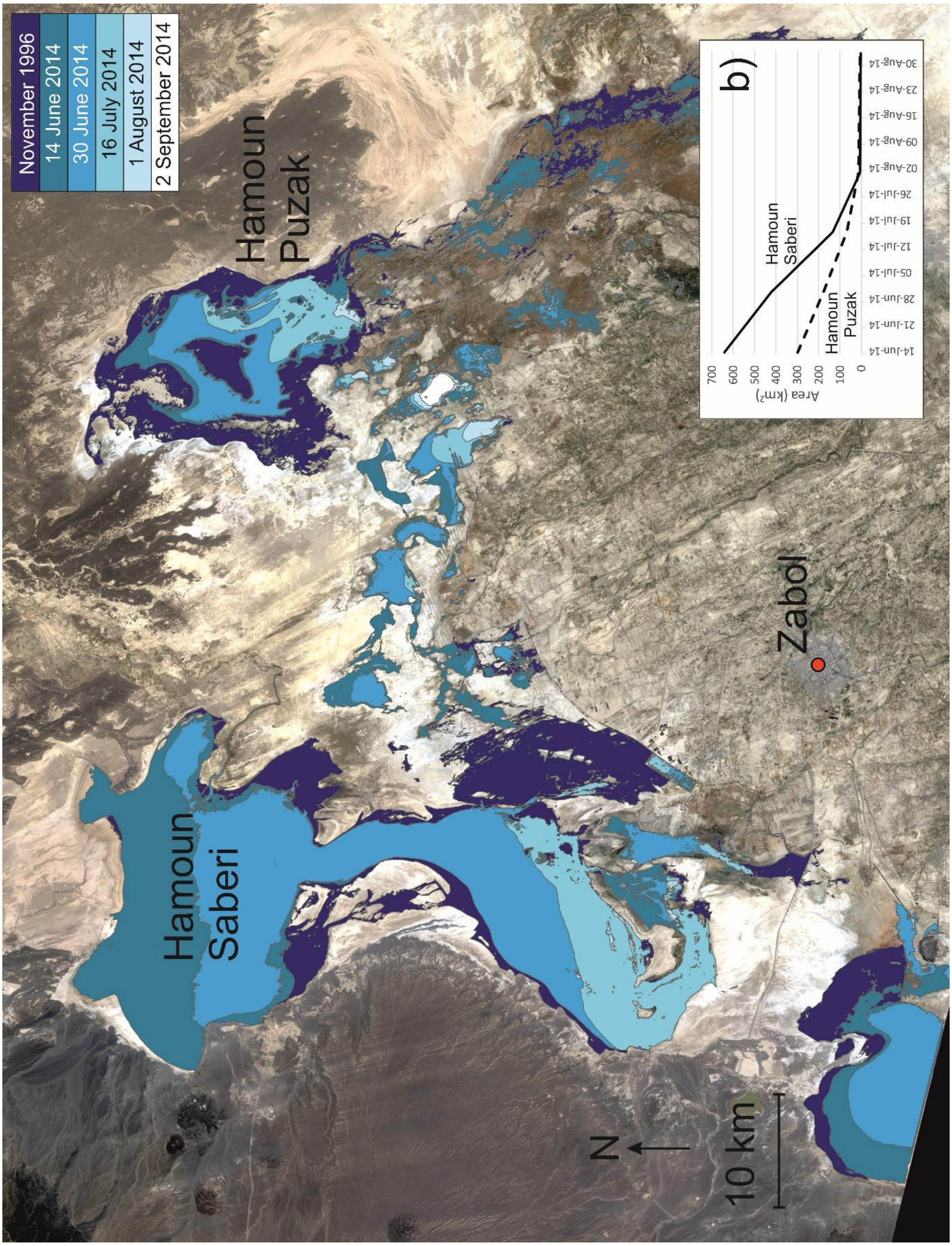

Figure 7. Changes in the surface extent of the Hamoun Lakes between June and September, 2014. Note the rapid desiccation during June and July, and resultant exposure of new surfaces for potential deflation.

5.2. Dust sources: dry beds of Hamoun Lakes

The GLUE results (Figs 4 and 5) reveal that the dominant source of dust collected at Zabol is Hamoun Puzak, and also that in general, the sources of the dust vary little over the three month period from June to the beginning of October, 2014. This finding is unexpected for a number of reasons. Firstly, with the wind at Zabol during this period coming from the northwest to north-northwest (327° ± 36°), the most obvious candidate source of the Zabol dust is the upwind Hamoun Saberi (Figures. 1 and 7). Yet, consistently, Hamoun Puzak contributes ~40-90% (uncertainties included) of the dust received at Zabol. Furthermore, given the rapid increase in Saberi's exposed dry bed during the early period of sampling, its contribution would be expected to increase proportionally over this period. But Saberi's contributions actually vary little during the season (Fig. 4). When the surface area of the lakes is considered, either relative to lake-full conditions (Figure 8a and 8c), or as absolute surface areas (Figure 8b and 8d), there is little temporal relationship with the relative dust contributions of the two lake beds.

Similarly, investigation of the meteorological conditions during June-October do not readily explain the dominance of Hamoun Puzak. There is no clear correlation with either wind magnitude, or direction, that can readily explain the dominance of Puzak, and no obvious explanation arise for the occasional excursions when other sources contribute markedly more. For instance, on August 27th replicate samples were collected (S33 and S34 in Fig. 4) and yield consistent results- note that, for consistency, only one of these samples is included in Figs. 8 and 9.

These result suggest that Hamoun Puzak – or at least the western margin of Hamoun Puzak, where the source samples were collected (Fig. 1) - is a prolific and persistent source of dust over Zabol irrespective of the existence of large adjacent alternative sources. Why is Hamoun Puzak such an effective dust emitter? And why, despite its size and position directly upwind of Zabol, does Hamoun Saberi contribute relatively little? We propose that several factors may be at play, as follows.

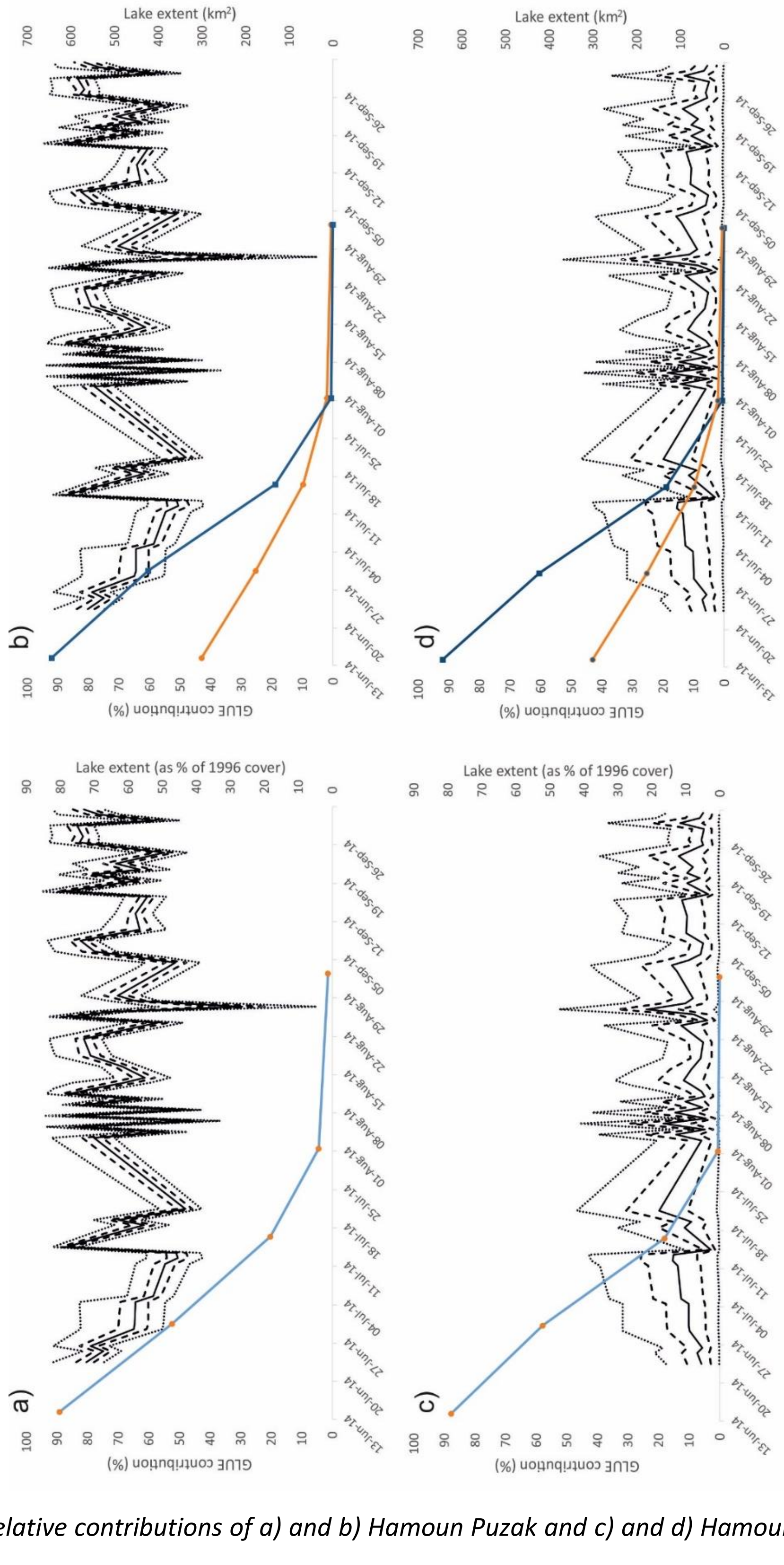

Figure 8. The relative contributions of a) and b) Hamoun Puzak and c) and d) Hamoun Saberi, plotted alongside a) and c) the surface area of the lakes (expressed as a percentage of the 1996 lake-full condtions) and b) and d) the absolute surface area of the lakes. There is no consistent trend in dust provenance, despite the changing area of the potential sources. Solid lines indicate the median estimate, dashed lines the first and third quartiles and dotted lines the 2.5% and 97.5% bounds.

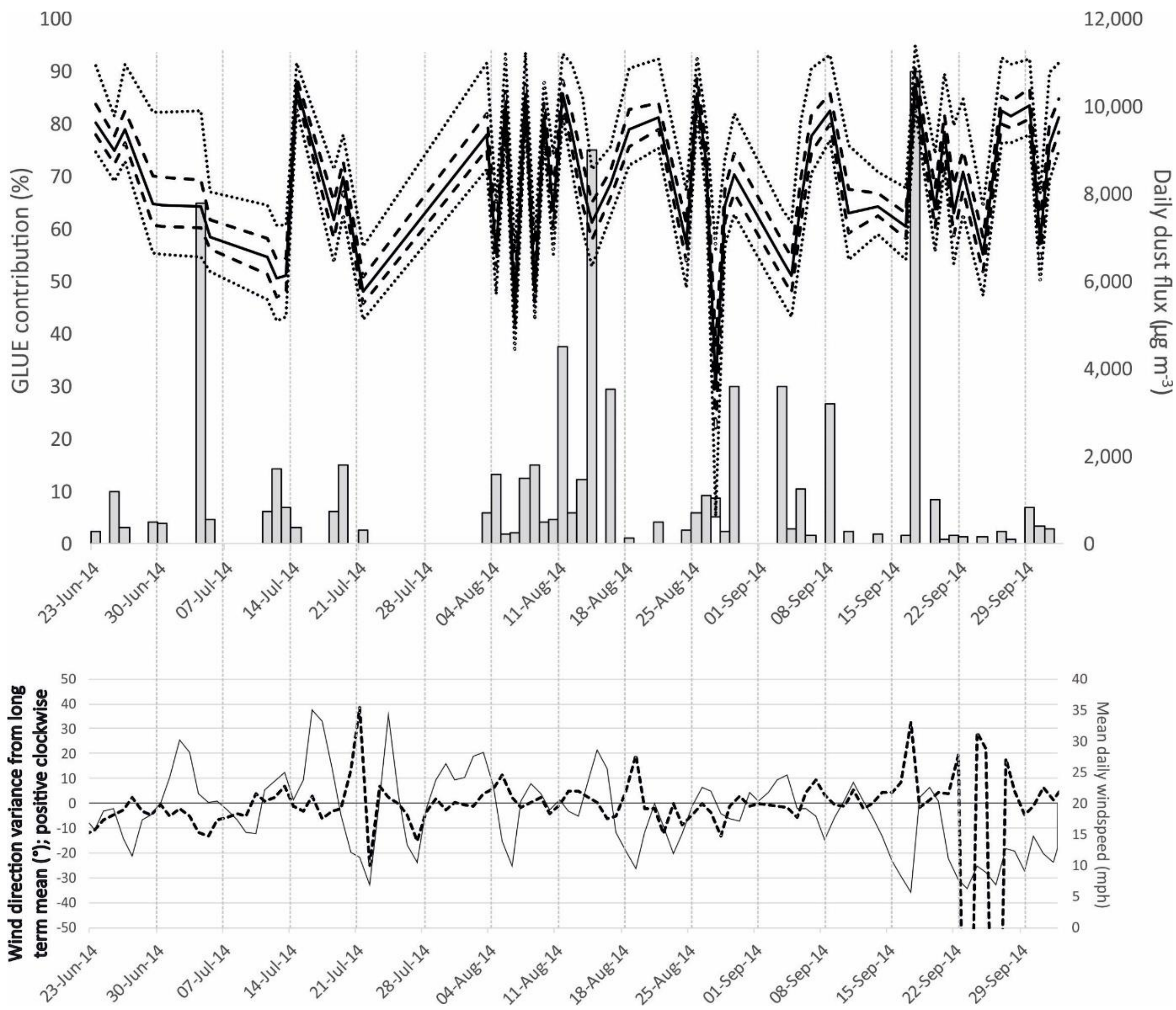

Figure 9. The dominant dust contribution from Hamoun Puzak (top; key for lines as in Figure 8), shown alongside the magnitude of dust collected at Zabol (bars), and the mean daily wind speed (thin solid line) and variance in mean wind direction (bold dashed line).

First, let us consider the hydrological setting and sedimentology of the area. The Hamoun Lakes are predominantly fed with water from the Hirmand River to the east, with the Khash River also feeding directly into Hamoun Puzak from the east. Hamoun Saberi is fed largely from the north by the Harut and Farah rivers. The sampled region at the western margin at Hamoun Puzak lies in series of channels and small closed basins which act as spillways connecting the lakes during lake-full episodes, and thus is likely to have distinctly different sedimentology from lake bed areas subject to direct lacustrine sedimentation (King et al., 2011, Sweeney et al., 2011). Evidence that these sediments are distinct from those of Hamoun Saberi is implicitly provided in the primary function of the DFA used here to define the characteristics of these dusts (Fig. 3). The wind regime necessary for aeolian transport to Zabol from western Hamoun Puzak is compatible with this zone being a dominant source, as northerly orientation lies well within the 327° ± 36° (one sigma) direction of the observed winds. It may also be that the transport pathways associated with the low-level jet from the north are strongly impacted by topography. We note that wind streaks evident on the satellite imagery of the region suggest that topography is steering and deflecting local winds in complex manner.

This orientation, however, raises another question: Why does Hamoun Saberi, which lies directly upwind of Zabol, not contribute more to Zabol's dust flux, especially during the latter part of our study when an additional 640 km^2 of dry lake bed became exposed? It is well-reported that dust production can be spatially highly variable, even at sub-basin scales (Mahowald et al., 2003, Reheis et al., 2002, Bullard et al., 2008). One possibility relates to the differing geochemistry of the sediments that can promote the formation of protective crusts. Field experiments have shown that, dry river courses that are replenished frequently with fine-grained fluvial sediment can be much more effective at producing deflatable dust relative to playas and, counter-intuitively, playa centres are relatively low emission sources (Sweeney et al., 2011, King et al., 2011). Dry lake-bed deposits from the Mojave, for instance, have been reported to yield less dust than those with fluctuating water-levels (Reynolds et al., 2007), and the progressive and rapid desiccation of Hamoun Saberi during the study period may have been simply unfavourable for the generation of deflatable dust.

Conversely, it may be that exogenic water supply from the northerly channels sufficiently dampened the surface to limit additional deflation. We note that shallow flooding was observed during the field sample collection, despite the lack of rain observed either in Zabol, or the fortnightly Landsat images. Over longer timescales, rates of aeolian erosion have been shown to inversely relate to soil moisture (Whitney et al., 2015). Although the whole region is sparsely vegetated, the role of vegetation in influencing surface roughness and thus susceptibility to aeolian erosion also cannot be overlooked (e.g. Cowie et al., 2013, Li et al., 2007). Lastly, we point to the cause of the additional 13% of variability, which was not well explained by the discriminant function analysis of the source sediments. This variability may imply that a significant component of Zabol dust derives from outside the immediate area of the Hamoun Lakes. Dust plumes transported from Karakum desert in Turkmenistan may affect the Sistan region (Kaskaoutis et al., 2015) and may be a source of exogenous dust not accounted for among the four potential sources we sampled. In short, further work is needed to identify precisely why Hamoun Puzak dominates the aeolian dust flux at Zabol.

5.3. Dust sources: cultivated and uncultivated land

The connection between land management, agriculture and aeolian dust emissions are well documented (Wiggs and Holmes, 2011, Okin et al., 2001), and the role of agriculture in exacerbating drought-driven dust events such as the decade-scale 'Dust Bowl' of 1930s USA is clearly established (Worster, 2004). We find that the cultivated cropland to the north of Zabol is the region's second-largest overall source of dust (Fig. 4), slightly out-stripping Hamoun Saberi, and contributing substantially more than uncultivated rangeland with sparse vegetation. Desertification (i.e. semi-arid and arid land degradation) has been recognized in other regions of Iran (Sepehr et al., 2007), and given the difficulties of agriculture in such an extremely dry and hot climate, it is unsurprising that sustainable land management is difficult to achieve. The spread of wind erosion is challenging land managers worldwide - from the Argentinian Pampas (Buschiazzo and Zobeck, 2008) to the Tibetan Plateau (Zhang et al., 2012); and even temperate regions such as southern Sweden (Barring et al., 2003). The findings here that cultivation-based farming is the second largest contributor to Zabol's dust flux (with median contributions of 4-25% for individual samples) highlights an anthropogenic dust source that may be quelled through more considered farming practices in the future.

6. Conclusion

Identifying source(s) of aeolian sediments (sand and dust) is essential to improved planning and management of arid and semi-arid regions. Here we present a quantitative sediment fingerprinting approach coupled with the GLUE methodology to quantify source contributions of dust to the city of Zabol in the Sistan-Hamoun region of south-east Iran. Zabol consistently ranks globally as one of the most susceptible to fine (PM2.5 and PM10) aerosol pollutants. Using GLUE, we have assigned quantitative estimates of the relative contributions of four potential dust sources; two dry lake beds (Hamoun Puzak and Hamoun Saberi), cultivated land, and sparsely-vegetated uncultivated rangeland. The dry bed of Hamoun Puzak is the major source supplying sediment for dust samples, with cultivated land contributing more than Hamoun Saberi or uncultivated areas. Robust estimates of uncertainty reveal that whilst the other three dust sources are broadly similar in magnitude, the western end of Hamoun Puzak is undoubtedly the main source.

The samples used for these analyses were collected over a three-month period, during the first half of which the surface water extent of both Puzak and Saberi lakes decreased by > 98%.Yet, the relative contributions from the different land classes remained remarkably consistent. We also note that despite a persistent seasonal wind bearing NW-NNW upon Zabol, the main dust source lies to the northerly segment of the winds observed. This suggests that either the median wind direction is not the most dust-bearing, or the transport pathways are more complex than suspected.

Our results demonstrate both the potential and the necessity of combining quantitative provenancing techniques with robust uncertainty methods and, ultimately, improved land management. The straightforward approach of linking the main wind direction to a large and rapidly-drying lake bed (Hamoun Saberi) does not yield a good outcome, in this case. Spatial variation in dust sources has been identified elsewhere, most strikingly at the Bodélé Depression in the Chadian Sahara (Washington et al., 2003); here we demonstrate the application of methods with the scope to identify such spatial variation from the point of receipt of the dust. We are unable to outline the exact reasons for Hamoun Puzak's susceptibility to aeolian erosion. However, we attribute notable influence to the geomorphological conditions of the western arm of the Puzak, with its array of interconnected small basins and spillways proving more prone to generating dusts emissions.

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Table 1: Summery characteristics of dust samples collected in Zabol during 21 June to 4 October, 2014.

dust sample no	Sampling date	W V* (m/s)	dust mass (ug/m³)	dust sample no	Sampling date	W V* (m/s)	dust mass (ug/m³)
1	23 June 2014	7.1	270.2	30	24 August 2014	6.5	300.15
2	25 June 2014	8.6	1180.5	31	25 August 2014	9	695.65
3	26 June 2014	6.3	380	32	26 August 2014	10.4	1100.15
4	29 June 2014	8.1	500.6	33	27 August 2014**	9.8	1052
5	30 June 2014	9	458	34	27 August 2014***	9.8	922.39
6	4 July 2014	9.5	7800	35	28 August 2014	8	606.29
7	5 July 2014	8.9	570.2	36	29 August 2014	7.4	271.3
8	11 July 2014	10.3	724	37	3 Sep 2014	10	3594.92
9	12 July 2014	10.5	1700.7	38	4 Sep 2014	11.4	3594.92
10	13 July 2014	10.5	831	39	5 Sep 2014	8.1	339.56
11	14 July 2014	8.9	370.3	40	6 Sep 2014	8.5	1243.88
12	18 July 2014	11.3	740.2	41	8 Sep 2014	6.1	186.21
13	19 July 2014	8.3	1800	42	10 Sep 2014	9.1	3188.85
14	21 July 2014	4.8	320.8	43	13 Sep 2014	7.4	296.8
15	3 August 2014	12.3	715.3	44	16 Sep 2014	4.6	213.77
16	4 August 2014	10	1600	45	17 Sep 2014	4.3	192.4
17	5 August 2014	5.1	216	46	19 Sep 2014	10.1	10785.5
18	6 August 2014	6.6	246.4	47	20 Sep 2014	8.5	1013.43
19	7 August 2014	10	1500	48	21 Sep 2014	4.9	111.76
20	8 August 2014	10	1803	49	22 Sep 2014	2.8	179.61
21	9 August 2014	10	480.04	50	24 Sep 2014	3.4	155.98
22	10 August 2014	8	560.04	51	26 Sep 2014	2.3	165.52
23	11 August 2014	9.1	4500	52	27 Sep 2014	5.5	274.83
24	12 August 2014	8.3	720	53	29 Sep 2014	4	90.31
25	13 August 2014	8.3	1480	54	30 Sep 2014	7.1	814.46
26	14 August 2014	11.5	9004.58	55	1 Oct 2014	5.1	413.15
27	16 August 2014	10.9	3529.4	56	2 Oct 2014	5.3	331.67
28	18 August 2014	5.1	126.74	57	3 Oct 2014	7.5	597.6
29	21 August 2014	8.3	498.5				

* W V indicates Wind Velocity; ** Sample collected on the day; *** Sample collected on the night.

Table 2: Results of a two-stage statistical process for selecting optimum composite fingerprints for distinguishing sources of dust.

Kruskal-Wallis H test			Stepwise DFA		
Fingerprint property	Chi-Square	P-value	Step	Entered fingerprint	Wilk's lambda
Trace elements			1	Fe	0.356
Au	6.79	0.079	2	Sr	0.188
Pt	1.58	0.664	3	Mn	0.081
Mg	20.83	0.000**	4	Cr	0.053
Al	1.48	0.686	5	Pb	0.033
Sr	20.6	0.000**			
Li	22	0.000**			
Fe	20.9	0.000**			
Cr	19.5	0.000**			
Cu	19.5	0.000**			
Zn	5.16	0.16			
As	9.9	0.019*			
Ni	20.75	0.000**			
Pb	9.58	0.022*			
Mn	20.24	0.000**			
Co	18.73	0.000**			
Sn	12.9	0.005**			
Ions					
Na^+	7.24	0.065			
NH^+_4	2.6	0.456			
K^+	4	0.254			
Cl^-	0.4	0.941			
NO^-_2	3.3	0.358			
NO^-_3	7.01	0.072			
Mg^{2+}	1.38	0.709			
Ca^+	14.5	0.002**			

*Statistically significant at P<0.05

** Statistically significant at P<0.01

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Using GLUE to pull apart the provenance of atmospheric dust

Reza Dahmardeh Behrooz[1], Hamid Gholami[2*], Matt W. Telfer[3*], John D. Jansen[4*], Abolhassan Fathabadi[5]

1. Department of Environmental Sciences, Faculty of Natural Resources, University of Zabol, Zabol, Sistan, Iran
2. Department of Natural Resources Engineering, University of Hormozgan, Bandar-Abbas, Hormozgan, Iran.
3. School of Geography, Earth and Environmental Sciences, Plymouth University, Plymouth, Devon PL4 8AA, UK
4. Department of Geoscience, Aarhus University, Aarhus, 8000, Denmark.
5. Department of Range and Watershed Management, University of Gonbad-e-Kavoos, Gonbad-e-Kavoos, Golestan, Iran

Corresponding Authors: Hamid Gholami, hgholami@hormozgan.ac.ir; Matt W Telfer, matt.telfer@plymouth.ac.uk; John Jansen, jdj@geo.au.dk

HIGHLIGHTS

- The GLUE model is applied to reveal the provenance of aeolian dust in Zabol, Iran.
- Quantitative estimates and uncertainties of four different dust sources are discriminated.
- One dry lake, Hamoun Puzak, is by far the dominant dust source, despite others nearby.
- The major dust source is insensitive to large changes in exposed local dry lake beds.
- Cultivated land is the second most important dust source, ahead of some playas.

Using GLUE to pull apart the provenance of atmospheric dust

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Abstract

Identifying the sources of aeolian dust is a crucial step in mitigating the associated hazards. We apply a Generalized Likelihood Uncertainty Estimation (GLUE) model to constrain the uncertainties associated with sediment fingerprinting of atmospheric dust in the Sistan region on the Iran-Afghanistan border, one of the world's dustiest places. 57 dust samples were collected from the rooftop of the Zabol Department of Environmental Protection during a summer dusty period from June to October 2014, in addition to 31 surface soil samples collected from potential sources nearby, including cultivated land (n=8), uncultivated rangeland (n=7), and two dry lakes: Hamoun Puzak (n=10) and Hamoun Saberi (n=6). Dust and soil samples were analyzed for 24 tracers including 16 geochemical elements and 8 water-soluble ions. Five optimum composite fingerprints (Fe, Sr, Mn, Cr and Pb) were selected for discriminating sources by a two-stage statistical process involving a Kruskal-Wallis test and stepwise discriminant function analysis (DFA). Uncertainty ranges for source contributions of dust determined by the GLUE model showed that the dry lake Hamoun Puzak is the dominant source for all dust samples from Zabol and cultivated land is a secondary source. We found marked spatial variance in the importance of regional dry lake beds as dust sources, and temporal persistence in dust emissions from Hamoun Puzak, despite very large areas of adjacent lake beds drying during the study period. Aeolian sediment fingerprinting studies can benefit considerably from the constraints provided by modelling frameworks, such as GLUE, for quantifying the uncertainty in dust provenance data.

KEY WORDS: Sediment fingerprinting, uncertainty, GLUE, atmospheric dust, Iran

1. Introduction

Constraining the source of atmospheric dust particles circulating in the ancient past as well as the present-day is essential to understand the manifold implications of dust in the Earth system (Ridgwell, 2002; Goudie and Middleton, 2006; Shao et al., 2011). Ancient dust deposits are also archives of long-term environmental change (Dietze et al., 2016); the best-known and longest being the >8 Myr loess record in China (Sun & Zhu 2010). Present-day dust storms trigger a series of negative off-site and on-site repercussions (Goossens, 2003). Off-site effects include respiratory disease in humans and animals, contamination of food and water supplies, and interference with traffic safety, machinery, and electronics. On-site effects include the loss of soil organic matter, nutrients, and overall agricultural productivity (Goudie and Middleton, 2006). From this perspective, identifying sources of dust and quantifying multi-source contributions and their uncertainties is a key step towards hazard mitigation, especially in drylands.

A diverse range of techniques have been employed for tracing sources of atmospheric dust, including isotopic ratios (e.g., Krom et al., 1999; Nakano et al., 2004; Grousset and Biscaye, 2005; Chen et al., 2007; Cao et al., 2008; Wang et al., 2005; Rio-Salas et al., 2012; Yang et al., 2009); mineralogical and chemical characteristics (Shen et al., 2009); meteorological data (Rezazadeh et al., 2013; Nabavi et al., 2016; Ge et al., 2016; Rashki et al., 2017); synthesis of isotopic and geochemical data (e.g., Aarons et al., 2017; Wei et al., 2017; Chavagnac et al., 2008); synthesis of trace element and water-soluble ion analyses (Dahmardeh Behrooz et al., 2017a,b); numerical simulation (Hamidi et al., 2014; Nabavi et al., 2017); satellite data (Long et al., 2016; Cherboudj et al., 2016; Schepanski et al., 2012); and multidisciplinary approaches (Yan et al., 2015; Cao et al., 2015). While most of the studies listed above are highly successful at inferring dust sources, we note that in many cases the uncertainties associated with ascribing provenance are not considered formally. This is an important omission for two reasons: 1) airborne dust is commonly generated simultaneously from multiple populations and areas of fine-grained particles; and 2) these multiple populations are, in turn, typically an amalgam generated from different sources and mixed to differing degrees over timescales ranging from geological to individual storm events. In other words, dust provenance presents a very challenging mixing problem and hence uncertainty is fundamental. These two points ultimately stem from geomorphic processes of fine-particle production, transport, deposition, and reworking.

Sediment fingerprinting is widely used to quantify source contributions of fluvial sediments (e.g., Collins et al., 1997; Walling, 2005; Stone et al., 2014; Zhou et al., 2016a; and Manjoro et al., 2017) and its application to aeolian studies is growing (e.g., Liu et al., 2016; and Gholami et al., 2017a,b). Moreover, the uncertainties involved with this method are gaining increased attention (Walling, 2013). In order to manage and quantify the uncertainty in fluvial sediment fingerprinting, some studies have applied a Monte Carlo simulation framework (e.g., Motha et al., 2003; Collins et al., 2012; Voli et al., 2013; Smith and Blake, 2014; Stone et al., 2014; Sherriff et al., 2015; and Vale et al., 2016). Similarly, Bayesian approaches are also applied to fingerprinting aeolian sands (Gholami et al., 2017b) and fluvial sediments (e.g., Massoudieh et al., 2013; Cooper et al., 2014; Cooper et al., 2015; Stewart et al., 2015; and Abban et al., 2016). Yet, several challenges remain in adequately capturing the uncertainty associated with diverse aeolian dust sources and pathways (Walling, 2013) and we suggest that techniques developed in other disciplines may offer a way forward (Gholami et al., 2017b).

First proposed for hydrological modelling by Beven and Binley (1992), GLUE (Generalized Likelihood Uncertainty Estimation) has gained much favour as a tool for evaluating uncertainty estimates (e.g., Hassan et al., 2008; Zhou et al., 2016b; Viola et al., 2009; Mantovan and Todini, 2006; Gong et al., 2011). Here, we apply GLUE to the problem of dust provenance in the Sistan-Hamoun region on the Iran-Afghanistan border. Since it constitutes a major dust source for south-west Asia, Sistan has been the focus of numerous previous investigations (e.g., Goudie and Middleton, 2006; Rashki et al., 2012, 2013 a,b, 2015; Alizadeh Choobari et al., 2014). Recent work has generated an important dataset of dust samples from a meteorological station at Zabol, which has been analyzed geochemically to provide qualitative estimates of source (Dahmardeh Behrooz et al., 2017a) and the temporal variability of dust emissions (Dahmardeh Behrooz et al., 2017b). Here we provide the first attempt to formally quantify aeolian dust provenance and associated uncertainties with this dataset using GLUE.

2. Study area

The Sistan-Hamoun study area (Fig. 1) straddles the border between Afghanistan and the Sistan and Baluchestan province of south-eastern Iran (30˚5′ to 31˚28′ N and 61˚15′ to 61˚50′ E) (Rashki et al., 2012, 2013a). The Hamoun Lakes complex comprises three main lakes: Hamoun Hirmand, Hamoun Saberi, and Hamoun Puzak, which are recharged primarily from Afghanistan by the Hirmand (Helmand) River with smaller contributions from streams to the north and west (Esmaeili and Omrani, 2007). Following exceptionally high runoff, the lakes form a single body of water ~5700 km^2 in area and ~13 Mm^3 in volume (Sharifikia, 2013), though such events have become rare in recent decades while dust emissions have grown correspondingly in magnitude (Goudie and Middleton, 2006; Rashki et al., 2012).

[Approximate location of figure 1]

The climate in the Sistan region is arid to hyper-arid, and land-use is chiefly linked to agriculture and fishing. At Zabol meteorological station (Fig. 1), mean rainfall is 55 mm y^{-1} and mean evaporation is >4000 mm y^{-1} (Moghaddamnia et al., 2009). The prevailing wind is the notorious Levar or "Wind of 120 Days" from the north, which in the summer is accelerated into a Low-Level Jet (LLJ) by a persistent high-pressure system over the Hindu Kush and the channeling effect of the surrounding topography (Alizadeh-Choobari et al., 2014; Kaskaoutis et al., 2016, 2017). As a result, the city of Zabol and its ~135,000 inhabitants experience dust storms of catastrophic proportions, resulting in Zabol ranking as the world's most polluted city for particulate matter less than 2.5 µm (PM2.5) in size (World Health Organisation, 2016).

3. Methods

3.1 Field sampling

We set out to characterise the soil materials for four different potential sources of atmospheric dust emissions to the north of Zabol city (Fig. 1): 1) the dry lake-bed of Hamoun Puzak (Fig. 2a); 2) the dry lake-bed of Hamoun Saberi (Fig. 2b); 3) cultivated arable farmland generally without crop-cover in summer (Fig. 2c); and 4) bare rangeland surfaces with sparse to negligible natural vegetation cover (Fig. 2d). A total of 31 surficial soil samples (<5 cm depth in a 30 cm^2 area) were collected from the four potential sources (Table 1). We sieved the soil samples with a 400-mesh sieve, retaining particles with a nominal geometric diameter of < 38.5 µm, which is roughly equivalent to the aerodynamic diameter of dust (Cao et al., 2008). After sieving, we retained about 5 g of dust-sized material from each sample.

[Approximate location of Figure 2]

During an exceptionally dusty summer period in Zabol (23 June to 4 October 2014), 57 atmospheric dust samples were collected at one- to four-day intervals (Table 1) with sampling apparatus fitted to the rooftop of the Department of Environmental Protection (5 m above ground level, 31˚N, 61.3˚E) in an outer suburban area with no major industrial activities nor local fugitive dust sources. Our two dust samplers (Model Chrono, Zambelli, Milan) were equipped with cyclones operating at a flow rate of 16.7 L/min as per the EU norms (Dahmardeh Behrooz et al, 2017a; 2017b). Total suspended-particle

(TSP) samples were collected in Teflon filters (0.45 µm pore size and 47 mm diameter) and then desiccated for 24-hours at 25 °C. Dust mass concentrations were measured gravimetrically by weighing the Teflon filters before and after sampling using an analytical balance (Adam model) with ±0.1 mg precision. We refrigerated all dust samples at 4˚C until chemical analysis (Dahmardeh Behrooz et al., 2017 a).

3.2 Laboratory analysis of water-soluble ions and trace elements

We measured the concentrations of 8 water-soluble ions in our samples (viz., Na^+, NH^+_4, K^+, Ca^{2+}, Mg^{2+}, Cl^-, NO^-_3, NO^-_2). Three cations (Na^+, NH^+_4 and K^+) were measured with a Shim-pack IC-C1 (Shimadzu DGU-12A) using 5-mM HNO_3 solution as eluent. Three anions (Cl^-, NO^-_3 and NO^-_2) were measured with a Shim-pack ICA1 (Shimadzu DGU-12A), using 2.5–mM phthalic acid combined with 2.4-mM tris-(hydroxymethyl) aminomethane as eluent (Lin, 2002). Two cations (Ca^{2+} and Mg^{2+}) were measured via flame atomic absorption spectrometry (Philips, PU9400X, England).

After acid digestion, all samples were analyzed to determine the concentrations of 16 trace elements (viz., Al, As, Au, Co, Cr, Cu, Fe, Li, Mg, Mn, Ni, Pb, Pt, Sn, Sr, and Zn) via Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-OES, Perkine Elmer, Optima 2000, USA). Further details of sampling and laboratory procedures are given in Dahmardeh Behrooz et al. (2017a, b).

3.3 Two-stage method: Kruskal-Wallis test and discriminant function analysis

The measurements of 8 water-soluble ions and 16 trace elements form the basis of the sediment fingerprinting method aimed at identifying the source contribution of the Zabol dust samples. We adopt a two-stage statistical procedure following the approach of Collins et al. (1997) to identify the most suitable tracers for source discrimination. In stage one we tested the primary ability of tracers to discriminate dust sources using the Kruskal-Wallis H test. Tracers with critical values at the 95 % confidence levels or better were taken to the second stage in which we identified optimum composite fingerprints using a stepwise discriminant function analysis based on minimization of Wilk's lambda.

3.4 Generalised Likelihood Uncertainty Estimation (GLUE)

GLUE was first devised by Beven and Binley (1992) as a means of sensitivity analysis and uncertainty estimation in environmental model outputs. We use GLUE to quantify the uncertainty in the sediment fingerprinting results via the following five steps:

1) Random sampling of parameter sets (300,000 iterations) are conducted using the Latin Hypercube Sampling (LHS) method (Zhou et al, 2016b), by assuming source contributions from each source are non-negative and that total contributions sum to unity. Due to the lack of prior information, we used a uniform distribution as the prior distribution for all parameters.

2) Selection of a likelihood function and behavioral parameter thresholds. Here, we adopt the Nash–Sutcliffe coefficient (ENS) as the likelihood function (Jin et al, 2010):

$$ME = 1 - \frac{\sum(O_{obs} - O_{sim})}{\sum(O_{obs} - \hat{O}_{obs})} = 1 - \frac{\sigma_i^2}{\sigma_{obs}^2} \qquad \text{(eq.1)}$$

where $\underset{\cdot}{O}_{obs}$ is the mean value of the observed tracer concentration; O_{sim} is the simulated tracer concentration; O_{obs} is the observed tracer concentration; σ^2_i is the error variance for the *i*th model (i.e., the combination of the model and the *i*th parameter set) and σ^2_{obs} is the variance of the observations.

3) Sampled parameter sets from step 1 are input to the mixing model (equation 2) and the likelihood function is calculated for each parameter set as:

$$C_{dust} = C_{Sources} \times P \qquad \text{(eq. 2)}$$

where P is an m dimensional column vector of sources contribution (sampled parameter sets), C_{dust} is an n-dimensional column vector of element concentration in sediment sample, $C_{Sources}$ is an n×m-dimensional matrix representing mean tracer concentration in sources (each row represents mean tracer concentration in each source), where n is the number of optimum composite fingerprints (n=5) and m is the number of dust sources (m=4).

4) Parameter sets are divided into behavioural and non-behavioural types with respect to a threshold value (Zhou et al, 2016). In this step, those parameter sets that have likelihood functions greater than a threshold value were classified as behavioural parameter sets. For the next step, non-behavioural parameter sets were discarded.

5) For behavioural parameter sets, likelihood weights are rescaled such that they sum to one, then each parameter is sorted and we calculate cumulative distributions for each parameter. Quintiles and uncertainty intervals are calculated via the cumulative distributions.

3.5 Geospatial analysis and climate data

Landsat data were downloaded from the United States Geological Survey's Earth Explorer, and all analysis was conducted within ArcGIS 10.3. Quantitative analysis of water extent was conducted using a modified Normalized Difference Water Index (NDWI), based on the green (Band 3) and short-wave infrared (Band 6) bands of Landsat 8 data (Xu, 2006). Climate data are taken from the Hourly Global Surface Data (DS3505) dataset for the Zabol station (World Meteorological Organization ID: 40829), accessed via the legacy Climate Data Online (CDO) portal of the National Oceanic and Atmospheric Administration's (NOAA) National Climatic Data Centre (NCDC) online (available at https://www7.ncdc.noaa.gov/CDO/dataproduct). To calculate back-trajectories for the air mass affecting Zabol during selected peak dust events of the observation period, we use NOAA's Hysplit model (summarized in Stein et al., 2015). Ensemble runs for the 24 hours preceding peak dust events were run based on airflow at 50, 100 and 500 m above the land surface. Additional remote sensing data regarding atmospheric dust flux and dust deposition was derived from NASA's Earthview platform in the form of a) visible MODIS imagery from July 2014, b) the MERRA-2 monthly PM2.5 dust deposition product for July 2014 (see Gelaro et al., 2017) and c) the AURA Ozone Monitoring Instrument (OMI) Aerosol Index product for July 2nd (Torres, 2006), near the start of the observed period of study.

4. Results

4.1 Kruskal-Wallis test and discriminant function analysis

The results of the Kruskal-Wallis tests (Table 2) indicate that among the twenty-four measured properties (8 water-soluble ions and 16 element concentrations), thirteen trace elements (Mg, Sr, Li, Fe, Cr, Cu, As, Ni, Pb, Mn, Co, and Sn) and one ion (Ca^{2+}) show statistically significant differences at the 95 %-level between the four potential dust sources (the two dry lake beds, cultivated farmland, and rangeland areas of natural vegetation cover). Trace elements clearly out-performed water-soluble ions for tracking spatial sources of dust. Thirteen trace elements were passed to stage-two for stepwise discriminant function analysis (DFA). The DFA yielded five trace elements (Fe, Sr, Mn, Cr and Pb) with optimum composite fingerprints that correctly discriminate 87 % of our source samples (Table 2 and Fig. 3).

[Approximate location of Figure 3]

4.2 Using GLUE to constrain uncertainty in the source contributions of dust

Uncertainty intervals of source contributions estimated by the GLUE-mixing model at the 95 % confidence level are presented in Figure 4. These results show that the most important dust source is clearly Hamoun Puzak (Figs. 4a and 5). Median contributions from this lake-bed span 29 to 88 % (samples 33 and 45, respectively). Hamoun Saberi is a less important source for our samples. Median contributions from this lake bed span 3 to 24 % (samples 11 and 33, respectively) (Fig. 4b). The sparsely vegetated rangeland is the least active dust source. Median contributions span 2 to 22 % (samples 45 and 34, respectively) (Fig. 4c). Cultivated farmland is recognized as the second-most important source for all of 57 samples. Median contributions from farmland span 4 to 25 % (samples 23 and 34, respectively) (Figs. 4d). For most samples, the lower-limit of predicted uncertainty is zero for contributions from the three sources other than Hamoun Puzak (Figs. 4b-d). Figure 5 presents an overview of the source contributions with all samples plotted together as a frequency histogram.

Overall, a strong correlation (Pearson's $r = 0.783$, $p < 0.001$) can, unsurprisingly, be seen between dust flux and the logarithm of the windspeed, with a threshold of around $10\ m\ s^{-1}$. However, no significant correlation is observed between the relative contribution of Hamoun Puzak and Saberi, and either the windspeed (Pearson's $r = -0.06$, $p = 0.66$) or wind direction (Pearson's $r = 0.238$, $p = 0.07$).

[Approximate location of Figure 4]

[Approximate location of Figure 5]

5. Discussion

Sediment fingerprinting is a highly effective technique for quantifying source contributions of fluvial sediments (e.g. Collins et al., 1997; Walling, 2005; Stone et al., 2014; Zhou et al., 2016a; Manjoro et al., 2017) and aeolian sands (e.g. Liu et al., 2016; Gholami et al., 2017a,b). Here we build upon this approach by exploring the potential of the GLUE methodology for distinguishing spatially proximal aeolian dust sources with similar underlying geology and geomorphology. We demonstrate its efficacy at formally quantifying the uncertainty distributions associated with aeolian dust fingerprinting due to spatial and temporal variation in the dust cycle. Further, we use the method to reveal spatial complexity—alongside an unexpected lack of temporal complexity—in the nature of the dust sources.

5.1 Environmental context of dust emissions

The strong correlation between dusty days and the surface area of exposed lake floors indicates that i) dust storms in the Sistan-Hamoun region are directly related to the dryness of the Hamoun Lakes, and ii) these lake beds are the main source of dust emissions (Goudie and Middleton 2006; Rashki et al., 2012; 2013a; 2013b; 2015). Such relationships are not uncommon, as worldwide observations suggest that exposed dry lake beds can govern the frequency and intensity of dust storms; for example, at Owens Lake, USA (Reheis et al., 2009); Aral Sea, Uzbekistan (Breckle et al., 2012); Makgadikgadi pan complex and Etosha Pan, southern Africa (Prospero et al., 2002; Mahowald et al., 2003; and Washington et al., 2003); and Kata Thandi-Lake Eyre, Australia (Baddock et al., 2009).

The frequency and magnitude of dust emissions from the Hamoun Lakes has also been related to annual/decadal scale variations in the surface area of the lakes, which varies dramatically. At its maximum extent, observed following the 1998 spring-melt Hirmand River floods (Rashki et al., 2012a), the Hamoun lake complex forms a single body of water ~4500 km^2 in area. This is comprised of Hamoun Hirmand (~1400 km^2), Hamoun Saberi (~1400 km^2) and Hamoun Puzak (~1700km^2), with Hamoun Baringak forming a series of smaller lakes and spillways between Saberi and Puzak. During more typical lake-full episodes, these bodies of water are not conjoined; for instance, during the spring of 1996 (Figure 6a), Hamoun Saberi spanned ~815 km^2 and Hamoun Puzak spanned 375 km^2 (Hamoun Baringak is here considered part of the western extent of the Puzak basin). Hamoun Hirmand lies mostly downwind of Zabol, and hence is not considered further. Between 1999 and 2010, a prolonged drought likely related to the El Nino Southern Oscillation resulted in the rapid and sustained desiccation of the Hamoun Lakes, with a concomitant increase in the frequency of dusty days (Rashki et al., 2012; 2013b). Since 2010, lake levels have been highly variable (Fig. 6), with a return to lake-full conditions experienced around 2011, and a subsequent return to large areas of exposed dry lake beds in more recent years.

Such changes can also develop rapidly. Within the timeframe of this study (June-October 2014), Hamoun Puzak and Hamoun Saberi lost around 295 km^2 and 640 km^2 of water surface area, respectively (i.e. 98.5% and 99.9% of their extent on June 14th) (Fig. 7). The small, shallow Hamoun Baringak lakes, directly to the north of Zabol, dried especially early in the season, with almost all water lost by July 2014. This desiccation affected Hamoun Saberi and Puzak proportionally at very similar rates, but Saberi's larger surface area at the start of this study led to greater absolute change in the lake floor exposure area. During this period, there was no rainfall recorded at the Zabol meteorological station, but the persistent 'Wind of 120 Days' blew from 327° ± 36° during June-October, with daily average windspeeds up to 15 m s^{-1} (~35 mph) and 50 of the 138 days of the study period exceeding daily averages of ~9 m s^{-1} (~20 mph).

[Approximate location of Figure 6]

[Approximate location of Figure 7]

5.2. Dust sources: dry beds of Hamoun Lakes

The GLUE results (Figs 4 and 5) reveal that the dominant source of dust collected at Zabol is Hamoun Puzak, and also that in general, the dust sources vary little over the three-month period from June to the beginning of October, 2014. This finding is unexpected, for a number of reasons. Firstly, as the wind at Zabol during this period comes from the northwest to north-northwest (327° ± 36°), the most obvious candidate source of Zabol dust is the upwind Hamoun Saberi (Figures. 1 and 7). Yet, consistently, Hamoun Puzak contributes ~40-90% (uncertainties included) of the dust received at Zabol. Furthermore, given the rapid increase in Saberi's exposed dry bed during the early period of sampling, its contribution would be expected to increase proportionally over this period. But Saberi's contributions actually vary little during the season (Fig. 4). When the surface area of the lakes is considered, either relative to lake-full conditions (Figure 8a and 8c), or as absolute surface areas (Figure 8b and 8d), there is little temporal relationship with the relative dust contributions of the two lake beds.

[Approximate location of Figure 8]

Similarly, investigation of the meteorological conditions during June-October do not readily explain the dominance of Hamoun Puzak. There is no clear correlation with either wind magnitude, or direction, that can readily explain the dominance of Puzak, and no obvious explanation for the occasional excursions when other sources contribute markedly more. For instance, on August 27th replicate samples were collected (S33 and S34 in Fig. 4) and yield consistent results (note that, for consistency, only one of these samples is included in Figs. 8 and 9). Satellite aerosol observations, modelled dust deposition, and aerosols indices (Figure 10), whilst confirming the regional importance of the Hamoun Lakes, do not readily identify localized sources due to their relative spatial coarseness.

These results suggest that Hamoun Puzak—or at least Hamoun Baringak at the western margin of Hamoun Puzak, where the source samples were collected (Fig. 1)—is a prolific and persistent source of dust over Zabol irrespective of the existence of large adjacent alternative sources. Why is Hamoun Puzak such an effective dust emitter? And why, despite its size and position directly upwind of Zabol, does Hamoun Saberi contribute relatively little? We propose that several factors have an influence, as follows.

[Approximate location of Figure 9]

[Approximate location of Figure 10]

First, let us consider the hydrological setting and sedimentology of the area. The Hamoun Lakes are predominantly fed with water from the Hirmand River to the east, with the Khash River also feeding directly into Hamoun Puzak from the east. Hamoun Saberi is fed largely from the north by the Harut and Farah rivers. The sampled region around Hamoun Baringak and Hamoun Puzak lies in series of channels and small closed basins which act as spillways connecting the lakes during lake-full episodes. Such areas are likely to have distinctly different sedimentology from lake bed areas subject to direct lacustrine sedimentation (King et al., 2011, Sweeney et al., 2011). Evidence that these sediments are distinct from those of Hamoun Saberi is implicitly provided in the primary function of the DFA used here to define the characteristics of these dusts (Fig. 3). The wind regime necessary for aeolian transport to Zabol from Hamoun Baringak is compatible with this zone being a dominant source, as northerly orientation lies well within the 327° ± 36° (one sigma) direction of the observed winds. It may also be that the transport pathways associated with the low-level jet from the north are strongly impacted by topography. We note that wind streaks evident on the satellite imagery of the region (visible in the northern and western part of Figure 7) suggest that topography is steering and deflecting local winds in a complex manner. For instance, topographic roughness caused by rocky hills around Hamoun Saberi may reduce effective winds at the surface. Further detailed analysis of the possible role of topographic steering of the winds in this, and other comparable studies, is clearly desirable, but does require high-resolution data and sophisticated modeling of the wind field to achieve substantial results.

The mean orientation of the seasonal winds, however, raises another question: Why does Hamoun Saberi, which lies directly upwind of Zabol, not contribute more to Zabol's dust flux, especially during the latter part of our study when an additional 640 km^2 of dry lake bed became exposed? It is well-reported that dust production can be spatially highly variable, even at sub-basin scales (Mahowald et al., 2003, Reheis et al., 2002, Bullard et al., 2008). One possibility relates to the differing geochemistry of the sediments that can promote the formation of protective crusts. Field experiments have shown that dry river courses when replenished frequently with fine-grained fluvial sediment can be much more effective at producing deflatable dust relative to playas and, counter-intuitively, playa centres are relatively low emission sources (Sweeney et al., 2011, King et al., 2011). Dry lake-bed deposits from the Mojave in the western US, for instance, have been reported to yield less dust than those with fluctuating water-levels (Reynolds et al., 2007), and the progressive and rapid desiccation of Hamoun

Saberi during the study period may have been simply unfavourable for the generation of deflatable dust.

Conversely, it may be that exogenic water supply from the northerly channels sufficiently dampened the surface to limit additional deflation. We note that shallow flooding was observed during the field sample collection, despite the lack of rain observed either in Zabol, or the fortnightly Landsat images. Over longer timescales, rates of aeolian erosion have been shown to inversely relate to soil moisture (Whitney et al., 2015). Although the whole region is sparsely vegetated, the role of vegetation in influencing surface roughness and thus susceptibility to aeolian erosion also cannot be overlooked (e.g. Cowie et al., 2013, Li et al., 2007). Lastly, we point to the cause of the additional 13% of variability, which was not well explained by the discriminant function analysis of the source sediments. This variability may imply that a significant component of Zabol dust derives from outside the immediate area of the Hamoun Lakes. Dust plumes transported from the Karakum desert in Turkmenistan are also known to affect much of southwest Asia, including the Sistan region (Kskaoutis et al., 2015), and may be a source of exogenous dust not accounted for among the four potential sources we sampled. To address these questions, we use the Hysplit model to calculate back-trajectories for three intervals characterized by high dust deposition during the study period (Figure 11). These confirm variability in both localized and regional wind trajectories for the high dust-flux days during period July to September 2014. The role of the Karakum in contributing long-distance flux is supported, over the 24-hour transport window modelled here (Figure 11a). However, there is also evidence of local and regional variability, with July and August winds coming locally via a north-northwesterly track (over a still largely inundated Hamoun Saberi), and the September peak in dust emission driven near Zabol by a due northerly wind, tracking directly over a desiccated Hamoun Baringak and Puzak (Figure 11b). This is likely related to seasonal variation of the Caspian Sea – Hindu Kush Index (CasHKI), a broadly east-west atmospheric pressure dipole between 40–50°N, 50–55°E and 35–40°N, 70–75°E (Kaskaoutis et al., 2016, 2017). This suggests that the interplay of atmospheric and hydrological controls on dust emission is crucial in this region.

[Approximate location of Figure 11]

5.3. Dust sources: cultivated and uncultivated rangeland areas

The connections between land management, agriculture and aeolian dust emissions are well documented (Wiggs and Holmes, 2011, Okin et al., 2001), and the role of agriculture in exacerbating drought-driven dust events such as the decade-scale ‘Dust Bowl’ of 1930s USA is clearly established

(Worster, 2004). We find that the cultivated cropland to the north of Zabol is the region's second-largest overall source of dust (Fig. 4), slightly out-stripping Hamoun Saberi, and contributing substantially more than uncultivated rangeland with sparse vegetation. Desertification, by which we mean mainly arid land degradation, has been recognized in other regions of Iran (Sepehr et al., 2007), and given the difficulties for agriculture in such an extremely dry and hot climate, it becomes apparent that sustainable land management is difficult to achieve. The spread of wind erosion is challenging land managers worldwide - from the Argentinian Pampas (Buschiazzo and Zobeck, 2008) to the Tibetan Plateau (Zhang et al., 2012) and even temperate regions such as southern Sweden (Barring et al., 2003). The findings here that cultivation-based farming is the second largest contributor to Zabol's dust flux (with median contributions of 4-25% for individual samples) highlights an anthropogenic dust source that could be quelled with alternative farming practices.

6. Conclusion

Identifying source(s) of aeolian sediments (sand and dust) is essential to improve planning and management of arid and semi-arid regions. Here we present a quantitative sediment fingerprinting approach coupled with the GLUE methodology to quantify source contributions of dust to the city of Zabol in the Sistan-Hamoun region of south-east Iran. Zabol consistently ranks globally as one of the most susceptible to fine (PM2.5 and PM10) aerosol pollutants. Using GLUE, we have assigned quantitative estimates of the relative contributions of four potential dust sources: two dry lake beds (Hamoun Puzak and Hamoun Saberi), cultivated land, and sparsely-vegetated uncultivated rangeland. The dry bed of Hamoun Puzak is the major source supplying sediment for dust samples, with cultivated land contributing more than Hamoun Saberi or rangeland areas. Robust estimates of uncertainty reveal that whilst the other three dust sources are broadly similar in magnitude, the western end of Hamoun Puzak (Hamoun Baringak) is undoubtedly the main source.

The samples used for these analyses were collected over a three-month period, during the first half of which the surface water extent of both Puzak and Saberi lakes decreased by > 98%. Yet, the relative contributions from the different land classes remained remarkably consistent. We also note that despite a persistent seasonal wind bearing NW-NNW upon Zabol, the main dust source lies to the northerly segment of the winds observed. This suggests that either the median wind direction is not the most dust-bearing, or the transport pathways are more complex than suspected. Hysplit analyses suggest important temporal variations during the windy season.

Our results demonstrate both the potential and the necessity of combining quantitative provenancing techniques with robust uncertainty methods and, ultimately, improved land management. The straightforward approach of linking the main wind direction to a large and rapidly-drying lake bed (Hamoun Saberi) does not yield a good predictive outcome, in this case. Spatial variation in dust sources has been identified elsewhere, most strikingly at the Bodélé Depression in the Chadian Sahara (Washington et al., 2003); here we demonstrate the application of methods with the scope to identify such spatial variation from the point of receipt of the dust. We are unable to outline the exact reasons for Hamoun Puzak's susceptibility to aeolian erosion. However, we attribute notable influence to the geomorphological conditions of the western arm of the Puzak, with its array of interconnected small basins (Hamoun Baringak) and spillways proving more prone to generating dust emissions.

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Figure Captions

Figure 1: Sampling sites in the Sistan region: the dry-bed of Hamoun Puzak (HP); the dry-bed of Hamoun Saberi (HS); uncultivated range Land (RL); and cultivated Land (CL). Inset shows the hourly averaged wind regime for the period June-October 2014 (data source: National Climatic Data Centre, Climate Data Online).

Figure 2. Typical examples of the land surfaces we sampled. a) Hamoun Puzak dry lake-bed, b) Hamoun Saberi dry lake-bed, c) close-up view of cultivated agricultural land surface during the summer months, and d) sparsely vegetated, uncultivated rangeland.

Figure 3. Scatterplot constructed from the first and second functions derived from a stepwise DFA for the source groups including the four land (i.e. Hamoun Puzak (HP), Hamoun Saberi (HS), cultivated land (CL) and uncultivated rangeland (RL)). Five optimum fingerprints (Fe, Sr, Mn, Cr and Pb) were used to construct the scatterplot and 87% of the source samples are discriminated correctly.

Figure 4. GLUE results for dust source contributions yielding 95% confidence limits (with percentiles 2.5, 25, 50, 75 and 97.5). A) Hamoun Puzak; B) Hamoun Saberi; C) uncultivated rangeland; and d) cultivated land.

Figure 5. Summary of all source contributions plotted as a probability density function.

Figure 6. Decadal scale changes in the Hamoun Lakes. Following lake-full conditions in the late 1990s (a), a sustained decade of drought resulted in the exposure of large areas of dry lake-beds (b and c) and therefore potential dust sources. Since then, levels have varied and often changed rapidly (d, e and f). All images are infrared/red/green composites based on Landsat 5 and 8 imagery, using Bands 4/3/2 and 5/4/3 respectively. Vegetation is shown as red tones.

Figure 7. Changes in the surface extent of the Hamoun Lakes between June and September, 2014. Note the rapid desiccation during June and July, and resultant exposure of new surfaces for potential deflation.

Figure 8. The relative contributions of a) and b) Hamoun Puzak and c) and d) Hamoun Saberi, plotted alongside a) and c) the surface area of the lakes (expressed as a percentage of the 1996 lake-full condtions) and b) and d) the absolute surface area of the lakes. There is no consistent trend in dust provenance, despite the changing area of the potential sources. Solid lines indicate the median estimate, dashed lines the first and third quartiles and dotted lines the 2.5% and 97.5% bounds.

Figure 9. The dominant dust contribution from Hamoun Puzak (top; key for lines as in Figure 8), shown alongside the magnitude of dust collected at Zabol (bars), and the mean daily wind speed (thin solid line) and variance in mean wind direction (bold dashed line).

Figure 10. The dust plume affecting Zabol from the north is evident in remotely-sensed data from July 2014. a) The MODIS Corrected Reflectance Imagery (Red:Green:Blue) 500 m colour composite clearly shows the plume as a brown streak emanating from near the Hamoun Lakes and tracking southeastwards. b) The MERRA-2 PM2.5 monthly dust deposition reanalysis data also highlight the plume to the southeast of the Hamoun Lakes, as does c) the AURA OMI UV-derived Aerosol Index for July 4^{th}, 2014.

Figure 11. Hysplit back trajectories for three dust events during the observation period in summer 2014 (4^{th} July – white triangles, 14^{th} August – grey circles, 4^{th} September – black squares). a) Regional tracks over the 24 hours preceding the observations demonstrate the long-distance transport of dust, with a likely source in the Karakum Desert of Turkmenistan. b) Local dust transport pathways over the

Hamoun Lakes to Zabol demonstrate different pathways during the summer dust season, with early summer pathways (white triangles and grey circles) routing over the still-inundated Hamoun Saberi, and the September trajectory (black squares) coming over the desiccated Hamoun Baringak and Puzak.

Table 1: Summary characteristics of dust samples collected in Zabol during 21 June to 4 October, 2014.

dust sample no	Sampling date	W V* (m/s)	dust mass (ug/m^3)	dust sample no	Sampling date	W V* (m/s)	dust mass (ug/m^3)
1	23 June 2014	7.1	270.2	30	24 August 2014	6.5	300.15
2	25 June 2014	8.6	1180.5	31	25 August 2014	9	695.65
3	26 June 2014	6.3	380	32	26 August 2014	10.4	1100.15
4	29 June 2014	8.1	500.6	33	27 August 2014**	9.8	1052
5	30 June 2014	9	458	34	27 August 2014***	9.8	922.39
6	4 July 2014	9.5	7800	35	28 August 2014	8	606.29
7	5 July 2014	8.9	570.2	36	29 August 2014	7.4	271.3
8	11 July 2014	10.3	724	37	3 Sep 2014	10	3594.92
9	12 July 2014	10.5	1700.7	38	4 Sep 2014	11.4	3594.92
10	13 July 2014	10.5	831	39	5 Sep 2014	8.1	339.56
11	14 July 2014	8.9	370.3	40	6 Sep 2014	8.5	1243.88
12	18 July 2014	11.3	740.2	41	8 Sep 2014	6.1	186.21
13	19 July 2014	8.3	1800	42	10 Sep 2014	9.1	3188.85
14	21 July 2014	4.8	320.8	43	13 Sep 2014	7.4	296.8
15	3 August 2014	12.3	715.3	44	16 Sep 2014	4.6	213.77
16	4 August 2014	10	1600	45	17 Sep 2014	4.3	192.4
17	5 August 2014	5.1	216	46	19 Sep 2014	10.1	10785.5
18	6 August 2014	6.6	246.4	47	20 Sep 2014	8.5	1013.43
19	7 August 2014	10	1500	48	21 Sep 2014	4.9	111.76
20	8 August 2014	10	1803	49	22 Sep 2014	2.8	179.61
21	9 August 2014	10	480.04	50	24 Sep 2014	3.4	155.98
22	10 August 2014	8	560.04	51	26 Sep 2014	2.3	165.52
23	11 August 2014	9.1	4500	52	27 Sep 2014	5.5	274.83
24	12 August 2014	8.3	720	53	29 Sep 2014	4	90.31
25	13 August 2014	8.3	1480	54	30 Sep 2014	7.1	814.46
26	14 August 2014	11.5	9004.58	55	1 Oct 2014	5.1	413.15
27	16 August 2014	10.9	3529.4	56	2 Oct 2014	5.3	331.67
28	18 August 2014	5.1	126.74	57	3 Oct 2014	7.5	597.6
29	21 August 2014	8.3	498.5				

* W V indicates Wind Velocity; ** Sample collected on the day; *** Sample collected on the night.

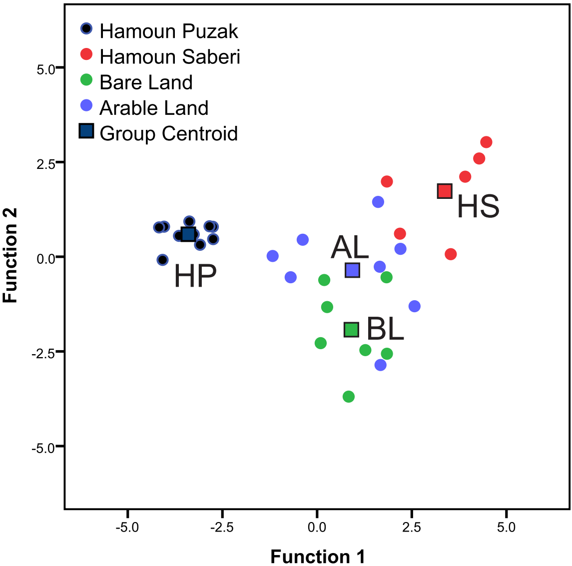
Table 2: Results of a two-stage statistical process for selecting optimum composite fingerprints for distinguishing sources of dust.

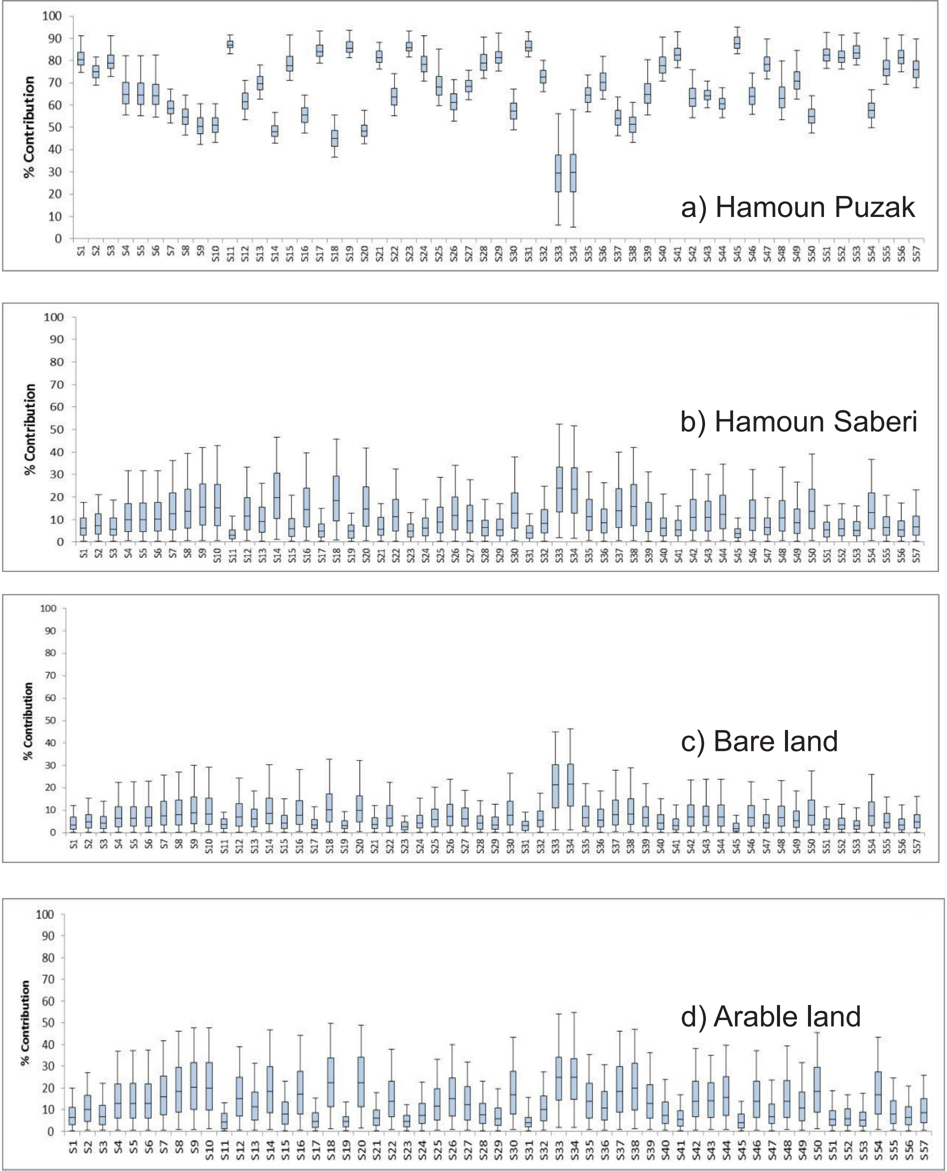
Kruskal-Wallis H test		
Fingerprint property	Chi-Square	P-value
Trace elements		
Au	6.79	0.079
Pt	1.58	0.664
Mg	20.83	0.000**
Al	1.48	0.686
Sr	20.6	0.000**
Li	22	0.000**
Fe	20.9	0.000**
Cr	19.5	0.000**
Cu	19.5	0.000**
Zn	5.16	0.16
As	9.9	0.019*
Ni	20.75	0.000**
Pb	9.58	0.022*
Mn	20.24	0.000**
Co	18.73	0.000**
Sn	12.9	0.005**
Ions		
Na^+	7.24	0.065
NH_4^+	2.6	0.456
K^+	4	0.254
Cl^-	0.4	0.941
NO_2^-	3.3	0.358
NO_3^-	7.01	0.072
Mg^{2+}	1.38	0.709
Ca^{2+}	14.5	0.002**

Stepwise DFA		
Step	Entered fingerprint	Wilk's lambda
1	Fe	0.356
2	Sr	0.188
3	Mn	0.081
4	Cr	0.053
5	Pb	0.033

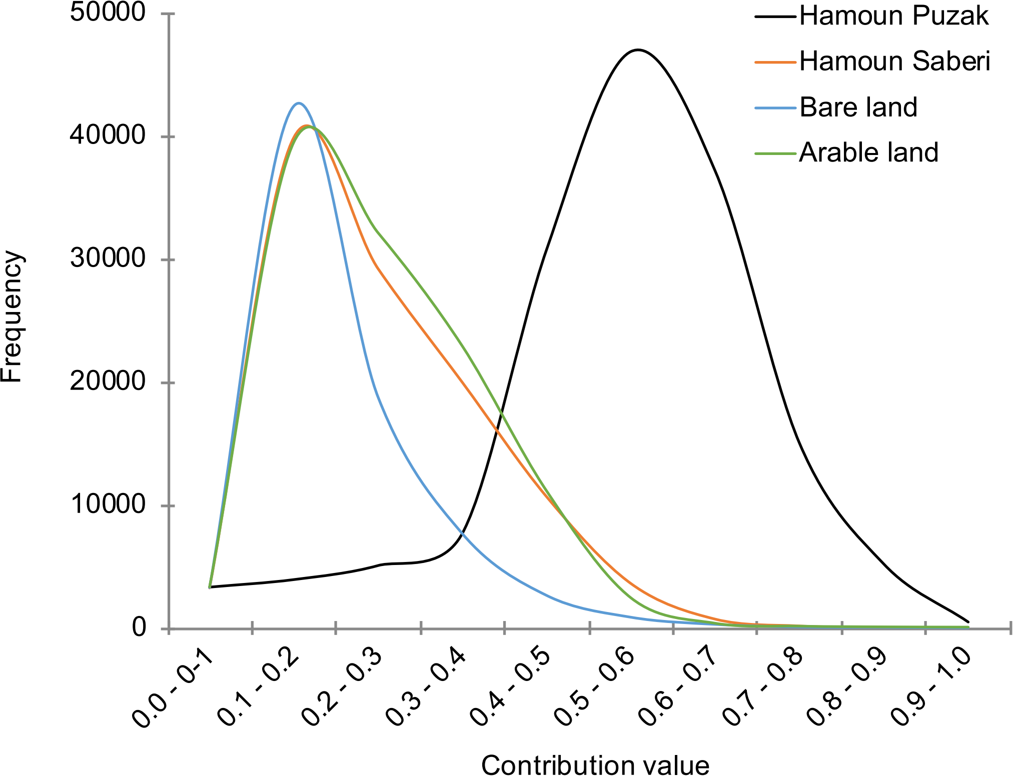
*Statistically significant at P<0.05

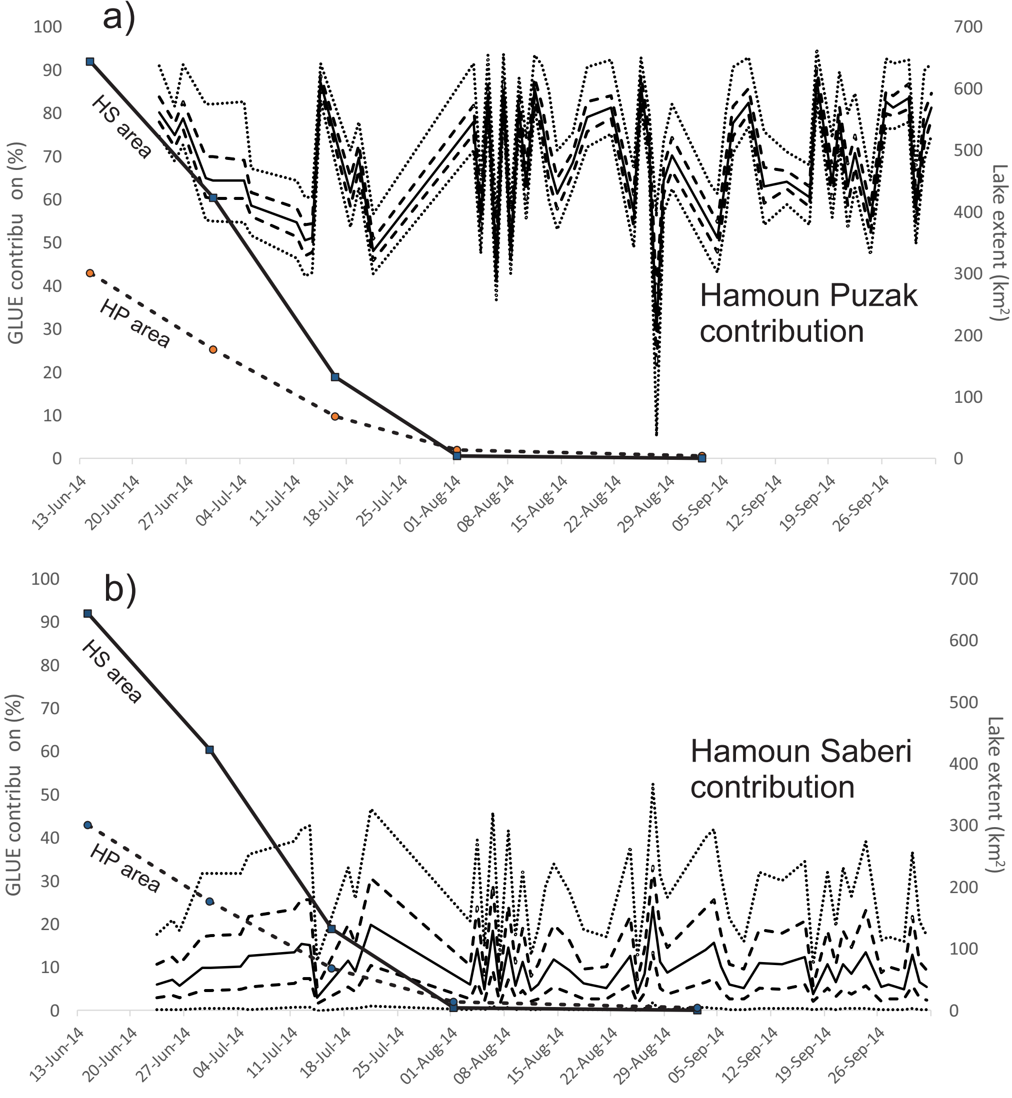
** Statistically significant at P<0.01

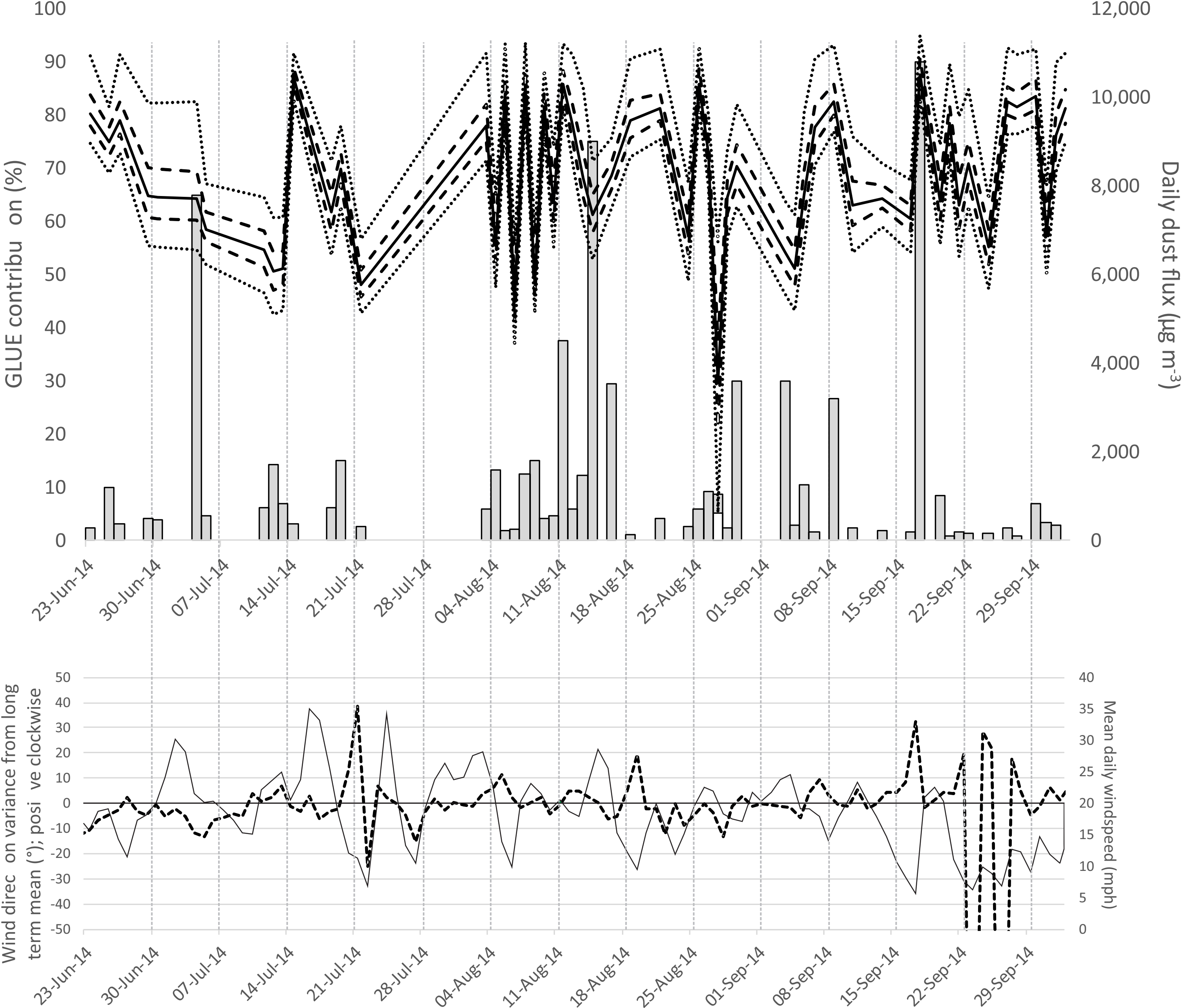

Hamoun Puzak
Hamoun Saberi
Bare Land
Arable Land
Group Centroid
HP
AL
HS
BL
5.0
2.5
0.0
-2.5
-5.0
Function 2
-5.0
-2.5
0.0
2.5
5.0
Function 1



a) Hamoun Puzak
b) Hamoun Saberi
c) Bare land
d) Arable land
% Contribution
0 10 20 30 40 50 60 70 80 90 100
S1 S2 S3 S4 S5 S6 S7 S8 S9 S10 S11 S12 S13 S14 S15 S16 S17 S18 S19 S20 S21 S22 S23 S24 S25 S26 S27 S28 S29 S30 S31 S32 S33 S34 S35 S36 S37 S38 S39 S40 S41 S42 S43 S44 S45 S46 S47 S48 S49 S50 S51 S52 S53 S54 S55 S56 S57


Hamoun Puzak
Hamoun Saberi
Bare land
Arable land
Frequency
50000
40000
30000
20000
10000
0
0.0 - 0-1
0.1 - 0.2
0.2 - 0.3
0.3 - 0.4
0.4 - 0.5
0.5 - 0.6
0.6 - 0.7
0.7 - 0.8
0.8 - 0.9
0.9 - 1.0
Contribution value


a)
100
90
80
70
60
50
40
30
20
10
0
700
600
500
400
300
200
100
0
GLUE contribu on (%)
Lake extent (km2)
HS area
HP area
Hamoun Puzak
contribution
13-Jun-14
20-Jun-14
27-Jun-14
04-Jul-14
11-Jul-14
18-Jul-14
25-Jul-14
01-Aug-14
08-Aug-14
15-Aug-14
22-Aug-14
29-Aug-14
05-Sep-14
12-Sep-14
19-Sep-14
26-Sep-14
b)
GLUE contribu on (%)
Lake extent (km2)
HS area
HP area
Hamoun Saberi
contribution



GLUE contribu on (%)
Daily dust flux (µg m-3)
100
90
80
70
60
50
40
30
20
10
0
12,000
10,000
8,000
6,000
4,000
2,000
0
23-Jun-14
30-Jun-14
07-Jul-14
14-Jul-14
21-Jul-14
28-Jul-14
04-Aug-14
11-Aug-14
18-Aug-14
25-Aug-14
01-Sep-14
08-Sep-14
15-Sep-14
22-Sep-14
29-Sep-14
Wind direc on variance from long term mean (°); posi ve clockwise
Mean daily windspeed (mph)
50
40
30
20
10
0
-10
-20
-30
-40
-50
35
25
15
5

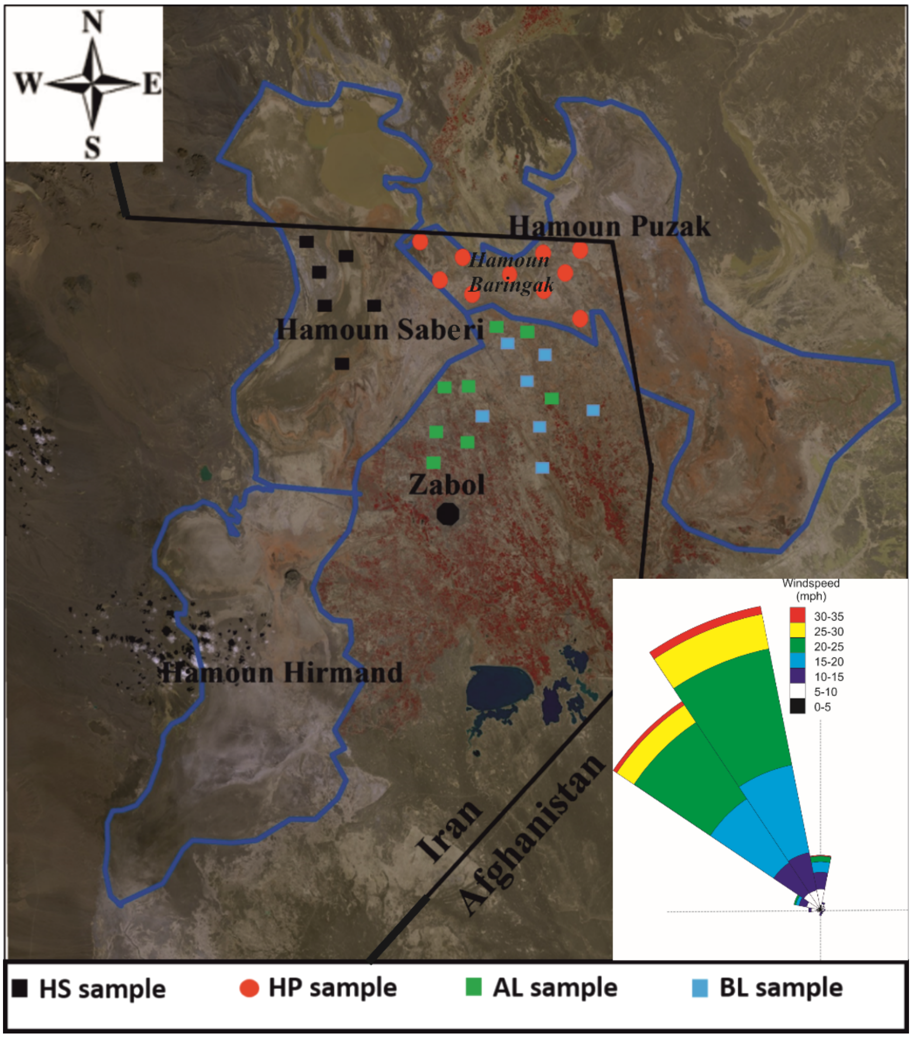

N
W
E
S
Hamoun Puzak
Hamoun
Baringak
Hamoun Saberi
Zabol
Hamoun Hirmand
Iran
Afghanistan
Windspeed
(mph)
30-35
25-30
20-25
15-20
10-15
5-10
0-5
HS sample
HP sample
AL sample
BL sample

Figure 1

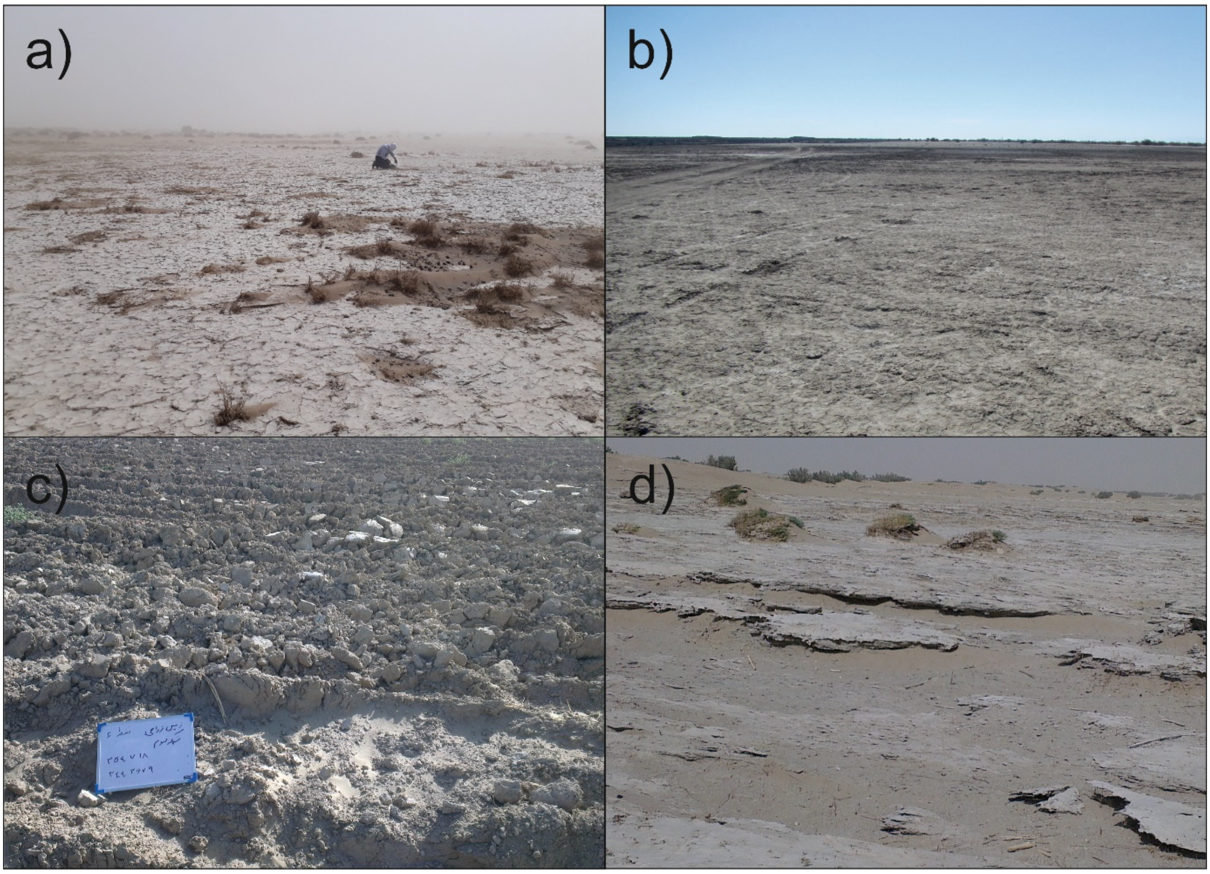
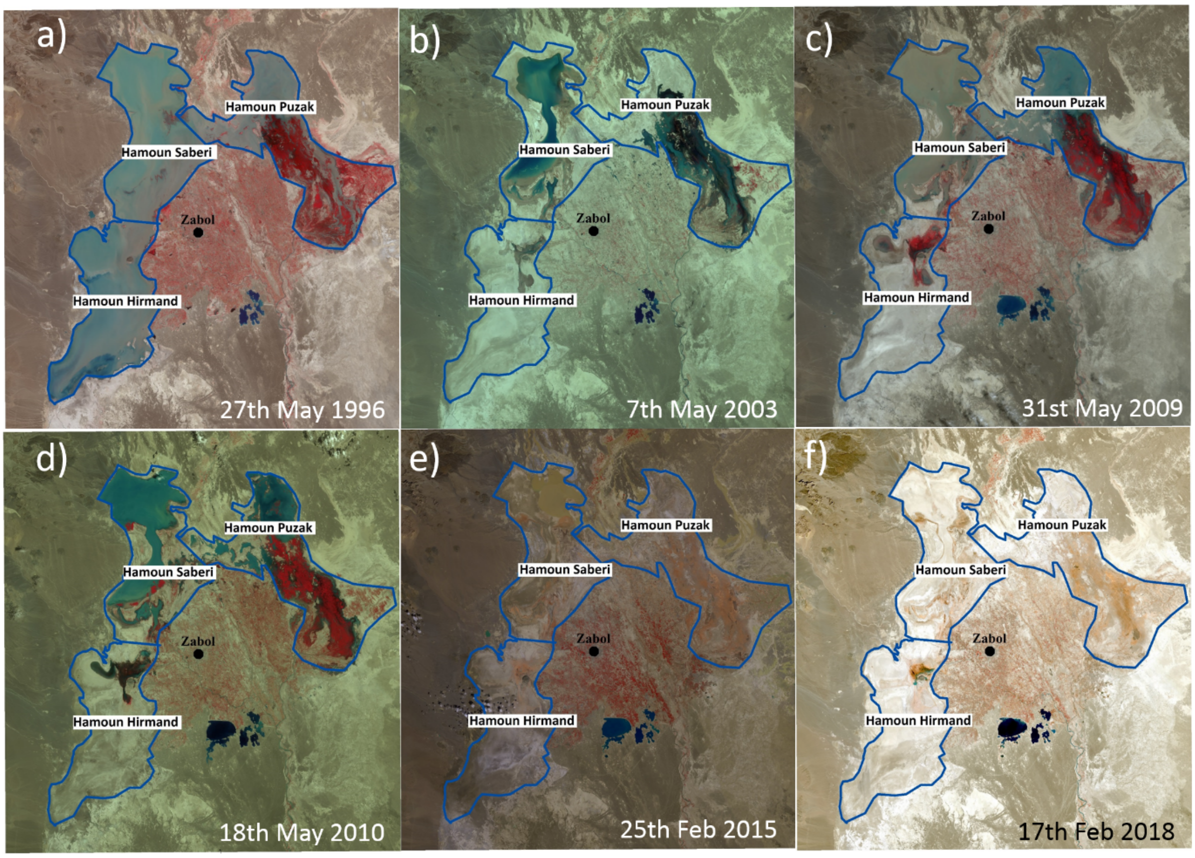


Figure 2.



a)
Hamoun Puzak
Hamoun Saberi
Zabol
Hamoun Hirmand
27th May 1996
b)
Hamoun Puzak
Hamoun Saberi
Zabol
Hamoun Hirmand
7th May 2003
c)
Hamoun Puzak
Hamoun Saberi
Zabol
Hamoun Hirmand
31st May 2009
d)
Hamoun Puzak
Hamoun Saberi
Zabol
Hamoun Hirmand
18th May 2010
e)
Hamoun Puzak
Hamoun Saberi
Zabol
Hamoun Hirmand
25th Feb 2015
f)
Hamoun Puzak
Hamoun Saberi
Zabol
Hamoun Hirmand
17th Feb 2018

Figure 6.

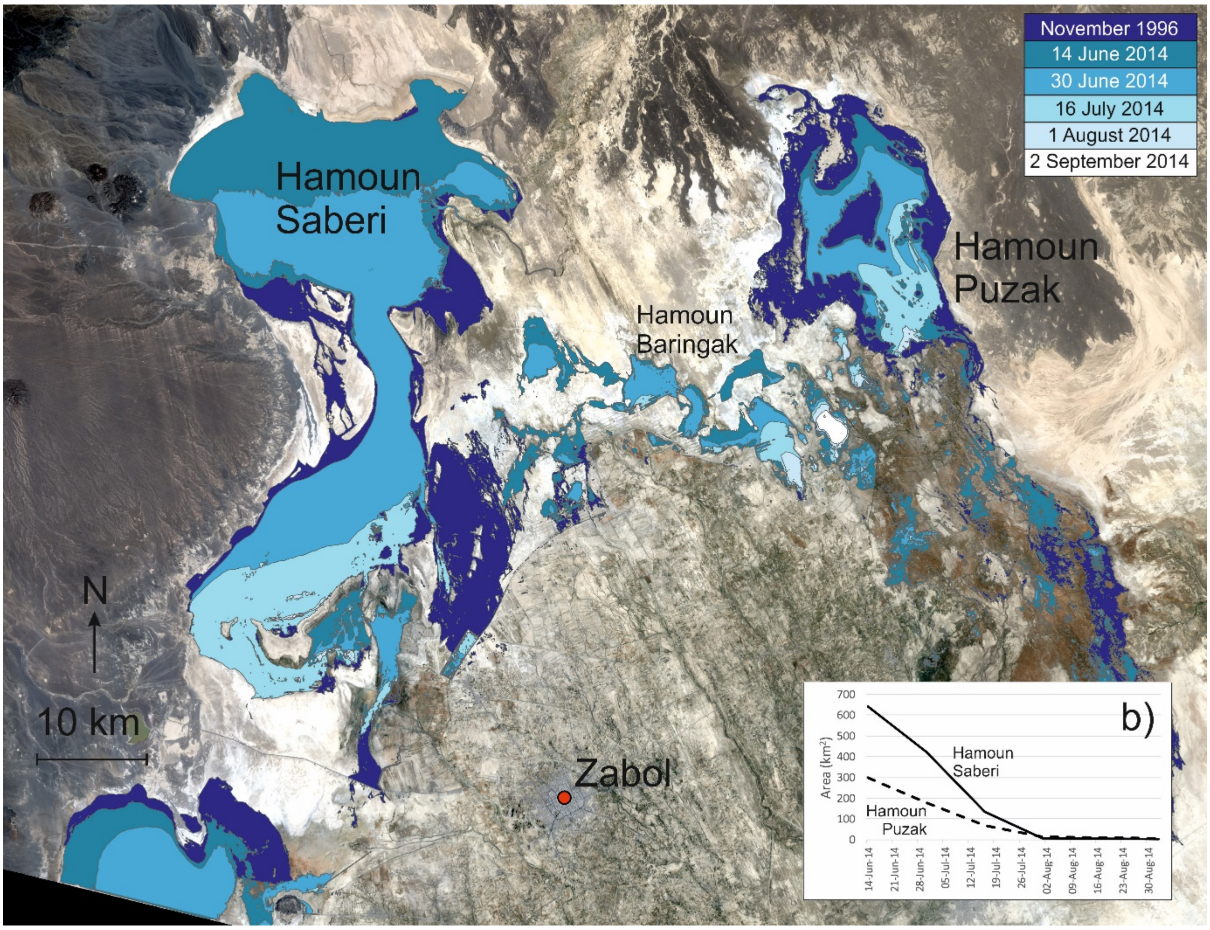

November 1996
14 June 2014
30 June 2014
16 July 2014
1 August 2014
2 September 2014
Hamoun Saberi
Hamoun Puzak
Hamoun Baringak
N
10 km
Zabol
b)
Area (km²)
700
600
500
400
300
200
100
0
Hamoun Saberi
Hamoun Puzak
14-Jun-14
21-Jun-14
28-Jun-14
05-Jul-14
12-Jul-14
19-Jul-14
26-Jul-14
02-Aug-14
09-Aug-14
16-Aug-14
23-Aug-14
30-Aug-14

Figure 7.

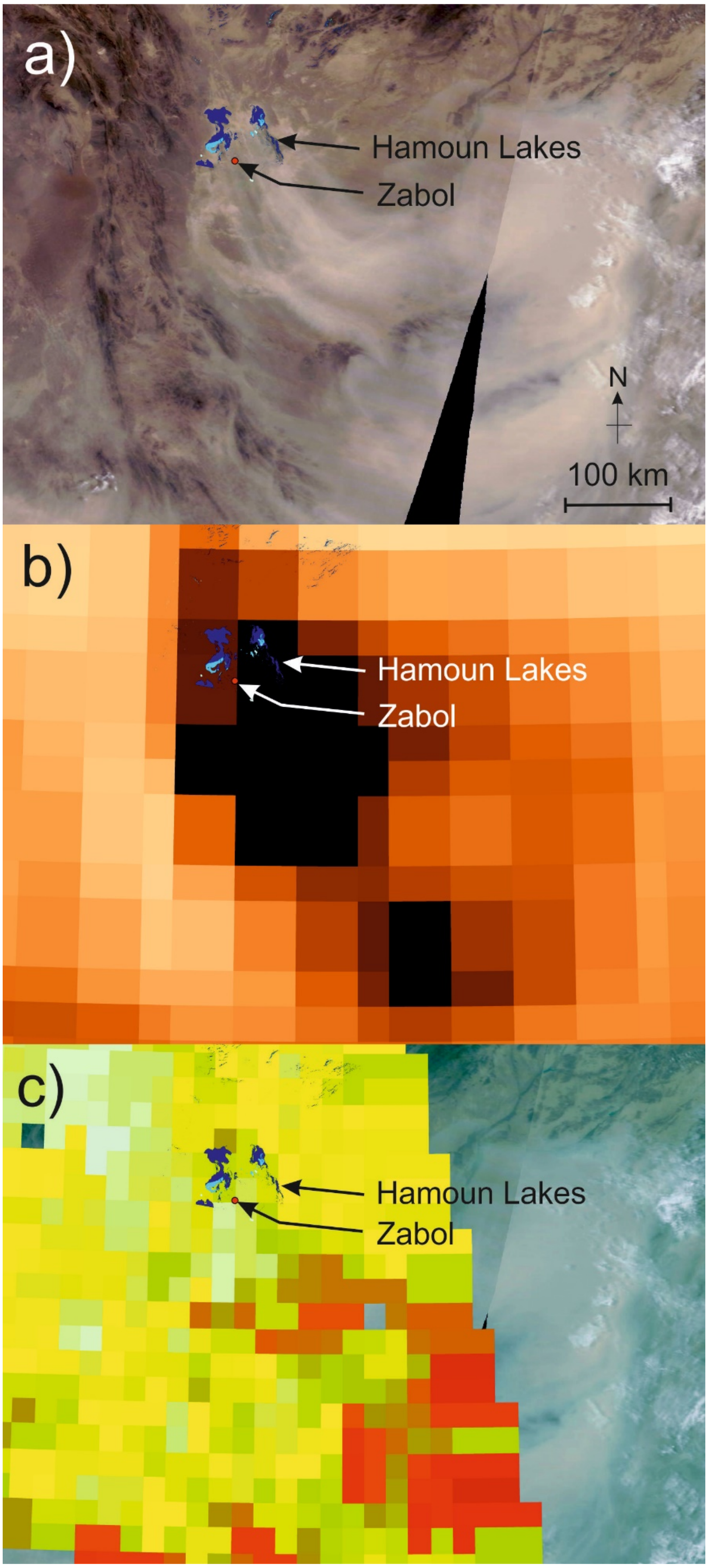

a)
Hamoun Lakes
Zabol
N
100 km
b)
Hamoun Lakes
Zabol
c)
Hamoun Lakes
Zabol

Figure 10.

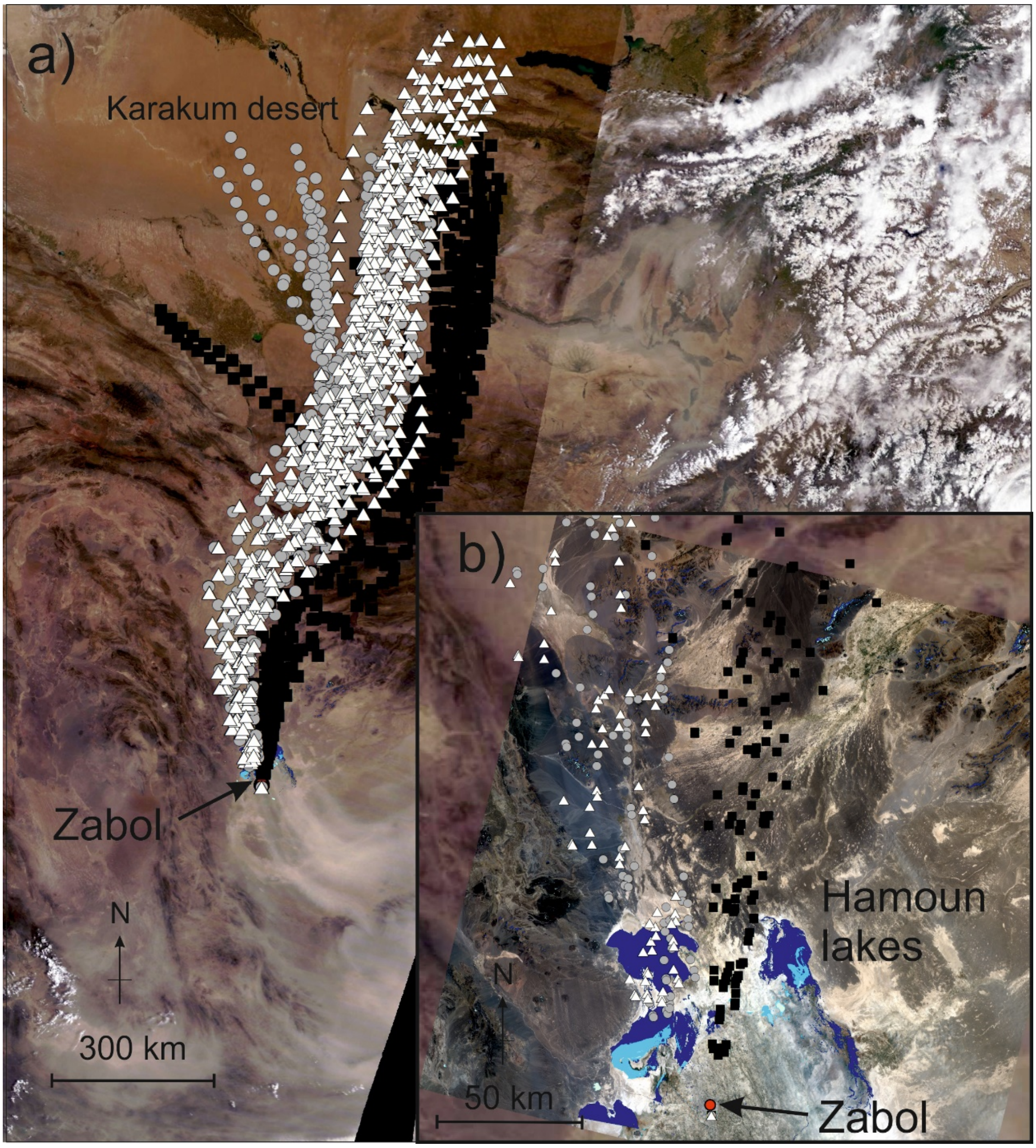


Figure 11.

CRediT Author statement

Reza Dahmardeh Behrooz: Software, Formal Analysis, Investigation, Visualization, Writing – Original Draft Preparation. **Hamid Gholami:** Software, Formal Analysis, Investigation, Visualization , Writing – Original Draft Preparation Supervision, Project Administration. **Matt W. Telfer:** Formal Analysis, Investigation, Visualization , Writing – Review & Editing. **John D. Jansen:** Investigation, Visualization, Writing – Review & Editing. **Abolhassan Fathabadi**: Conceptualization, Supervision, Project Administration